

It may be noted from that the vacuum polarization may be considered to have a range near $\lambda/2\pi$, or 4×10^{-11} cm. This is much larger than the nuclear radius, but considerably smaller than the average radius of a $2p$ electron orbit.

Although the expression (27) for V_p is accurate only for $\alpha Z \ll 1$, it was used to obtain a rough magnitude for the effect of vacuum polarization on the $2p_{3/2}$ - $2p_{1/2}$ fine structure. The results showed that vacuum polarization gives an effect on the fine structure which varies with Z much like the effect of the finite nuclear radius, and about 40% as large as that expected from a nuclear radius of $1.5 \times 10^{-13} A^{1/3}$. Since the potential is attractive, however, it is in the wrong direction to account for the anomalously large effect of the "finite nuclear radius"

observed above. Wichmann and Kroll¹⁴ have recently succeeded in making a much more complete calculation of vacuum polarization without requiring that $\alpha Z \ll 1$. They find an effect on the x-ray fine structure which is not very different from that given here.

Since vacuum polarization gives the equivalent of an attractive potential, its effect is to give too small an effective radius for the nucleus rather than the observed large radius. The Lamb shift, on the other hand, does correspond to a repulsion and hence gives an effect of the correct sign. Its calculation for the case when αZ is not very small is difficult and has not yet been done. When it is accomplished, the observed fine structure anomaly may afford a good method of testing these electrodynamic corrections for large Z .

¹⁴ E. Wichmann and N. M. Kroll, Phys. Rev. (to be published).

Radioactive Ca⁴⁷†

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Ca⁴⁷ (4.5 day) has been investigated by using beta-gamma coincidence measurements and NaI gamma-ray spectrometry. Three gamma rays of energies 1.29 Mev, 0.812 Mev, and 0.500 Mev are present with absolute intensities of (71±6)%, (5±0.5)%, and (5±0.5)%, respectively. These gammas are in coincidence with a 0.70±0.02 Mev beta group. In addition, (24±6)% of the decay is through a 1.9±0.2 Mev beta group directly to the ground state.

INTRODUCTION

CALCIUM-47 has been the subject of several recent investigations; the results of these studies, however, indicate considerable uncertainty exists concerning the decay characteristics of this nuclide. Cork *et al.*¹ prepared Ca⁴⁷ by an (n,γ) reaction on enriched (9.6%) Ca⁴⁶. Marquez² prepared Ca⁴⁷ by irradiating K₂Cr₂O₇ in the 450-Mev synchrocyclotron at the University of Chicago.

Aten, Grevell, and Van Dijk³ prepared Ca⁴⁷ by the reaction Ca⁴⁸(d,dn)Ca⁴⁷ and Ti⁵⁰($d,\alpha p$)Ca⁴⁷ using 26-Mev deuterons. Table I is a summary of data obtained by these workers.

From Table I, it is apparent that some serious discrepancies appear in the beta energies and that the gamma spectrum is in some doubt. Because of the possible medical and biological interest in Ca⁴⁷, it seemed worthwhile to attempt clarification of the decay scheme.

† Work performed under contract to the U. S. Atomic Energy Commission.

¹ Cork, LeBlanc, Brice, and Nester, Phys. Rev. **92**, 367 (1953).

² Luis Marquez, Phys. Rev. **92**, 1511 (1953).

³ Aten, Grevell, and Van Dijk, Physica **19**, 1049 (1953).

PREPARATION AND PURIFICATION OF THE SAMPLE

Calcium-47 was prepared in two different ways:

(a) In the 86-inch cyclotron at Oak Ridge National Laboratory (ORNL) by irradiation of CaO with ~14-Mev protons:



(b) By irradiation with thermal neutrons on enriched (9.6%) Ca⁴⁶ in the low-intensity test reactor at ORNL: Ca⁴⁶(n,γ)Ca⁴⁷.

Gamma ray and decay data obtained from Ca⁴⁷ prepared in each manner were in agreement. Because of the larger amounts of calcium in the proton-irradiated material, 4π beta counting was not tried with this source. Chemical processing of the sample was the same for both methods of preparation and is described below.

The calcium oxide was dissolved in dilute nitric acid and the pH adjusted to ~2.0. The daughter Sc⁴⁷ activity, together with other Sc activities was removed by successive extractions with 0.5M thenoyltrifluoroacetone (TTA) in xylene.⁴

To the aqueous solution, iron (III) carrier was added

⁴ W. S. Lyon and B. Kahn, Phys. Rev. **99**, 728 (1955).

TABLE I. Decay characteristics of Ca⁴⁷.

| | Cork <i>et al.</i> ^a | Marquez ^b | Aten <i>et al.</i> ^c |
|----------------------------|--|-----------------------------------|---|
| Beta energy and intensity | 0.46 Mev 1.4 Mev | 0.685 Mev (81%) 2.06 Mev (19%) | 0.8±0.2 Mev 2.0 Mev |
| Gamma energy and intensity | 1.30, 0.800, 0.495, 0.234, 0.150 Mev | | 1.3±0.2 Mev γ (3.4 times intensity of 2.0-Mev beta group) |
| Half-life | 5.35±0.1 day | 4.3 day | |

^a See reference 1.
^b See reference 2.
^c See reference 3.

and a series of Fe(OH)₃ precipitation scavengings were performed by making the solution basic. Finally the calcium was precipitated as calcium oxalate by addition of ammonium oxalate to the solution at pH~10. The calcium oxalate was filtered, washed, ignited to calcium oxide in a furnace at 800°C and the CaO dissolved in dilute nitric acid and made to volume. Aliquots were then taken from this solution for counting. The time elapsed between final separation of the Ca⁴⁷ from the 3.5-day Sc⁴⁷ daughter and counting was between one and two hours.

Aliquots of the Ca⁴⁷ solution were used for decay measurements, 4π-coincidence counting, aluminum beta absorption counting, NaI gamma-ray spectrometry, and beta-gamma coincidence measurements using an external beta counter.

EXPERIMENTAL RESULTS

Examination of the gamma-ray spectrum (Fig. 1) of the Ca⁴⁷ using a 3 in.×3 in. NaI(Tl) gamma-ray spec-

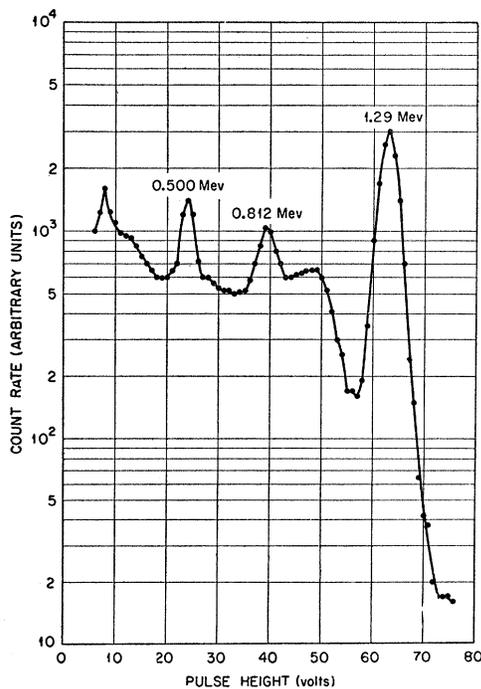


FIG. 1. Gamma-ray spectrum of Ca⁴⁷.

trometer indicates the presence of three gamma rays associated with Ca⁴⁷. By comparison of the pulse-height voltage of these peaks with the pulse-height voltages of known gamma-ray energy standards the gamma-ray energies have been found to be 1.29 Mev, 0.812 Mev, and 0.500 Mev. The small peak at 160 kev is associated with the 3.5-day Sc⁴⁷ daughter. The absolute intensities of the three Ca⁴⁷ gamma rays were measured by integrating beneath a linear pulse height vs count rate plot of the gamma-ray spectrum and dividing by known efficiency factors for the three gamma-ray energies. These efficiency factors have been obtained in a manner similar to that described by Kahn and Lyon.⁵

The absolute disintegration rate of the Ca⁴⁷ solution was obtained by 4π-coincidence counting. Here an aliquot of the Ca⁴⁷ was evaporated to dryness on a thin (25 μg/cm²) plastic film 2.3 cm in diameter and coated with 25 μg/cm² Al to make it conducting. This film, supported by an aluminum holder, was placed in a 4π counter, and the counter was evacuated, filled to one-half atmosphere with methane, and used as the beta detector. The 3 in.×3 in. NaI gamma spectrometer was used as the gamma detector. The pulse-height selector of the spectrometer was set so as to accept only counts from the 1.29-Mev gamma-ray photopeak. The disintegration rate of the Ca⁴⁷ source was then found as:

$$N_0 = C_\beta \times C_\gamma / C_{\beta\gamma} \tag{1}$$

where C_β=observed beta counts per minute, C_γ=observed gamma counts per minute, C_{βγ}=observed beta-

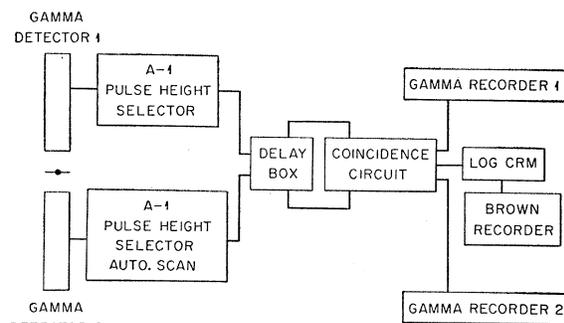


FIG. 2. Block diagram of γ-γ coincidence spectrometer assembly.

⁵ B. Kahn and W. S. Lyon, Nucleonics 12, 26 (1953).

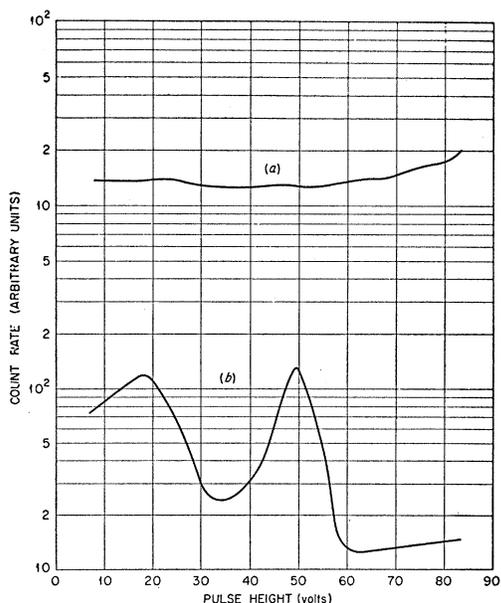


FIG. 3. Ca^{47} Gamma-gamma coincidence data: (a) γ - γ coincidence spectrum of 1.29-Mev γ ray. (b) γ - γ coincidence spectrum of 0.812-Mev γ ray.

gamma coincidence counts per minute after correction for background and random coincidence counts was made, and N_0 = number of disintegrations per minute of Ca^{47} .

In addition, the disintegration rate of the source was determined by setting the pulse-height selector so as to accept gamma pulses only from the 0.812-Mev γ -ray photopeak and then from the 0.500-Mev gamma ray photopeak. The values obtained agreed with each other.

The fraction of decay proceeding through each gamma ray was then found by dividing the absolute intensities of the gamma rays by the absolute disintegration rate. These data indicate the decay to proceed $(71 \pm 6)\%$ through the 1.29-Mev gamma ray and $(5 \pm 0.5)\%$ through the 0.812-Mev and 0.500-Mev gamma ray. Approximately 24% of the total beta disintegrations are unaccompanied by gamma radiation.

Through the kindness of P. R. Bell and N. H. Lazar, several sources of Ca^{47} were examined by using a calibrated 3 in. \times 3 in. NaI(Tl) crystal and a 40-channel pulse analyzer. The results are in agreement with those given above.

As a further check on the absolute disintegration rate of the Ca^{47} 4π source, the source was removed from the 4π chamber and after several weeks, the gamma-ray spectrum was again obtained using the spectrometer. The amount of Sc^{47} 0.160-Mev gamma ray present was then obtained by graphical integration. With knowledge of the efficiency of the spectrometer for this gamma ray, the branching ratio of Sc^{47} , and the half-life of Sc^{47} ,⁴ together with the half-life of Ca^{47} , the amount of Ca^{47} parent can be calculated. This calculation was made for several different preparations and was found to agree

with the 4π -coincidence value within experimental error.

Decay of the purified Ca^{47} was followed on a well-type NaI scintillation counter equipped with an ~ 5 g/cm² Pb absorber so as to absorb essentially all of the 160-keV gamma rays from the daughter Sc^{47} activity. Decay followed over 5 half-lives indicates the Ca^{47} half-life to be 4.5 days.

Because of the equal intensities of the 0.812-Mev gamma and the 0.500-Mev gamma, it appeared reasonable that they were in coincidence, and that the 1.29-Mev gamma ray was a cross-over gamma ray. To establish this, gamma-gamma coincidence data were obtained. The assembly used is shown schematically in Fig. 2. Gamma detector number 1 was a 3 in. \times 3 in. NaI(Tl) crystal, and gamma detector number 2 was a 1 in. \times 1½ in. NaI(Tl) crystal. When the source is placed between the two detectors, pulses from each crystal go through $A-1$ amplifiers and the single-channel analyzers and thence to a variable delay box where the delay in the two circuits is matched. Events occurring within the resolving time of the instrument and recorded as coincident are fed into a log count rate meter and thence to a Brown recorder. The pulse-height selector in the number 1 circuit was first set on the 1.29-Mev γ peak and the slit width opened to its full value of 10 volts. By use of the automatic-scan pulse-height selector in the number 2 circuit, the coincidence counting rate as a function of pulse height (using a 5-

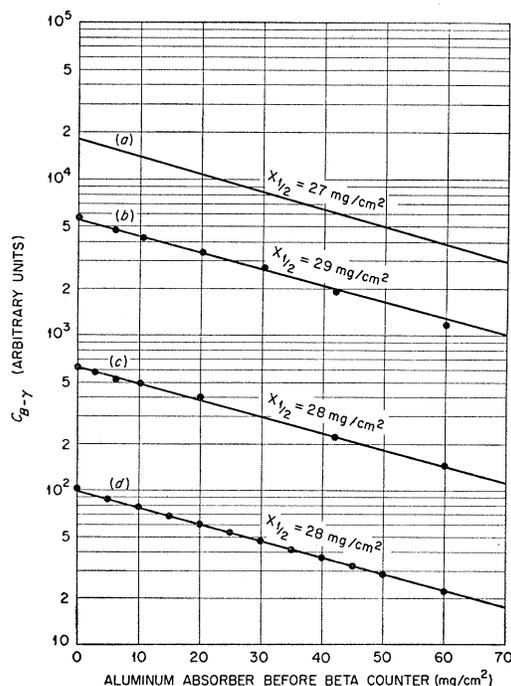


FIG. 4. Beta-gamma coincidence rate as a function of Al absorber placed before the beta counter. (a) Ca^{47} β -1.29-Mev γ coinc. (b) Ca^{47} β -0.81-Mev γ coinc. (c) Ca^{47} β -0.50-Mev γ coinc. (d) Rb^{86} 0.68-Mev β -1.08-Mev γ coinc.

volt slit) in the number 2 circuit was observed. The speed of the Brown recorder was synchronized to that of the variable-scan pulse-height selector in circuit 2.

Figure 3 is a replot of data obtained by using this assembly. The upper curve is a plot of coincidence counting rate *vs* pulse height in circuit 2 when the invariant pulse height in circuit 1 was set on the 1.29-Mev photopeak. It is evident that there are no coincidences between the 1.29-Mev γ and the other two Ca⁴⁷ gamma rays. The lower curve is a replot of data obtained when the invariant pulse-height selector was set on the 0.812-Mev photopeak. Here it is apparent that the 0.500-Mev gamma ray is in coincidence with the 0.812-Mev gamma ray as evidenced by the peak in coincidence counting rate at about 0.500 Mev. In addition, coincidences between the 0.812-Mev photopeak and the 0.500-Mev Compton distribution are seen. Thus, the two lower-energy gammas are shown to be coincident.

By replacing the NaI crystal and amplifier and pulse-height selector in Fig. 2 with an end-window beta proportional counter and scaler, and by feeding the coincidence counts into a mechanical recorder, it is possible to obtain beta-gamma coincidence data. Here the gamma detector is set on the photopeak of a particular gamma ray and the coincidence counting rate as a function of aluminum absorber placed before the beta detector is obtained.

Aluminum absorption data were obtained with the gamma detector set first at 1.29 Mev, then at 0.812, and finally at 0.500 Mev. Figure 4 is a replot of these data; here the coincidence rate is plotted as a function of aluminum absorber. The similarity in slopes suggests that the low-energy gamma rays and the 1.29-Mev gamma ray are in coincidence with the same beta group.

By comparison of the slope of these curves with slopes of known energy beta-gamma coincidence curves, the maximum energy of this beta group may be set.

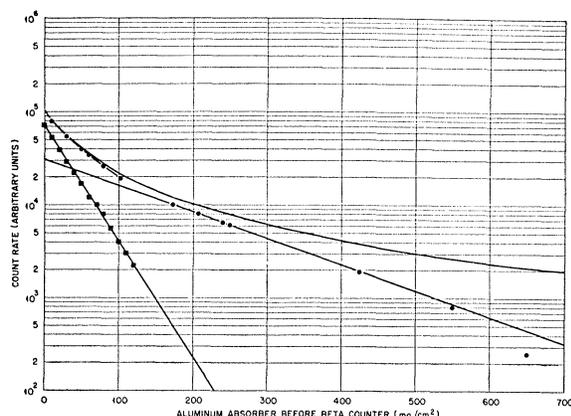


FIG. 5. Aluminum absorption data of Ca⁴⁷ count rate as a function of absorber thickness.

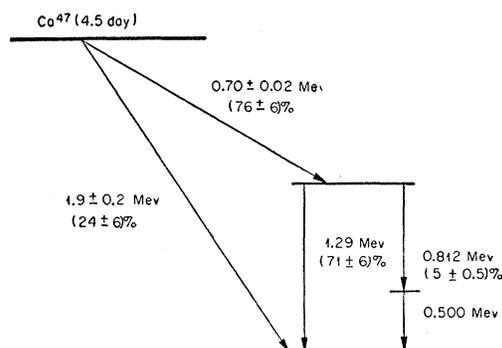


FIG. 6. Decay scheme of Ca⁴⁷.

The 0.68-Mev beta-gamma coincidence spectrum of Rb⁸⁶ is also shown in Fig. 3. It seems apparent that the beta energy of the lower energy Ca⁴⁷ is close to that of Rb⁸⁶, and we have assigned a value of 0.70 ± 0.02 Mev to this group. The beta counting aluminum absorption data indicate the presence of a more energetic beta group (Fig. 5). Resolution of the beta absorption curve indicates the more energetic beta group comprises about 24% of the total number of beta disintegrations. This is in agreement with the fraction of beta decay unaccompanied by any gamma. A search was made for a lower-energy gamma ray corresponding to those previously reported.¹ None was found. In addition, attempts were made to find coincidences between the hard-beta group and any low-energy gamma ray; no statistically significant coincidences were obtained. Thus, it appears that this beta group goes directly to the ground state. The maximum beta energy of the more energetic beta-ray group was found by plotting and analyzing the aluminum absorption data using the method of Harley and Hallden,⁶ and the value obtained was $1.93 \text{ Mev} \pm 0.2 \text{ Mev}$.

CONCLUSIONS AND ACKNOWLEDGMENTS

Based on the experimental findings reported above, a decay scheme (Fig. 6) can be drawn which appears consistent with the data. These results are in fair agreement with those of reference 3. The sequence of the 0.8 and 0.5 γ rays is presently unknown.

Note added in proof.—The $\log ft$ values for the 1.9-Mev and 0.70-Mev beta transitions as calculated by the method of Moszkowski [Phys. Rev. **82**, 35 (1951)] are 8.2 and 6.0, respectively. These are much too high to be in agreement with spin and parity assignments predicted by the single particle model. At present, no explanation for this discrepancy can be offered.

The authors are indebted to Farno Green who originally suggested the investigation and supervised the cyclotron irradiations and to P. R. Bell and N. H. Lazar for several helpful discussions and suggestions.

⁶ J. H. Harley and N. Hallden, Nucleonics **13**, No. 1, 32 (1955).