Decay Chain Ca^{49}-Sc^{49}-Ti^{49}

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The decays of Ca^{49} and its daughter have been studied by beta- and gamma-ray scintillation spectroscopy. Ca^{49} was observed to decay with a half-life of 8.75±0.20 minutes, exciting gamma-ray transitions in Sc^{49} of 3.10±0.03, 4.05±0.05, and 4.68±0.05 Mev energy and relative intensities of 1.0, 0.11±.02, and 0.0038 ±0.0010. Two beta-ray groups were resolved from the Ca^{49} spectrum of energy 1.95±0.05 and 0.89±0.15 Mev. In agreement with previous work, Sc^{49} was found to be a pure beta emitter. The beta-ray energy of Sc^{49} was measured as 2.05±0.05 Mev, and the half-life was found to be 57.2±0.7 minutes. Spin assignments are proposed on the basis of the single-particle model.

INTRODUCTION

RADIOACTIVE decay processes exciting nuclei having one nucleon more or less than a closed shell are of interest since the excited states may be studied from the point of view of the single-particle model, and may be compared with similar situations in neighboring nuclei. Such a case is the chain Ca^{49}-Sc^{49}-Ti^{49}. Here Ca^{49}, with its closed proton shell and odd neutron, excites energy levels in Sc^{49}, where the neutron shell is closed and a single odd proton past a closed shell is available for excitation.

Early work by Walke[1] assigned two isomers to Ca^{49} of half-life 30 minutes and 2.5 hours. Sc^{49} was found to have a half-life of 57±2 minutes and a beta-particle energy of 1.8 Mev. Irradiating enriched Ca^{48} with slow neutrons, der Mateosian and Goldhaber[2] assigned an 8.5-minute period to Ca^{49} and demonstrated that the Sc^{49} of about one-hour half-life was the radioactive daughter of the Ca^{49}. Aluminum absorption of the beta-rays indicated approximate energies of 2.7 and 2.4 Mev for Ca^{49} and Sc^{49}, respectively. Although these experiments did not disclose any isomer of Ca^{49}, hard gamma-rays were seen in about equal intensity to that of the beta-particles. Recent measurements of Sc^{49} by Koester[3] confirm Walke's half-life of 57 minutes, and indicate a beta-particle energy of 2.0 Mev. A limit to the gamma radiation was given by Koester as $E_\gamma/\beta<0.05$ Mev.

For the investigation reported here, scintillation spectroscopy of both beta and gamma radiation was performed, and decay schemes for both Ca^{49} and Sc^{49} were evolved.

EXPERIMENTAL METHODS

Source Preparation and Purity

The Ca^{49} activity was prepared by slow neutron irradiation of either analytical reagent grade $Ca(NO_3)_2$ or spectroscopically pure $CaCO_3$ enriched in Ca^{48} to 51.4 percent. Before irradiation, the target materials were handled with great care to prevent the introduction of any foreign substances, since no chemical purification of the Ca^{49} was made after bombardment. Irradiations were performed in the rapid transfer pneumatic tube of the ORNL Graphite Reactor for 8.5 minutes. After irradiation, samples were usually inserted in the appropriate scintillation spectrometer within 2 to 4 minutes after removal from the reactor. All single-crystal beta- and gamma-ray spectroscopy measurements were made using the enriched $CaCO_3$ target material. Several spectra were run during a period of three to five half-lives for each gamma-ray sample, and it was established that all portions of the spectra decayed with the same half-life.

Since the Ca^{49} rapidly decays into Sc^{49}, beta decay curves of Ca^{49} are complex and the half-life of Ca^{49} is difficult to measure with precision by this method. A half-life measurement was therefore made using a single-channel analyzer set on the 3.1-Mev gamma-ray peak of Ca^{49}, which yielded a half-life of 8.75±0.20 minutes over 8 half-lives.

Samples of Sc^{49} were chemically separated from Ca^{49} sources prepared by irradiating several grams of analytical reagent $Ca(NO_3)_2$ in the ORNL Graphite Reactor for 10 minutes. The $Ca(NO_3)_2$ was then dissolved in 0.01*M* HCl containing ~1 mg Sc^{+++} carrier, and stirred for another 10 minutes with 0.1*M* thenoyltrifluoroacetone (TTA) in xylene. After washing the organic phase twice with water, the Sc was back-extracted into 1.0*M* HCl, and finally precipitated as the hydroxide. The measurements described below were performed on small portions of the $Sc(OH)_3$ precipitate.

The half-life of Sc^{49} beta particles was followed for 11 half-lives on a 2π, flow-type proportional counter, and a value of 57.2±0.7 minutes was obtained from a least squares analysis of the data.

Gamma-Ray Spectroscopy

All gamma-ray studies used scintillation spectrometers composed of 3-inch high, 3-inch diameter, thallium-activated sodium iodide crystals attached to DuMont type 6363 photomultiplier tubes in the manner described by Lazar and Klema.[4] Sources were placed on

[1] H. Walke, Phys. Rev. **51**, 439 (1937); **52**, 777 (1937); Walke, Thompson, and Holt, Phys. Rev. **57**, 163 (1940); **57**, 171 (1940).
[2] E. der Mateosian and M. Goldhaber, Phys. Rev. **79**, 192 (1950).
[3] L. Koester, Z. Naturforsch. **9A**, 104 (1954).
[4] N. H. Lazar and E. D. Klema, Phys. Rev. **98**, 710 (1955).

the axis of the crystal, 9.3 cm from the top surface of the crystal. Polystyrene absorbers 1.3 g/cm^2 were placed between the source and crystal to remove the beta rays. A twenty-channel analyzer designed by Bell, Kelley, and Goss[5] was used for pulse-height analysis. The spectrometer was calibrated with gamma rays from Na^{24} (2.76 Mev), Y^{88} (0.908 and 1.85 Mev), and ThC″ (2.62 Mev) sources.

The low-energy portion of a typical gamma-ray spectrum from Ca^{49} is shown in Fig. 1. The peak at 0.51 Mev is attributed to pair production in the lead shield surrounding the scintillation detector by the energetic gamma rays. An approximate calculation showed the intensity of this peak could be satisfactorily explained in this manner. The full energy peak and the middle and lower pair peaks for a gamma ray at 3.10 Mev are clearly shown. Several experiments under conditions similar to those of Fig. 1 failed to disclose any further peaks which were reproducible enough for interpretation, particularly in view of the rapid buildup of bremsstrahlung produced in the absorber by the 2.05-Mev beta ray from the decay of the daughter, Sc^{49}. Any gamma radiation in the energy range of 0.7 to 1.9 Mev must have an intensity less than five percent that of the 3.10-Mev gamma ray or it could have been seen in these measurements.

The high-energy gamma-ray spectrum of Ca^{49} is displayed in Fig. 2, showing full energy peaks at 4.05 and 4.68 Mev in addition to the 3.10 Mev peak already shown in Fig. 1. By using as a guide the shape of the spectrum from the 4.44-Mev gamma ray in the reaction $N^{15}(p,\alpha\gamma)C^{12}$ and the spectrum from the 2.76-Mev transition in Na^{24}, the complex spectrum of Fig. 2 was analyzed as shown. Relative gamma-ray intensities were then calculated from the full-energy peak areas and from the intrinsic peak efficiences determined experimentally using high-energy γ rays in certain nuclear reactions. Table I summarizes the data on the Ca^{49} gamma rays, including the intrinsic peak efficiencies used.

A careful measurement of the number of beta-ray transitions per gamma ray was undertaken to check the hypothesis that ground-state beta transitions in Ca^{49} are forbidden and are therefore present in very low intensity. A few tenths of a milligram of enriched $Ca^{48}CO_3$ was irradiated in the ORNL Graphite Reactor for one minute. As soon as possible after the irradiation, the sample was placed on a Formvar-polystyrene film about 25 micrograms/cm^2 thick, and covered with a similar film. With this source placed 9.1 cm above a 3-inch high, 3-inch diameter sodium iodide crystal spectrometer, a measurement of the 3.10-Mev peak was made at a carefully determined time. The source was then transferred to a 4π beta counter[6] and the decay of Ca^{49} and the growth-decay of Sc^{49} was followed in the 4π counter for about 105 minutes. The Sc^{49} counting rates at six times between 70 and 105 minutes after the end of the bombardment were analytically extrapolated to the mid-time of the bombardment as zero time, using

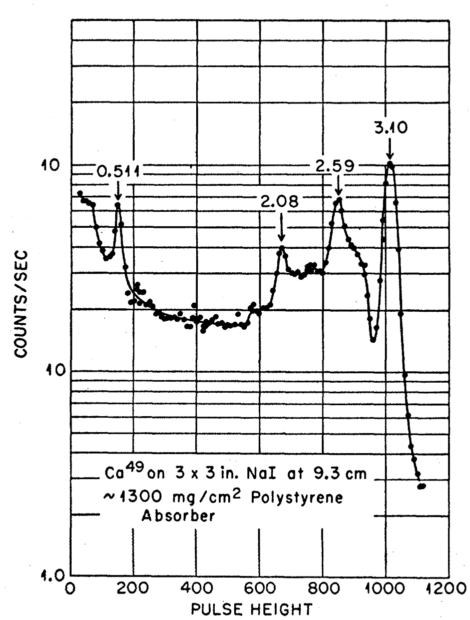

FIG. 1. Gamma-ray spectrum from Ca^{49} on 3-inch diameter, 3-inch high NaI(Tl) crystal to about 3.4 Mev.

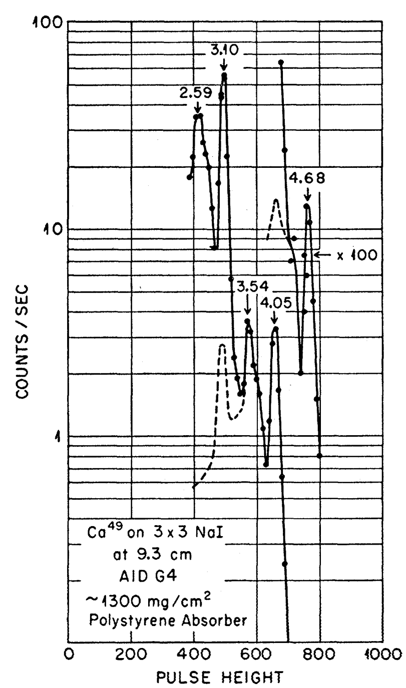

FIG. 2. Gamma-ray spectrum from Ca^{49} on 3-inch diameter, 3-inch high NaI(Tl) crystal above 2.5 Mev.

[5] Bell, Kelley, and Goss, Oak Ridge National Laboratory Report ORNL-1278, 1951 (unpublished).

[6] The authors are indebted to A. R. Brosi for the use of his 4π counting equipment.

TABLE I. Energies and intensities of Ca^{49} gamma rays.

Gamma-ray energy, Mev	Intensity relative to 3.10-Mev gamma ray	Intrinsic peak efficiency used
3.10 ± 0.03	1.00	0.076
4.05 ± 0.05	0.11 ± 0.02	0.048
4.68 ± 0.05	0.0038 ± 0.0010	0.041

the 57.2-minute half-life reported above. The six values of the Sc^{49} rates at zero time agreed with their mean value to 1 percent. From the ratio of the Sc^{49} and Ca^{49} half-lives and an analytical decay correction for the time between zero and the gamma-ray analysis, the beta disintegration rate for the Ca^{49} sample was calculated. The absolute number of 3.10-Mev gamma rays was obtained from the area of the full-energy gamma-ray peak corrected for the Compton and pair distribution from the 4.05-Mev gamma ray, applying the usual exponential correction for absorption in the 2 g/cm^2 of polystyrene absorber placed between source and detector, and dividing by the product of geometry and intrinsic peak efficiency. From this measurement $N_\beta/N_\gamma=0.95\pm0.10$, where the quoted error reflects primarily the authors' uncertainty in the sodium iodide peak efficiency for this high-energy gamma ray. It is concluded that all beta-ray transitions occur to excited states, within the experimental error.

Gamma-gamma coincidence spectroscopy measurements on Ca^{49} showed that neither the 4.05- nor 4.68-Mev gamma rays were in coincidence with the 3.10-Mev gamma ray, and that any gamma-ray transitions between the 4.05- and 3.10-Mev states were present in less than two percent of the intensity of the 3.10-Mev transition.

A search was made for gamma radiation from Sc^{49}. A strong source of Sc^{49} was located 9.3 cm above a 3-inch×3-inch sodium iodide spectrometer. Directly beneath the source was placed a polystyrene absorber 2 g/cm^2 thick. No peaks were seen, only the typical bremsstrahlung distribution with the approximately exponential decrease in counting rate with pulse height. The data from this experiment were used to set a limit on the number of 1.35-Mev gamma-ray transitions from the known first excited state[7,8] of Ti^{49}. For this calculation it was assumed that a peak whose area was about 1.5 percent of the area of the bremsstrahlung spectrum would have been easily discerned. Using an intrinsic peak efficiency of 0.174 for the 1.35-Mev gamma ray and 1.0 for the bremsstrahlung, the ratio of gamma ray to bremsstrahlung intensity becomes about 0.1. If the further assumption is made that the ratio of bremsstrahlung photons to beta disintegrations in this geometry is approximately 5×10^{-3}, then the maximum number of 1.35-Mev gamma rays per beta disintegration is estimated to be about 5×10^{-4}.

[7] Bretscher, Alderman, Elwyn, and Shull, Phys. Rev. **96**, 103 826A (1954).
[8] W. W. Pratt, Phys. Rev. **97**, 131 (1955).

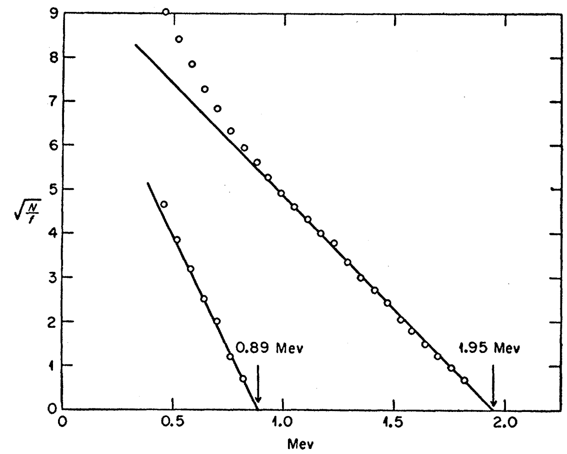

FIG. 3. Fermi analysis of the Ca^{49} beta-ray spectrum.

Beta-Ray Spectroscopy

Scintillation methods were employed for studies of the beta spectra from Ca^{49} and Sc^{49}. The scintillation spectrometer used a 1-inch diameter, $\frac{1}{2}$-inch high anthracene crystal attached to the face of a DuMont 6292 photomultiplier tube by means of silicone stopcock grease. The beta particles from the source were collimated into a well in the top face of the crystal, forming a hollow crystal spectrometer of the type described by Bell.[9] The measured resolution was 14 percent (full width at half-maximum counting rate) for the 0.625-Mev internal conversion electron line from Ba^{137m}. Energy calibration for the spectrometer was obtained using the 0.625-Mev line of Ba^{137m} and the 0.976-Mev line of Bi^{207}.

Sources of Ca^{49} for measuring the beta-ray spectra were prepared by placing about 0.2 mg of irradiated $Ca^{48}CO_3$ between the Formvar-polystyrene films, each about 25 μg/cm^2 thick. The total source thickness was approximately 1 mg/cm^2. A Fermi plot of the beta distribution is shown in Fig. 3, after subtracting the gamma-ray background obtained by placing a thick copper absorber over the collimating hole of the spectrometer. The figure shows the data resolved into components of 1.95 ± 0.05 and 0.89 ± 0.15 Mev. The β-ray intensity ratio obtained was $I_{0.9}/I_{1.95}=0.14$. This may be compared with the ratio of the intensities of the 4.05 and 3.10 gamma rays, namely 0.11, since it has previously been shown that all beta decays are to excited states in Sc^{49}, and no gamma-ray cascades were seen. However, branching calculations using the gamma-ray intensities are to be preferred, since uncertainties in the amount of scattering of beta rays and the relatively poor resolution of the β spectrometer make intensity determinations from these beta-ray spectra somewhat tenuous.

[9] P. R. Bell, *Beta- and Gamma-Ray Spectroscopy*, edited by K. Siegbahn, (Interscience Publishers, Inc., New York, 1955) Chap. 5, p. 136.

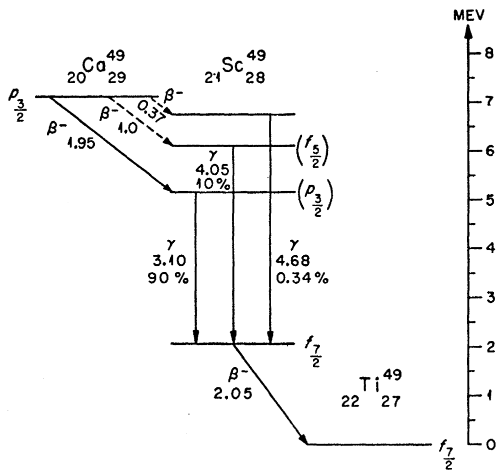

FIG. 4. Proposed decay scheme for the decay chain $Ca^{49}-Sc^{49}-Ti^{49}$.

The Sc^{49} beta-ray energy was measured in a similar fashion, except that the source was considerably thicker. An energy of 2.05±0.05 Mev was obtained from a Fermi analysis of the data.

A summary of the beta-ray data is presented in Table II, where abundances of the Ca^{49} beta groups and energies of the two lowest energy beta groups are inferred from the gamma-ray studies.

DISCUSSION

Mass Differences

The results of the experimental measurements described above have been incorporated into the decay scheme shown in Fig. 4.

Excellent confirmation of the decay scheme proposed for Ca^{49} is seen by comparing the $Ca^{49}-Sc^{49}$ ground state mass difference measured here with that calculated from the other data[10–13] shown in Table III. The measured mass difference of 5.05±0.05 Mev agrees very well with the calculated value of 5.21 Mev in view of the large uncertainty in the Q-values of the $Ca^{48}(d,p)Ca^{49}$ reaction.

The mass differences between Ca^{49}, Sc^{49}, and Ti^{49} may be compared with other Ca-Sc and Sc-Ti mass differences, plotting the data after the manner of Way and Wood.[14] Such a correlation is shown in Fig. 5, where the experimental points are labeled according to the nucleus whose decay measures the mass difference. Nuclides reported here are underlined. The beta-decay energies used in the diagrams were obtained from the table of King,[15] except for the Ca^{47} point, which was measured by Lyon[16] and independently by Lidofsky *et al.*[17] The curves for the Ca-Sc and Sc-Ti mass differences show a marked nonlinearity as a function of neutron number, unlike the more nearly linear dependence exhibited by the nuclides of higher atomic number. Such deviations from linearity appear more conspicuously among the light elements.[14] The Sc^{49} point falls on a smooth curve, but the Ca^{49} decay energy shows the expected jump past the closed shell at 28 neutrons.

Spin Assignments

In the region of the closed shells, single-particle spin assignments are probably fairly reliable. Since Ca^{49} has a 20-proton closed shell, and contains one neutron more than the 28-neutron, $f_{7/2}$ closed shell, the possibilities for the ground-state spin are $f_{5/2}$, $p_{3/2}$, and $p_{1/2}$. Experimentally, it appears the $p_{3/2}$ spin falls lower than the others in nuclides having 29 odd nucleons.[18] Recent $Ca^{48}(d,p)Ca^{49}$ angular distribution measurements by Braams[19] suggest $l=1$ for the ground state of Ca^{49}, which is consistent with the proposed $p_{3/2}$ configuration.

Because of the 28-neutron closed shell in Sc^{49} and its 21 protons, the ground state is almost surely $f_{7/2}$, in common with other nuclei having 21 nucleons. The first two excited states would be expected to arise from $p_{3/2}$ and $f_{5/2}$ orbitals. These assignments are in agreement with the log ft values for the beta transitions feeding the levels. An indication that the $f_{5/2}$ state lies above the $p_{3/2}$ state is seen in the calculated gamma-ray transition probability for decay of the second excited state to the ground state relative to a decay to the first excited state. Both transitions are $M1$ according to the proposed decay scheme, and using single particle estimates[20] the probability for the 0.95-Mev transition is only about 0.02 that of the 4.05-Mev gamma ray, or about 0.2 percent the intensity of the 3.10-Mev gamma ray, and thus would not have been seen in the measurements described above. If the orbital assignments were reversed the cascade gamma ray would retain its $M1$

TABLE II. Summary of beta-decay data on Ca^{49} and Sc^{49}.

Nuclide	Beta-ray energy, Mev	Abundance[a]	log ft
Ca^{49}	(0.37)[a]	0.0034	4.5
Ca^{49}	(1.00)[a]	0.10	4.6
Ca^{49}	1.95±0.05	0.90	4.9
Sc^{49}	2.05±0.05	1.00	5.7

[a] Calculated from gamma-ray data.

[10] N. S. Wall, Ph. D. thesis, Massachusetts Institute of Technology, 1953 (unpublished), and private communication to D. M. van Patter and W. Whaling, reported in Revs. Modern Phys. **26**, 426 (1954).
[11] Collins, Nier, and Johnson, Phys. Rev. **84**, 717 (1951).
[12] T. Okuda and K. Ogata, Phys. Rev. **60**, 690 (1941).
[13] J. Mattauch and R. Bieri, Z. Naturforsch. **9A**, 303 (1954).
[14] K. Way and M. Wood, Phys. Rev. **94**, 119 (1954).

[15] R. W. King, Revs. Modern Phys. **26**, 336–8 (1954).
[16] W. S. Lyon (to be published).
[17] Lidofsky, Benczer, and Fischer, Phys. Rev. **99**, 658(A) (1955).
[18] R. H. Nussbaum, thesis, University of Amsterdam, 1955 (unpublished).
[19] C. M. Braams (private communcation to R. H. Nussbaum). See reference 18.
[20] S. A. Moszkowski, *Beta- and Gamma-Ray Spectroscopy*, edited by K. Siegbahn, (Interscience Publishers, Inc., New York, 1955), Chap. 13.

character, but the ground-state transition would be $E2$. Again from the single particle formulas, the 0.95-Mev gamma ray would be about 40 times as intense as the 4.05-Mev gamma ray, and would have been easily discerned in the experiments described here.

No assignments have been made for the excited state at 4.68 Mev. That this level cannot be the expected $p_{1/2}$ state is established by the absence of gamma-ray transitions cascading from this state. If the 4.68-Mev level were $p_{1/2}$, then the ground-state transition would be $E3$, and the 1.58-Mev transition to the $p_{3/2}$ level would be $M1$ and in much higher intensity than the 4.68-Mev gamma ray.

The high energy of the first excited state in Sc^{49} (3.10 Mev) is consistent with Nussbaum's observation[18] that in nuclides with 21 or 27 identical nucleons the first excited state lies more than 1 Mev above the $f_{7/2}$ ground state. One might expect the level scheme of Ca^{41} to show some similarity to that of Sc^{49}, since Ca^{41} has 20 protons and 21 neutrons. From angular distribution measurements of the $Ca^{40}(d,p)Ca^{41}$ reaction, Holt and Marsham[21] assigned an orbital angular momentum $l=3$ to the ground state of Ca^{41}, and $l=1$ to the first two excited states. Similar measurements by Black[22] confirmed the ground state and first excited state l-values. Because of strong capture gamma rays to the first two excited states in Ca^{41}, Kinsey *et al.*[23] concluded that these states should have a spin of either $3/2^-$ or $1/2^-$. The above data strongly indicate that the ground state can be attributed to an $f_{7/2}$ configuration and that the first excited state is $p_{3/2}$. It is surprising that the 3.10-Mev $p_{3/2}$ state in Sc^{49} lies so much higher than the corresponding 1.9-Mev state in Ca^{41}, since 20 is generally thought to be more "magic" than 28 and so the ground-state excited-state energy separation in that case might be expected to be more pronounced.

TABLE III. Data from various sources used for calculation of $Ca^{49}-Sc^{49}$ mass difference.

	Mass difference, Mev	Experimental uncertainty, Mev	Reference
1	$Ca^{48}+D^2=H^1+Ca^{49}+2.80$	0.30	10
2	$4(C^{12})=Ca^{48}+44.32$	0.09	11
3	$Ti^{49}=4(C^{12})+H^1-55.82$	0.05	12
4	$2(H^1)=D^2+1.441$	0.001	13
5	$Sc^{49}=Ti^{49}+2.05$	0.05	This work
6	$Sc^{49}=Ca^{49}-5.21$		Sum of lines 1, 2, 3, 4, 5

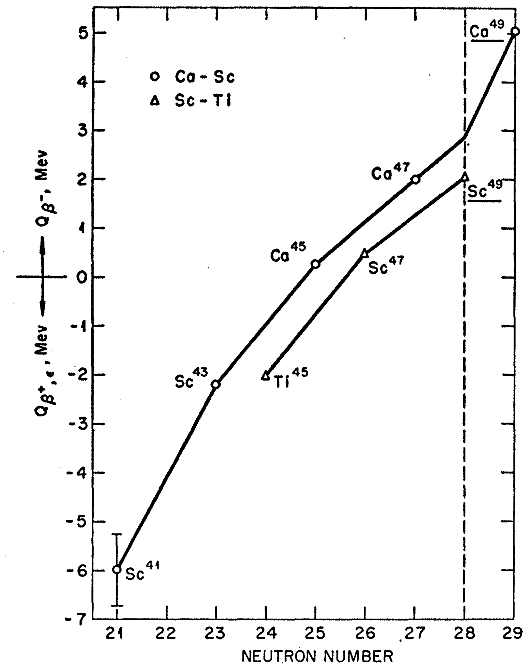

FIG. 5. Correlation of Ca−Sc and Sc−Ti mass differences from beta decay data. Decay energies reported here are underlined.

Jeffries[24] has measured the ground-state spin of Ti^{49} as 7/2, and from studies of the reaction $Ti^{48}(d,p)Ti^{49}$ Bretscher *et al.*[7] assign $l=3$ to this state of Ti^{49}. Therefore the ground state is assigned an $f_{7/2}$ orbital, again consistent with systematics in this region. The first excited state in Ti^{49} is at 1.35 Mev[7] but a gamma ray of this energy is not seen in the decay of Sc^{49}. The probable assignment for this state is $p_{3/2}$,[7] and so the beta-ray transition to this state would be second forbidden. Angular distribution measurements of the stripping reaction on Ti^{48} also indicate that the second excited state at 1.70 Mev is a p-state.[7] Thus the pure beta emission observed from Sc^{49} is consistent with the known facts about Ti^{49}.

ACKNOWLEDGMENTS

The authors are indebted to Q. V. Larson for assistance in many phases of the experimental work reported here.

[21] J. R. Holt and T. N. Marsham, Proc. Phys. Soc. (London) **A66**, 565 (1953).
[22] C. F. Black, Phys. Rev. **90**, 381(A) (1953).
[23] Kinsey, Bartholomew, and Walker, Phys. Rev. **85**, 1012 (1952).
[24] C. D. Jeffries, Phys. Rev. **92**, 1262, 1096(A) (1953).