## $\beta^+$ decay and cosmic-ray half-life of <sup>54</sup>Mn

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<sup>54</sup>Mn has previously been observed in cosmic rays where is it expected to decay by  $\beta^+$  and  $\beta^-$  transitions with a half-life of a few million years. The present experiment to measure the  $\beta^+$  decay branch in the laboratory establishes an upper limit of  $4.4 \times 10^{-8}$  for the branching ratio, and a lower limit of 13.3 for the log ft value of this second forbidden unique transition. If it is assumed that the  $\beta^-$  decay branch has the same log ft value, then its partial half-life must be greater than  $4 \times 10^4$  years.

#### I. INTRODUCTION

<sup>54</sup>Mn is an odd-odd nucleus which decays in the laboratory with a half-life of 312 days via an allowed electron capture transition to the 835 keV level in <sup>54</sup>Cr (Ref. 1). In the cosmic rays, <sup>54</sup>Mn is believed to be produced through spallation of primary iron nuclei by interstellar hydrogen. As a high-energy cosmic ray,<sup>54</sup>Mn would be stripped of all its atomic electrons, thus preventing its decay by electron capture. However, as shown in Fig. 1, it is energetically possible for it to decay via second forbidden unique transitions to the ground states of <sup>54</sup>Cr and <sup>54</sup>Fe by positron or negatron emission, respectively. The only previous experiment of which we are aware, by Berényi *et al.*,<sup>2</sup> established a lower limit of  $1 \times 10^6$  years for the  $\beta^+$  decay partial half-life of <sup>54</sup>Mn. From studies of the decays of <sup>10</sup>Be, <sup>22</sup>Na, and <sup>26</sup>Al, the log*ft* values for such transitions have been found<sup>3,4</sup> to be between 13.9 and 15.7, yielding estimates of the partial half-lives for these decay modes in <sup>54</sup>Mn of  $8.4 \times 10^7$  y  $< t_{1/2}(\beta^+) < 6.0$  $\times 10^9$  y and  $9.2 \times 10^5$  y  $< t_{1/2}(\beta^-) < 6.5 \times 10^7$  y. Because of these long  $\beta^+$  and  $\beta^-$  half-lives, <sup>54</sup>Mn has been proposed as a cosmic-ray chronometer and a probe of models of the interstellar medium and cosmic-ray propagation<sup>5</sup> More recently, the presence of a long-lived radioactive isotope of manganese in the cosmic rays has been experimentally confirmed,<sup>6</sup> with a half-life estimated to be (1 to  $2) \times 10^6$  y in order to explain the measured abundance as a function of cosmic-ray energy.<sup>7</sup>

While the dominant decay mode for fully ionized <sup>54</sup>Mn is expected to be  $\beta^-$  decay, it is extremely hard to experimentally isolate this small branch in the laboratory from the deexcitation  $\gamma$  rays from the  $\approx 100\%$  EC decay branch. Instead, we have searched for the even smaller, but easier to detect,  $\beta^+$  decay branch by measuring its energy spectrum with a silicon particle detector telescope in coincidence with the back-to-back 511-keV positron annihilation  $\gamma$  rays. Our experimental limit for the branching ratio and its corresponding log ft value for this  $3^+$ -0<sup>+</sup> transition then allowed us to constrain the partial half-life for  $\beta^-$  decay, which is also a  $3^+$ -0<sup>+</sup> transition, by assuming the same log ft value.

#### II. EXPERIMENTAL METHOD A. Source preparation

Since the  $\beta^+$  decay branching ratio is expected to be less than  $10^{-6}$ , any radioactive contaminant in the <sup>54</sup>Mn source with a  $\beta^+$  activity at or greater than this level would mimic the sought after effect. Detection and elimination of such contaminants at this minute level is thus extremely important. 100  $\mu$ Ci of <sup>54</sup>Mn dissolved in HCl were purchased from the New England Nuclear Company and used approximately one year later, allowing any short-lived activity to decay away. To our knowledge, there are no long-lived isotopes that undergo only ground-state to ground-state  $\beta^+$  decay. Therefore, the contaminants of interest were identified by detecting the deexcitation  $\gamma$  rays from a sample of the <sup>54</sup>Mn solution by means of a shielded high resolution pileup suppressed 110 cm<sup>3</sup> intrinsic Ge detector. The original sample contained  ${}^{65}$ Zn,  ${}^{60}$ Co, and  ${}^{22}$ Na at a level of (30 to 50)  $\times 10^{-6}$ times the activity of the <sup>54</sup>Mn. The <sup>65</sup>Zn contaminant is particularly troublesome because it positron decays to the ground-state of <sup>65</sup>Cu with a branching ratio of  $1.45 \times 10^{-2}$  and a 227 day half-life.<sup>1</sup> The end-point for this positron spectrum is 325 keV, which makes it almost indistinguishable in a low statistics experiment from the expected 355-keV end-point  $\beta^+$  decay of <sup>54</sup>Mn. In fact, a source of <sup>65</sup>Zn similar to the <sup>54</sup>Mn source was used to calibrate the efficiency of the apparatus.

The <sup>54</sup>Mn solution was further diluted and passed through columns of hydrated antimony pentoxide and AG1-X8 anion exchange resin to remove the sodium, and cobalt and zinc, respectively. After two passes, the solution containing the manganese fraction was boiled to reduce its volume and a few drops were deposited and heated to dryness on a thin polyethylene backing to make a thin bare source for the positron decay experiment. Similar sources were prepared from commercially obtained <sup>65</sup>Zn and <sup>22</sup>Na solutions for energy and efficiency calibrations of the apparatus. A small sample of the chemically purified manganese was also counted with the high resolution Ge detector system for approximately 10 days. After background subtraction and correction for



FIG. 1. Decay scheme of <sup>54</sup>Mn.

the relative efficiencies of the  $\gamma$  rays, the amounts of  $^{65}$ Zn,  $^{60}$ Co, and  $^{22}$ Na relative to the  $^{54}$ Mn activity were found to be less than  $2 \times 10^{-6}$ ,  $1.1 \times 10^{-6}$ , and  $1.4 \times 10^{-6}$ , respectively.

#### **B.** Detector setup

A schematic view of the arrangement for detecting positrons emitted by the source is shown in Fig. 2. A 7.3  $\mu$ Ci <sup>54</sup>Mn source was mounted in front of a two-element silicon surface-barrier detector telescope to detect the emitted positrons. The front and rear elements of the telescope had thicknesses of 75 and 1000  $\mu$ m and active areas of 50 and 100 mm<sup>2</sup>, respectively. The use of a particle detector telescope was dictated by the results of an earlier experiment where a single  $1000-\mu$ m-thick silicon detector was used. The relatively large number of background events, which limited the sensitivity of this mode of acquiring data, was attributed to internal bremsstrahlung (IB) photons. This process produces photons with a continuum of energies up to 542 keV and with an emission probability of  $1.7 \times 10^{-4}$  per EC decay, coincident with the 835-keV  $\gamma$  rays. In those rare cases where the total electromagnetic energy of up to 1377 keV was captured by the three detectors, the requirement that 1022 keV be deposited in the NaI detectors caused the appearance of these spurious events with energies up to 355 keV in the gated energy spectrum of the silicon detector. Ancillary experiments to detect the IB photons and to mea-



FIG. 2. Schematic view of experimental setup for detecting positrons.



FIG. 3. Spectrum of positrons from the decay of  $^{65}$ Zn. This is a projection of the summed telescope energy spectrum when 511 keV is deposited in both halves of the Nal detector.

sure the corresponding response of the silicon and NaI detectors confirmed this interpretation. The use of the detector telescope provided far superior discrimination against the detection of IB photons but reduced the overall efficiency for detecting positrons.

The source and telescope were mounted in the center of an 8.25-cm hole in an annular 30-cm×30-cm<sup>2</sup> NaI detector that was optically divided into two halves. The back-to-back 511-keV  $\gamma$  rays from positrons annihilating in the back element of the  $\beta^+$  telescope were detected in these two halves of the annular detector. A fourfold coincidence between the telescope elements and each half of the annular NaI detector was required in the electronic hardware trigger. For each such trigger, the energy signals from all four detectors, the summed energy in the telescope, timing signals between the thick silicon detector and each of the other three detectors, and the timing signal between the two halves of the NaI annulus were recorded. The trigger rate with the <sup>54</sup>Mn source was approximately  $1/3 \text{ sec}^{-1}$ . The efficiency of the apparatus for detecting positrons from the decay of <sup>54</sup>Mn was measured by detecting positrons from a  $^{65}$ Zn source mounted in the same geometry as the  $^{54}$ Mn. A spectrum from  $^{65}$ Zn of positrons in coincidence with the 511-511-keV  $\gamma$ rays peaks in the NaI detectors is shown in Fig. 3. Spectra from a <sup>22</sup>Na source were also recorded in order to correct for the possible <sup>22</sup>Na contamination in the manganese sample.

#### **III. ANALYSIS AND RESULTS**

The nine parameter event-by-event data from the positron decay experiment was sorted off-line with a variety of software gates. The candidate positron spectrum was extracted by projecting out the summed telescope energy spectrum in coincidence with 511-keV  $\gamma$  rays detected in both halves of the annular NaI detector within the prompt timing requirement for all detectors. Background spectra were extracted by projecting the summed telescope energy spectrum in coincidence with combinations of gates set above and below 511 keV in both halves of the NaI annulus. Background events due to random coincidences were determined to be negligible by setting



FIG. 4. Projections of summed telescope energy spectra observed with the <sup>54</sup>Mn source. (a) 511-511-keV gate. (b) and (c) less than 511-keV and greater than 511-keV gates in both halves of the NaI detector with a summed energy of 1022 keV.

gates off the prompt peak in each of the timing spectra. In a total running time of 360.4 h over a period of 21 days, no excess counts were obtained in the back-to-back prompt 511-keV gated spectrum over those in the background gates. The spectrum of possible positron events obtained together with two typical background spectra are shown in Fig. 4. The background spectra shown here were obtained by requiring coincidences in the two halves of the NaI annular detector between  $\gamma$  rays whose energies were greater and less than 511 keV, but whose sum was 1022 keV.

In the 511-511-keV gate of interest, we observed  $208\pm14.4$  events in the first 355 keV of the summed telescope energy spectrum, whereas the average number of events in all the background gates was  $213\pm5.6$ . Thus the  $1\sigma$  upper limit on the number of <sup>54</sup>Mn positrons detected was 15.5. It is possible that these events were caused by residual interactions of the coincident 835-keV and internal bremsstrahlung photons in our detectors. It is apparent that no positrons were detected from the previously mentioned possible <sup>22</sup>Na contaminant. Using our experimentally determined efficiency of 0.10% for detecting positrons, this results in an upper limit of  $4.4 \times 10^{-8}$ for the  $\beta^+$  decay branching ratio of <sup>54</sup>Mn, which corresponds to a lower limit of  $2.0 \times 10^7$  years for the partial half-life of this decay channel. This result clearly rules out the possibility that  $\beta^+$  is the dominant decay mode for the <sup>54</sup>Mn nuclei found in cosmic rays. Using the accepted value of 1377 keV for the mass difference between <sup>54</sup>Mn and <sup>54</sup>Cr, we obtain a lower limit of 13.3 for the  $\log ft$  value of this transition. Using this  $\log ft$  value for the  $\beta^-$  decay branch with 697 keV of available energy, we obtain a lower limit of  $4 \times 10^4$  years for the partial half-life of this decay mode. This value is  $\approx 25$  times shorter than the estimated half-life of <sup>54</sup>Mn in cosmic rays and thus consistent with  $\beta^-$  as the dominant channel for this decay. Experiments to measure the  $\beta^-$  decay rate of <sup>54</sup>Mn are now in progress.

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