Search for the β^+ decay of ⁵⁴Mn

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We have performed a search for the β^+ decay of ⁵⁴Mn by looking for back-to-back 511-keV γ rays in two high-purity Ge detectors. No excess of events above background was observed, and a limit of 5.7×10^{-7} % has been established for the β^+ branch. The significance of this result for the use of 54 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 diferent 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 β^+ and/or β^- 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 54Mn is shown in Fig. 1. β^+ decay is energetically allowed only to the ground state of ⁵⁴Cr through a second-forbidden unique transition with an end-point energy of 355 keV. β^- decay to the ground state of $54Fe$ 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 β^- decay is expected to be approximately two orders of magnitude larger than that for β^+ decay, assuming the nuclear-matrix elements of the β^- and β^+ transitions are the same. However, the measurement of the β^- decay is very dificult, due to electron backgrounds superimposed on the β decay events. The estimate of the intensity of the β^- branch is of the order of $10^{-4}\%$. This is more than a hundred times less intense than the internal conversion α 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 β^- spectrum. Compton-ejected electrons can also interfere with the β^- 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 511 keV annihilation photons, as done by Sur et al. [3], or alternatively through the measurement of only the an-

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nihilation photons, as we describe below. If a positive result is observed, it must be shown that it really comes from the 54 Mn decay and not from contaminants. A careful γ -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 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 β^+ emitters. The isotopes of major concern were ²²Na and ⁶⁵Zn. 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^2 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 γ counting at a low-background counting facility at Lawrence Berkeley Laboratory, where we searched for the 1115 keV γ ray from ⁶⁵Zn and for the 1275 keV γ ray from ²²Na. No evidence of either line was seen and the following upper limits were established for the activities of 65 Zn and 22 Na relative to the 54 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 cm³ volume placed face to face at the center of an 8.25-cm hole in a 30×30 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(T1) 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 110kHz. The Ge-Ge coincidence rate was about 30 Hz. The majority of the coincidence events from 54 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 γ ray. The 511-511 keV efficiency was measured by placing a 0.044 μ Ci ⁶⁵Zn source inside the apparatus together with the 54 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 65 Zn source, removing the 54 Mn and remeasuring the $511-511$ keV coincidence efficiency. The overall efficiency, including dead-time effects, for detecting 511-511keV photopeak events in the Ge detectors under our experimental conditions was $(0.80 \pm 0.03)\%$.

We measured 54 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 $22Na$ and 65 Zn sources. For the 54 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 ± 10.9 . In the surrounding regions with the same total deposited energy there was an average of 78.4 ± 6.5 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 5.7×10^{-7} % for the β^+ branching ratio and a lower limit of 14.1 for the logft of this transition. This corresponds to a lower limit on the β^+ decay half-life of 1.50×10^8 years. Assuming the same $\log\!f\!t}$ for the β^- decay branch, we obtain an upper limit of 2.9×10^{-4} % for its intensity and a lower limit on the cosmic-ray half-life of 2.95×10^5 years for the decay of the bare nucleus of 54 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 53 Mn and 55 Mn, and a level of ⁵⁴Mn consistent with zero. The ratios of $\frac{53,54,55}{1.28+0.32}$, respec-
from his measurement are $1: < 0.25: 1.28^{+0.32}_{-0.25}$, respectively. From the observed fraction of 54 Mn, Leske set a lower limit for the cosmic-ray confinement time $\tau_{\rm esc}$, of lower limit for the cosmic-ray confinement time $\tau_{\rm esc}$, of $\tau_{\rm esc} > 50 \times T_{\beta}$ (⁵⁴Mn). If we now insert our lower limit for the ⁵⁴Mn β^- half-life of 2.95 \times 10⁵ years in this in-
equality, we get $\tau_{\text{esc}} > 15$ million years. Although this is
callity, we get $\tau_{\text{circ}} > 15$ million years. Although this is only a lower limit, it is the first application of 54 Mn as a cosmic ray chronometer. This result for the confinement time is consistent with the value of 15^{+7}_{-4} million years

determined from measurements of the 10 Be abundance in the cosmic rays [6].

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