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β^+ Decay and Cosmic-Ray Half-Lives of ^{143}Pm and ^{144}Pm

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ABSTRACT

The positron decay partial half-lives of ^{143}Pm and ^{144}Pm are needed to assess the viability of elemental Pm as a cosmic-ray clock. We have conducted experiments to measure the β^+ branches of these isotopes; we find β^+ branches of $<5.7\times10^{-6}$ for ^{143}Pm and $<8\times10^{-7}$ for ^{144}Pm. Though these branches are a factor of 20 lower than the previous experimental limits, the resulting partial half-lives are still too uncertain to permit any firm conclusions.

1. INTRODUCTION

The mean confinement time of cosmic rays within the Galaxy can be determined by comparing the cosmic-ray abundances of suitably long-lived radioactive isotopes to their stable neighbors. For heavy elements the current state of the art in cosmic-ray mass measurement is not sufficient to resolve a radioactive isotope from its immediate neighbors. This difficulty prompted Drach and Salamon (Drach and Salamon, 1987) to consider the possibility of using the *elemental* abundances of Tc and Pm, which have no stable isotopes, as cosmic-ray clocks. They were able to make reasonable estimates of the cosmic-ray half-lives of ^{95}Tc and ^{96}Tc, based on the average value of log ft for all well-established second-forbidden non-unique transitions (see also Hindi *et al.*, elsewhere in these proceedings). For ^{144}Pm, however, the relevant β^+ decay is a third-forbidden non-unique transition, and there was only one transition of similar forbiddenness on which to base the β^+ half-life. Accordingly the half-life estimates for Pm were too uncertain to permit any conclusions about the suitability of Pm as a cosmic ray clock. The current experimental limits on the β^+ partial half-lives of ^{143}Pm and ^{144}Pm are an order of magnitude lower than the most conservative lower limits considered by Drach and Salamon. If these half-lives turn out to be (much) longer than the mean confinement time of the cosmic rays, then all the (cosmic-ray) Pm isotopes would be electron-capture-decay-only isotopes and their abundance could then be used as a probe of cosmic-ray acceleration and of density variations of the medium traversed by the cosmic rays (Drach and Salamon, 1987, and references therein). Prompted by these considerations, we have attempted to improve on the current experimental limits on the β^+ branching ratios of ^{143}Pm and ^{144}Pm.

2. EXPERIMENTAL PROCEDURE

2.1 Source Preparation

The ^{143,144}Pm source was produced by bombarding a 0.25-mm-thick Pr foil with a beam of 30-MeV α particles from Lawrence Berkeley Laboratory's 88-Inch Cyclotron. The beam current was 4 μA and the duration of the bombardment was 5 hours. Two weeks after the irradiation, the target was dissolved in concentrated HCl; a few drops of concentrated HNO_3 were added and the solute was run through an anion exchange column of AG1-X8 resin. The Pm was precipitated from the resulting solution with HF. Two samples were prepared, one which was counted at that point, and another which was counted approximately three years later.

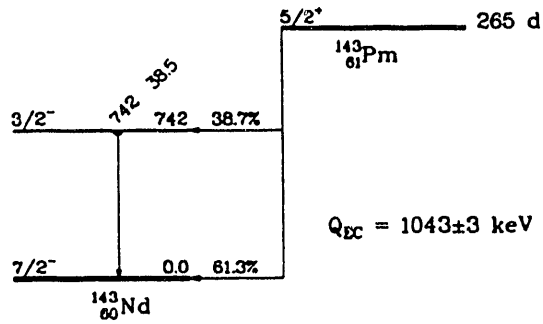

Fig. 1: Decay scheme of ^{143}Pm. Level energies are in keV.

The setup and procedure for the two sets of measurements were very similar. Here we give the details of the experimental procedure for the last set measurements.

2.2 Decay Rate Measurements

The decay schemes of ^{143}Pm and ^{144}Pm are shown in Figs. 1 and 2, respectively. For ^{143}Pm we sought to measure the positron decay rate to the ground sate by recording coincidences between the back-to-back 511-keV annihilation photons; for ^{144}Pm we sought to measure the β^+ decay rate to the 697-keV state of ^{144}Nd by recording coincidences between the back-to-back 511-keV annihilation photons and the 697-keV γ ray. The source was placed between two intrinsic Ge (Gamma-X) detectors placed face to face. The Ge detectors were positioned at the center of an 8.25-cm hole in a 30x30-cm NaI detector. To reduce the count rate due to Nd x rays a 1 mm Cu absorber was placed in-front of each of the Ge detectors. The attenuation of Nd x rays also prevented the summing of the 477-keV γ ray (with the Nd x rays) into the 511-keV region. To reduce coincidences due to γ ray scattering from one detector into another, 3 mm-thick Pb sheets were wrapped around the Ge detectors and placed in between them.

The energy and fast timing signals for Ge-Ge and Ge-Ge-NaI coincidences were digitized and recorded event-by-event on magnetic tape for later analysis. The source was counted in the above configuration for 14.5 days; after that period the source was removed and background was counted for 6.8 days.

The efficiency of the system for detecting back-to-back 511-keV photons and 511-511-697 coincidences was determined using calibrated ^{22}Na, ^{60}Co, and ^{137}Cs sources. The resulting efficiency for 511-511 coincidences was (1.37±0.12)%, and that for 511-511-697 coincidences was (0.389±0.034)%.The absolute decay rates of the ^{144}Pm and ^{143}Pm activities were determined in separate singles runs with the source at a distance of ≈15 cm from the detector. Figure 3 shows a sample singles spectrum of the source. For these singles runs the efficiency of the detector was obtained using calibrated ^{133}Ba, ^{60}Co and ^{152}Eu sources. At the start of the last set of coincidence measurements the activity of ^{144}Pm was (0.180±0.005) μCi and that of ^{143}Pm was (0.526±0.032) μCi. For the first set of measurements the activity of ^{144}Pm was (1.4±0.1) μCi and that of ^{143}Pm was (8.1±0.5) μCi.

3. DATA ANALYSIS AND RESULTS

The magnetic tapes were replayed to generate five two dimensional (2D) coincidence histograms: (1) NaI-Ge gated by 511-keV γ rays (detected in the other Ge detector), (2) NaI-Ge gated by 618-keV γ rays, (3) NaI-Ge gated by 697-keV γ rays, (4) Ge-Ge, and (5) Ge-Ge vetoed by the NaI detector. From the three NaI-Ge 2D spectra one can then obtain (project) NaI spectra gated by two γ rays (one detected in each Ge detector), one with an energy of 511, 618, or 697 keV, and the other with any desired energy *E*. Figure 4 shows NaI spectra gated by (a) 511-511 (b) 511-495, (c) 511-527, and (d) 618-477-keV γ rays. Figure 4 (d) demonstrates the ability to isolate the 697-keV γ ray in the NaI by detecting it in coincidence with accompanying radiation. The spectrum plotted in figure 4(a) shows no conspicuous evidence for the 697-keV γ ray gated by two 511 keV γ rays. The peak at 1274 keV in this spectrum arises from a small (42±4 pCi) ^{22}Na contaminant in the source. The

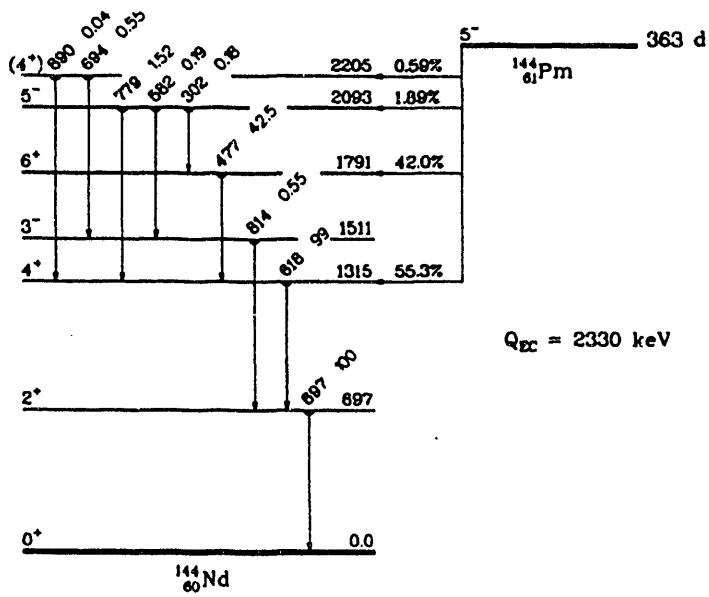

Fig. 2: Decay scheme of ^{144}Pm. Level energies are in keV. The weak (6×10^{-4}%) 1396-keV line from the 2093 (5^-) to the 697 (2^+) states is not shown.

lines below ≈ 1000 keV are not associated with two 511-keV γ rays, but rather with the continuum falling within the two 511-keV gates in the Ge detectors (fig. 3). This is demonstrated by figs. 4(b) and 4(c), which show that essentially the same lines appear when one of the Ge gates is set, respectively, below and above the 511-keV region. The continuum in the 511-keV region arises from Comptons of the 697-keV γ ray (Compton edge at 509 keV) and γ rays of higher energy produced in the ^{144}Pm decay, and from the summing of the 477-keV or the 618-keV γ ray with the Comptons of any of the other γ rays produced in the cascade.

A NaI spectrum in coincidence with true 511-511 coincidences (*i.e.*, with the contribution of the continuum underneath the 511's subtracted out) was generated. The area of the 697-keV peak was extracted from a fit with a Gaussian plus linear background. The resulting β^+ branch for ^{144}Pm is $(0.9\pm1.3)\times10^{-6}$.

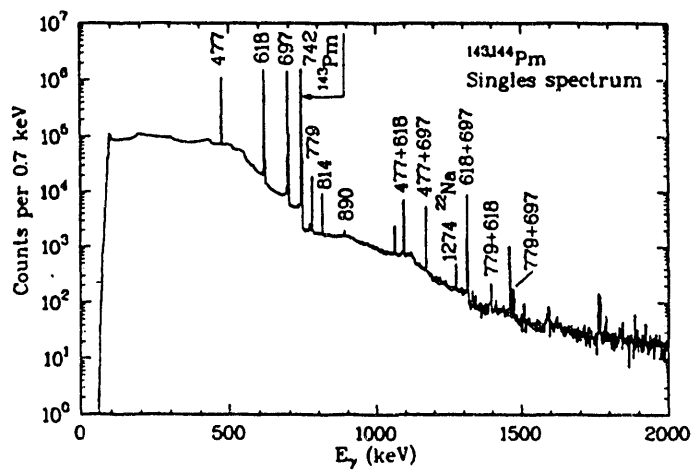

Fig. 3: A sample singles spectrum of the ^{143,144}Pm source.

The β^+ branch for ^{143}Pm was extracted from the number of 511-511 coincidences recorded in the Ge detectors, vetoed by the NaI. After subtracting the remaining contribution of the ^{22}Na contaminant and the background 511-511 coincidences, we obtain a β^+ branch of $(4.9\pm5.6)\times10^{-7}$.

The β^+ branches extracted from this weak source confirm the results obtained from the first set of measurements, which were $<5.7\times10^{-8}$ for ^{143}Pm and $<8\times10^{-7}$ for ^{144}Pm. For ^{143}Pm the limit is determined by background and unvetoed (contaminant) ^{22}Na 511-511 coincidences; since the relative contribution of these was smaller for the stronger source, the extracted limit was correspondingly lower. For ^{144}Pm, on the other hand, the limit is dominated by coincidences between Comptons of γ rays which arise form ^{144}Pm decay, (i.e., the "background" scales with the source strength) and hence the stronger source did not give a correspondingly lower limit.

From the lower set of limits on the β^+ branches one obtains β^+ partial half-lives of $> 1.3\times10^7$ yr for ^{143}Pm and $> 1.2\times10^6$ yr for ^{144}Pm. Although our limits on the branches are approximately a factor of 20 lower than the previous experimental limits (Varga, Berenyi, et al., 1967), the resulting β^+ half-lives are still too uncertain to establish Pm as a good cosmic-ray clock, or as an EC-decay-only cosmic-ray element.

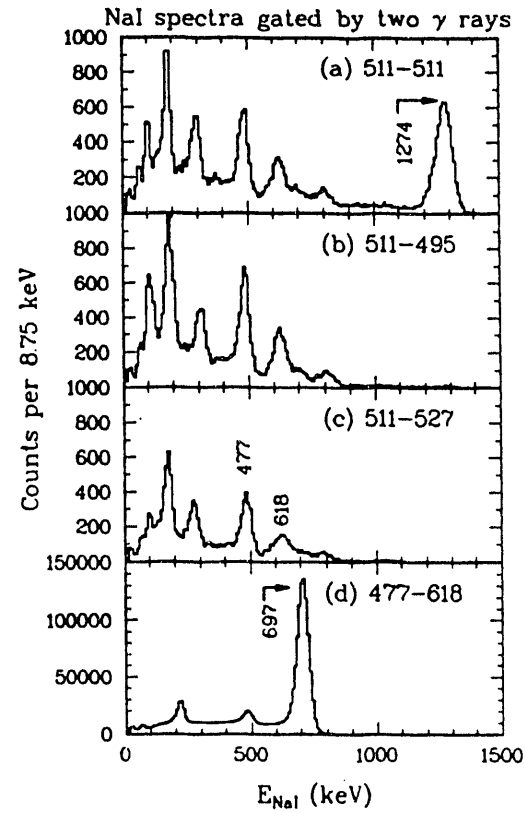

Fig 4: Spectra recorded in the NaI detector, in coincidence with gamma rays in each of the two Ge detectors. The gamma ray energies in the Ge detectors are (in keV): (a) 511-511, (b) 511-495, (c) 511-527, and (d) 477-618.

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