

## Low-Intensity First-Forbidden Beta-Decay Branch in $\text{Ca}^{45}\dagger$

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A weak  $[(1.7_{-0.3}^{+0.8}) \times 10^{-6}$  per decay] inner beta branch in the decay of 165-day  $\text{Ca}^{45}$  populates the  $12.47_{-0.08}^{+0.14}$  keV state in  $\text{Sc}^{45}$ , as established by the observation of the  $K$  conversion electrons of this transition, superimposed on the  $258 \pm 2$  keV allowed ground-state beta continuum. The  $\log f_{it}$  inferred for this probably first-forbidden unique ( $\frac{7}{2}^- \rightarrow \frac{3}{2}^+$ ) inner group is  $9.28_{-0.18}^{+0.06}$ . No certain evidence was found for the  $L$  lines; the  $K/L$  ratio is given as  $2 \leq K/L \leq \infty$ , with a most likely value of 5.

### INTRODUCTION

IN recent studies at this Laboratory<sup>1</sup> of "hole" states in odd-mass isotopes in the  $f_{7/2}$  shell, a 12.7-keV gamma-emitting state of 0.44-sec half-life was found in  $\text{Sc}^{45}$  by ( $p, p'$ ) reaction on  $\text{Sc}^{45}$ . The transition was judged to be  $M2$ , from the  $d_{3/2}$  state (formed by promoting a proton from the  $d_{3/2}$  shell to pair with the  $f_{7/2}$  proton) to the  $f_{7/2}$  ground state.

Such a low-energy slow transition is potentially a very favorable case for Mössbauer studies if it can be abundantly supported by a long-lived, readily producible radioactive parent.  $\text{Ca}^{45}$  (165-day half-life), although well known to decay by allowed ( $f_{7/2} \rightarrow f_{7/2}$ ) negaton decay to the  $\text{Sc}^{45}$  ground state, seemed worth examining as such a source. A beta transition to the 12.7-keV ( $\frac{3}{2}^+$ ) state would be first-forbidden unique ( $\Delta J=2$ , yes). In this mass region such transitions have, typically,  $\log f_{it} \sim 8.5$ . A beta branching ratio for this transition of  $10^{-3}$ - $10^{-4}$  would thus be expected, and if realized, could perhaps serve the indicated purpose. In competition with the known 0.25-MeV ground-state decay, such a weak transition to a state so low in energy could easily have escaped attention.

The 12.7-keV gammas were looked for directly in the  $\text{Ca}^{45}$  decay<sup>2</sup> with a proportional counter, but the relatively intense inner bremsstrahlung (I.B.) from the ground-state beta decay masked their presence, despite use of an anticoincidence plastic scintillator beta detector sandwiching the source, to eliminate the I.B.

A further consideration, which, *a priori*, reduces the likelihood of the existence of a useful intensity of unconverted gamma ray is the high conversion expected. By extrapolation from the tables of Rose, the  $K$  conversion coefficient of a 12-keV  $M2$  transition is 260, implying a corresponding reduction in gamma abundance. Nevertheless, it was deemed worth searching for the 8-keV  $K$  conversion electrons of the 12.7-keV transition in the  $\text{Ca}^{45}$  decay, as a measure of the intensity of the beta branch feeding the metastable state. This branching ratio is currently of interest in connection

with considerations<sup>3</sup> of hole states in mass 45 in the shell model (see Discussion).

### EXPERIMENT

Sources for beta spectroscopy were prepared in the Argonne electromagnetic isotope separator. High-specific-activity  $\text{Ca}^{45}$  from Oak Ridge was used as charge material. The ions were focused at 50 kV, then decelerated to 300 V and electrostatically focused to a  $\sim 2$ -mm diam spot through a 3-mm mask onto a double  $20 \mu\text{g}/\text{cm}^2$  graphite film. Taking into account the spread in ion energies from the plasma source, we estimate the impact energies of the ions ranges from zero to  $\sim 300$  eV, at which energy the penetration into the graphite lattice is  $\ll 1 \mu\text{g}/\text{cm}^2$ .<sup>4</sup> Source strength was  $\sim 100 \mu\text{Ci}$ ; nominal source thickness was  $\sim 0.1 \mu\text{g}/\text{cm}^2$ .  $\text{Ca}^{44}$  contamination is deemed negligible.

However, the ion beam in the separator had a relatively intense component of mass 45 as  $\text{Si}^{29}\text{O}^{16}$ , from the quartz source tube in the plasma-ion-source oven. We estimate the mass of  $\text{Si}^{29}\text{O}^{16}$  collected as  $0.25 \pm 0.2 \mu\text{g}$ , giving a total source thickness of  $2\text{--}15 \mu\text{g}/\text{cm}^2$ . The beam impact area could be seen on the graphite film.

The beta spectrum was surveyed in the Argonne iron-free toroidal beta spectrometer<sup>5</sup> at a transmission of 10% and instrumental resolution of 0.3%. The earth's magnetic field was compensated<sup>6</sup> to  $10^{-4}$  G, sufficient to maintain the transmission constant (to one part in  $10^2$ ), down to 1 keV.

The detector was a bare anthracene crystal, 1 mm  $\times$  18 mm, mounted directly on the EMI 9536S photomultiplier tube.<sup>7</sup> Light collection from the edge was enhanced with a shaped Lucite ring surrounding the crystal. Figure 1 exhibits the pulse amplitude distribution from 7.9-keV electrons focused on the detector. The indicated integral discrimination level gave an efficiency of  $\sim 90\%$  at this value with a background of

<sup>3</sup> R. Lawson (private communication).

<sup>4</sup> I. Bergstron *et al.*, Nucl. Instr. Methods **21**, 249 (1963).

<sup>5</sup> M. S. Freedman *et al.*, Nucl. Instr. Methods **8**, 255 (1960); Bull. Am. Phys. Soc. **8**, 526 (1963); and Nucl. Instr. Methods (to be published).

<sup>6</sup> M. S. Freedman, F. Wagner, and F. T. Porter, Nucl. Instr. Methods (to be published).

<sup>7</sup> I. S. Sherman, M. S. Freedman, F. T. Porter, and F. Wagner, AIEEE Transactions, NS-11, 20 (1964); and Nucl. Instr. Methods (to be published).

<sup>†</sup> Based on work performed under the auspices of the U. S. Atomic Energy Commission.

<sup>1</sup> R. E. Holland, F. J. Lynch, and K.-E. Nystén, Phys. Rev. Letters **13**, 241 (1964) and private communication.

<sup>2</sup> R. E. Holland (private communication).

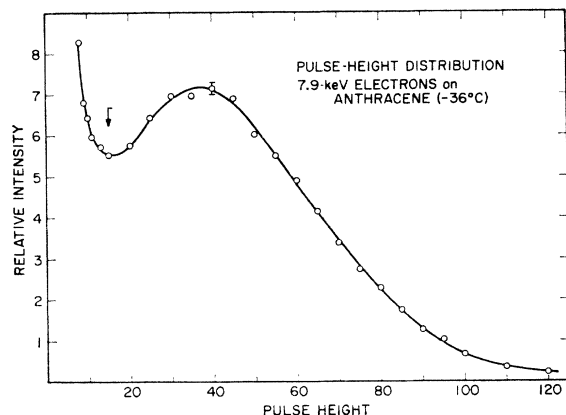


FIG. 1. Pulse-height distribution of 7.9-keV electrons on anthracene. The photomultiplier (EMI9536S) and scintillator were cooled to  $-36^{\circ}\text{C}$ . The background (i.e., no electrons striking the detector) has been subtracted. The low amplitude rise is caused by the long-lived states in anthracene excited by the impinging electrons (See Ref. 7). The arrow indicates the discriminator level which for this photomultiplier and temperature yielded a background of approximately 180 counts/min.

180 counts/min, compared with rates of  $\sim 13\,000$  counts/min in the region of interest with the  $\text{Ca}^{45}$  source. We believe the efficiency of the detector is constant within 10% over the region 7–9 keV which was examined in detail. The photomultiplier and crystal were cooled to  $-36^{\circ}\text{C}$ , at which temperature the evaporation of the anthracene is effectively suppressed ( $< .01$  mm/year from vapor-density measurements at this Laboratory).

Since the energy of the transition was known within a fraction of a keV,<sup>2</sup> the spectrum was surveyed in detail only in the region of the  $K$  conversion electrons. The position of the expected line on the total  $\beta$  spectrum is indicated in detail in the momentum plot of Fig. 2(a). Figure 2(b) shows the preliminary survey with  $\sim 8 \times 10^4$  counts per point. This survey was made with 1-min counts, in repeated sweeps over the indicated range. The total sweep times are short compared to possible drifts in electronic gains and, of course, the half-life of  $\text{Ca}^{45}$ ; therefore, no corrections have been applied to the data.

Figure 2(c) indicates on an expanded scale more detailed counting of the limited region at the  $K$  line with  $(0.7-2) \times 10^6$  counts per point, extending over 30 h. From Fig. 2(b) it is clear that the counting rate is linear with momentum in this limited range and that the slope is well determined. The solid line of Fig. 2(c) is fitted to the points above and below the line and has the known slope of the continuum. Figure 2(d) shows the subtraction of the solid line from the total spectrum.

The line is clearly broader than the expected line (0.3%) for this source diameter and the spectrometer settings used. Our judgment is that this is the result of the source thickness of 2–15  $\mu\text{g}/\text{cm}^2$  of  $\text{Si}^{29}\text{O}^{16}$  which is present in the isotope separator oven as mentioned above. This is consistent with our many observations on

line broadening at similar energies in 10–20  $\mu\text{g}/\text{cm}^2$  sources in other isotopes. The area under the line was compared with the area of the total beta spectrum on a momentum plot  $[(\text{counts}/\text{min})/H\rho$  versus  $H\rho]$ . This ratio is

$$\frac{e_K \text{ of the 12.5-keV transition}}{\text{Total } \beta \text{ spectrum}} = (1.4_{-0.3}^{+0.8}) \times 10^{-5}.$$

In other cases in which we have observed such severe line broadening at similar energies, we have been able to reveal the presence of line tails extending far below

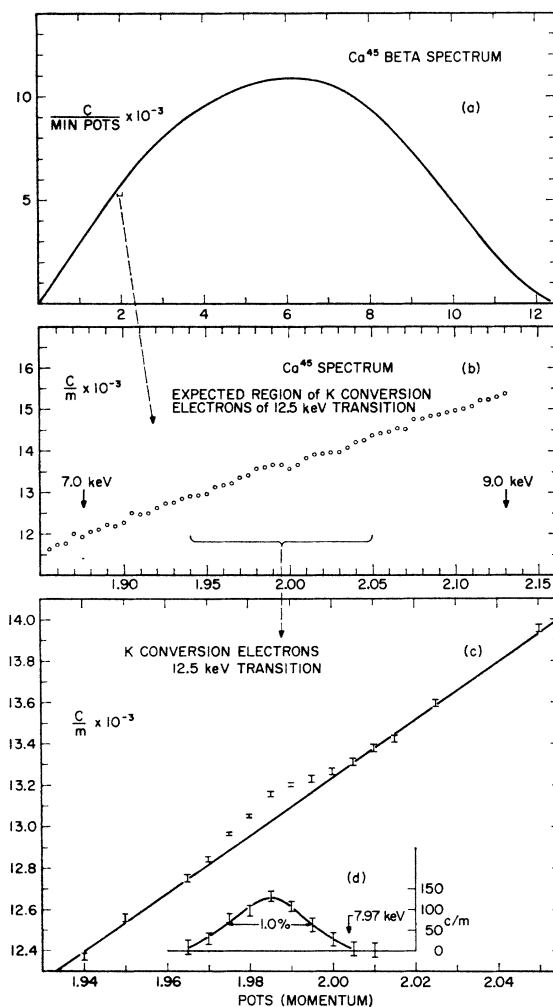


FIG. 2. (a) is the beta spectrum of  $\text{Ca}^{45}$  (momentum spectrum). The indicated region is shown expanded in the count plot of (b) which is a preliminary survey of the region where the  $K$  electrons of the 12.5-keV transition were expected. Errors in (b) are approximately the size of the circles. Part (c) is a further expanded count plot concentrating with additional counting on the immediate region of the  $K$  line. The solid curve in (c) is an "eye" fit to the points on the continuum. Insert (d) is a count plot of the difference between the experimental points and the solid line of (c). Since the line width is between 3 and 4 times the instrumental resolution (because of source thickness, see text) a high-energy edge extrapolation has been used as the line position.

the line, of intensity so low as to be indistinguishable from underlying continuous distributions, by tracing out the line profile in coincidence with a high-energy line, in our double toroidal coincidence spectrometer. Based on this experience, we assign the large positive uncertainty in the line area in this case, in which it is hardly practical to further reduce the statistical uncertainties in the line area by more extended counting.

Since the  $K$  conversion coefficient for an  $M2$  transition of this energy in scandium is estimated<sup>8</sup> to be  $\simeq 260$  we can neglect the gamma intensity. The  $K/L$  ratio, again by extrapolating theoretical curves,<sup>8</sup> is estimated to be  $\simeq 10$ . Experimentally, we find a best value of 5 (see below). We conclude therefore that  $\text{Ca}^{45}$  decays via the weak beta branch to the 12.5-keV state in  $\text{Sc}^{45}$ ,  $(1.7_{-0.3}^{+0.8}) \times 10^{-5}$  per decay.

The energy of the line is  $7.97_{-0.3}^{+1.4}$  keV and of the transition,  $12.47_{-0.3}^{+1.4}$  keV, the uncertainty arising mainly from the line broadening. Comparison to the calibration standard  $\text{Bi}^{207}$  was made using extrapolated leading-edge intercept to the continuum, because of the gross line broadening.

A search was made for the  $L$  conversion electrons of the 12.5-keV transition involving the accumulation of  $\sim 10^7$  counts at the line. No convincing evidence for the  $L$  lines was found, although the data indicated the possible presence of the line. From this we establish a most likely  $K/L$  ratio of 5, with a 68% confidence limit range from 2 to infinity.

## DISCUSSION

If we take the half-life of  $\text{Ca}^{45}$  as 165 days<sup>9</sup> and the ground-state beta-decay energy as 258 keV,<sup>10</sup> and use the nomograms of Moszkowski,<sup>11</sup> we find  $\log f_{0l}$  for the ground-state transition is 6.0 and for the transition to the 12.5-keV state  $\log f_{1l} = 9.28_{-0.18}^{+0.06}$ .<sup>12</sup> This  $f_{1l}$  value is  $\sim 10$  times the mean of other first-forbidden unique transitions, but a few have been observed<sup>13</sup> even above  $\log f_{1l} = 10$ .

The retardation of this first forbidden unique transition has been discussed recently by Cohen and Lawson.<sup>14</sup> They point out that if  $\text{Ca}^{45}$  is assigned the normal shell-model state  $(\pi d_{3/2})^4(\nu d_{3/2})^4(\nu f_{7/2})^5 \equiv \psi_1$  and if the 12.5-keV state in  $\text{Sc}^{45}$  is the one-proton hole state  $(\pi d_{3/2})^3(\nu d_{3/2})^4(\pi f_{7/2})^2(\nu f_{7/2})^4$ ,<sup>15</sup> the beta

transition to the 12.5-keV state is strictly forbidden by ordinary single-particle beta-decay operators since both a  $\nu f_{7/2} \rightarrow \pi f_{7/2}$  and a  $\pi d_{3/2} \rightarrow \pi f_{7/2}$  transition are required. They propose an admixture of  $(\pi d_{3/2})^2(\nu d_{3/2})^4(\pi f_{7/2})^2(\nu f_{7/2})^5 \equiv \psi_2$  double hole state in the  $\text{Ca}^{45}$  ground state and calculate from the experimental value of  $f_{1l}$  the extent of the mixing. If  $\alpha$  is defined in the wave function for the ground state of  $\text{Ca}^{45}$ ,

$$\psi_{\text{Ca}^{45}} = (1-\alpha)^{1/2}\psi_1 + \alpha^{1/2}\psi_2,$$

their calculations yield  $\alpha = 0.19$ , for assumed positive nuclear deformation, or  $\alpha = 0.11$  for negative deformation.

From a strict experimental point of view there is no direct evidence that the 12.5-keV state in  $\text{Sc}^{45}$  is  $\frac{3}{2}+$ , i.e., that the transition is  $M2$ , although, as indicated above<sup>15</sup> the  $l=2$  character of angular distributions in  $(p,\alpha)$  and  $(d,\text{He}^3)$  reactions yielding this state indicate a  $d$  orbital. The conversion coefficient or conversion ratios of the transition are unknown. The measured lifetime<sup>1</sup> (0.44 sec) combined with estimates of the conversion coefficients yielded a partial  $\gamma$ -ray mean life which is  $\sim 200$  times the single-particle estimate for an  $M2$  transition although this has been interpreted in terms of large probabilities for core excitation.<sup>16</sup> The possibility exists that the 12.5-keV state is significantly populated in the  $\text{Ti}^{45}$  decay. Although the ground-state decay has  $\log f_{0l} = 4.6$ , a normal allowed case, the configuration of  $\text{Ti}^{45}$ ,  $(\pi d_{3/2})^4(\nu d_{3/2})^4(\pi f_{7/2})^2(\nu f_{7/2})^3$ , does permit a first forbidden unique transition to the 12.5-keV state in  $\text{Sc}^{45}$  by a  $(\pi d_{3/2}) \rightarrow (\nu f_{7/2})$  decay, not retarded as from the normal configuration of  $\text{Ca}^{45}$ . Thus, although the branching ratio to the 12.5-keV state might again be  $\sim 10^{-4}$  to  $10^{-5}$  (owing to the fast ground-state decay), the absence of the beta minus continuum ( $\text{Ti}^{45}$  decays by  $\beta+$  and electron capture) may allow the observation of the  $L$  conversion electrons from the 12.5-keV transition. The inferred multipolarity from the  $K/L$  or  $L$  subshell ratio could be used to test the  $\frac{3}{2}+$  assignment. We are preparing to make these measurements.

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<sup>16</sup> R. D. Lawson and M. H. Macfarlane, Phys. Rev. Letters 14, 152 (1965); R. K. Bansal and J. B. French, Phys. Letters 14, 230 (1965).

<sup>8</sup> M. E. Rose, *Internal Conversion Coefficients* (North-Holland Publishing Company, Amsterdam, 1958).

<sup>9</sup> J. P. Cali and L. F. Lowe, Nucleonics 17, 86 (1959); C. F. G. Delaney and J. H. J. Poole, Phys. Rev. 89, 529 (1953).

<sup>10</sup> P. Macklin, L. Feldman, L. Lidofsky, and C. S. Wu, [Phys. Rev. 77, 137 (1950)] give  $254 \pm 3$  keV. We find  $258 \pm 2$  keV.

<sup>11</sup> S. A. Moszkowski, Phys. Rev. 82, 35 (1951).

<sup>12</sup> J. P. Davidson, Jr., Phys. Rev. 82, 48 (1951).

<sup>13</sup> See, for example, the summary by P. Lipnik and J. W. Sunier, Nucl. Phys. 56, 241 (1964).

<sup>14</sup> S. Cohen and R. D. Lawson, Phys. Letters 17, 299 (1965).

<sup>15</sup> Such a  $d_{3/2}$  proton hole state is assigned to the first excited state in  $\text{Sc}^{46}$  and other odd Sc isotopes to account for the  $l=2$