## Search for the $\beta^+$ decay of <sup>54</sup>Mn

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We have performed a search for the  $\beta^+$  decay of <sup>54</sup>Mn by looking for back-to-back 511-keV  $\gamma$  rays in two high-purity Ge detectors. No excess of events above background was observed, and a limit of  $5.7 \times 10^{-7}$ % has been established for the  $\beta^+$  branch. The significance of this result for the use of <sup>54</sup>Mn as a cosmic ray chronometer is discussed.

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Radioactive nuclei that decay in the laboratory via electron capture can have very different half-lives as cosmic rays. This is because during the acceleration and propagation of cosmic rays, these nuclei will be stripped of their atomic electrons. While some of them become stable, others have sufficient decay energies to undergo  $\beta^+$  and/or  $\beta^-$  decays. If the resulting cosmic-ray halflives are of the order of millions of years, the isotopic composition of the corresponding elements in the cosmic rays can be of help in determining their confinement time in the interstellar medium [1].

The decay scheme of  ${}^{54}\text{Mn}$  is shown in Fig. 1.  $\beta^+$  decay is energetically allowed only to the ground state of <sup>54</sup>Cr through a second-forbidden unique transition with an end-point energy of 355 keV.  $\beta^-$  decay to the ground state of <sup>54</sup>Fe is possible through a second-forbidden unique transition with a 697-keV end-point energy. Due to larger available phase space, the probability of the  $\beta^-$  decay is expected to be approximately two orders of magnitude larger than that for  $\beta^+$  decay, assuming the nuclear-matrix elements of the  $\beta^-$  and  $\beta^+$  transitions are the same. However, the measurement of the  $\beta^-$  decay is very difficult, due to electron backgrounds superimposed on the  $\beta$  decay events. The estimate of the intensity of the  $\beta^-$  branch is of the order of  $10^{-4}$ %. This is more than a hundred times less intense than the internal conversion electrons from the 835-keV transition and the probability of shakeoff [2] associated with the electron capture transition. These internal conversion and shakeoff electrons will produce a low energy tail in charged particle detectors which hides the  $\beta^-$  spectrum. Compton-ejected electrons can also interfere with the  $\beta^-$  detection. Much simpler to detect, but also requiring a careful search, is the positron decay, which can be measured by looking for the positrons in coincidence with back-to-back 511keV annihilation photons, as done by Sur et al. [3], or alternatively through the measurement of only the an-

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result is observed, it must be shown that it really comes from the <sup>54</sup>Mn decay and not from contaminants. A careful  $\gamma$ -ray measurement can help in determining the level of positron emitting contaminants present in the source. This experiment was designed to improve on the limit

nihilation photons, as we describe below. If a positive

obtained by Sur *et al.* of  $4.4 \times 10^{-6}$ %. They used a Sitelescope inside a NaI(Tl) annulus, and the events of interest were the detection of the positron in both Si surface-barrier detectors ( $\Delta E$  and E), with annihilation inside the E detector, and the 511-keV photons being detected on both halves of the NaI annulus. Their overall detection efficiency was 0.10%.

The radioactive source used in the present experiment was purchased from New England Nuclear Co. and chemically purified in order to minimize the presence of other  $\beta^+$  emitters. The isotopes of major concern were <sup>22</sup>Na and <sup>65</sup>Zn. The chemistry was done by mixing the liquid source with a mixture of DOWEX 1-X8 anion-exchange resin, HAP (hydrated-antimony pentoxide) and concentrated HCl. Zinc ions attach to the resin, and sodium ions to the HAP. The Na-HAP precipitate and the resin were then removed from the liquid phase by centrifuging. The liquid source was dried on small pieces of filter paper, and sealed with several layers of adhesive tape.



FIG. 1. Decay scheme of  $^{54}$ Mn. The dashed lines indicate the yet unobserved branches.

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These materials were thick enough to stop the positrons. The finished source was 1 cm<sup>2</sup> in area and had an activity of  $(3.8 \pm 0.2)\mu$ Ci at the beginning of the experiment. The level of contaminants was checked throughout the experiment and also by  $\gamma$  counting at a low-background counting facility at Lawrence Berkeley Laboratory, where we searched for the 1115 keV  $\gamma$  ray from <sup>65</sup>Zn and for the 1275 keV  $\gamma$  ray from <sup>22</sup>Na. No evidence of either line was seen and the following upper limits were established for the activities of <sup>65</sup>Zn and <sup>22</sup>Na relative to the <sup>54</sup>Mn source:  $6.5 \times 10^{-5}\%$  and  $1.9 \times 10^{-6}\%$ , respectively.

The experimental setup shown in Fig. 2 was composed of two high-purity Ge detectors of 110 and 200 cm<sup>3</sup> volume placed face to face at the center of an 8.25-cm hole in a  $30 \times 30$  cm annular NaI(Tl) detector. The source was placed between the Ge detectors at close geometry, with the external annulus acting as an anti-Compton and anti-coincidence shield. A standard fast-slow electronic coincidence was set to produce a master gate every time there was a coincidence between the Ge detectors. For each event the following parameters were recorded: the energy signals from each Ge detector and each half of the NaI(Tl) annulus, the time between one Ge detector signal and each of the other signals, and the pile up rejection inhibit signals from the ORTEC 572 amplifiers corresponding to each Ge detector.

The singles detection rates of the Ge detectors were 15 and 19 kHz, that of the annulus was 110 kHz. The Ge-Ge coincidence rate was about 30 Hz. The majority of the coincidence events from <sup>54</sup>Mn decay were produced by 835-keV photons that Compton backscatter from one detector into the other. To minimize the rate of such events, we set the discrimination thresholds on each Ge detector above 200 keV, the position of the backscatter peak associated with the 835-keV  $\gamma$  ray. The 511-511 keV efficiency was measured by placing a 0.044  $\mu$ Ci <sup>65</sup>Zn source inside the apparatus together with the <sup>54</sup>Mn source. The overall effect of dead time and pile up from the electronics and acquisition system for the events of interest was 65%, measured by keeping the <sup>65</sup>Zn source, removing the <sup>54</sup>Mn and remeasuring the 511-511 keV coincidence efficiency. The overall efficiency, including dead-time effects, for detecting 511-511 keV photopeak events in the Ge detectors under our experimental conditions was  $(0.80 \pm 0.03)\%$ .

We measured <sup>54</sup>Mn for 520.6 h, recording about five events per day in the 511-511 keV coincidence region. We then removed the source and collected 305.5 hours of background data.

The search for the 511-511 keV signal was done by



FIG. 2. Experimental setup.

setting gates in one spectrum at the 511 keV position and also at the higher- and lower-energy sides of the expected peak and then looking at the coincident spectrum in the other detector. The same procedure was adopted for the background runs, in order to subtract it from the raw data. The position and width of the true coincidence time peak were determined with the <sup>22</sup>Na and <sup>65</sup>Zn sources. For the <sup>54</sup>Mn data set, the time spectrum showed no peak, thus indicating that essentially all the events we observed were random coincidences. After the background subtraction, the net signal was calculated by taking the number of events in the 511-511 keV position and subtracting from it the average of the two neighboring regions corresponding to the same total energy, 1022 keV.

There is no statistically significant structure in either of the Ge detector spectra in the 511-keV region. Figure 3 shows the region around 511 keV in both Ge detectors, for the source and background measurements. After background subtraction, the total number of counts in the region where the annihilation peak was expected was  $76.7 \pm 10.9$ . In the surrounding regions with the same total deposited energy there was an average of  $78.4\pm6.5$ counts. We then establish an upper limit of 11.9 counts (68.3% CL, [4]). Using the average source strength and efficiency we set an upper limit of  $5.7 \times 10^{-7}$ % for the  $\beta^+$  branching ratio and a lower limit of 14.1 for the log*ft* of this transition. This corresponds to a lower limit on the  $\beta^+$  decay half-life of  $1.50 \times 10^8$  years. Assuming the same log ft for the  $\beta^-$  decay branch, we obtain an upper limit of  $2.9 \times 10^{-4}$ % for its intensity and a lower limit on the cosmic-ray half-life of  $2.95 \times 10^5$  years for the decay of the bare nucleus of <sup>54</sup>Mn.

Recent measurements of the isotopic abundances of Fe-group elements at energies of approximately 325 MeV/nucleon by Leske [5], show that there are



FIG. 3. Expanded views of the Ge detector spectra. These spectra are both gated by 511 keV deposited on the other Ge detector and a time gate at the position of the true-coincidence peak. (a) and (b) are the total sums of source in. (c) and (d) are the total sums of the background. The arrows indicate the 511-keV regions.

roughly equal amounts of <sup>53</sup>Mn and <sup>55</sup>Mn, and a level of <sup>54</sup>Mn consistent with zero. The ratios of <sup>53,54,55</sup>Mn from his measurement are 1 : <0.25 :  $1.28^{+0.32}_{-0.25}$ , respectively. From the observed fraction of <sup>54</sup>Mn, Leske set a lower limit for the cosmic-ray confinement time  $\tau_{\rm esc}$ , of  $\tau_{\rm esc} > 50 \times T_{\beta^-}$  (<sup>54</sup>Mn). If we now insert our lower limit for the <sup>54</sup>Mn  $\beta^-$  half-life of  $2.95 \times 10^5$  years in this inequality, we get  $\tau_{\rm esc} > 15$  million years. Although this is only a lower limit, it is the first application of <sup>54</sup>Mn as a cosmic ray chronometer. This result for the confinement time is consistent with the value of  $15^{+7}_{-4}$  million years

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determined from measurements of the  $^{10}$ Be abundance in the cosmic rays [6].

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