Nuclear Energy Levels of Tl^{203}†

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Using scintillation spectrometry and coincidence counting techniques, Tl^{203}, formed by orbital electron capture in Pb^{203}, has been shown to emit gamma rays of energies 280, 403, and 683 kev. The 403- and 280-kev gamma rays were found to be in sequence; the 683-kev gamma ray represents the associated cross-over transition. By energy and conversion coefficient measurements, it has been shown that the same 280±2 kev level of Tl^{203} is excited in the decay of both Hg^{203} and Pb^{203}. A coincidence method for the measurement of the conversion coefficients of the 403-kev gamma rays is described. The K-shell conversion coefficient of the 403-kev gamma ray has been measured as 0.076, the $K/(L+M)$ ratio being 3.7. The angular correlation function for the 403-kev−280-kev cascade has the form $W(\theta)=1-(0.152\pm0.007)\cos^2\theta$. For the known multipole mixture of the 280-kev quantum, the correlation measurements indicate the 403-kev transition to be 76 percent $E2$ and 24 percent $M1$. The interference phases of the $E2$ and $M1$ matrices of both transitions are the same in Lloyd's notation. The angular correlation studies and the conversion coefficient measurements indicate the orbitals of the levels of Tl^{203} to be $d_{5/2}$, $d_{3/2}$, and $s_{1/2}$, in order of decreasing excitation energy. The spin of the ground state of Pb^{203} is shown to be 5/2(−).

INTRODUCTION

THE 46-day Hg^{203} has been shown[1–3] to decay by beta emission to a level at 280 kev in Tl^{203}. Early investigations of Pb^{203} showed[4] it to decay to Tl^{203} by orbital electron capture and subsequent emission of gamma rays of energies 270 and 470 kev. Magnetic spectrographic studies[5] of the radiations of the radio isotopes of lead have indicated that gamma rays of energies 153, 269, and 423 kev are emitted in the decay of the 52-hour Pb^{203}. From the energy and the measured K/L ratio of the 269-kev gamma ray, it was concluded[5] that the 269-kev level is different from the 280-kev level of Tl^{203} observed in the decay of Hg^{203}. It was also suggested by the same author that the 153- and 269-kev gamma rays are in cascade and that the 423-kev quantum is emitted in the cross-over transition.

In order to obtain further data concerning the levels of Tl^{203}, the radiations of Pb^{203} have been reinvestigated with thallium activated sodium iodide and anthracene scintillation spectrometers and coincidence methods.

CHEMICAL PROCEDURE

Pb^{203} was produced by the bombardment of metallic thallium by 14-Mev deuterons. To separate out the radioactive lead, the irradiated sample of thallium was dissolved in concentrated sulfuric acid; the sulfuric acid was then diluted to a 10N solution, and the lead sulfate thus precipitated was centrifuged out and washed with dilute sulfuric acid and a small quantity of water. The lead sulfate was then dissolved in an aqueous solution of ammonium acetate. Whenever a source in the solid state was needed, lead chromate was precipitated from the solution by addition of potassium chromate. In the course of some measurements, the lead chromate precipitate was dissolved in concentrated nitric acid with the addition of hydrogen peroxide.

ENERGIES AND RELATIVE INTENSITIES OF THE GAMMA RAYS

A strong source of Pb^{203} in solution was placed at a distance of 75 cm from a crystal of NaI(Tl) of thickness 3.5 cm which was mounted on a photomultiplier tube, RCA-5819. In Fig. 1 is plotted the pulse-height distribution generated in the crystal by the gamma rays of Pb^{203}. The source distance was large to avoid "pile up" of pulses arising from any coincident gamma rays. From the spectrogram it is clear that the quantum radiations of Pb^{203} consist of the 70-kev x-rays of thallium and gamma rays of energies 280, 403, and 683 kev. The small rise in counting rate at ~150 kev is produced by Compton secondaries in the crystal of the

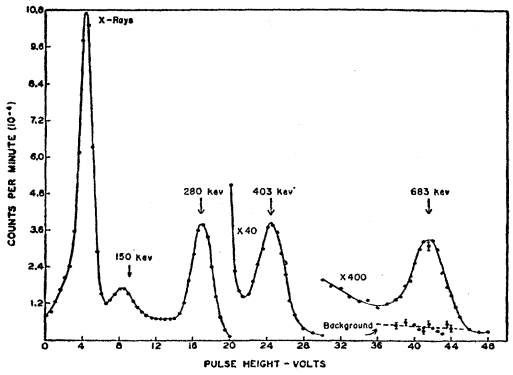

FIG. 1. Pulse-height distribution generated by gamma rays from Pb^{203} in a NaI(Tl) crystal of dimensions 3.5 cm×3.5 cm diameter.

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[2] H. W. Wilson and S. C. Curran, Phil. Mag. **42**, 762 (1951).

[3] Cork, Martin, LeBlanc, and Branyan, Phys. Rev. **85**, 386 (1952).

[4] Lutz, Pool, and Kurbatov, Phys. Rev. **65**, 61 (1944).

[5] G. D. O'Kelley, Ph.D. thesis, University of California Radiation Laboratory Unclassified Report UCRL 1243, May, 1951 (unpublished).

280-kev quantum and by recoil photons resulting from backscattering in the source.

An experiment was performed to determine accurately the energy of the 280-kev gamma ray and to compare it with that of the gamma ray emitted in the decay of Hg^{203}. The photopeak produced in the crystal by the gamma ray of Hg^{203} was scanned just before and immediately after observing the same peak associated with a source of Pb^{203}. The same geometry was maintained throughout the course of these comparative measurements, and the sources of Hg^{203} and Pb^{203} were approximately equal in strength. To avoid changes in pulse height resulting from fluctuations in room temperature or line voltage, the measurements were performed in a relatively short time. As shown in Fig. 2, the energies of the gamma rays at about 280 kev emitted in the case of both Hg^{203} and Pb^{203} differ by not more than 0.5 kev. This small limit of difference sug-

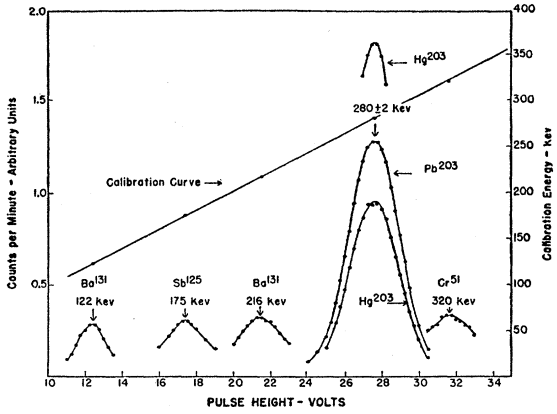

FIG. 2. Photopeaks due to gamma rays from Hg^{203} and Pb^{203}. The photopeak due to gamma ray from Hg^{203} was observed immediately before and after observing the peak due to gamma ray from Pb^{203}. The photopeaks due to gamma rays from Cr^{51}, Ba^{131}, and Sb^{125} were measured for calibration of the spectrometer.

gests that the same 280-kev level of Tl^{203} is excited in the decay of either radionuclide. From the calibration of the scintillation spectrometer, the energy of the gamma ray is 280 ± 2 kev.

To show that the rise in counting rate in the region of ~150 kev (Fig. 1) does not constitute evidence of the presence of the previously reported 153-kev radiation, a comparison was made between the gamma spectra of Hg^{203} and Pb^{203} using thin sources in which little backscattering occurred. In these measurements the peak at ~150 kev was found to be greatly reduced as compared with the intensity indicated in Fig. 1 and approximately what should be expected as the normal Compton contribution of a 280-kev gamma ray. The spectra of both radioelements were practically identical below 280 kev in energy, further substantiating that no appreciable 153-kev radiation is emitted by Pb^{203}, because Hg^{203} has been previously shown to emit a single gamma ray at 280 kev. From the ratio of the counting

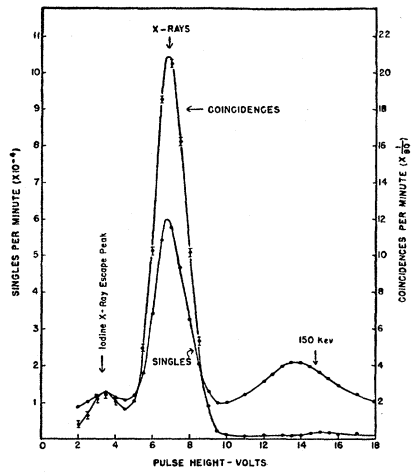

FIG. 3. Coincidence rate between the 280-kev gamma ray and the low-energy radiations from Pb^{203} where the statistical error, not shown, is less than the size of the point.

rate at ~150 kev to that at the photopeak, it was estimated that the 153-kev quantum could have an intensity of not more than two percent of that of the 280-kev quantum. Additional evidence of the absence of any 153-kev radiation was obtained from coincidence measurements limited to energies of 180 kev or less. A source of Pb^{203} was placed between the NaI(Tl) crystals of two coincident scintillation spectrometers. The channel of one of the spectrometers was fixed at the photopeak of the 280-kev gamma ray, while the channel of the other was allowed to traverse the interval of pulse heights corresponding to gamma-ray energies 20 and 180 kev. To avoid spurious coincidences arising from the backscattering of the 403- and 683-kev gamma rays from one crystal into the other, a lead absorber of thickness 2 g/cm² was placed between the source and the crystal of the spectrometer set at the photopeak of the 280-kev gamma ray. The presence of the lead absorber increased appreciably the single counting rate at ~150 kev. Only in the case of the 403-kev gamma ray does this scattering in the lead give rise to coincidences (it will later be shown that the 280- and 403-kev gamma rays are in cascade) at ~150 kev as indicated by the slight rise in the coincidence rate in that region of energy as shown in Fig. 3, where a spectrogram of quanta coincident with the 280-kev radiation is presented along with the single counting rate in the same energy interval. Any gamma ray of energy 153 kev of intensity even as little as one percent of that of the 280-kev line would have yielded a distinct photopeak in the coincidence rate.

The relative intensities of the gamma rays from Pb^{203} were obtained from the areas of the photopeaks (really "full energy" peaks) shown in Fig. 1. In making the calculations, the variation with energy of the efficiency of the crystal for absorption of the full energy of the

gamma rays was taken into account.[6,7] From these measurements, the relative intensities of the 683-, 403-, and 280-kev gamma rays were estimated to be 1:5.1:120. To calculate the relative intensities of the corresponding transitions, the total conversion coefficients of the gamma rays must also be considered. Taking the conversion coefficients of the 280- and 403-kev gamma rays to be 0.24[2] and 0.098 (the measurement of this latter value will be discussed in the text) and that of the 683-kev quantum to be negligible, the ratio of the transition intensities was found to be 1:5.6:150.

With every disintegration of Pb^{203} through capture of a K-shell electron (about 80 percent of the transitions[8,9] occur by K capture) there is associated a K-series x-ray of thallium. Other quanta of 70-kev thallium x-rays may arise from the K-shell conversion of the several gamma rays emitted. The contribution to the x-ray intensity arising from internal conversion is appreciable only in the case of the 280-kev line. Because of the low intensities and small conversion coefficients, the remaining two gamma rays contribute little to the x-ray intensity. The Auger effect[10] reduces the intensity of the x-rays from thallium by about five percent. If one takes into account the efficiency of the crystal for full energy absorption of the 280-kev gamma ray and the 70-kev x-rays, the K-shell conversion coefficient of the 280-kev gamma ray, absorption of the 70-kev x-rays in the aluminum-magnesium oxide housing and in the source itself, and the Auger effect, a comparison of the areas under the 70-kev and 280-kev photopeaks indicates that with each disintegration (K and L capture included) there is associated roughly one 280-kev transition. On considering all possible errors resulting from the several corrections applied, it was concluded that perhaps as many as ten percent of the K-capture disintegrations might proceed to the ground state. However, as will be shown later in the text, coincidence measurements yield a much lower value for the upper limit of the probability of the ground-state transition.

From the pulse-height spectrum of the conversion electrons emitted from a source of Pb^{203} and observed in an anthracene crystal, the $K/(L+M)$ ratio for the 280-kev gamma ray was found to be 2.5. This value is in good agreement with recent measurements[11] of the corresponding ratio for the decay of Hg^{203}. Because the intensity of the conversion electrons related to the 403-kev gamma ray is quite low, its $K/(L+M)$ ratio could not be determined by this method.

COINCIDENCE MEASUREMENTS

To determine the relationship between the 280- and 403-kev gamma rays, coincidences between the two quanta were studied. Two NaI(Tl) scintillation spectrometers were placed in coincidence, and in the crystal of either spectrometer there was generated a pulse-height distribution like the one shown in Fig. 1 when a source of Pb^{203} was interposed between the two crystals. The channel of one of the spectrometers was set at the photopeak of the 280-kev gamma ray while the channel of the other was moved in steps of ~9 kev from 335 to 445 kev. In Fig. 4 is plotted the coincidence rate (represented by open circles and calculated statistical probable errors) as observed in the above-cited energy interval. The points are to be compared with the single counting rate (represented by the solid curve). From the close agreement between the coincidences and the single counting rate, it can be concluded that the 403- and 280-kev gamma rays are indeed in cascade and that the 683-kev radiation is emitted in the cross-over transition. This is supported by the fact that coincidences were observed between the 683-kev gamma rays and the x-rays. No other quantum radiations were found to be coincident with the 683-kev gamma ray.

A coincidence method was employed in a further attempt to estimate the number of electron capture disintegrations of Pb^{203} which proceed directly to the ground state of Tl^{203}. A source of Pb^{203} was placed before the adjacent NaI(Tl) crystals of two scintillation spectrometers in coincidence. The crystals were shielded from each other by a thickness of one centimeter of lead.

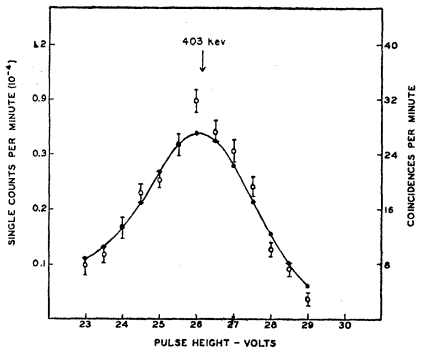

FIG. 4. Coincidence rate between the 280-kev gamma ray and the gamma spectrum at 403 kev. The solid curve represents the single counting rate. The points, shown with their statistical error, represent the coincidence rate in each position of the channel.

[6] The efficiency of a crystal for absorption of the total energy of a gamma ray is given by $E=RS$, where S is the total interaction cross section of the crystal for the gamma ray, and R is the ratio of the area of the full energy peak to that of the total contribution to the counting rate from the gamma ray. Both R and S vary with quantum energy and crystal size. The value of S was obtained from the curve given by Jordan,[7] while R was evaluated as a function of energy by observing the pulse-height distribution generated in the crystal by several different monoenergic gamma-ray sources. R is a function of the distance and geometry of the source, but for the gamma rays shown in Fig. 1, the relative values of R will change but little as the source geometry and distance is altered.

[7] W. H. Jordan, Ann. Rev. Nuclear Sci. **1**, 221 (1951).

[8] R. E. Marshak, Phys. Rev. **61**, 431 (1942).

[9] M. E. Rose and J. L. Jackson, Phys. Rev. **76**, 1540 (1949).

[10] E. H. S. Burhop, *The Auger Effect* (Cambridge University Press, Cambridge, 1952).

[11] F. R. Metzger (private communication, 1953). Data relating to the conversion coefficients of the gamma ray from Hg^{203} are $\alpha_K=0.163\pm0.006$, $K/L=3.5$, $L/M=3.2$.

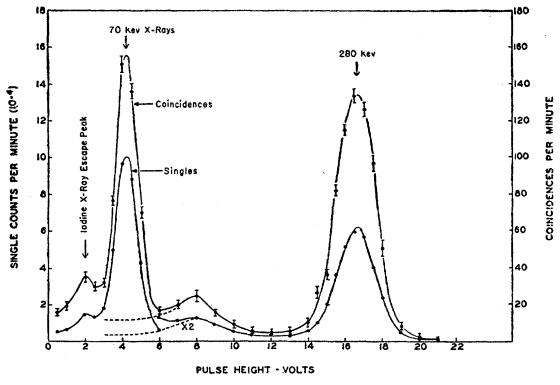

FIG. 5. Coincidence rate between Tl K x-rays and the low-energy gamma spectrum from Pb^{203}. The curve with points shown with statistical error represent the coincidence rate. The single counting rate is also plotted.

The channel of one spectrometer (hereafter denoted by the subscript "1") was fixed at the photopeak of the 70-kev x-rays while that of the other (henceforth to be indicated by the subscript "2") was moved over the region of energy lying between 20 kev and 350 kev. The single counting rate as well as the coincidence rate was recorded for each pulse-height setting of the channel as shown in Fig. 5. The two peaks in the coincidence rate at 70 and 280 kev represent coincidences between x-rays and x-rays, and x-rays and gamma rays.

If N_{xx} represents the number of coincidences between x-rays (two x-rays will be emitted when, after K capture, the 280-kev gamma ray is converted), $N_{x\gamma}$ the number of coincidences between 70-kev x-rays and 280-kev gamma rays, N_{x2} the number of single counts due to x-rays, and $N_{\gamma 2}$ the number of single counts arising from 280-kev gamma rays recorded in the second spectrometer while measuring N_{xx} and $N_{x\gamma}$, an extension of the treatment given by Dunworth[12] yields

$$\frac{N_{xx}}{N_{x2}}\cdot\frac{N_{\gamma 2}}{N_{x\gamma}}=\frac{2\alpha_K F}{K+K\alpha_T+\alpha_K F}, \tag{1}$$

where α_T and α_K are the total and the K-shell conversion coefficients of the 280-kev gamma ray, K the fraction of the disintegrations of Pb^{203} occurring through K-shell electron capture, and F the fraction of all the disintegrations of Pb^{203} in which the 280-kev transition is involved. From the calculations of Marshak[8] and the tables of Rose and Jackson,[9] the value of K is found to be 0.80 for the first forbidden transitions in question. The single counting rates at the energies of the x-ray peak and the 280-kev gamma-ray peak were properly corrected by extrapolations shown in Fig. 5 for the presence of counts arising from the Compton effect of radiations of higher energies. A similar extrapolation was necessary to remove x-ray—gamma coincidences from the apparent (x-ray)-(x-ray) coincidences. The extrapolation indicated in Fig. 5 corrects for coincidences between x-rays detected in spectrometer "1" and γ rays detected by way of the Compton effect in spectrometer "2." An additional correction is necessary to take into account $N_{x_2\gamma_1}$ the coincidences between gamma rays detected in spectrometer "1" and x-rays in spectrometer "2." The latter correction was obtained by observing the spectrum of "singles" in spectrometer "1" and extrapolating as in Fig. 5 to find the contributions of the gamma rays to the single counting rate in the x-ray peak; then $N_{x_2\gamma_1}$ can easily be calculated. In particular,

$$\frac{N_{x_2\gamma_1}}{N_{x_1\gamma_2}}=\frac{N_{x_2}}{N_{\gamma_2}}\cdot\frac{N_{\gamma_1}}{N_{x_1}}. \tag{2}$$

In practice, the actual calculation of the quantity $(N_{xx}/N_{x2})(N_{\gamma 2}/N_{x\gamma})$ was made only at the three highest points of the peaks of Fig. 5. The average of the three computations 0.30 ± 0.010, was then employed to obtain F from Eq. (1), which becomes

$$F\alpha_K/(0.80+0.80\alpha_T+\alpha_K F)=0.150\pm0.005. \tag{3}$$

The conversion coeficients are taken to be those[11] of the 280-kev gamma ray of Hg^{203}, it being assumed that the gamma rays of this energy emitted by Hg^{203} and Pb^{203} are identical. This assumption appears valid from a consideration of the energies and of the $K/(L+M)$ ratios of the two transitions. From Eq. (3), the calculated value of F is 1.06 ± 0.05. Thus, the fraction of the total number of disintegrations proceeding to the ground state is small.‡

To determine the conversion coefficient of the 403-kev gamma ray, the coincidence rate between the conversion electrons and the 280-kev gamma ray from Pb^{203} was studied. To make these measurements a source of Pb^{203} of thickness about 1 mg/cm^2 was placed at a distance of 0.08 cm from an anthracene crystal 2.5 cm in diameter and 1 cm in thickness, mounted upon a Dumont 6292 photomultiplier tube. The conversion electrons were recorded over an energy interval extending from 250 to 450 kev. The gamma rays were detected in a NaI(Tl) scintillation spectrometer, the channel of which was set at the photopeak of the 280-kev gamma ray.

In Fig. 6 is plotted the value of $N_{\gamma e^-}/N_\gamma$ as a function of the pulse-height setting of the channel of the anthracene spectrometer. $N_{\gamma e^-}$ is the number of coincidences between the 280-kev gamma rays and the conversion electrons of the 403-gamma ray and N_γ is the number of single counts recorded simultaneously with the coincidences in the gamma-ray spectrometer fixed at the photopeak of the 280-kev gamma ray. To obtain the coincidence rate of Fig. 6, the coincidences due to Compton recoils produced in the anthracene crystal by the 403-kev gamma ray were subtracted from the observed coincidence rate. This Compton contribution to

[12] J. W. Dunworth, Rev. Sci. Instr. **11**, 167 (1940).

‡ Since this paper was submitted for publication, a more detailed analysis of the statistics of the measurements has indicated that the relative probability is one-half that the value of F lie between 0.98 and 1.00.

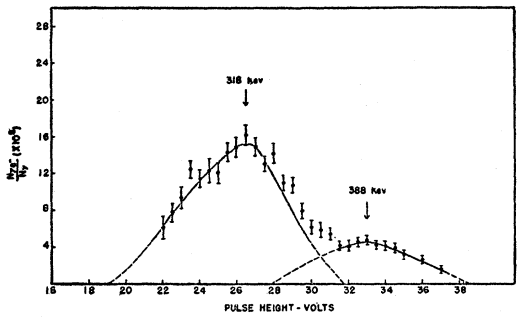

FIG. 6. Coincidence rate between the 280-kev gamma ray and the electrons from the conversion of the 403-kev gamma radiation.

the coincidence rate was obtained by interposing 0.08 centimeter of aluminum between the source and the anthracene crystal. In the coincidence curve of Fig. 6, the peaks arising from the K and L shell conversion electrons are well-resolved. The conversion electrons in the group centered at 388 kev include also those originating in the M shell. The slight asymmetry of the peak of K-shell electrons may be ascribed to non-uniformities in the anthracene crystal and to energy loss in the source. An extension of the treatment given by Dunworth[12] results in the equation

$$(N_{\gamma e^-}/N_\gamma)_\Sigma = f\omega_e\alpha_K/(1+\alpha_T), \tag{4}$$

where f is the fraction of the 280-kev transitions in sequence with the 403-kev radiation, converted and unconverted, ω_e the fractional solid angle subtended at the source by the anthracene crystal, and $(N_{\gamma e^-}/N_\gamma)_\Sigma$ the area under the K-shell peak of Fig. 6. From Fig. 1, f is estimated to be $5.1(1+\alpha_T)/150$, and ω_e is calculated to be 0.47. From Eq. (4) and the quantities defined above, α_K is calculated to be 0.063. An attempt was made to take into account backscattering in the source and at the surface of the crystal. The resulting value of α_K was found to be about 20 percent greater. This correction increases the value of α_K to 0.076.

A comparison of the areas of the two conversion electron peaks of Fig. 6 gives the ratio

$$\alpha_K/(\alpha_L+\alpha_M)=3.7. \tag{5}$$

The results of these conversion coefficient measurements were such as to suggest that the 403-kev transition is a mixture of $M1$ and $E2$ radiations. From the K-shell conversion coefficient and the curves of Rose *et al.*,[13] the 403-kev radiation is found to be a mixture, 74 percent $E2$ and 26 percent $M1$. By comparing the $K/(L+M)$ ratio with curves given by Goldhaber and Sunyar,[14] a similar mixture is obtained.

The value of the conversion coefficient of the 280-kev gamma ray identifies it also as a mixture of $E2$ in $M1$, 74.7 percent $E2$ and 25.3 percent $M1$, so that the ratio of the intensities is

$$E2/M1=2.95^{+0.31}_{-0.25}. \tag{6}$$

The levels of Tl^{203}, concerned in the decay of Hg^{203}, have been shown to have spin values of $3/2$[1,11,15] for the first excited state and $1/2$[16,17] for the ground state.§ Since the lowest multipole order of the 403-kev transition is $M1$, the 683-kev level of Tl^{203} could have spin values of 1/2, 3/2, or, 5/2 with the same parity as that of the two lower levels, which, according to the shell model,[18] have even parity.

ANGULAR CORRELATION STUDIES

To obtain more evidence concerning the spin values of the excited states of Tl^{203} excited in the decay of Pb^{203}, the spatial correlation of the two cascade gamma rays was investigated. Taking the orbital of the first excited level of Tl^{203} to be 3/2, the calculations of Ling and Falkoff[19] and Lloyd[20] show that the angular correlation function should contain only a $\cos^2\theta$ term in addition to the isotropic contribution; that is, it should be of the form

$$\omega(\theta)=1+a_2\cos^2\theta. \tag{7}$$

Because of this fact, the asymmetry in correlation was measured only at 135° and 180°. The experiment was performed with two NaI(Tl) scintillation spectrometers in coincidence. In one channel of the coincidence circuit, all pulses above the pulse height corresponding to a gamma-ray energy of 180 kev were accepted. In the other channel, all pulse heights greater than that of a pulse of a 330-kev gamma ray were accepted. The source employed in the measurements was in the form of powdered lead chromate. A "least squares" fit of the data yielded for $\omega(\theta)$

$$\omega(\theta)=1-(0.147\pm0.007)\cos^2\theta. \tag{8}$$

When corrected for the finite angular resolution of the detectors, the correlation function becomes

$$\omega(\theta)=1-(0.152\pm0.007)\cos^2\theta. \tag{9}$$

The asymmetry at 180° was observed using solutions of lead chromate in nitric acid and lead sulfate in an aqueous solution of ammonium acetate. The asymmetry after the resolution correction was found to be de-

[12] J. W. Dunworth, Rev. Sci. Instr. **11**, 167 (1940).
[13] Rose, Goertzel, and Perry, Oak Ridge National Laboratory Unclassified Report, ORNL-1023, July, 1951 (unpublished).
[14] M. Goldhaber and A. W. Sunyar, Phys. Rev. **83**, 906 (1951).
[15] Hollander, Perlman, and Seaborg, Revs. Modern Phys. **25**, 469 (1953).
§ Assignment of spin 3/2 to the first excited state of Tl^{203} is further supported by the asymmetric correlation function described in the next section.
[16] H. Schüler and J. E. Keystone, Z. Physik **70**, 1 (1931).
[17] D. A. Jackson, Z. Physik **75**, 223 (1932).
[18] Maria G. Mayer, Phys. Rev. **78**, 16, 22 (1950).
[19] D. S. Ling, Jr. and D. L. Falkoff, Phys. Rev. **76**, 1639 (1949).
[20] S. P. Lloyd, Ph.D. thesis, University of Illinois, 1951; see also Phys. Rev. **85**, 904 (1952).

scribed by $a_2=-0.175\pm0.028$ in the case of the lead sulfate solution and $a_2=-0.154\pm0.023$ for the lead chromate solution. The value of a_2 indicated in Eq. (9) and these latter values overlap when their respective statistical probable errors are considered, showing that the correlation function is independent of the physical state of the source.

To obtain the spin of the 683-kev level of Tl^{203} from the correlation measurements, the theoretical values of a_2 were obtained for dipole-mixture and quadrupole-mixture correlations from relations by Ling and Falkoff[17] for the possible spins of the second excited level, 1/2, 3/2, and 5/2. The mixture of the 280-kev transition was taken to be 74.7 percent $E2$ and 25.3 percent $M1$. Using these values of a_2, the mixture-mixture correlations for all the above mentioned spins were calculated for all possible mixtures in the 403-kev transition. In calculating the mixture-mixture correlation functions, the expressions given by Lloyd[20] were used. From these calculations, it can be seen that the observed correlation can be explained by any one of the following sets of conditions:

(a) The spin of the second excited state is 1/2 and the 403-kev gamma ray is a mixture of $E2$ and $M1$ such that $E2/M1=320$ or 0.16.

(b) The spin of the second excited state is 3/2 and the 403-kev transition is a mixture such that $E2/M1$ is approximately zero or greater than 800.

(c) The spin of the second excited level is 5/2 and the mixture of $E2$ with $M1$ in the 403-kev transition is such that $E2/M1=22$, 0, 0.11, or 3.1.

The fact that the previously discussed conversion coefficient measurements show the 403-kev transition to be 74 percent $E2$ and 26 percent $M1$ ($E2/M1=2.85$) rules out all conditions listed above except (c) where the spin of the 683-kev level is 5/2 and the 403-kev gamma ray is a mixture of $E2$ and $M1$ such that $E2/M1=3.1$. The conditions (a) and (b) are ruled out

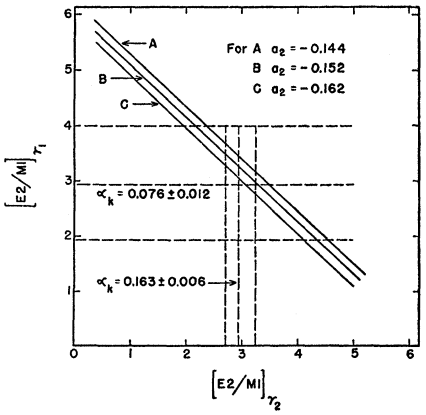

FIG. 7. The dependence of multipole mixture ($E2/M1$) in the first transition on the multipole mixture ($E2/M1$) in the second transition for the observed correlation function. The $E2/M1$ ratio allowed by the values of the conversion coefficients and their errors is shown by the dotted lines.

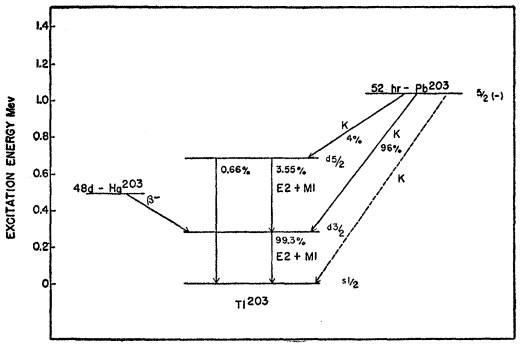

FIG. 8. Level scheme of Tl^{203}.

also by the observed intensities of the electron capture transitions and by the observed relative intensities of gamma rays originating from the second excited state of Tl^{203}.

For any given correlation function, the calculated ratio of $E2$ to $M1$ in the first transition is, of course, a function of the same ratio for the second transition. In Fig. 7 is shown the variation of the mixture in the first transition with the assumed mixture for the second transition, the correlation function being that observed in the present measurements. The curve was calculated taking as 180° (Lloyd's notation), the interference phase between $E2$ and $M1$ components of both transitions. The mixing ratio of the 403-kev transition can also be obtained from the curve of Fig. 7. The known mixture of the 280-kev transition and the observed correlation function give a value of 3.2 for $E2/M1$ in the case of the 403-kev gamma ray.

DISCUSSION

The angular correlation and conversion coefficient measurements establish the spins of the levels of Tl^{203} as 5/2, 3/2, and 1/2 in order of decreasing excitation energy. The parity for all the levels is even. In Tl^{203}, the number of protons is just one short of a magic number; so it is appropriate to write the orbitals as $d_{5/2}$, $d_{3/2}$, and $s_{1/2}$ in accord with the single-particle assignments of the shell model. The fact that few or no electron capture transitions to the ground state of Tl^{203} are observed excludes the possibility that the ground-state spin of Pb^{203} might be 1/2 or 3/2. If the spin of the ground state of Pb^{203} is assumed to be 7/2 or more, it is impossible to explain the relative intensities of the electron capture transitions which lead to the $d_{5/2}$ and $d_{3/2}$ levels of Tl^{203}. However, a spin of 5/2(−) does satisfactorily explain the relative intensities of the capture transitions forming Tl^{203}.

If the ft values of the transitions to the 683- and 280-kev levels are taken to be about equal, the energy release of the capture process terminating at the ground state of Tl^{203} is calculated[21] to be 1.03 Mev. For an electron capture transition to the ground state of Tl^{203}

[21] S. A. Moszkowski, Phys. Rev. 82, 35 (1951).

of relative intensity two percent, logft is found to be 8.0. The same quantity for transitions to the excited levels has the value 6.5. These values of logft suggest that all the transitions to Tl^{203} are first forbidden[22] ($\Delta I=0\pm1$; ±2, yes!); this is additional confirmation that the ground state of Pb^{203} should be $5/2(-)$.

The complete level scheme of Tl^{203}, as derived from the present measurements, is shown in Fig. 8. There is a similarity between the level scheme of Tl^{203} and those of odd-A isotopes of Au and Ir.[23] Other recent measurements[24,25] as well as these seem to indicate that $M1+E2$ mixtures are a general feature of transitions in odd-Z—even-N nuclei.

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Disintegration of Y^{91}

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The radiations of the fission product Y^{91} have been investigated with beta- and gamma-scintillation spectrometers. In addition to the well-known 1.55-Mev ground-state beta transition, there is a weak (0.3 percent) beta transition of energy 0.330 Mev which is followed by a gamma ray of energy 1.22 ± 0.01 Mev. Definite assignment of these radiations to Y^{91} was established through exacting chemical procedures, measurement of the half-life of the gamma ray, and energy balance considerations. There is reason to believe that the 1.22-Mev level of $_{40}Zr^{91}$ is not a simple single-particle state.

INTRODUCTION

THERE has been considerable confusion in the literature as to whether or not there is weak γ radiation associated with the 60-day Y^{91} activity.[1–3] The recent measurements of Boley and Dunavan[3] suggest that the 1.2-Mev gamma ray ascribed to Y^{91} by Langer and Price is actually due to an impurity whose half-life is 160 ± 30 days. Boley and Dunavan base their impurity argument not only on the half-life discrepancy but also on the fact that they found a beta-to-gamma ratio of ~20, whereas Langer and Price had found this ratio to be >1000. However, since an examination of highly purified Y^{91} (prepared at this laboratory) indicated the presence of 1.2-Mev gamma rays, it seemed worthwhile to investigate in detail the source of this gamma radiation.

APPARATUS

Scintillation spectrometers were used for the measurement of the beta- and gamma-ray spectra. NaI(Tl) crystals 2 inches in diameter and 2 inches thick were used for gamma detection, and a Pilot Plastic Scintillator-B[4] phosphor $1\frac{3}{4}$ inches in diameter and $\frac{3}{4}$ inch thick was used for beta detection. The scintillators were coupled directly to Dumont type 6292 photomultiplier tubes. Each NaI(Tl) crystal was surrounded by a layer of MgO reflector. The MgO was held in place by a thin aluminum cup. The beta phosphor was covered with aluminum 0.0005 inch thick which served both as a light shield and a reflector.

A block diagram of the electronic circuitry used to obtain coincidence spectra is shown in Fig. 1. The twofold coincidence circuit has a resolving time of about 0.2 microsecond.

GAMMA-SPECTRUM

Preliminary examination of the gamma-ray spectrum of Y^{91} with the scintillation spectrometer revealed a weak photopeak at about 1.2 Mev and an intense, hyperbolic-shaped distribution at lower energy which had an apparent endpoint in the neighborhood of 1 Mev. The bulk of the latter distribution was attributed to internal and external bremsstrahlung resulting from

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