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Spins of Excited States of Tl^{208} from Alpha-Gamma Angular Correlations*†

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Measurements have been made of the angular correlations of the 5.60-MeV α ray occurring in the decay of Bi^{212} to the fourth excited state of Tl^{208} with the subsequent 452-keV γ ray, and of the 5.76-MeV α ray occurring in the decay to the second excited state with the subsequent competing 288- and 328-keV γ rays. The detectors employed were a 2-in.× 2-in. NaI(Tl) crystal for γ rays, and a silicon surface barrier counter for α rays. The results of the angular correlation measurements were

Transition	$W(\theta)$
5.76-MeV α ray—288-keV γ ray	$1-(0.182\pm0.018)P_2(\cos\theta)$
5.76-MeV α ray—328-keV γ ray	$1+(0.537\pm0.042)P_2(\cos\theta)$
5.60-MeV α ray—452-keV γ ray	$1-(0.101\pm0.017)P_2(\cos\theta)$ $+(0.070\pm0.023)P_4(\cos\theta)$

The ratio of the intensity of the 2.88-keV γ ray to the intensity of the 328-keV γ was found to be 2.87 ± 0.22. The multipolarities of the 288- and 328-keV transitions have been confirmed to be predominantly $M1$. A unique spin assignment of 5^+ has been made to the 328-keV second excited state of Tl^{208}. Moreover, consistent assignments of 4^+, 3^+, and 6^+ are made to the third through fifth excited states.

INTRODUCTION

THE levels of nuclides in the region of doubly magic Pb^{208} have received a considerable amount of theoretical attention.[1–6] Pryce has calculated the expected spins of excited states of nuclides differing from Pb^{208} by two nucleons, or holes, in terms of the one-particle model.[7] He assumed the doubly closed shell has a spherically symmetric field which gives rise to one-particle states; a strong spin-orbit interaction component is included in the field. The interaction between the 2 odd nucleons was treated as a perturbation weak when compared with the spin-orbit interaction. The strengths of the singlet and triplet interactions were included as parameters in the perturbation. The ground state and first excited level of Tl^{208} were predicted to form a doublet with spins 5^+ and 4^+, respectively. Previous experiments appear to have fulfilled the prediction. The next four excited states were predicted to form a quartet with spins 5^+, 4^+, 6^+, and 3^+, respectively. Experimental information has been insufficient to test this prediction.

In the experiment to be described the angular correlations of α rays from Bi^{212} to the second and fourth excited states of Tl^{208} with the subsequent γ rays have been measured. This experiment differs from earlier $\alpha-\gamma$ angular correlation measurements in that a solid-state counter was used for the detection of α particles. The energy resolution achieved is not comparable with that of magnetic devices, but the advantages of simplicity of equipment and larger attainable solid angles are gained. Signal-rise times may be achieved that compare favorably with those attained by plastic scintillators and the best photomultipliers presently available.

BACKGROUND

This experiment consists of the measurement of the angular correlations of the α rays from Bi^{212} to the second and fourth excited states of Tl^{208}, with the succeeding γ rays. The interpretation of the results depends to a large extent upon existing information with regard to the spins and parities of the initial state of Bi^{212}, and the final states, the ground state, and first excited level of Tl^{208}. This information and previous experimental work bearing directly on the spins and parities of the higher excited states of Tl^{208} are discussed below. Figure 1 illustrates the principal decay features of ThB(Pb^{212}). Average values of log*ft* for the β-ray transitions, as found by various experimenters, are also shown.[8–17]

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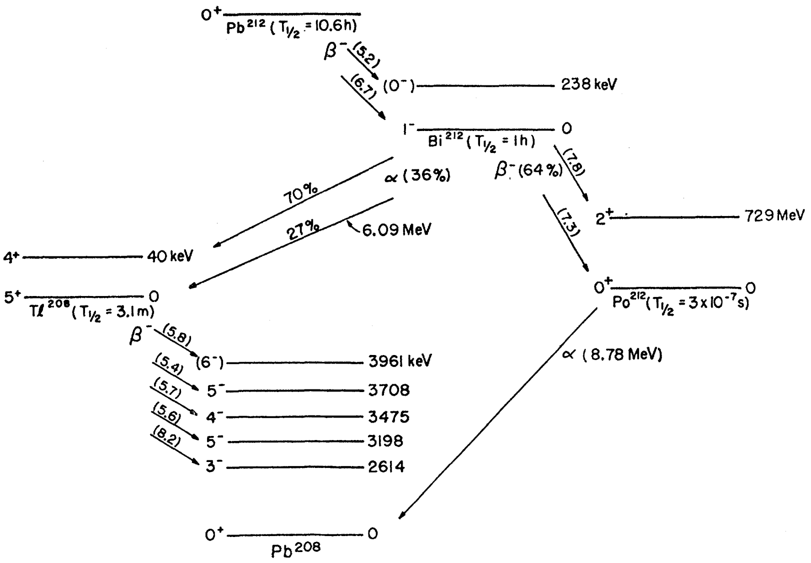

FIG. 1. Principal decay schemes of ThB active deposit. Values of log ft for β decays are shown in parentheses.

Bi^{212}

The spin and parity of Bi^{212} are tabulated as 1^-[18,19]; the following discussion outlines the justification for this assignment.

It is first assumed that both Pb^{212} and Po^{212} have 0^+ ground-state spins. It is then observed that the 238-keV transition in Bi^{212} has been identified as $M1$, by measurements of internal conversion coefficients.[9,20–24] The 729-keV level in Po^{212} has been assigned a spin 2^+, as the radiation from the level has been found to be $E2$ on the basis of measurements of conversion coefficients.[13,14,21] This assignment is supported by the requirement that[25] $\mathrm{Po}^{212(1)}$ has spin 1^-, 2^+, 3^-, $\cdots$ due[16] to the existence of long-range α particles from this level to $\mathrm{Pb}^{208(0)}$, and the calculated relative intensities of γ- and α-ray emissions which indicate that the spin is $2+$.[26] It follows from the above, and from Fig. 1, that the β-ray transitions in the $\mathrm{Pb}^{212}-\mathrm{Po}^{212}$ decay chain must all be first forbidden, or all allowed. The former conclusion is more likely due to the values of logft for 3 of the transitions, which would limit the possible spin assignments for Bi^{212} to 0^-, 1^-, or 2^-. The low value for logft then indicated for the transition to $\mathrm{Bi}^{212(1)}$ agrees with findings elsewhere in the region[16,27] $A=208$, $\Delta I=0$, ± 1; these low values have been justified theoretically by King.[11] In fact, a $0^+ \rightarrow 0^-$ transition to $\mathrm{Bi}^{212(1)}$ is suggested by the value of logft, which would then require Bi^{212} to have spin 1^-, due to the $M1$ radiation to the ground state. The unlikely alternate conclusion, all the β-ray transitions are allowed, would require the spin of Bi^{212} to be 1^+. As will be seen, a positive-parity ground state is unlikely, from shell-model considerations.

Spin 0 is unlikely for Bi^{212} because the $M1$ nature of the 40-keV transition in Tl^{208} would require $\mathrm{Tl}^{208(0)}$ and $^{(1)}$ to have the same spin and parity.[20,28–31] If this were so, it would be expected that the intensity of the α decay to the ground state would be twice as strong as that to the 40-keV level.[9] Spin 2^- assignment to Bi^{212} was excluded by Burde and Rozner,[12] from their determination of the shape of the most energetic portion of the β-ray spectrum of Bi^{212}; 0^- was similarly excluded from examination of the β-ray transition to $\mathrm{Po}^{212(1)}$. Horton has measured the asymmetry of the angular correlation of the 6.05-MeV α rays to $\mathrm{Tl}^{208(1)}$ with the subsequent 40-keV γ ray.[32] A spin of 1 for Bi^{212} was the only value consistent with his results and the β decay of Tl^{208}. These results confirmed and refined the experiment performed by Weale.[30]

Shell-model considerations favor the 1^- assignment to Bi^{212}. A portion of the sequence of single particle

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levels, as proposed by Bergström and Andersson,[5] is shown in Fig. 2. A positive parity ground state for Bi^{212} would be very difficult to understand. Krisyouk *et al.*[8] considered the ground state of the 83rd proton most likely $h_{9/2}$, as in Bi^{209},[33,34] and the odd neutrons $g_{9/2}$ or $i_{11/2}$. A ground-state configuration $[(h_{9/2})_p(i_{11/2})_n{}^3]_{1^-}$ for Bi^{212} was favored by the above authors. Bi^{214} is noted to have a 1^- ground state[35] and Lee-Whiting has shown that Bi^{210} has a ground-state configuration $[(h_{9/2})_p(i_{11/2})_n]_{1^-}$.[33]

The Ground State and First Excited Level of Tl^{208}

The spins of the ground state and first excited level of Tl^{208} are tabulated as 5^+ and 4^+, respectively.[18,19] The assignment of 4^+ by Demichelis and Ricci to the ground state on the basis of a β-γ angular correlation is not in agreement with generally accepted conclusions.[17]

The work of Elliott, Graham, Walker, and Wolfson[36,37] in measuring internal conversion coefficients and γ-γ angular correlations established the spins of the first four excited states of Pb^{208}. The assignments have been confirmed by Wood and Jastram, who measured γ-γ angular and polarization-direction correlations.[38] Additional confirmation has been supplied by Emery and Kane[13] and Krisiuk *et al.*,[15,39] with further determinations of internal conversion coefficients. The latter group has proposed a 6^- assignment to the 3.961-MeV level of Pb^{208} on the basis of conversion of transitions from this level. This assignment is supported by Simons *et al.* who have performed a γ-γ angular correlation measurement on a cascade from the 3.961 level.[40] The most consistent interpretation of the values of log*ft* listed in Fig. 1 for the β-ray transitions to the first four excited states of Pb^{208} is that all transitions are first forbidden and that the spin of Tl^{208} is 4^+ or 5^+. Acceptance of the 6^- assignment to the 3.961-MeV level of Pb^{208} favors the 5^+ assignment to Tl^{208}. It is unlikely that Tl^{208} has spin 3^+ due to the failure to observe a β-ray transition to the ground state of Pb^{208} and to the value of log*ft* for the transition to the 3^- level.

Horton,[32] mentioned previously, assigned spins of 5^+ and 4^+, respectively, to Tl$^{208(0)}$ and $^{(1)}$ on the basis of his experiment. Assignments of 4^+ and 3^+, respectively, were consistent only if his data suffered from large statistical deviations. Siekman and de Waard[41] have measured the mean lifetime of the 40-keV level of Tl^{208}, and have applied corrections[42] to the same measurement performed by Burde and Cohen.[43] The results, $\tau_\gamma = 1.2 \pm 0.5 \times 10^{-10}$ sec and $1.8 \pm 0.9 \times 10^{-10}$ sec, respectively, are in fair agreement with the calculations of de-Shalit,[44] assuming a $(s_{1/2})_p(g_{9/2})_n$ configuration for the first two states of Tl^{208}. His results are listed in Table I. If the first 2 states do not belong to the same configuration, it is difficult to explain the short lifetime. Of the possibilities listed in Table I, only the first 2 provide reasonable agreement with lifetime measurements, and the second of these is in

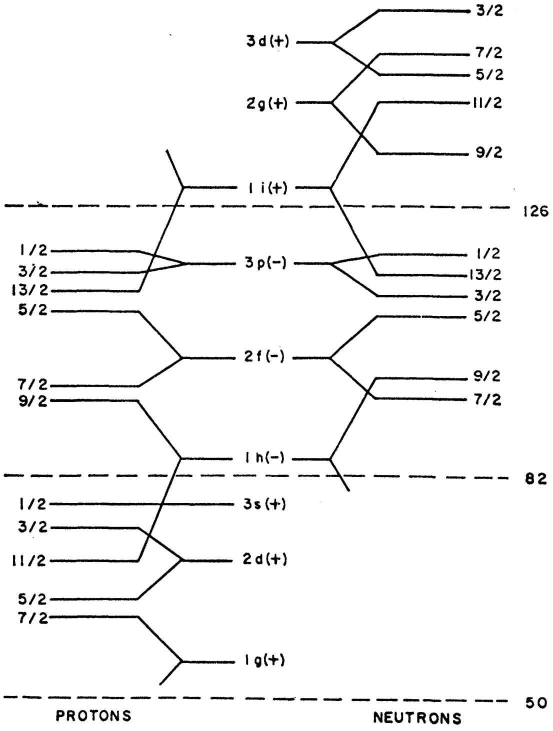

FIG. 2. Single particle levels.

TABLE I. Calculated lifetimes for 40-keV γ-ray transition for four configurations of first two states of Tl^{208}.

Configuration	Spin and parity of Tl$^{208(1)}$	Spin and parity of Tl$^{208(0)}$	Mean lifetime in 10^{-10} sec
$(s_{1/2})_p(g_{9/2})_n$	4^+	5^+	1.8
$(s_{1/2})_p(i_{11/2})_n$	6^+	5^+	2.5
$(d_{3/2})_p^{-1}(g_{9/2})_n$	4^+	5^+	80
$(d_{3/2})_p^{-1}(i_{11/2})_n$	6^+	5^+	25

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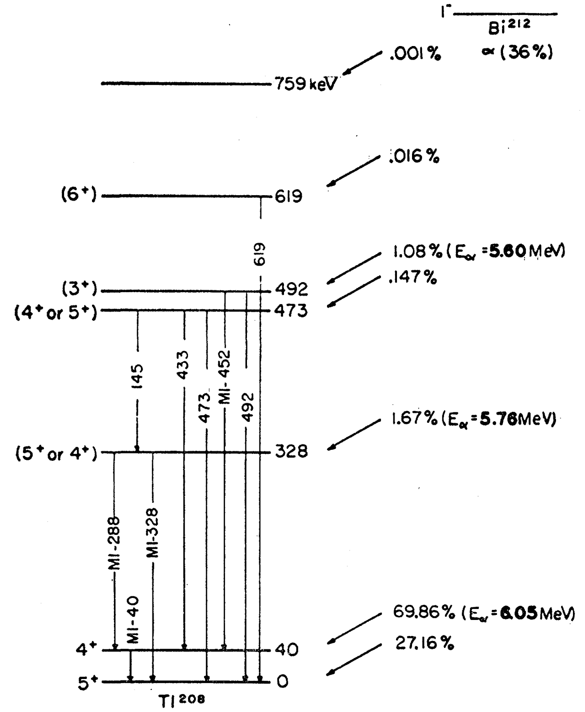

FIG. 3. Excitation of levels of Tl^{208} by α-decay of Bi^{212}.

disagreement with Horton's α-γ angular correlation measurement with regard to the spin assignments.

The shell model predicts Tl^{208} to have positive parity which supports the assumption that the β-ray transitions to Pb^{208} are first forbidden. The odd neutron in Pb^{209} is expected to have a spin of $g_{9/2}$.[5,7,34,45–48] The proton hole of Tl^{207} is predicted to have a ground state of $s_{1/2}$ and first excited state of $d_{3/2}$.[5,7,46] These assignments have been accepted.[34,49] Pryce has predicted that the ground state and 40-keV level of Tl^{208} form a $(s_{1/2})_p(g_{9/2})_n$ doublet with spins 5^+ and 4^+, respectively,[7] in agreement with the modified Nordheim's "weak" rule.[50,51]

The Higher Excited States of Tl^{208}

Additional details of the level scheme of Tl^{208} are provided by Fig. 3. The energy levels are chosen to agree with the α-ray disintegration energies found by Briggs,[52] except for the level at 759 keV which is due to Walen and Bastin-Scoffier.[53] The intensities of the α-ray transitions were determined by Rytz.[54] The following discussion presents the bases for the probable spin assignments shown for the second through fifth excited states.

Conversion electron intensities in coincidence with α decay to the higher excited states of Tl^{208} were measured by Nielsen.[28] With the use of known α-ray intensities, the 288-, 328-, and 452-keV transitions were shown to be predominantly $M1$ on the basis of K/L ratios and K-conversion coefficients. The 433- and 473-keV transitions were also deduced to be $M1$ on the basis of their observed K-conversion electron intensities. The tentative assignments of 4^+ or 5^+ to both the 328- and 473-keV levels were made accordingly. The assignment of 3^+ to the 492-keV level was made on the basis of the failure to observe a K-conversion line corresponding to a 492-keV γ ray, and the $M1$ assignment to the 452-keV transition. No K-conversion line was observed for a transition from the 492 level to the 328-keV level. Sergeev *et al.* have extended the work of Nielsen in observing conversion electron spectra.[29] As a result of their experiments,[55,56] they agreed with the previous conclusions; in addition weak K-conversion lines were found for a 492-keV transition to the ground state of Tl^{208}, and for a 145-keV transition to the 328-keV level. The latter transition was deduced to be $M1$.[57] The observation of a 619-keV transition and the absence of an observable 579-keV transition caused these experimenters to favor the assignment of 6^+ to the 619-keV level. Emery and Kane have measured the intensities of the 288-, 328-, and 452-keV γ rays by external photoelectric conversion.[13,58] Using the conversion electron intensities of Sergeev *et al.*, the $M1$ natures of these transitions were verified.[59] Bertolini *et al.* have measured γ-ray intensities coincident with α decay to the higher excited states of Tl^{208}, using a sodium iodide detector to observe γ rays.[60] Conversion coefficients were calculated. Their conclusions were the same as those previously made regarding the spins of $Tl^{208(2)}$ and $^{(4)}$.

Pryce has predicted on the basis of the shell model that the second to the fifth excited states of Tl^{208} form a

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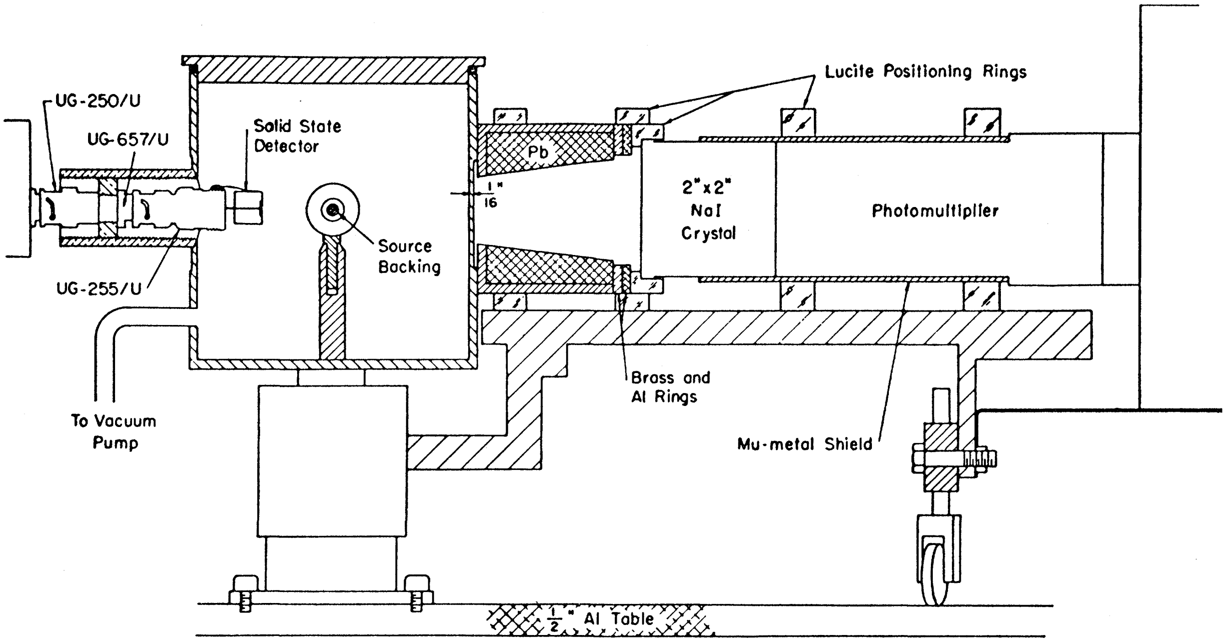

FIG. 4. Mechanical apparatus for $\alpha-\gamma$ angular correlation measurement.

$(d_{3/2})_p^{-1}(g_{9/2})_n$ quartet, with an expected sequence of spin assignments[7] of 5^+, 4^+, 6^+ and 3^+.

Korolev *et al.*[61] have measured the α-γ angular correlations of the α rays proceeding to $Tl^{208(2)}$ and $Tl^{208(4)}$ with the coincident γ rays. In the interpretation of the results, however, spins of 1^+ or 2^+ were considered for Bi^{212}. In addition, no effort was made to distinguish between the 288- and 328-keV γ rays. Their experimental results will be discussed below.

Summary

As a result of the findings mentioned in the previous discussion, the following assumptions are made: Bi^{212} has spin 1^-, and $Tl^{208(0)}$ and $^{(1)}$ have spins 5^+ and 4^+, respectively; and the 288-, and 328-, and 452-keV transitions in Tl^{208} are predominantly $M1$.

SOURCE

A 1.5-mCi dry source of radiothorium (RdTh) in barium stearate, prepared as described by Wahl,[62] was made available by Dr. H. W. Kraner.[63] This preparation has been shown to emanate with an efficiency of 100% at room temperature.[64] ThB (Pb^{212}) sources were prepared by passing dry nitrogen through a chamber containing the RdTh and depositing the thoron decay products electrostatically on nickel foil. Circular foils 2 mils in thickness and $\frac{3}{16}$ in. in diameter were used as backings for the ThB sources in the experiment. No effort was made to prevent the escape of source material from the backing during runs. It was determined that after a source had been in use for 10 h, about 0.05% of the α-ray counting rate was due to source material which had escaped from the foil backing. About 70% of these counts were due to activity deposited on the solid-state detector. This would be expected to enhance the coincidence counting rate with the γ-ray detector at the 180° position, but the correction is negligible. Source strengths were determined from the counting rate in the 8.78-MeV α-ray peak and the known solid angle of the solid-state detector. This gave good agreement with the activity as determined by the 2.62-MeV γ-ray counting rate measured with an 8-in. ×4-in. NaI crystal, and comparison with a calibrated aged thorium standard. It was shown that the source material was approximately evenly distributed over the surface of the foil, to validate corrections made for the finite size of the source.

APPARATUS

Figure 4 shows the mechanical apparatus with which angular correlation measurements were made. The angular position of the vacuum chamber is fixed reproducibly with respect to the table by a tapered pin; this holds the solid-state detector at 0°. One variation of mounting the solid-state counter, used for the detection of α rays, on a standard connector is shown.

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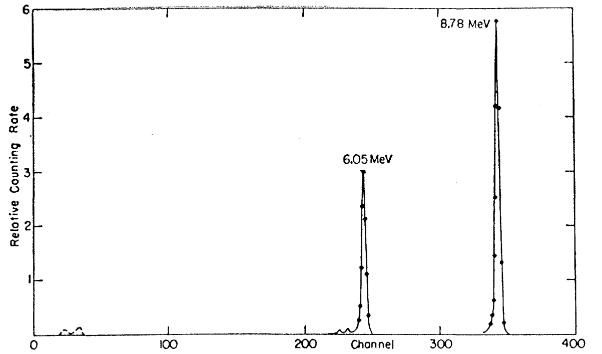

FIG. 5. α-ray spectrum of ThB active deposit.

The detector is $1\frac{1}{16}$ in. from the center of the source; this corresponds to $\Omega_\alpha(\mathrm{sr})/4\pi=0.00246$. The ThB ($\mathrm{Pb}^{212}$) source foil is placed on a Mylar backing. This in turn is attached to a $\frac{1}{32}$-in-thick aluminum ring which forms the top of the removable source holder. The carriage supporting the sodium iodide detector, collimator, photomultiplier, and preamplifier pivots around the vertical axis of the vacuum chamber. The face of the sodium iodide detector is about $4\frac{1}{2}$-in. from the source; the physical solid angle intercepted by the detector corresponds to $\Omega_\gamma(\mathrm{sr})/4\pi=0.0069$. The evacuated brass chamber is $4\frac{1}{2}$ in. high and has a 4-in. diameter.

A cathode follower and preamplifier arranged in series provide fast and slow signals from the solid-state counter. The fast signal from the cathode follower, typically about 6 mV, with a rise time varying from 4 to 15 nsec, depending on the detector, is amplified by 3 wide band amplifiers. The amplified signal cuts off a limiter which provides the pulse for the fast coincidence. The 5-nsec rise time output of the limiter is clipped to 20 nsec. The solid-state detector preamplifier provides a high-resolution signal to an amplifier and differential discriminator. Figure 5 illustrates the α-ray spectrum of a source of ThB (Pb^{212}) active deposit; Fig. 6 shows the peaks due to Bi^{212} α decay. A resolution of 100 keV is achieved in this case at a counting rate of 5700 cps. The photomultiplier used is the EMI 9536S. A signal from the anode cuts off a limiter similar to that used in the α-ray signal channel. A signal taken from the sixth dynode is used for spectral analysis of the γ rays by a multichannel analyzer. The outputs of the 2 limiters are fed to a fast coincidence circuit. A resolving time 9×10^{-9} sec was utilized. A slow coincidence, resolving time 1 μsec, is performed with the output of the fast coincidence circuit and the differential discriminator. Coincident events provide a suitable gating signal for the multichannel analyzer.

A Harshaw 2-in. $\times$2-in. NaI(Tl) crystal was used for the detection of γ rays. Figure 7 shows the γ-ray spectrum of the ThB source in equilibrium with its daughters. All solid-state detectors used in this work were of the surface barrier type, manufactured from 5-mm squares of n-type silicon semiconductor material. One Ortec counter was used; the remainder were made available by Dr. H. W. Kraner.[65] The above apparatus is described more completely elsewhere.[66,67]

The shape of the spectrum of accidental coincidences experienced in observing α-γ coincidences in the decay of Bi^{212} to Tl^{208} was determined. A cable length was inserted between the photomultiplier limiter and the fast coincidence circuit to eliminate true coincidences. The accidental coincidence spectrum and the γ-ray spectrum of the source agreed in shape down through the 80-keV x-ray peak, except for a small degradation in resolution of the peaks and a small energy shift upwards. The angular distribution of accidental coincidences was also measured and found to be isotropic, to a statistical accuracy of better than 1%. This check was repeated several times. The rate of singles γ-ray counting at the various γ-ray detector positions was checked frequently to determine that the effective solid angle was the same at all positions; in each case this was verified.

The angular correlation of the 729-keV γ ray to the ground state of Po^{212}, occurring in the β decay of Bi^{212}, with the subsequent α ray to the ground state of Pb^{212}

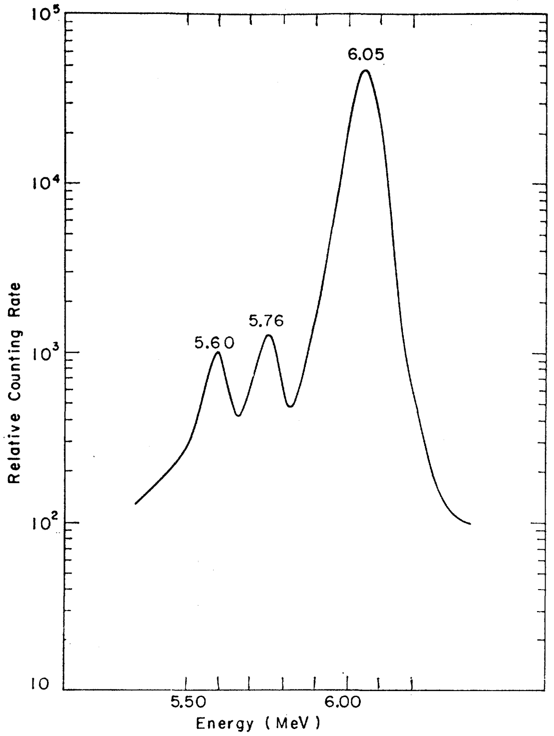

FIG. 6. Spectrum of Bi^{212} α-decay (5700 counts/sec).

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[65] H. W. Kraner, E. E. Hanson, and W. R. Neal, *Radium and Mesothorium Poisoning and Dosimetry and Instrumentation Techniques in Applied Radioactivity*, Annual Report of Radioactivity Center, M.I.T., Contract AT(30-1)-952, May 1, 1961.

[66] W. C. Cobb, Ph.D. thesis, Massachusetts Institute of Technology, 1962 (unpublished).

[67] W. C. Cobb, Nucl. Instr. Methods (to be published).

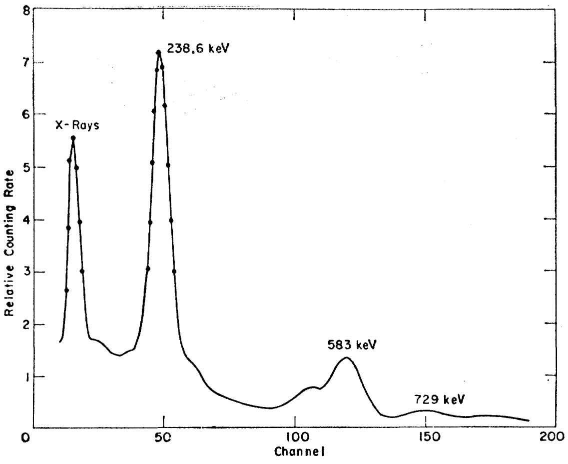

FIG. 7. γ-ray spectrum of ThB active deposit.

was examined for asymmetry; none should be expected since the spin of Po^{212} is zero. The limiter in the α-ray signal channel was modified to achieve a resolving time of 0.4 μsec. Data were taken with the γ-ray detector at 180° and at 90° and 270°. The results at 90° and 270° were equal within expected statistical deviations. The observed asymmetry for all true coincident γ rays giving rise to pulses in the region of the 729-keV photopeak was

$$A=W(180°)/W(90°)-1=-0.006\pm0.027.$$

It was estimated that about 7 to 10% of these counts was due to photons of higher energy, which are also expected to show no asymmetry.

The dependence of the gain of the photomultiplier upon its orientation in the magnetic field of the earth was determined. To better than 1 part in 600, there was no variation of gain at any of the angles to be observed during the experiment.

THEORETICAL ANGULAR CORRELATIONS

The theoretical α-γ angular correlation functions with which results were compared were calculated from the tables of Wapstra, Nijgh, and Van Lieshout.[68]

The theoretical angular correlation

$$W(\theta)=\sum_\nu a_\nu P_\nu(\cos\theta)$$

was corrected for the effect of the finite solid angles subtended by the detectors and the finite extent of the source.[66] Frankel[69] has shown that for circular detectors and a point source, the observed correlation may be written in the form

$$\bar{W}(\theta)=\sum_\nu a_\nu g_\nu P_\nu(\cos\theta).$$

The approximation was made that the corrections for finite source and the detector solid angles occur independently and additively. This is justified by the work of Feingold and Frankel,[70] and Lawson and Frauenfelder.[71]

The observed correlation is therefore of the form

$$\bar{W}(\theta)=\sum_\nu a_\nu[g_\nu P_\nu(\cos\theta)+f_\nu(\theta)],$$

where $f_\nu(\theta)$ is the correction for the finite size of the source. The correction $f_\nu(\theta)$ was found by integration over the source. Correction for the finite solid angles subtended by the detectors was done by the method of Rose,[72] with some modification due to the square shape of the solid-state counter. The values of $[g_\nu P_\nu+f_\nu]$ are attended by errors due to inexact measurement of the dimensions of the apparatus; the effect of these errors was calculated to be negligible.

REORIENTATION

The observed angular correlations may be attenuated by interactions induced by the recoil from α decay, by the magnetic field of the unpaired electron in Tl^{208}, and by precession of the intermediate state nucleus in other fields of unknown magnitude. Horton[32] has performed calculations regarding the effects of recoil for Tl^{208} which are applicable in the present case. His conclusions indicate that the effect of transitions induced by recoil is small. Alder[73] has calculated the effect upon angular correlations of a magnetic hyperfine interaction with the fields of unpaired electrons. The maximum "hard-core" values of attenuation indicate that significant perturbations in the correlations here observed are unlikely. However, as Flamm and Asaro[74] have observed experimentally, it is possible for attenuations to exceed hard-core predictions, probably dependent upon the lifetimes of the intermediate states.

It is difficult to predict the amount of attenuation that exists in this experiment; however, from a phenomenological point of view, the large asymmetry observed in the angular correlation of the 328-keV γ rays indicates that the attenuation cannot be large.

MIXTURE OF α-RAY ORBITAL ANGULAR MOMENTA

In general, it is possible for α rays to be emitted with more than one value of orbital angular momentum. In this experiment the case arises when the spin of the intermediate state is even, and specifically, 4^+. Figures 8 and 9 illustrate the effect of considering admixture of α-ray orbital angular momenta in the cases of interest. δ_α^2 is the ratio of the intensity of $L_\alpha=5$ emission to the intensity of $L_\alpha=3$ emission. It is seen that fortuitously α-ray orbital angular momenta admixture is relatively

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[68] A. H. Wapstra, G. J. Nijgh, and R. Van Lieshout, in *Nuclear Spectroscopy Tables* (North-Holland Publishing Company, Amsterdam, 1958).

[69] S. Frankel, Phys. Rev. **83**, 673 (1951).

[70] A. M. Feingold, and S. Frankel, Phys. Rev. **97**, 1025 (1955).

[71] J. S. Lawson and H. Frauenfelder, Phys. Rev. **91**, 649 (1953).

[72] M. E. Rose, Phys. Rev. **91**, 610 (1953).

[73] K. Alder, Phys. Rev. **83**, 1266 (1951).

[74] E. Flamm and F. Asaro, Phys. Rev. **129**, 290 (1963).

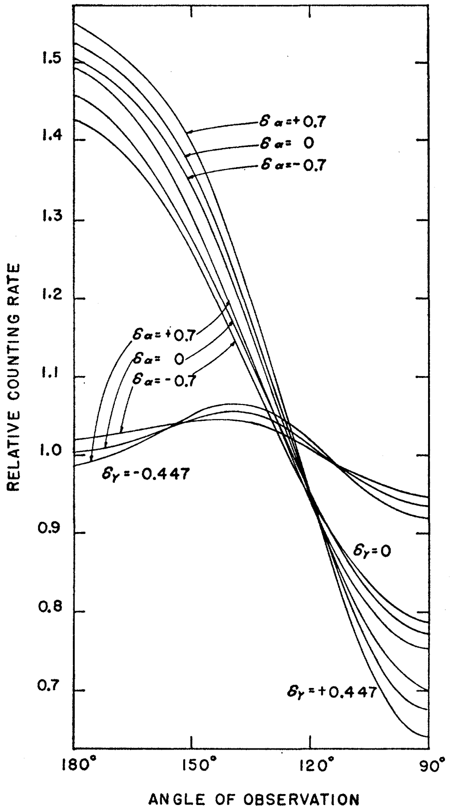

FIG. 8. Effect of α- and γ-ray orbital angular momenta admixture on angular correlation for $\alpha-\gamma$ transition sequence: $1(l=3,5)4(l=1,2)4$.

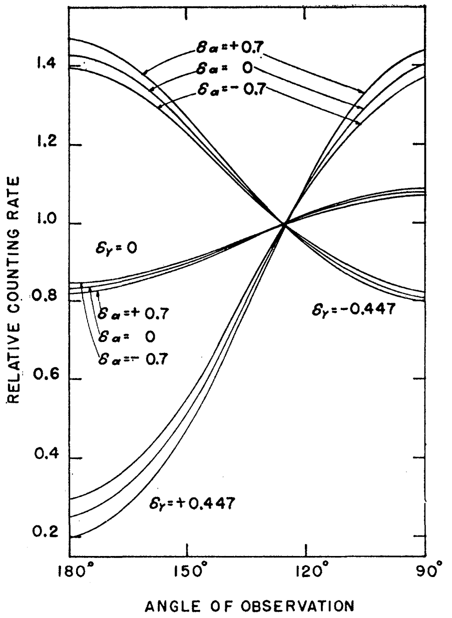

FIG. 9. Effect of α- and γ-ray orbital angular momenta admixture on angular correlation for $\alpha-\gamma$ transition sequence: $1(l=3,5)4(l=1,2)5$.

unimportant. Therefore, in order not to complicate the presentation of data, it was assumed that α rays are emitted with the lowest allowable value of angular momentum.

PROCEDURE

Initial activities of about 60 μCi were used. With these activities the counting rate in the α-ray signal channel was about 6000 counts per second and in the γ-ray signal channel about 20 000 counts per second. Less than 1% of the counting rate in the γ-ray signal channel was due to external background. Runs were not commenced until a source was sufficiently aged so that the ThB was essentially in transient equilibrium with its daughters. The differential discriminator was set to provide a pulse when signals were received from the solid-state detector preamplifier corresponding to the 5.60- (or to the 5.76-) MeV α rays originating from Bi^{212} decay to $\mathrm{Tl}^{208(4)}$ [or $\mathrm{Tl}^{208(2)}$]. The multichannel analyzer recorded pulses from the γ-ray detection system when gated by the slow coincidence circuit.

A typical set of runs was made as follows. The source was oriented with the active side toward the solid-state detector, and the normal to the source at 315°. A 10-min observation was made with the γ-ray detector at each angle proceeding from 90° to 180° in 30° steps, and the observations were then repeated in the reverse direction. After two or three such sequences were completed, the source holder was rotated 90° clockwise, and observations were made from 180° to 270°, recording an approximately equal total number of coincidences. Checks were made on the differential discriminator and the gain setting for the γ-ray spectrum every 40 min. A window width of approximately 300 keV was employed with the differential discriminator. The effect of this large setting, which caused some overlap between the 5.76- and 5.60-MeV α rays, was to require a correction for the observations on the 5.76-

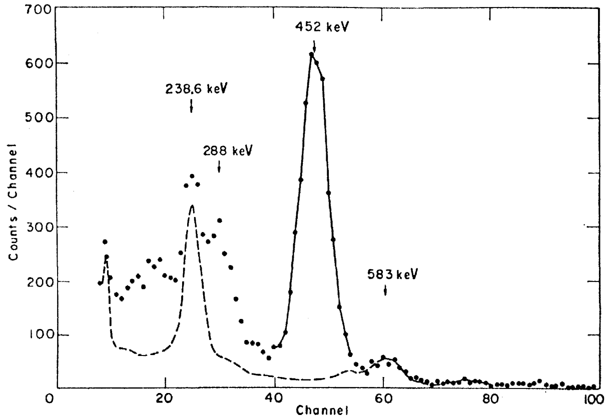

FIG. 10. Observed γ-ray spectrum coincident with 5.60-MeV α ray of Bi^{212}, $\theta=90°$. Correction for accidental coincidences shown.

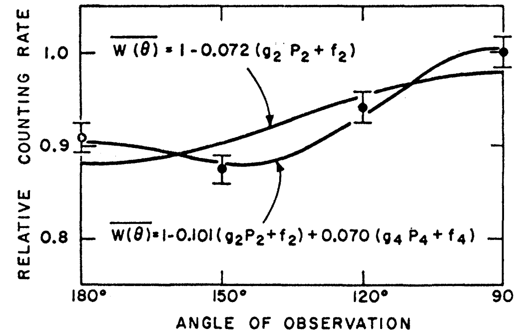

FIG. 11. Angular distribution of 452-keV γ ray coincident with 5.60-MeV α ray from Bi^{212}.

MeV α ray. The positions of the 238.6- and 583.0-keV γ-ray[75] peaks were used to establish the correct gain setting of the multichannel analyzer.

RESULTS

Tl$^{208(4)}$

In observing the γ-ray spectrum coincident with the 5.60-MeV α ray, an average of about 2000 counts was recorded, at each angle of observation, in the 452-keV total energy peak. The data from 90° to 180° agreed, within expected statistical deviations, with the data from 270° to 180°; these data were, therefore, combined. The correction for accidental coincidences was based on counts observed at energies higher than the 452-keV total energy peak and amounted to $7\frac{1}{2}\%$ of the counts observed in the 452-keV total energy peak. The width of the differential discriminator setting allowed some 288- and 328-keV γ rays to appear in the coincident spectrum. The γ-ray spectrum recorded in coincidence with the 5.60-MeV α ray at $\theta = 90°$ (combined with the data at $\theta = 270°$) is shown in Fig. 10.

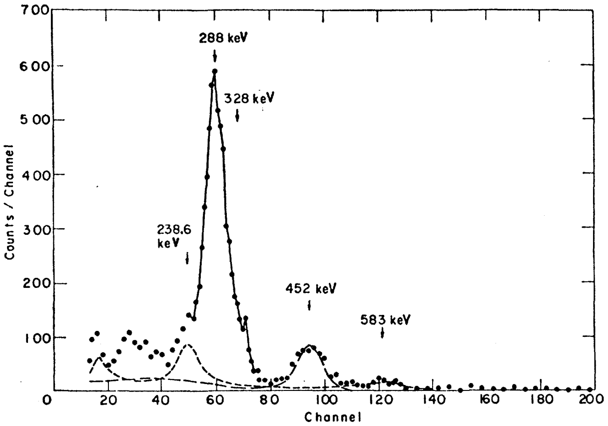

FIG. 12. Observed γ-ray spectrum coincident with 5.76-MeV α ray of Bi^{212}, $\theta = 90°$. Corrections for accidental coincidences and 452-keV γ rays shown.

The angular distribution of the corrected counts in the 452-keV total energy peak was calculated by the least-squares method. Best fits were made to the experimental observations with a function containing P_0 and P_2 terms only, and with one containing a P_4 term as well. The results of the calculations for the angular distribution of the 452-keV γ ray were

$$(1)\quad W(\theta) = 1 - (0.072 \pm 0.015) P_2(\cos\theta) \text{ and}$$

$$(2)\quad W(\theta) = 1 - (0.101 \pm 0.017) P_2(\cos\theta) + (0.070 \pm 0.023) P_4(\cos\theta).$$

The deviations represent one statistical standard deviation. Figure 11 shows the two least-squares fits to the observed data. The functions used in making the fits included the geometric corrections to the angular correlations. The statistical goodness of fit is better with a P_4 term included in the calculated distribution.

The finding of Korolev *et al.*[61] for the angular correlation of the 5.60-MeV α ray with the 452-keV γ ray was $W(\theta) = 1 - (0.083 \pm 0.016) P_2(\cos\theta)$. They found no P_4 term to be present in their observations.

Tl$^{208(2)}$

In observing the γ-ray spectrum coincident with the 5.76-MeV α ray, a little less than 3000 counts were recorded at each angle, in the 288–328-keV total energy peaks. The 90°–180° and 270–180° data were combined as on the Tl$^{208(4)}$ runs. Correction for accidental coincidences was also made in the same way and amounted to 5% of the total counts observed in the 288–328-keV total energy peaks. Correction was also made for counts due to the 452-keV γ rays in the coincident spectrum. This correction amounted to $3\frac{1}{2}\%$ of the counts in the 288–328-keV total energy peak. The γ-ray spectra recorded in coincidence with the 5.76-MeV α ray at 2 angles are shown in Figs. 12 and 13.

The 288–328-keV total energy peak recorded at each angle was analyzed to determine the contribution from each component. This was done by the method of least

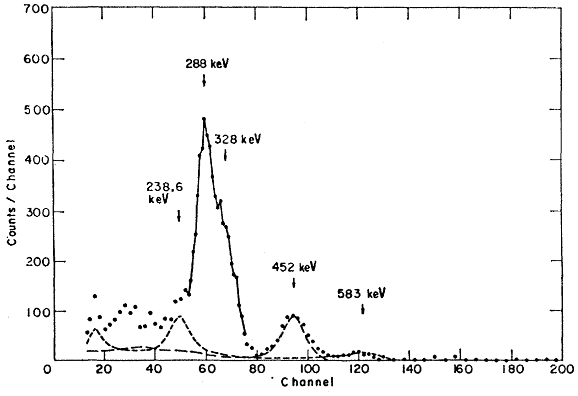

FIG. 13. Observed γ-ray spectrum coincident with 5.76-MeV α ray of Bi^{212}, $\theta = 180°$. Corrections for accidental coincidences and 452-keV γ rays shown.

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[75] D. E. Muller, H. C. Hoyt, D. J. Klein, and J. W. M. DuMond, Phys. Rev. 88, 775 (1952).

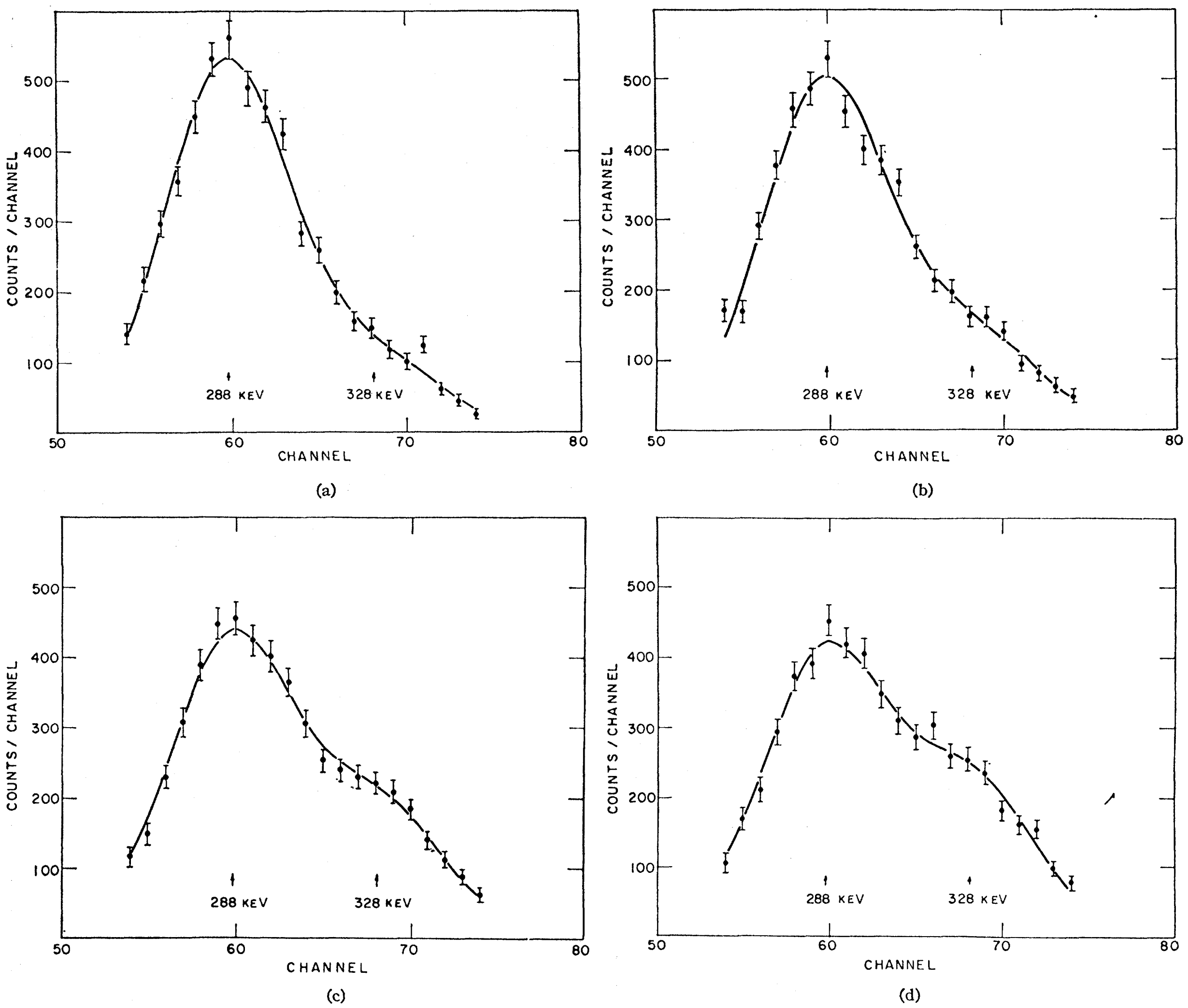

FIG. 14(a) Least-squares fit to 288–328 keV total energy peak, $\theta=90°$. (b) Least-squares fit to 288–328 keV total energy peak, $\theta=120°$. (c) Least-squares fit to 288–328 keV total energy peak, $\theta=150°$. (d) Least-squares fit to 288–328 keV total energy peak, $\theta=180°$.

squares as described by Rose.[66,72] A Gaussian shape was assumed for each component of the peak, except that the tails were modified in accordance with observations on a Cr^{51} ($E_\gamma=323$ keV) source. Assumptions were made regarding the resolutions and center channels of each component. Variations from these assumptions were considered. The least-squares fits to the corrected total energy peaks are shown in Figs. 14(a), (b), (c), and (d).

The calculated angular distributions of both the 288- and 328-keV γ rays showed a very good fit with only P_0 and P_2 terms included. The results of the calculations were for the 288-keV γ ray:

$$W(\theta)=1-(0.182\pm0.018)P_2(\cos\theta);$$

for the 328-keV γ ray:

$$W(\theta)=1+(0.537\pm0.042)P_2(\cos\theta).$$

The standard deviations reported above include, for the 288- and 328-keV γ-ray distributions, respectively, statistical standard deviations of $\pm0.016P_2$ and $\pm0.034P_2$, and deviations based on uncertainties in the correct resolutions and center channels of $\pm0.008P_2$ and $\pm0.025P_2$. Figures 15 and 16 show the least-squares fits to the observed angular distributions.

Korolev *et al.*[61] have also measured the angular correlation of the 5.76-MeV α ray with the succeeding γ rays. However, no attempt was made to resolve the 288- and 328-keV γ rays. Their finding for the combined angular distribution of the 288- and 328-keV γ rays was

$$W(\theta)=1-(0.063\pm0.017)P_2(\cos\theta)+(0.105\pm0.021)P_4(\cos\theta).$$

A value of 2.87±0.22 was obtained for the ratio of the intensity of the 288-keV γ ray to the intensity of the 328-keV γ ray. The deviation reported includes a statistical standard deviation of ±0.07 and an estimated ±0.21 based on the uncertainty in resolutions and center channels used in the analysis of the 288–328-keV γ-ray total energy peaks. A correction was made for the differences in photofraction and transmission through the chamber walls. The finding of Emery[58] for this ratio was 2.2, accurate to 30%, and a value of 2.59±0.24 is given by Bertolini *et al.*[60]

INTERPRETATION OF RESULTS

$Tl^{208(2)}$

Bi^{212} has been assumed to have spin 1^- and $Tl^{208(0)}$ and $^{(1)}$ to have spins 5^+ and 4^+, respectively. $Tl^{208(2)}$ has been presumed to have spin 5^+ or 4^+ due to the predominantely $M1$ nature of the 288- and 328-keV γ-ray transitions.

The theoretical angular correlation for a transition of the type 1 (3) 4 (1,2) 5 is a function of the mixing ratio δ_γ where δ_γ^2 is the ratio of the intensity of $E2$ emission to the intensity of $M1$ emission. Based on measured values of conversion coefficients,[13,28,29,66] a reasonable upper limit to multipole mixing for the 288- and 328-keV γ-ray transitions appeared to be given by $\delta_\gamma^2=0.20$.

The angular correlations of the competing 288- and 328-keV γ rays with the 5.76-MeV α ray to the second excited state of Tl^{208} were found to be

$$288\text{-keV } \gamma \text{ ray: } W(\theta)=1-(0.182\pm0.018)P_2(\cos\theta);$$

$$328\text{-keV } \gamma \text{ ray: } W(\theta)=1+(0.537\pm0.042)P_2(\cos\theta).$$

Figures 17 and 18 illustrate the best fits that can be made, by varying δ_γ, to the observed data with a 4^+ assignment to $Tl^{208(2)}$. The geometric corrections are included in the calculated distributions. These correspond, for the 288-keV γ ray, to

$$W(\theta)=1-0.113P_2(\cos\theta)-0.142P_4(\cos\theta)$$

with

$$\delta_\gamma=-0.70$$

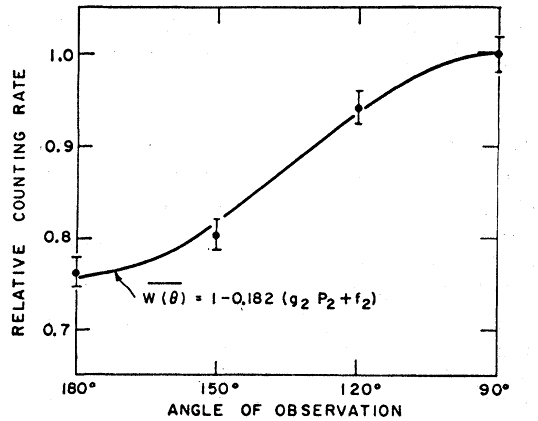

FIG. 15. Angular distribution of 288-keV γ ray coincident with 5.76-MeV α ray from Bi^{212}.

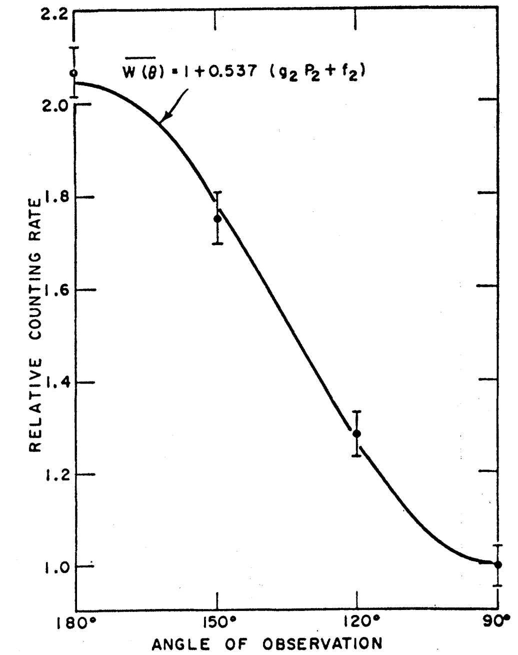

FIG. 16. Angular distribution of 328-keV γ ray coincident with 5.76-MeV α ray from Bi^{212}.

and for the 328-keV γ ray, to

$$W(\theta)=1+0.517P_2(\cos\theta)+0.050P_4(\cos\theta)$$

with

$$\delta_\gamma=-0.65.$$

Best fits were also made choosing $\delta_\gamma=-0.447$, $\delta_\gamma^2=0.20$. (The fits became progressively worse as δ_γ^2 was decreased.) These are also shown in Figs. 17 and 18. These calculated angular correlations correspond, for the 288-keV γ ray, to

$$W(\theta)=1+0.075P_2(\cos\theta)-0.073P_4(\cos\theta)$$

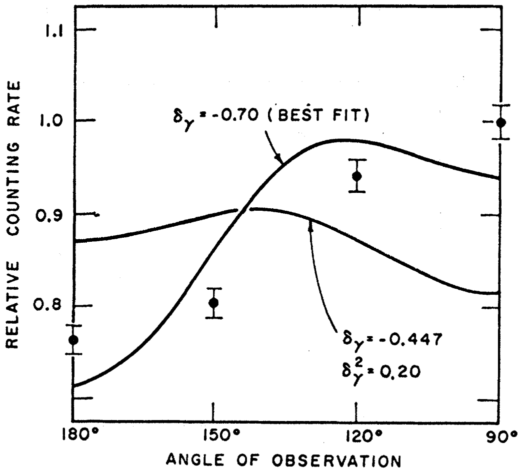

FIG. 17. Comparison of observed angular distribution of 288-keV γ ray to distributions based on 4^+ spin assignment to second excited state of Tl^{208}.

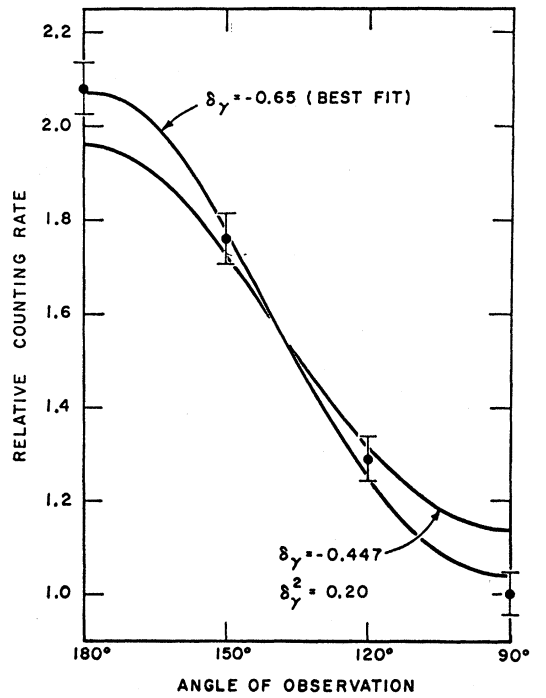

FIG. 18. Comparison of observed angular distribution of 328-keV γ ray to distributions based on 4^+ spin assignment to second excited state of Tl^{208}.

and for the 328-keV γ ray, to

$$W(\theta)=1+0.401P_2(\cos\theta)+0.028P_4(\cos\theta).$$

Best fits to the observed data were also made by assuming a 5^+ assignment to $Tl^{208(2)}$ leading to a calculated correlation, for the 288-keV γ ray,

$$W(\theta)=1-0.182P_2(\cos\theta)+0.002P_4(\cos\theta),$$
$$\delta_\gamma=0.0617$$

and for the 328-keV γ ray,

$$W(\theta)=1+0.536P_2(\cos\theta)-0.021P_4(\cos\theta),$$
$$\delta_\gamma=0.234.$$

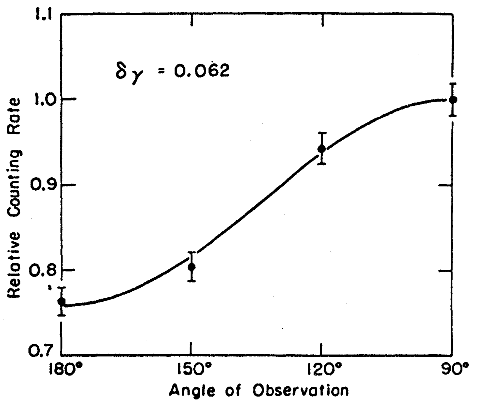

FIG. 19. Comparison of observed angular distribution of 288-keV γ ray to distribution based on 5^+ spin assignment to second excited state of Tl^{208}.

These distributions represent good fits to the observed data, and are compared with the experimental data on Figs. 19 and 20.

The experimental data lead to the conclusion that the spin of the second excited state of Tl^{208} is 5^+. The observed angular correlations also confirm the predominantly $M1$ nature of both transitions. It was estimated that for the 288-keV transition, δ_γ^2 is no larger than 0.01, and for the 328-keV transition no larger than 0.25.

$Tl^{208(4)}$

The angular correlation of the 452-keV γ ray with the 5.60-MeV α ray to the fourth excited state of Tl^{208} was found to be

$$W(\theta)=1-(0.101\pm0.017)P_2(\cos\theta)+(0.070\pm0.023)P_4(\cos\theta).$$

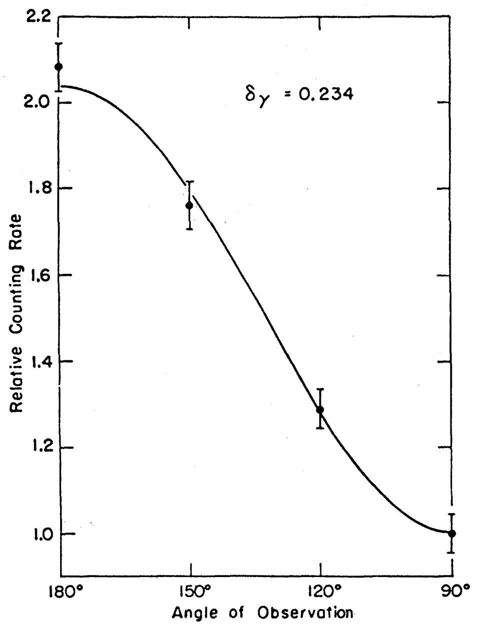

FIG. 20. Comparison of observed angular distribution of 328-keV γ ray to distribution based on 5^+ spin assignment to second excited state of Tl^{208}.

This distribution includes an error not precisely known due to the presence in the 452-keV γ-ray total energy peak of 433- and 473-keV γ rays which occur in coincidence with the 5.62-MeV α ray to the third excited state. These γ rays were not excluded by the differential discriminator used to select α-ray energies. The intensity of the 5.62-MeV α ray is about 13.5% of the intensity of the 5.60-MeV α ray. There is also a 145-keV transition which competes with the 433- and 473-keV transitions. In addition there is present a 492-keV γ ray which is also in coincidence with the 5.60-MeV α ray. The K-conversion line corresponding to this transition is reported by Sergeev *et al.*[29] to have about 8% of the intensity of the K-conversion line corresponding to the 452-keV transition. Nielsen[28] deduced the same ratio to be less than 5%, based on his failure to observe the 492-keV transition. The 433- and 473-keV γ rays are both thought to be $M1$, but their intensities relative to the 452-keV γ rays, and the values of the multipole mixing ratios for the 2 transitions are not known. Similarly, the relative intensity of the 492-keV γ ray is not known accurately. It is thought, however, that the conclusions that will be drawn from the above observed angular correlation will not be in error due to these unknown factors. The 452-keV transition is presumed to be predominantly $M1$ based on measurements of internal conversion coefficients. Because of the $M1$ nature of this transition, the spin of $\mathrm{Tl}^{208(4)}$ must be 3^+, 4^+, or 5^+.

The best fits to the observed data that can be made with the correlations based on 3^+, 4^+, and 5^+ assignments to $\mathrm{Tl}^{208(4)}$ are shown in Figs. 21 and 22. The calculated correlations are for an assignment of

$$3^+:\quad W(\theta)=1-0.072P_2(\cos\theta),$$

$$4^+:\quad W(\theta)=1-0.008P_2(\cos\theta)-0.101P_4(\cos\theta),$$

$$5^+:\quad W(\theta)=1-0.073P_2(\cos\theta)+0.006P_4(\cos\theta).$$

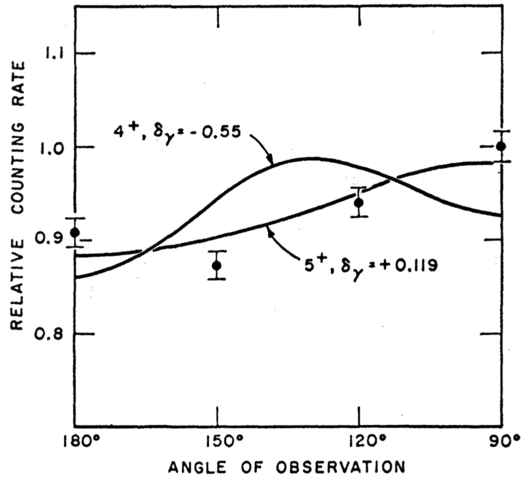

FIG. 21. Comparison of observed angular distribution of 452-keV γ ray to best fits attainable with distributions based on 4^+ and 5^+ spin assignments to fourth excited state of Tl^{208}.

The theoretical angular correlation corresponding to a 6^+ assignment to $\mathrm{Tl}^{208(4)}$ is

$$W(\theta)=1+0.437P_2(\cos\theta)-0.212P_4(\cos\theta).$$

The fits based on the 4^+ and 6^+ assignments are sufficiently poor that these assignments are unlikely. The experimental data indicate that the spin of $\mathrm{Tl}^{208(4)}$ is 3^+ or 5^+. The experimental result of Korolev *et al.*[61] leads to the same conclusion.

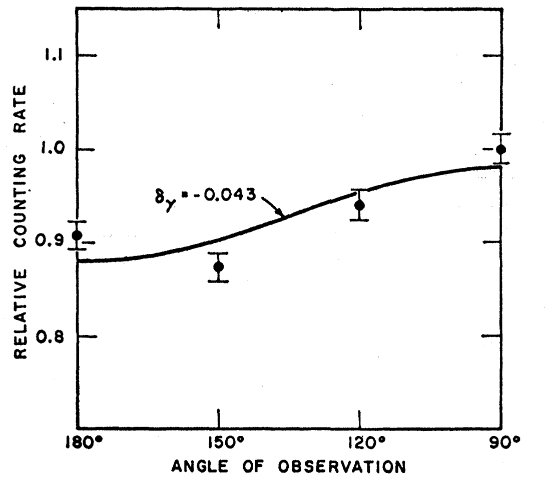

FIG. 22. Comparison of observed angular distribution of 452-keV γ ray to best fit attainable with distribution based on 3^+ spin assignment to fourth excited state of Tl^{208}.

CONCLUSIONS

It has been predicted by Pryce[7] that the second through fifth excited states of Tl^{208} form a $(d_{3/2})_p^{-1}(g_{9/2})_n$ quartet; his calculations indicate the sequence of spins is 5^+, 4^+, 6^+, and 3^+. Previous experiments indicate spin assignments of 5^+ and 4^+, respectively, to $\mathrm{Tl}^{208(0)}$ and $^{(1)}$, and a 1^- assignment to Bi^{212}.

$\mathrm{Tl}^{208(2)}$

A spin assignment of 5^+ or 4^+ is indicated on the basis of the predominantly $M1$ nature of the 288- and 328-keV transitions from this level. The present experiment indicates that 5^+ is the correct assignment.

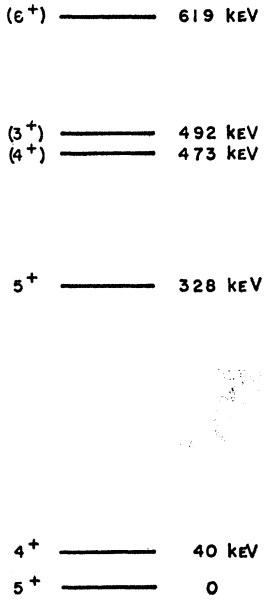

FIG. 23. Proposed level scheme for Tl^{208}.

$Tl^{208(4)}$

A spin of 3^+ has previously been suggested for this level on the basis of the $M1$ transition to the spin 4^+ first excited state and the weakness of the transition to the spin 5^+ ground state. This assignment is also consistent with the failure to observe a transition to the second excited state, which has here been found to have spin 5^+. This assignment is not in disagreement with the present experiment, which favors an assignment of 3^+ or 5^+. The 6^+ assignment suggested by the calculations of Pryce is inconsistent with the present experiment.

$Tl^{208(3)}$

A spin of 5^+ or 4^+ has been indicated by the observation of conversion electron intensities, which suggest that the transitions observed to the three lower levels are all $M1$. The 4^+ assignment is suggested by Pryce and is consistent with the 5^+ assignment that has been made to $Tl^{208(2)}$.

$Tl^{208(5)}$

The observation of conversion electrons corresponding to a transition to the spin 5^+ ground state, but not to the spin 4^+ first excited state, is consistent with a 6^+ assignment. Since 3^+ is most likely the spin of $Tl^{208(4)}$, 6^+ is the only unused spin assignment of the $(d_{3/2})_p^{-1}(g_{9/2})_n$ quartet.

Level Scheme

The measured angular correlations have contributed directly to the identification of the spins of the second and fourth states, and indirectly to the third and fifth states by induction from theory and previous experiment. A level scheme for Tl^{208} compatible with present information is presented in Fig. 23.

M1 Transitions from the Higher Excited States of Tl^{208}

The multipolarities of the 288- and 328-keV transitions have been confirmed to be predominantly $M1$. It is implied that there is configuration mixing in the ground and first excited state doublet $(s_{1/2})_p(g_{9/2})_n$ and the 4^+ and 5^+ states of the $(d_{3/2})_p^{-1}(g_{9/2})_n$ quartet, as otherwise these $M1$ transitions would be strictly l forbidden. This is consistent with the identification of the 452-keV transition as predominantly $M1$ from measurement of conversion coefficients, and the relative weakness of the 492-keV transition. For if the 452-keV transition contained a large $E2$ component, it would appear that the 492-keV transition should exhibit comparable strength.

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