Energy Levels of Ti^{48} from the Decay of Sc^{48} and V^{48}

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The gamma rays following the decay of Sc⁴⁸ and V⁴⁸ have been investigated by means of threefold coincidence techniques. In the case of Sc48, it was found that within the limits of experimental error all of the decays occur via a three-gamma cascade. In V48, although the decay occurs primarily via a two-gamma process, a small three gamma contribution was detected. By twofold coincidence measurements, the 2.2-Mev gamma ray of V48 decay was found to be in cascade with the 0.99-Mev gamma ray. Level schemes are proposed for both decays.

I. INTRODUCTION

R ECENTLY the simple decay schemes which were suggested by the early work with Sc48 and V48 (both of which decay to levels in Ti⁴⁸) have undergone considerable modification. The previous experimental work¹⁻³ indicated a cascade of two gamma rays, 0.99 Mev and 1.32 Mev in each case, with a third gamma ray of low intensity observed in the V48 decay and believed to be the crossover (i.e., energy = 2.31 Mev). Typical pulse-height distributions, covering the region 0.3 to 1.4 Mev, are shown in Fig. 1.

In the case of the Sc⁴⁸ decay, the first suggestion that more than two gamma rays might be involved came in 1952 from Kurath,⁴ who predicted a third gamma ray of energy about 1 Mev in the cascade. Hamermesh et al.⁵ provided experimental support for this prediction by analyzing the gamma-ray pulse-height distribution observed with a sodium-iodide scintillation spectrometer and by investigating twofold gamma-ray coincidences from a Sc48 source.

In the decay of V⁴⁸, the weak high-energy gamma ray originally thought to be the crossover for the 0.99-Mev and the 1.32-Mev gamma cascade was bracketed in energy between the thresholds for photoneutron production in deuterium and beryllium, i.e., between 1.63 and 2.23 Mev, by Fluharty and Deutsch.⁶

The work of Ticho et al.,7 using a scintillation spectrometer, accurately determined the energy of this gamma ray to be 2.22 Mev and hence clearly showed that it could not be the crossover. This result was soon confirmed by other workers.^{8,9} The ray described, undetected in Sc⁴⁸ decay, has been estimated to occur in about 2 percent of the V48 decays.7,8

The Indiana group reported that they had observed coincidences between the 2.2-Mev gamma ray and

- ² Good, Peaslee, and Deutsch, Phys. Rev. 69, 313 (1946).
- ⁸ Robinson, Ter-Pogossian, and Cook, Phys. Rev. 75, 1099 (1949)
- D. Kurath, Phys. Rev. 87, 528 (1952)
- ⁵ Hamermesh, Hummel, Goodman, and Engelkemeier, Phys. Rev. 87, 528 (1952).
- ⁶ R. G. Fluharty and M. Deutsch, Phys. Rev. 76, 182 (1949).
 ⁷ Ticho, Green, and Richardson, Phys. Rev. 86, 422 (1952).
 ⁸ M. M. Miller, Phys. Rev. 88, 516 (1952).
 ⁹ Roggenkamp, Pruett, and Wilkinson, Phys. Rev. 88, 1262 (1952). (1952).

annihilation radiation and, in addition, detected a weak high-energy positron by beta spectrometer measurements.^{8,9} The decay schemes proposed by this group for the decay of Sc48 did not take into account the threegamma cascade and involved levels whose spins and parities were difficult to reconcile with the shell model.

The present work was undertaken in an attempt to check some of the previous reports with the aim of determining more satisfactory decay schemes for these mass 48 nuclei. The results of our investigation are represented in the proposed decay scheme shown in Fig. 2.



FIG. 1. Typical Sc⁴⁸ and V⁴⁸ gamma-ray pulse-height distributions in region 0.3-1.4 Mev. The curves have been normalized so that the 1.32-Mev photopeaks have the same height.

¹ W. C. Peacock and M. Deutsch, Phys. Rev. 69, 306 (1946).



FIG. 2. Proposed decay scheme for Sc^{48} and V^{48} to levels in Ti^{48} .

II. SOURCE PREPARATION

The Sc⁴⁸ used in this work was prepared by bombardment of Ti⁴⁸ metal with high-energy neutrons produced in the Argonne cyclotron by bombardment of a beryllium target with 22-Mev deuterons. A few hours' bombardment supplied a sufficient amount of the desired 44-hour half-life without significant impurities. By checking the half-life and investigating the pulse-height distribution, we found the sources pure enough not to require chemical separation.

The V⁴⁸ was produced by bombardment of a chromium-plated copper target with 22-Mev deuterons in the cyclotron. After bombardment the plating was dissolved and the V⁴⁸ chemically separated. The V⁴⁸ was divided into sources of various strengths, each of which was inserted in a small glass bottle whose walls were thick enough to stop the positrons without appreciably absorbing the gamma rays.

III. TRIPLE-COINCIDENCE MEASUREMENTS

The apparatus consisted of three single-crystal NaI(Tl) scintillation spectrometers employing singlechannel, pulse-height analyzers whose output pulses were fed into a triple coincidence circuit. The analyzers could be operated either as differential or integral discriminators. By means of a switch, the coincidence circuit could be altered to detect twofold coincidences.

The crystals, each of diameter 1.25 in. and thickness 0.5 in. and used in conjunction with RCA-5819 photomultipliers, were mounted symmetrically with their cylindrical axes in a horizontal plane. The sources were located about 2 cm from the center of each crystal. Since the coincidence arrangement was relatively slow (resolving time about 2 μ sec) and conveniently strong sources always gave an appreciable number of accidental coincidences, the true coincidence rates were established in the following way:

The observed triple coincidence rate T can be expressed mathematically in the form $T=aS+bS^2+cS^3$. In this expression S is the singles counting rate in each detector, and a, b, and c are factors involving the detection efficiency for each gamma ray and the number of gamma rays in cascade. The term aS is associated with real triple coincidences; the term bS^2 comes from a real double coincidence combining with a third independent pulse to give an accidental triple coincidence; and the term cS^3 arises from the accidental production of a coincidence by three pulses. Since correction for the omission of the cS^3 term never exceeded a few percent for the sources used in the present work, the above relation was written in the approximate form

$$T/S = a + bS. \tag{1}$$

The triple coincidence experiments consisted of dividing the source material into sources of various convenient strengths, determining singles and coincidence counting rates for each, and plotting (T/S) vs S. Straight lines were observed in each case, and a nonzero intercept, a, was a measure of the occurrence of a three-gamma, or higher-order, cascade. Experiments performed with Co⁶⁰ and Sc⁴⁶ sources produced straight lines passing through the origin. The results for Sc⁴⁸, Co⁶⁰, and Sc⁴⁶ are shown in Fig. 3.

The possibility that one or two pulses might occasionally feed through the electronic circuitry to produce an apparent triple-coincidence count (and hence lead to a false value for the intercept, a) was separately investigated, and a null result was obtained.

From the data taken on twofold coincidences, it was possible, in conjunction with the observed intercept, a, to estimate the number of disintegrations involving a three-gamma cascade relative to the total number of disintegrations. For Sc⁴⁸, a ratio of 1.05 ± 0.07 was obtained. Since beta-ray measurements on Sc⁴⁸ decay



FIG. 3. Observed ratio of triples to singles counting rates as function of singles counting rate for Sc⁴⁸, Co⁶⁰, and Sc⁴⁶.

have detected only one branch, it seems reasonable to conclude that essentially all of the disintegrations are associated with a three-gamma cascade.

In the case of V⁴⁸, in which the same operating conditions were used as for Sc48, it became immediately apparent from the small observed value of the intercept a, that the major mode of decay involved a two-gamma cascade.

It was found that part of this observed intercept was associated with triple coincidence pulses with one of the three component pulses produced by the combination (in one of the crystals) of an annihilation quantum and a Compton quantum escaping from one of the other crystals. An investigation of double coincidences with Na²² showed that this effect could be eliminated by shielding the crystals from each other with $\frac{1}{8}$ in. of lead and raising the integral discriminator bias settings to 890 kev from the previously used levels of about 720 kev.

The new bias settings reduced the detection efficiencies to the extent that a month of almost continuous counting was required in the V^{48} experiment. The stability of the equipment was checked periodically against the 890-kev photoline of a Sc46 source. The intercept obtained under these conditions was, as before, nonvanishing, implying the presence of a three gamma cascade. Unlike those in the Sc48 case, the gamma ray energies involved in the V48 three gamma cascade are not known unambiguously, as may be seen from the proposed decay scheme. If it is assumed that the energies are the same as the ones associated with Sc48 decay (i.e., involving the 3.35-, 2.31-, 0.99-, and 0.0-Mev levels of Ti⁴⁸), then a comparison of the V⁴⁸ intercept with the Sc^{48} intercept gives the result that the triple cascade in V⁴⁸ occurs in 7 ± 3 percent of the disintegrations. On the other hand, if it is assumed that the 3.22, 2.31, 0.99, and 0.0 levels are responsible for all of the threefold coincidences in V48, our efficiency for detecting the cascade will be lower, and a result of 10 ± 5 percent will be obtained.

IV. POSITRON EMISSION VS ELECTRON CAPTURE IN V48

Since 95 ± 5 percent of the decays of Na²² occur by positron emission² and all of the decays are associated with the emission of a single 1.28-Mev gamma ray, the number of positrons per 1.28-Mev gamma ray is 0.95 ± 0.05 . If this number and the observed Na²² and V⁴⁸ pulse-height distributions are used, an estimate can be made of the number of positrons per 1.32-Mev gamma ray in V48. Comparing the annihilation and 1.28-Mev photopeaks in the one case with the annihilation and 1.32-Mev photopeaks in the other, we obtained the result 0.49±0.04 positrons per 1.32-Mev gamma ray in V48. An empirical correction for the variation of photopeak magnitude with energy was employed. This result is an average of the values obtained by using four different NaI(Tl) crystals: three of our standard crystals (1.25 in. in diameter and 0.5 in. thick) and one larger crystal (1.5 in. in diameter and 1.0 in. thick). It is in good agreement with the value of 0.46 ± 0.09 obtained by the same method by Sterk et al.¹⁰ but only in fair agreement with the measurement of Good et al.² which gave 0.58 ± 0.04 positrons per disintegration.

V. ANALYSIS OF THE Sc48 GAMMA-RAY ENERGIES

By normalizing the Sc⁴⁸ and V⁴⁸ pulse-height distributions so that the 1.32-Mev photopeaks were of equal height and subtracting the V⁴⁸ from the Sc⁴⁸ distribution, Sterk et al.¹⁰ determined the energy of the third gamma ray in Sc48 to be 1.05 Mev. By the same method we have obtained 1.04 ± 0.02 Mev.

VI. THE 2.2-MEV GAMMA RAY OF V48

From V⁴⁸ gamma-ray pulse-height distributions, we have been able to estimate the energy and relative intensity of the 2.2-Mev gamma ray, obtaining 2.23 ± 0.03 Mev for the energy and 2.3 ± 0.5 percent for the intensity relative to the 1.32-Mev gamma ray. These results are in good agreement with those obtained by others.7,8

The weak high-energy branch shown in the positron spectrum of the Indiana group⁹ did not appear to be established with certainty but indicated an energy difference between the two branches of about 120 kev although only about 90 kev had been expected from the gamma-ray measurements. The positron spectrum has recently been reinvestigated by Marquez¹¹ at the University of Chicago. The high-energy branch was not visible in the positron spectra of Marquez, and an upper limit of about 1 percent was estimated for the branching ratio for an energy difference greater than 70 kev. For an energy difference of 120 kev, an upper limit of 0.3 percent was assigned. These limits are well below the 5-percent intensity estimated by the previous workers.

We have reinvestigated the pulse-height distribution in coincidence with the 2.2-Mev gamma ray using a ten-channel pulse-height analyzer¹² which was gated to register pulses in coincidences with pulses from a singlechannel analyzer.

The single-channel analyzer received pulses from a NaI(Tl) crystal RCA-5819 photomultiplier tube circuit and was initially adjusted to accept gamma-ray pulses between 1.9 and 2.4 Mev in height. The ten-channel analyzer was associated with another crystal photomultiplier circuit and covered the region of pulse heights between 0.4 and 1.4 Mev. The V⁴⁸ source was mounted a few millimeters above the ten-channel crystal and about 10 cm from the single-channel crystal in such a position that no straight line passing through the source could intersect both crystals. Studies of the singles

¹⁰ Sterk, Wapstra, and Kropveld, Physica **19**, 135 (1953). ¹¹ We are indebted to Dr. L. Marquez for communicating his

results in advance of publication. ¹² S. B. Burson (to be published).



FIG. 4. Typical normalized ten-channel coincidence pulse-height distributions for V^{48} with single-channel detector counting pulses from (a) the 1.32-Mev photopeak and (b) the 2.2-Mev photopeak.

counting rate as a function of the distance between the source and the single-channel crystal indicated that about 8 percent of the counts in the 1.9–2.4 Mev window were produced by "sum-line" pulses. Since these pulses arose from the combination of pulses from the 0.99- and 1.32-Mev gamma rays, they caused the appearance of some annihilation pulses in the coincidence spectrum. Corrections were made for this effect and for accidental coincidences.

The coincidence spectra were compared with "noncoincidence" spectra and with additional coincidence spectra which had been obtained with the single-channel analyzer adjusted to count pulses from the 1.32-Mev photopeak. On normalizing the latter spectra to correspond to the same number of single-channel analyzer pulses as were obtained when the single-channel analyzer was set for the 2.2-Mev region, it was found that the 0.99-Mev photopeak was the same height in both cases within a statistical uncertainty of about 15 percent. The annihilation and 1.32-Mev photopeaks were both zero within the statistical errors in the 2.2-Mev coincidence spectrum. A typical pair of normalized distributions corrected for background, accidentals, and sum lines are plotted in Fig. 4. Three pairs of this sort were obtained in the course of the experiment. From these measurements we conclude that the 2.2-Mev gamma ray is in cascade with the 0.99-Mev gamma ray with no more than 15 percent of the 2.2-Mey gamma rays in coincidence with positrons.

The conclusion of the Indiana group^{8,9} that the 2.2-Mev gamma ray is in coincidence with annihilation radiation disagrees with the present result. A calculation based on the results presented in Fig. 2 of their paper⁶ indicates that the relative probability for producing a coincidence between a pulse from the annihilation channel and a 2.2-Mev gamma ray is only 20 percent of the corresponding value for a coincidence of a pulse in the annihilation channel with a 0.99-Mev gamma ray. Since it is evident that about 20 percent of the pulses in the annihilation channel were Compton pulses from the 0.99-Mev gamma ray, this part of the coincidence data of the aforementioned workers would seem to be compatible with our conclusion that the 0.99 Mev, not the annihilation radiation, is in coincidence with the 2.2-Mev gamma ray. However the null result which they obtained for 0.99–2.2 Mev coincidences is inconsistent with our result.

VII. DECAY SCHEMES

In regard to the determination of decay schemes for Sc⁴⁸ and V⁴⁸, the following points, obtained from experiment, should be kept in mind: (1) Only one beta branch in Sc⁴⁸ and one positron branch in V⁴⁸ have been clearly established. (2) Both of these transitions are considered "allowed." (3) At least 95 percent of the disintegrations of Sc⁴⁸ are associated with a three-gamma cascade, the gamma-ray energies being 0.99, 1.04, and 1.32 Mev, with no other gamma rays (especially the 2.2-Mev gamma ray) having been detected. (4) The decay of V48 is associated primarily with a two-gamma cascade, the gamma-ray energies being 0.99 and 1.32 Mev. (5) The fraction of V⁴⁸ disintegrations which involves a threegamma cascade is 7 ± 3 percent if the cascade starts from the 3.35 Mev level and 10 ± 5 percent if the cascade starts from the 3.22-Mev level. The present experiment does not eliminate the possibility that both of these cascades may occur. (6) About 2 percent of the V^{48} decays involve gammas of energy 2.23 Mev of which at least 85 percent are in coincidence with 0.99-Mev gamma rays. (7) Angular correlation studies^{9,13} of the 0.99- and 1.32-Mev gamma rays of V48 give results suggesting the spin assignments 0^+ , 2^+ , and 4^+ for the ground and first two excited states respectively.

From the experimental evidence it is seen that the smallest number of energy levels in Ti48 which can account for the gamma rays observed in both the Sc48 and V^{48} decay is four, namely, 0.99, 2.31, 3.22, and 3.35 Mev above the Ti⁴⁸ ground state. The V⁴⁸ angular correlation work would then assign a spin of 2⁺ to the 0.99-Mev level and 4⁺ to the 2.31-Mev level. Since the Sc48 beta decay is allowed and goes to the 3.35-Mev level but not to the 2^+ or 4^+ levels, a lower limit of 6 is obtained for the Sc48 ground state and a lower limit of 5 for the 3.35-Mev level. The V⁴⁸ decay is also allowed and goes to the 4^+ level but not the 2^+ level. Hence the V⁴⁸ ground state must be 4⁺ or 5⁺. Values of 3⁺ or 4⁺ are assigned to the 3.22-Mev level since the transition occurs primarily to the 2⁺ level and not to the ground state, 0⁺. Values higher than 4⁺ may be excluded since in this case transitions to the 4⁺ level would be much more probable than those to the 2^+ level. (+) parities

¹³ P. S. Jastram and C. E. Whittle, Phys. Rev. 87, 1133 (1952).

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Upper Limits on the Neutrino Mass from the Tritium Beta Spectrum*

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The shape of the tritium beta spectrum near the end point has been investigated in a spherical electrostatic integral spectrograph with particular reference to the possible effects of a nonzero neutrino mass. It is shown that the source thickness of 100 micrograms/cm² may be satisfactorily taken into account in the last kilovolt of the spectrum, upon which the results are based. An upper limit to the neutrino mass of 500, 250, and 150 electron volts is found for the Dirac, Majorana, and Fermi forms, respectively, of the beta interaction.

INTRODUCTION

HE effect of a nonzero neutrino mass upon the shape of the beta spectrum is very marked within a distance of several neutrino rest masses from the end point of the spectrum,¹ and is hence best investigated with a spectrum of end point as small as possible. The present investigation uses tritium for this purpose. Since the original report on this work,² the results of a similar investigation by Langer and Moffat³ have appeared; their results are in essential agreement with our mass limits, although we prefer to theirs our method of taking into account the effects of spectrograph resolution (see below). References to earlier work on tritium appear in reference 3.

The spectrograph used has been reported upon elsewhere;⁴ electrons from a source at a center of hemispherical symmetry pass through a retarding field and those having more than a certain energy go to a collector thus giving an integral spectrum. The momentum resolution of 1/3 percent is an essential ingredient of the neutrino mass limits which can be set.

EFFECT OF SOURCE THICKNESS

In a spectrum of end point 18 kilovolts, source effects constitute a formidable problem which, however, diminishes if, as in the present instance, one is interested primarily in the last few kilovolts of the spectrum. It is shown in what follows that a thick source converts a differential spectrum to a simple integral spectrum near the end point, and that this process and a subsequent integration by the spectrograph do not blur the characteristic features which arise from finite neutrino mass. The source used in these measurements consisted of tritium soaked up in 100 micrograms/cm² of zirconium which in turn had been deposited on a tungsten button.⁵

We base our conclusions on the suitability of thick sources for our purpose upon two lines of evidence: the behavior of monoenergetic beams of electrons in passing through thin foils, as summarized, for example, by Klemperer;⁶ and the shape of conversion lines from sources of varying thicknesses, as observed and discussed by Owen and Primakoff.7

As to the first line of evidence: the work of Klemperer at 7-13 kilovolts⁶ and of White and Millington above 100 kilovolts^{6,8} are quantitatively consistent in indicating the existence of a "universal straggling curve" for the emergent electron spectrum when monoenergetic electrons are incident upon a foil. This universal straggling curve has several noteworthy features, as follows. The energy is peaked at a value less than the incident energy by an amount ΔV proportional to the thickness of the foil; at 18 kilovolts for Al absorber and not too badly for other substances, ΔV (kilovolts) $\approx 6 \times 10^{3} t$ (g/cm²); the width of the peak at half maximum value is nearly equal to ΔV ; the peak has a long low-energy tail, the average energy and the larger part

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 ⁴ D. R. Hamilton and L. Gross, Rev. Sci. Instr. 21, 912 (1950).

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