Galactic Confinement Time of Iron-Group Cosmic Rays Derived from the ^{54}Mn Chronometer

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The β-decay half-life of ^{54}Mn is needed to employ this isotope as a cosmic ray chronometer. We have determined the partial half-life of ^{54}Mn for positron emission by counting a highly purified 35-μCi source of ^{54}Mn in GAMMASPHERE to search for the astrophysically interesting β^+ decay branch through the observation of coincident positron-annihilation γ rays. A careful analysis of 97 hours of source counting and 61 hours of background shows a net signal of 24 ± 10 back-to-back 511-511 keV coincident events. Based on this result, the branch for this decay mode is $(2.2 \pm 0.9) \times 10^{-7}\%$. The implications of this result for the ^{54}Mn cosmic-ray chronometer problem are discussed. [S0031-9007(97)04630-9]

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In the leaky-box model, cosmic rays (CR) propagate through our galaxy along complex paths as a result of scattering by random magnetic fields. Eventually this random walk leads the CR out or the galaxy. The mean confinement time of CR within our Galaxy can be determined by comparing the CR abundances of suitably long-lived radioactive isotopes with those of their stable neighbors. Radioisotopes that have been used for determinations of the CR confinement time include ^{10}Be ($t_{1/2} = 1.6$ Myr [1,2,3]), ^{26}Al ($t_{1/2} = 0.87$ Myr [4,5]), and ^{36}Cl ($t_{1/2} = 0.3$ Myr [6]). Measurements of the abundances of these isotopes lead to CR confinement times in the range 10–20 Myr and imply a mean density of interstellar matter traversed by the CR of approximately 0.3 atoms/cm^3. This is substantially lower than the mean density found in the galactic disk, suggesting that the CR spend a substantial amount of time in the halo of our galaxy.

In addition to these light elements, it is of critical interest to understand the confinement time of the nuclei of the iron group, which are produced in explosive nuclear burning. Cassé [7] suggested that ^{54}Mn, which is a product of spallation of iron nuclei on interstellar hydrogen, might serve as a CR clock. Grove *et al.* [8] have emphasized the importance of knowledge of the ^{54}Mn half-life for understanding CR propagation. The ^{53,54,55}Mn spallation production cross sections from ^{56}Fe at an energy of 600 MeV/nucleon were measured by Webber *et al.* [9] to be 37.5, 42.3, and 40.0 mb (±6 mb), respectively. Thus, one might expect their cosmic-ray abundances to be nearly equal. However, recent measurements of cosmic-ray manganese by Leske [10] and DuVernois [11] show that the abundance of ^{54}Mn is much smaller than that of its neighboring isotopes. The decay of CR ^{54}Mn can possibly account for this discrepancy and thus provide the crucial datum point for the confinement time of iron-group nuclei.

In the laboratory, ^{54}Mn decays via electron capture with a half-life of 312 d. Because of their interactions with the interstellar medium, CR nuclei are fully stripped of their atomic electrons. This turns a nucleus such as ^{53}Mn, which decays in the laboratory via electron capture (EC), into a stable nucleus. As can be seen in Fig. 1, it is possible for ^{54}Mn to decay by second-forbidden unique β^- or β^+ transitions to the ground states of ^{54}Fe or ^{54}Cr, respectively. In the laboratory, these forbidden decays have to compete with the allowed (and therefore far more probable) EC decay. The EC decay mode is not available to a ^{54}Mn nucleus that has been fully stripped of its atomic electrons, and so the β^- and β^+ decays will determine the CR half-life of this isotope. Cassé [7] estimated the β^- and β^+ partial half-lives of ^{54}Mn to be about 2 Myr and 1 Gyr, respectively. Later, Wilson [12] suggested that the partial half-lives for these decays would be in the ranges 0.06–10 Myr (β^-) and 6–8000 Myr (β^+).

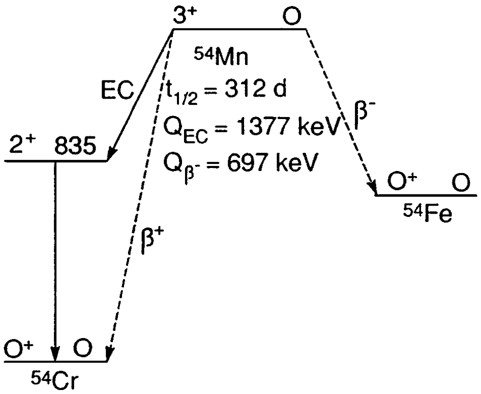

FIG. 1. The decay scheme of ^{54}Mn. Level and transition energies are given in keV.

Several attempts have been made previously to determine the β^- or β^+ half-life of ^{54}Mn. Kibédi *et al.* [13] searched for the β^- decays using a magnetic spectrometer to detect the electrons. They have established the lower limit of 2.2×10^4 yr for the partial half-life of ^{54}Mn due to the β^- decay mode. Assuming that the nuclear matrix elements for the β^- and β^+ transitions are the same, the larger available phase space for the β^- decay means that its probability will be much larger than that for β^+ decay. However, the observation of the β^- decay is very difficult owing to the electron background from γ-ray events superimposed on the β^- spectrum.

The weaker β^+ branch does not suffer from this problem, but its intensity is reduced compared to that of the β^- branch by a factor estimated to be about 500. Previous studies to determine the half-life of the β^+ decay mode were done by Sur *et al.* [14] and by da Cruz *et al.* [15]. In these experiments, the β^+ decay mode was to be identified through the observation of coincident 511-511 keV annihilation γ rays. These experiments used two 110-cm^3 germanium (Ge) detectors each surrounded by a 4π sodium iodide annular detector to search for the annihilation events. While neither experiment succeeded in observing this decay, da Cruz *et al.* [15] established a lower limit of 1.5×10^8 yr for the β^+ decay half-life of ^{54}Mn. In order to maximize their detection efficiency, these experimenters had to sandwich the source between the two Ge detectors. Because of this close geometry, a major limitation in these experiments was background and pileup produced by γ-ray Compton scattering from one Ge detector into the other. We have improved upon these previous searches by placing a relatively large source of ^{54}Mn into the GAMMASPHERE array of Compton-suppressed Ge detectors and searching for back-to-back 511-511 keV coincidences to determine the β^+ half-life.

The ^{54}Mn source material for this experiment was purchased from Isotopes Products Laboratories. Previous searches for the β^+ decay branch of ^{54}Mn have been hampered by the presence of minute amounts of positron-emitting contaminants such as ^{22}Na or ^{65}Zn in their source material. Thus, prior to the present β^+ decay search, we chemically purified the ^{54}Mn in order to remove any possible positron-emitting contaminants. To do so, a solution containing the ^{54}Mn was passed several times through AG1-X8 anion exchange resin columns pretreated with two different concentrations of HC1. The resulting ^{54}Mn was precipitated as manganese hydroxide. This precipitate was then centrifuged, decanted, redissolved, and reprecipitated 4 times in order to further purify it. The resulting manganese hydroxide was dried out and then sealed into the bottom of the plastic centrifuge tube for counting. The manganese hydroxide and its sealing material was thick enough to ensure that the positrons produced by the β^+ decay of ^{54}Mn stopped and annihilated within the source. The strength of the final source was determined to be 34.8 μCi by comparing the 835-keV γ-ray emission rate from this source to that observed from a calibrated ^{54}Mn standard.

The GAMMASPHERE array at the Lawrence Berkeley National Laboratory (LBNL) consists of 92 Compton-suppressed Ge detectors that surround and point inward toward a central source or target position [16]. In order to reduce the ambient background as much as possible, the normal GAMMASPHERE target chamber and beam line were removed for this experiment. Each Ge detector is approximately 275 cm^3 in volume and is surrounded by a hexagonally-shaped bismuth germanate (BGO) scintillator that is normally used to reject events in which a γ ray Compton scatters in the Ge crystal. Almost all of the Ge detectors are in pairs that are located diametrically opposite to one another. These pairs are ideally suited to searching for the back-to-back 511-511 keV γ rays produced by positron annihilation.

The present search consisted of 97 hours of counting the purified ^{54}Mn source and 61 hours of background counting with 43 pairs of Ge detectors. For all of these measurements, if two or more of the GAMMASPHERE Ge detectors fired within a time window of 1 μs and their immediately surrounding BGO shields did not fire, then the status of the entire array was read out and written to magnetic tape for subsequent off-line analysis. The information recorded for each event contains the energy signal in each Ge and each BGO element, the identification number of each element that fired, as well as the time when each element fired. The efficiency of this system for detecting back-to-back 511-511 keV coincidences was determined to be 2.97% by counting a calibrated 0.053-μCi source of ^{65}Zn and the ^{54}Mn source both placed at the source (target) position. An additional calibration test was performed by counting a calibrated 0.34-μCi source of ^{22}Na together with the ^{54}Mn at the source (target) position.

In the data analysis candidate positron annihilation events were initially identified by requiring that two back-to-back Ge detectors registered two γ rays in the range of 400–600 keV within a time window of 60 ns. Once an event containing this type of back-to-back γ-ray event was identified, the output of the entire GAMMASPHERE array was written into an ASCII file. The ASCII version of each event includes the number of Ge and BGO detectors fired, the universal time of the event, and the energy, position, and time of each Ge detector fired. All calibration data were also sorted in this same way. To minimize the risk of including positron annihilation events which might have been caused by isotopes other than ^{54}Mn, we restricted our final sort to two types of events: first, all of the events with exactly two Ge detectors (doubles) and second, all of the events with exactly three Ge detectors whenever the third detector registered the 835-keV γ ray (triples) within the 1 μs GAMMASPHERE trigger window. The second type of event was included in our analysis because of the high rate of random coincidences with the 835-keV γ ray;

that is, if we had accepted only twofold coincidences, we would have missed some back-to-back 511-511 keV events that happened to occur simultaneously with the detection of an 835-keV γ ray from an unrelated decay and that generated a threefold event.

From the time spectrum of ^{65}Zn calibration runs it was found that all "true" positron annihilation gammas occur in a time window of 15 ns and cover an energy range of 505.3 to 516.6 keV. Using the 15-ns time gate, a two-dimensional energy-energy array was formed. Within this array, the neighborhood of 511-511 keV was divided into 11.3-keV × 11.3-keV cells. To correct for random coincidences we formed another two-dimensional array in which the time gate was set off the time peak. After subtraction of randoms, in the data acquired with the ^{54}Mn source in place, we observed a total of 35 ± 8 events in the 511-511 cell of interest and an average of 6 events in the surrounding cells. In the background data, which were sorted in the same manner, all events had a multiplicity of seven or more and therefore did not contribute to the total number of events. Finally, we are left with a net 511-511 keV signal of 29 ± 9 events attributable to the ^{54}Mn source (Fig. 2). Five of these events are 511-511's that occurred within 1 μs of a random 835-keV γ ray. We call all events obtained by this method "double and triple Ge" events.

To decide if this signal is actually caused by the sought for β^+ decay of ^{54}Mn, it was necessary to check for other sources of 511-511 keV events. The only possible contaminants in our source that might produce similar signals are ^{22}Na and ^{65}Zn. While ^{65}Zn has a characteristic γ ray at 1115 keV (not in coincidence with positrons), the enormous rate of 835-keV γ rays emitted by the ^{54}Mn makes this small an activity level impossible to see via γ-ray singles counting. In order to determine if ^{65}Zn is contributing to our signal, we performed another chemical analysis of our sample after it had been counted. We redissolved the ^{54}Mn source in 2-M HC1 and ran it through a column of AG1-X8 anion exchange resin pretreated with 2-M HC1. Under such conditions, we had previously found that Zn sticks to the resin and can be quantitatively recovered by rinsing the column with H_2O. This was done, and the "Zn fraction" was counted at the LBNL Low Background Counting Facility. We searched for the characteristic 1115-keV line from ^{65}Zn. No peak was observed in a counting period of approximately 5 d, and a limit on the ratio of the ^{65}Zn activity to that of the ^{54}Mn in the original sample was established to be $\leq 2.3 \times 10^{-9}$. This implies that ^{65}Zn could contribute at most 1.5% to the signal we observed.

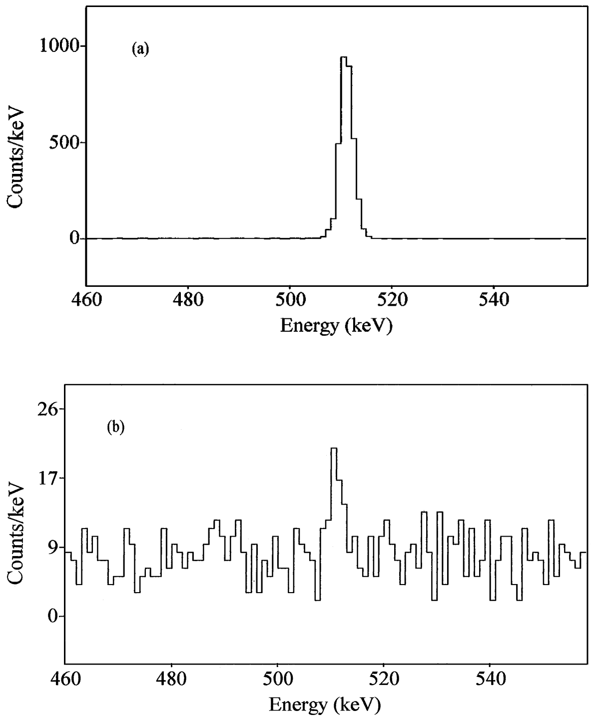

FIG. 2. Spectra of back-to-back coincident events seen in a germanium detector when 511 keV is observed in the opposite detector. (a) ^{65}Zn calibration source. (b) Background-subtracted ^{54}Mn data.

We have also examined our data carefully for the presence of ^{22}Na contamination by searching for 511-1275 keV and 511-511-1275 keV coincident events. We observed 2 ± 2 511-1275 events and zero 511-511-1275 events. To use these observations to determine the contribution of ^{22}Na to the 511-511 keV doubles and triples, we have measured the ratios of threefold events to twofold events from the ^{22}Na and ^{65}Zn calibration sources. By appropriately scaling these observations, we conclude that at most 4.7 events out of our total of 29 511-511 keV coincidences may be due to ^{22}Na. We therefore end up with a net signal of 24 ± 10 events that can be assigned to the β^+ branch of ^{54}Mn.

Based on these events, we find that the branching ratio for the β^+ decay of ^{54}Mn is $(2.2 \pm 0.9) \times 10^{-7}\%$. This corresponds to a partial half-life for β^+ decay of 3.9×10^8 yr and implies that the $\log ft$ for this second-forbidden-unique transition is 14.5. In reviewing the systematics of $\log ft$ values of nuclei that undergo both β^+ and β^- decay to neighboring ground states, we observe that in decays leading to nonclosed shell nuclei, $\log ft$ values in β^+ decays are generally 0.5 smaller than those in β^- decays for allowed transitions, while the two are roughly equal for forbidden transitions. Moreover, the $\log ft$ values in decays to closed-shell nuclei (such as the β^- decay to ^{54}Fe), are typically smaller by about 0.5. Since these two trends are essentially contradictory for the ^{54}Mn decay, we assume that the $\log ft$ values for the β^+ and β^- branches are equal. We correspondingly deduce that the β^- branch is $1.1 \times 10^{-4}\%$ (although our assumption about the equality of the $\log ft$ values may make this value uncertain by a factor of 2–3 larger or smaller), which implies that the cosmic-ray half-life of a bare ^{54}Mn nucleus is 7.6×10^5 yr. This result is in very good agreement with the value of $1-2 \times 10^6$ yr deduced by DuVernois [11] by combining his measurements of the isotopic composition of cosmic-ray manganese with the confinement time of 15×10^6 yr derived from the ^{10}Be, ^{26}Al, and ^{36}Cl chronometers. Thus it would appear that the confinement time of the iron-group cosmic rays is the same as that of the lighter cosmic-ray nuclei.

Direct measurements of the β^- decay branch of ^{54}Mn appear to be extremely difficult but could resolve the ambiguity introduced by the necessity to deduce the β^- $\log ft$ from our measured value for β^+. By counting the number of ^{54}Fe atoms produced as a result of the ^{54}Mn β^- decay, one could directly determine the rate for this process. Both accelerator mass spectrometry and neutron activation analysis appear to have the necessary sensitivity to determine the cosmic-ray half-life of ^{54}Mn.

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