Search for the β^- decay of ^{48}Ca

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(Received 24 June 1985)

Compressed pellets of $^{48}CaCO_3$ containing 3.71 g of ^{48}Ca were counted for 206 days in a well-type HPGe detector enclosed in a large NaI(Tl) detector. The cascade γ rays of 983.5, 1037.5, and 1312.1 keV in the β^- decay of 44-h ^{48}Sc, which would follow the β^- decay of ^{48}Ca, were searched for in the HPGe spectrum by requiring coincidences with summed γ rays in the NaI(Tl) detector. A 1900-2560-keV window on the NaI(Tl) included all three photopeak double sums. $\gamma\gamma$ coincidence efficiencies were determined with a calibrated ^{48}Sc source produced in the ^{48}Ca(p,n) reaction at $E_p = 12$ MeV. By combining the upper limits on the intensities of the three γ rays, the half-life for ^{48}Ca(β^-)^{48}Sc was found to be $T_{1/2} > 6\times10^{18}$ yr (95% confidence). For the $0^+ \rightarrow 5^+$ unique fourth-forbidden β^- branch of ^{48}Ca to the 131-keV first-excited state of ^{48}Sc, $\log f_0 t > 24.3$. Our experimental half-life limit is a factor of 300 longer than a previous result but is still two orders of magnitude shorter than a recent theoretical estimate.

I. INTRODUCTION

For many years ^{48}Ca has been the subject of searches for double β^- decay with the aim of determining the decay half-life and whether or not the process occurs with the emission of neutrinos. As may be seen in Fig. 1, an energy of 4271 keV is available for the $2\beta^-$ decay branch of ^{48}Ca to the ^{48}Ti ground state. Direct observation of the $2\beta^-$ decay of ^{48}Ca, or any other candidate such as ^{76}Ge, has yet to be achieved.

Also of theoretical interest is the rate for highly forbidden single β^- decay. Of the various naturally occurring nuclei for which double β^- decay is energetically allowed, only ^{48}Ca and ^{96}Zr can also decay by single β^- decay. However, the single β^- decay of ^{48}Ca has not received much experimental attention, the last search being reported[1] in 1952 by Jones and Kohman. In their work a chemical milking of Sc from 9 kg of natural calcium was carried out and β rays were measured with an end-window counter in an attempt to observe 44-h ^{48}Sc activity in the separated sample. They set a lower limit of $T_{1/2} > 2\times10^{16}$ yr on ^{48}Ca β^- decay. [Note that the value $T_{1/2} > 1.1\times10^{18}$ yr for ^{48}Ca β^- decay given in the Table of Isotopes[2] was incorrectly attributed to a β^- measurement by Awschalom[3] (1956) when that was, in fact, a $2\beta^-$ measurement.]

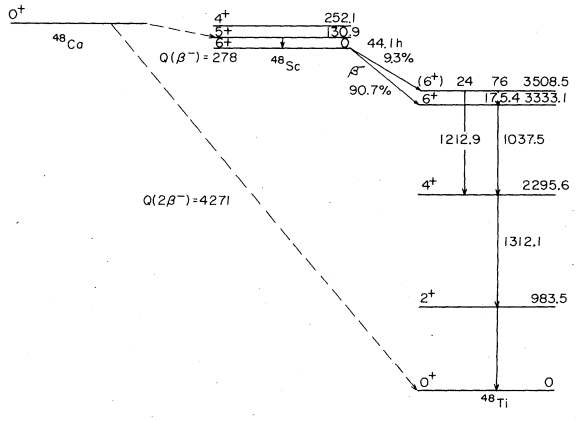

FIG. 1. Energy levels and γ-ray transitions relevant to the ^{48}Ca(β^-)^{48}Sc(β^-)^{48}Ti decay sequence. Energies are in keV.

With a decay energy[4] of $Q(\beta^-) = 278(5)$ keV there are three states in ^{48}Sc to which ^{48}Ca can decay. As shown in Fig. 1, the branch to the 6^+ ground state is nonunique sixth forbidden and would evidently have an extremely long partial half-life. While the decay to the 4^+ state at 252 keV is nonunique fourth forbidden, the decay energy of only 26 keV would make this branch very weak. This leaves the unique fourth-forbidden β^- transition to the $J^\pi = 5^+$ 131-keV level [$E_{\beta_{max}} = 147(5)$ keV] as the only significant possibility for ^{48}Ca decay.

Unique n-forbidden β^- decay was considered[5] theoretically by Warburton, Garvey, and Towner. However, at the time the present work was begun there was no reliable estimate for the particular case of ^{48}Ca unique fourth-forbidden β^- decay. When informed that our experiment was under way, Warburton[6] undertook a calculation and arrived at a half-life of 7.6×10^{20} yr (estimated uncertainty $\approx 70\%$) for the ^{48}Ca β^- branch to the $J^\pi = 5^+$ level of ^{48}Sc. As described in the next section, our experiment was designed to improve substantially on the result of Jones and Kohman, although the level of Warburton's prediction was very unlikely to be attained.

II. DESIGN OF THE EXPERIMENT

The general plan for trying to detect ^{48}Ca β^- decay was to search for the γ rays from ^{48}Sc decay in a ^{48}Ca sample under steady-state conditions. In order to achieve high counting efficiency and low background, a system was used that was recently applied to a search[7] for the decay of ^{180}Ta^m. This consisted of a well-type HPGe detector, having a volume of $\gtrsim 100$ cm^3 and an efficiency of 25.5%, enclosed in a 25.4-cm diam, 30.5-cm long NaI(Tl)

detector (see Ref. 7 for details). In the $^{180}Ta^m$ measurements the NaI(Tl) detector was operated in anticoincidence in order to reduce the cosmic-ray continuum background. But in the present work on ^{48}Ca-^{48}Sc, advantage was taken of the high γ-ray efficiency of the NaI(Tl) in a measurement of $\gamma\gamma$ coincidences.

As may be seen from Fig. 1 the dominant feature of ^{48}Sc decay is the emission of three principal cascade γ rays with energies of 983.5, 1037.5, and 1312.1 keV, and all of $>97\%$ intensity per decay. Summing of pairs of these γ's in the NaI(Tl) can be expected to occur with high probability. Since the γ energies are fairly close to each other, a single pulse-height window on the output of the NaI(Tl) can be set to include all three photopeak double sums. The plan was therefore to make a search in the HPGe γ spectrum for the ^{48}Sc γ rays in coincidence with the window on the NaI(Tl). It was anticipated that the only background contributions, under conditions when the total energy required for coincidences in the region of interest is ≈ 3 MeV, would be from the decay of naturally occurring ^{208}Tl and from cosmic rays. In ^{208}Tl decay the NaI(Tl) window includes a portion of the photopeak and Compton electron distribution due to the 2614-keV γ ray from the ^{208}Pb first-excited state, and thus one would expect to observe the various γ rays known to be in coincidence. Cosmic rays will always be able to deposit ≈ 2 MeV in the NaI(Tl) and ≈ 1 MeV in the HPGe in a continuum of coincidences.

III. SAMPLE PREPARATION AND EFFICIENCY CALIBRATION

The following two $^{48}CaCO_3$ samples were obtained on loan from Oak Ridge National Laboratory: (1) 5025 mg of $^{48}CaCO_3$, 97.27% enriched and containing 2167 mg of ^{48}Ca, and (2) 3864 mg of $^{48}CaCO_3$, 90.81% enriched and containing 1546 mg of ^{48}Ca. As verified by the tests described below, the highest γ-ray detection efficiency both in singles and coincidence was expected to be obtained for samples contained in the well of the HPGe detector. However, the delivered material was in the form of 9 g of $CaCO_3$ powder having an effective density of ≈ 1.0 g/cm^3 compared with the 3.93-cm^3 volume of the well (1.0 cm diam, 5 cm deep). In order to be able to get as much of the material as possible into the well, the $^{48}CaCO_3$ powder was compressed by using a hydraulic press and a die that produced pellets 9.14 mm in diameter. Pellets made with this equipment had a density of 2.2 g/cm^3 compared with the published compound density of 2.9 g/cm^3. Three pellets were made from the 97.3% sample having a total length of 3.43 cm and two were made from the 90.8% sample having a total length of 2.70 cm.

In order to locate the pellets in the well, yet keep the two samples physically separated, tubular containers were made from 0.05-mm thick Kapton®, a DuPont plastic. Two such tubes of lengths 3.50 and 3.0 cm, with 9.50-mm i.d., sealed and closed at one end with epoxy, were fabricated for holding the pellets of each enrichment. After installing the pellets in the tubes they were slid into a third tubular container 6 cm long and with 9.83 mm i.d., the 97.3% pellets being installed first. When the total sample was lowered into the well of the HPGe detector, about 1 cm of the topmost pellet protruded above the top of the well.

Prior to the measurements on the $^{48}CaCO_3$ sample, a ^{60}Co source was placed on top of the HPGe detector in order to match the gains of the NaI(Tl) photo-tube outputs and to determine the pulse-height gating conditions. The two ^{60}Co γ rays total 2.506 MeV, resulting in a prominent sum peak. By noting the peak channel and the upper and lower channels of a pulse-height window completely encompassing this peak, the corresponding channels were calculated for the ^{48}Sc γ-ray double sums. For the ^{48}Ca runs and ^{48}Sc calibrations, the calculated lower edge of the 983.5 + 1037.5 keV sum peak and the upper edge of the 1037.5 + 1312.1 keV sum peak were selected as 1900-2560-keV limits for gating by a single-channel analyzer. Gating conditions were checked with the ^{60}Co source halfway through the long run on ^{48}Ca and again at the end. Shifts in gain were minor and corresponded to an effect of $<10\%$ on the coincidence efficiency.

$\gamma\gamma$ coincidence efficiencies were determined under the above biasing conditions using a ^{48}Sc source. This was produced in the ^{48}Ca(p,n)^{48}Sc reaction by cyclotron bombardment of a small powder sample of $^{48}CaCO_3$ with 12-MeV protons. Runs were made on γ's in the HPGe detector both in singles and in coincidence with the NaI(Tl) for a number of positions of the ^{48}Sc source. One of the coincidence spectra is shown in Fig. 3(a). A separate well-calibrated Ge(Li) detector system was used to establish the absolute strength of the ^{48}Sc source. Yields of the various ^{48}Sc lines were then used to find the photopeak efficien-

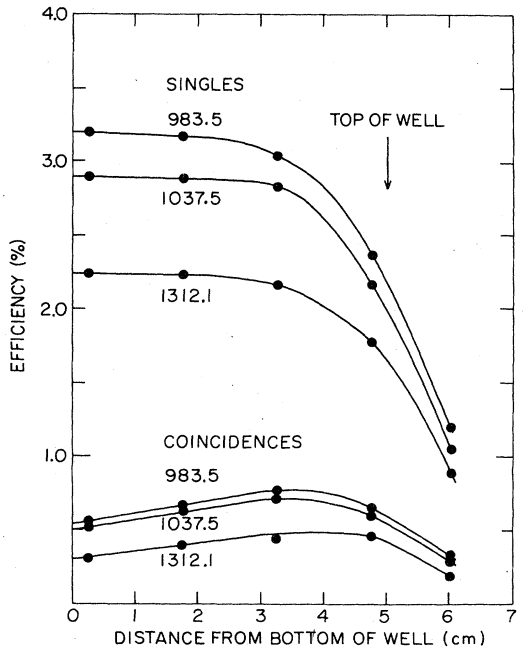

FIG. 2. Measured photopeak efficiencies (in %) for detecting the principal γ rays of ^{48}Sc vs the distance of the source from the bottom of the well in the HPGe detector: singles at the top, and coincidences with a 1900-2560-keV bias window on the output of the NaI(Tl) detector at the bottom.

cies both in singles and in coincidence. These results are shown in Fig. 2. The overall effective efficiences for the total $^{48}CaCO_3$ sample were then calculated from the averages of these curves, weighted according to the lengths and enrichments of the two separate samples.

The shapes of the efficiency curves show that the bottom of the well is not the location for the highest coincidence efficiency. As the sample is raised the singles rates are relatively constant, but there is less γ-ray absorption by the HPGe detector. This increases the summing effect in the NaI(Tl) thus raising the coincidence rate. When the source is at about 3.5 cm from the bottom of the well the coincidence rates reach maximum values and both the singles and coincidence efficiencies start decreasing.

IV. DATA ACCUMULATION AND ANALYSIS

As described in reports on two previous experiments,[7,8] a 4096-channel pulse-height analyzer was used to store the spectrum from the HPGe detector in an automatic mode such that each 24-h run was recorded on magnetic tape, the analyzer cleared, and a new run started. Since the singles counting rates in both detectors were only a few per second, it was sufficient to require coincidences by simply gating the multichannel analyzer with the 3.5-μsec wide output pulse from the single-channel analyzer that determined the window on the NaI(Tl) detector output. A ^{228}Th source was placed on top of the HPGe detector to check the gating operation and to ensure 100% coincidence efficiency.

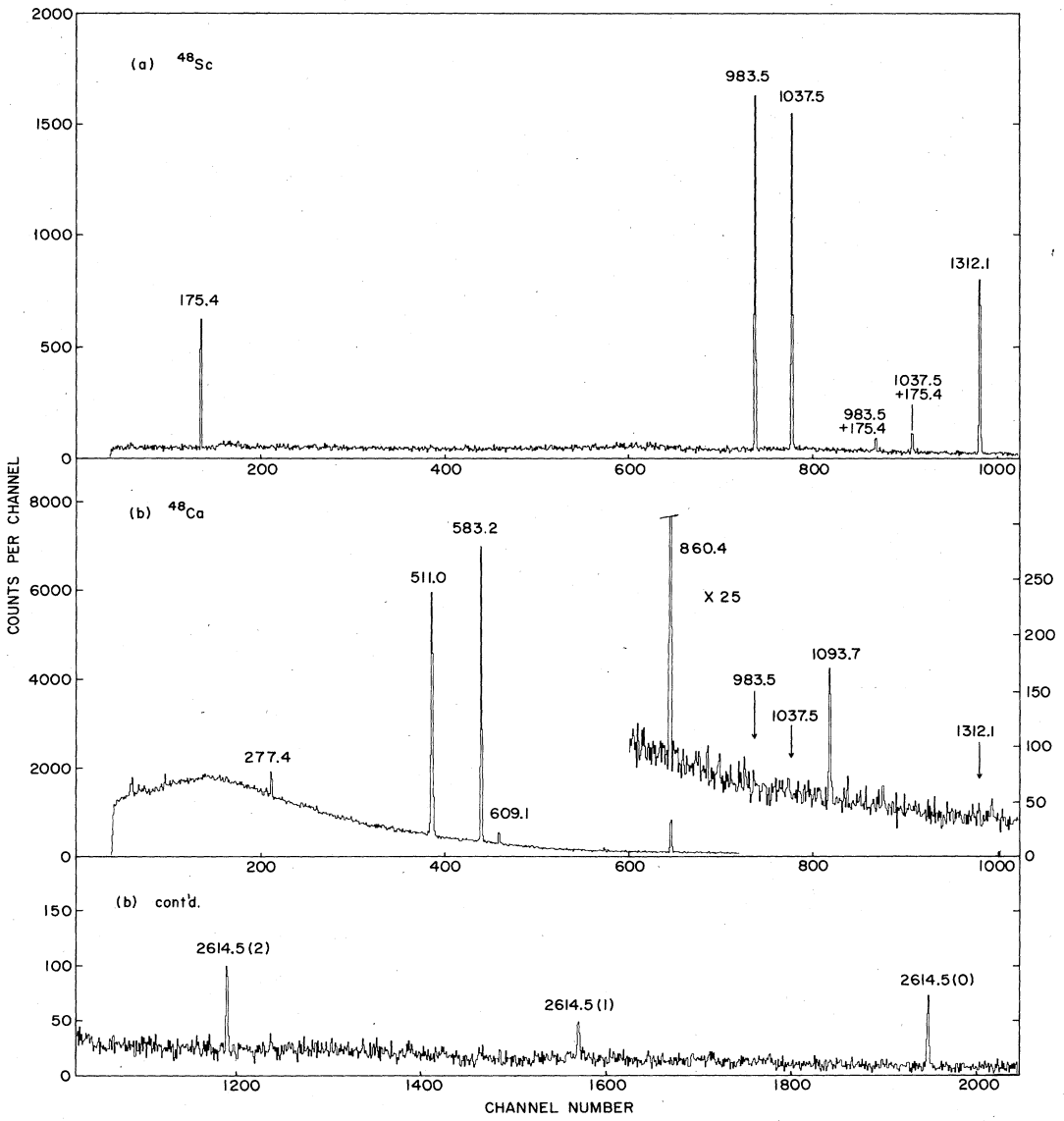

FIG. 3. (a) HPGe detector spectrum in coincidence with the NaI(Tl) detector for a ^{48}Sc source at a distance of 1.5 cm from the bottom of the well. (b) Coincidence spectrum under similar conditions as in (a) but from runs totaling 206 days with $^{48}CaCO_3$ pellets containing 3.71 g of ^{48}Ca. Data were actually recorded in 4096-channel spectra but were compressed here by a factor of 2.0 for more convenient display.

In spite of the very low coincidence rate with the $^{48}CaCO_3$ sample in place, enough counts were accumulated in the 511- and 583-keV ^{208}Tl peaks in 24 h (averages of 65 for the 511-keV peak and 52 for the 583-keV peak) to allow their positions to be determined by a computer fitting program to an accuracy of ≈ 0.2 channels. The various 24-h runs were matched by using a gain-shifting program so as to maintain good pulse-height resolution in the grand sum spectrum, as described[7,8] previously.

Figure 3(b) shows the coincidence spectrum with the $^{48}CaCO_3$ sample accumulated for a total of 206 days. All of the peaks can be attributed to ^{208}Tl decay,[2] but some of the lines, such as that at 1093.7 keV, show an enhancement because of summing effects. The 2614-keV full-energy-loss and one-escape peaks are probably mostly due to random coincidences, but the two-escape peak, relative to the full-energy peak, is much more intense than in singles. It therefore must be a real coincidence effect due to summing of the two associated annihilation quanta with cascade γ rays. Thus $2\times 511+1093.7$ keV falls within the window on the NaI(Tl) detector. There are no contaminant peaks expected in the ^{48}Ca spectrum near the positions of the ^{48}Sc 983.5, 1037.5, and 1312.1-keV γ rays [^{48}Sc spectrum, curve 3(a) and arrows in curve 3(b)]. Upper limits on their intensities were derived by standard statistical analysis. By combining these limits with the average efficiencies, the length of the run, and the number of ^{48}Ca atoms, half-life limits (95% confidence) were determined. Table I presents the resulting limits based on the γ-ray searches, and includes the effective γ-ray coincidence efficiencies derived from the data in Fig. 2.

By combining the half-life limits from Table I, the final value $T_{1/2} > 6\times 10^{18}$ yr is obtained for ^{48}Ca β^- decay. Corresponding to this limit is the limit $\log f_0 t > 24.3$ for the $0^+ \rightarrow 5^+$ transition, assuming that this is the only significant β^- branch.

Runs of 14 days each were made on the γ-ray singles spectrum from the HPGe detector, first with the $^{48}CaCO_3$ sample in place and then removed, but with the plastic tubular containers replaced in the well. A comparison of these spectra showed that the following weak contaminant radioactivities were present in the $^{48}CaCO_3$ samples: ^{235}U, ^{133}Ba, ^{152}Eu, and ^{146}Pm. All lines due to members of the ^{232}Th-^{208}Pb and ^{238}U-^{206}Pb decay chains, and peaks due to ^{40}K, ^{60}Co, and ^{137}Cs had the same intensities in both spectra. Based on an efficiency function determined[7] with a ^{176}Lu source the following approximate strengths were derived: ^{133}Ba, 1.8×10^{-5} μCi; ^{152}Eu, 1.0×10^{-6} μCi; and ^{146}Pm, 3.5×10^{-7} μCi. The ^{235}U lines in the ^{48}Ca spectrum were ≈ 14 times stronger than in the spectrum with the ^{48}Ca sample removed. From a comparison of the continuum levels in the regions of the three ^{48}Sc lines in singles and coincidence [Fig. 3(b)] it was shown that, in spite of the higher γ singles efficiencies (Fig. 2), the $T_{1/2}$ lower limit derived from the coincidence experiment is greater than could have been obtained from a singles run of the same duration by a factor of about 3.

TABLE I. ^{48}Sc $\gamma\gamma$ photopeak coincidence efficiencies and limits on $T_{1/2}$ for ^{48}Ca(β^-)^{48}Sc.

E_γ (keV)	$\gamma\gamma$ efficiency (%)	$T_{1/2}$ (yr)
983.5	0.648	$>3.8\times 10^{18}$
1037.5	0.598	$>3.6\times 10^{18}$
1312.1	0.402	$>3.0\times 10^{18}$

V. DISCUSSION

Our result of $T_{1/2} > 6\times 10^{18}$ yr for the half-life of ^{48}Ca(β^-)^{48}Sc decay exceeds the previous limit[1] of $T_{1/2} > 2\times 10^{16}$ yr by a factor of 300 but is still two orders of magnitude shorter than the theoretical estimate[6] of $T_{1/2} = 7.6\times 10^{20}$ yr. For a sample such as the one used here containing 3.71 g of ^{48}Ca, the disintegration rate corresponding to the predicted half-life is only 0.116 per day or 42 per yr. With our experiment arrangement a substantial improvement could be achieved by moving to a deep underground location, since the continuum background in Fig. 3(b) is clearly due almost entirely to cosmic rays. A larger quantity of $^{48}CaCO_3$ would not help very much in our setup since the $\gamma\gamma$ coincidence efficiency for material outside the well is relatively low (see Fig. 2). Note that the singles photopeak efficiency[8] for a very large sample surrounding this same detector was only $\approx 0.7\%$ at $E_\gamma = 1461$ keV. In order to push the $T_{1/2}$ limit on ^{48}Ca(β^-) substantially further, a combination of higher efficiency, lower background, and larger sample size will probably be needed.

ACKNOWLEDGMENTS

We would like to thank J. Hurst for the loan of the pellet-making equipment and W. B. Jones for producing the ^{48}Sc test source in the BNL 60-inch cyclotron. This research was supported by the U.S. Department of Energy, Division of Basic Energy Sciences, under Contract No. DE-AC02-76CH00016.

[1] J. W. Jones and T. P. Kohman, Phys. Rev. **85**, 941 (1952).
[2] *Table of Isotopes*, 7th ed., edited by C. M. Lederer and V. S. Shirley (Wiley, New York, 1978).
[3] M. Awschalom, Phys. Rev. **101**, 1041 (1956).
[4] A. H. Wapstra and G. Audi, Nucl. Phys. **A342**, 1 (1985).
[5] E. K. Warburton, G. T. Garvey, and I. S. Towner, Ann. Phys. (N.Y.) **57**, 174 (1970).
[6] E. K. Warburton, Phys. Rev. C **31**, 1896 (1985).
[7] J. B. Cumming and D. E. Alburger, Phys. Rev. C **31**, 1494 (1985).
[8] D. E. Alburger, E. K. Warburton, and J. B. Cumming, Phys. Rev. C **29**, 2294 (1984).