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Possibility of pions emitted in the spontaneous fission of ^{252}Cf

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We have performed a search for spontaneous fission of ^{252}Cf accompanied by neutral pions. We have placed an upper limit of one such decay per 10^9 fissions at the 90% confidence level.

The provocative suggestion of pionic radioactivity made by Ion, Ivascu, and Ion-Mihai[1] is supported by their considerations of the energetics of the process and by phase-space arguments based on statistical ideas proposed by Fermi.[2,3] Two-body pionic radioactivity, i.e., the spontaneous creation and emission of pions from the nuclear ground state, is forbidden by conservation of energy and momentum, but three- or more-body decay modes are possible. The total energy release in the fission of the heaviest elements exceeds 200 MeV, sufficient to create pions. This energy appears mainly in fragment kinetic energy, arising from Coulomb repulsion of the fission fragments. For pions to be created, the total energy release in normal two-body spontaneous fission, the fission Q value, must be reduced by the pion rest mass (134.96 MeV for a π^0). Pion emission is energetically possible for all elements with Z greater than about 80 where the two-body fission Q values exceed the pion rest mass.

At present, the dynamical details of how pion-accompanied spontaneous fission, with substantially reduced fragment kinetic energies, can actually occur are obscure. In the absence of detailed information, the application of statistical theory by Ion and co-workers[1] is appropriate for estimating the likelihood of this process. These authors have shown, as might be expected intuitively, that the probability for pion-accompanied spontaneous fission increases with increasing fission Q value. Thus the largest pion decay branching ratios are expected for the very heaviest elements, where the fission Q values are the largest. Ion and co-workers[1] have also given expressions for the pion momentum spectrum assuming statistical equilibrium in the three-body decay.

Inspired by Ref. 1 and by discussions with similarly inspired researchers at Los Alamos National Laboratory,[4] we have performed an experimental search for this decay mode in the spontaneous fission of 2.646-yr ^{252}Cf. This nuclide has a 3.1% branch for decay by spontaneous fission. Of all spontaneously fissioning nuclides available in sufficient quantity to perform a sensitive search for pion-accompanied fission, ^{252}Cf has the largest statistical branching factor for pion emission according to the estimates in Ref. 1.

Approximately 9 ng of isotopically pure (<99.99%) ^{252}Cf were electrodeposited onto a stainless-steel hemispherical target plate over an area of 0.125 cm^2. This target plate was incorporated into a fast fission-ionization chamber initially developed at Oak Ridge National Laboratory for the purpose of permitting fission measurements on intensely alpha-active isotopes like ^{252}Cf.[5,6] This chamber uses the "limited transverse range" principle to improve the discrimination between fission fragments and alpha particles. In this geometry, the ionization registered by the counter comes only from the initial portion of the particle track, where the energy deposited in the counter gas is much greater for fission fragments than for alphas. With the fast electronics designed especially for this chamber,[6] subnanosecond time resolution for fission fragments was realized; alpha-particle pileup pulses could not be discerned above electronic noise. The question of fission fragment detection efficiency arises because pion accompanied fission would be associated with reduced fragment kinetic energies. We note that the chamber is a ΔE detector, and, since fragment dE/dx values are primarily a function of velocity, not energy, the slightly reduced ΔE pulses from pion accompanied events would not have biased our experiment.

The chamber was filled to 2 atm of pure CH_4. The ^{252}Cf source gave approximately 5500 fissions/s. An additional target plate was placed in the ionization chamber containing 24 μg of ^{239}Pu to serve as an indication of induced fission activity, for example, from cosmic rays. No events above electronic noise that could be mistaken for fission fragments were detected from the ^{239}Pu source plate during the four-day data acquisition period.

Neutral pions decay with a mean life of 8.7×10^{-17} s, predominantly (98.799%) via the two-photon decay mode. Decay of a π^0 at rest produces a pair of back-to-back (180° correlated) 67.5 MeV photons. For near-symmetric fission of ^{252}Cf accompanied by π^0 emission, the maximum possible π^0 kinetic energy is about 97 MeV (for zero kinetic energy fragments). For this unrealistic limiting case, the correlation angle between the two photons would be reduced from 180° to approximately 70°.

The assembled ionization chamber had a total volume of approximately 200 cm^2 including the fast preamplifier/amplifier electronics. It was mounted in the center of the Holifield Heavy Ion Research Facility (HHIRF) spin spectrometer which served as the π^0 detector. The spin spectrometer, described in Refs. 7 and 8, is a 4π arrangement of 72 NaI(Tl) detectors approximating a hollow sphere with an inner radius of 17.8 cm and an outer radius of 35.5 cm. For our experiment, only 63 of the full complement of 72 detectors were available. The spin spectrometer enabled us to detect pairs of high energy photons in prompt time coincidence, measure both photon energies, and determine the correlation angle between the photons. The photon energies together with the correlation angle form an invariant mass relationship which should be centered about the π^0 rest mass for π^0 decay events:

$$M(\pi^0) = 2(E_{\gamma_1}E_{\gamma_2})^{1/2}\sin(\theta/2) ,$$

where E_{γ_1} and E_{γ_2} are the energies of the two detected photons and θ is the angle between them.

We have modeled the response of the NaI(Tl) elements of the spin spectrometer to 65–70 MeV photons which originate in the center of the array using the considerations outlines in Refs. 9–11. The 17.8-cm radial thickness of the detectors will contain >85% of the longitudinal electromagnetic shower generated by these photons. When the energy deposited in adjacent detectors is also included, >90% of the total energy should be deposited in active NaI(Tl). Each of the 63 NaI(Tl) detectors had a constant-fraction discriminator; for high energy photons, $E_\gamma > 20$ MeV, subnanosecond timing was achieved.

Event triggers were generated by the appearance of a fission event in the ion chamber, either from the ^{252}Cf or from the ^{239}Pu, in coincidence (± 100 ns) with any of the 63 NaI(Tl) detectors where the energy deposit exceeded 10 MeV. For these trigger events, we recorded in list mode the pulse height and the time relative to the fission event for all of the NaI(Tl) detectors, and the fission pulse height in the ion chamber. Most of the list mode events were histogrammed on-line to provide real-time event monitoring. For example, an on-line two-dimensional display of events in invariant mass versus correlation angle was provided for those events where two or more high energy photons ($E > 10$ MeV) were detected. Data acquisition was limited to four days because of the intensive use of the spin spectrometer in the HHIRF accelerator-based research program. More than 2×10^9 fission events were recorded in our experiment.

Analysis of the list-mode data was accomplished after the experiment was completed. Cluster sum energies were formed by finding the largest NaI pulse height present in an event and, adding to it any pulse heights in surrounding detectors. The next largest remaining pulse was then located and a cluster sum formed about it, and so on until all detectors which triggered were used. Only events containing exactly two clusters were considered for further processing. Vector sum pulse heights were generated for each cluster, as based on the known geometry, and used to calculate the correlation angle θ between two cluster events.

Cosmic-ray events were a major source of background. Minimum-ionizing cosmic-ray particles traversing one of the NaI detectors along its long dimension deposits ~60 MeV. This is uncomfortably close to the energy of a single photon expected from the decay of a π^0. We show in Fig. 1 the two-dimensional invariant mass versus correlation angle for all events observed in our experiment. These events correspond to the detection of two or more high energy events with energy deposit greater than 15 MeV occurring within the fission trigger time window of ± 100 ns. Cosmic-ray events are clearly observable in the region with invariant mass < 100 MeV and with small photon correlation angles, $\theta < 50°$. This region is outside the acceptable range for π^0 decay events from ^{252}Cf.

Exploiting the subnanosecond time resolution of the

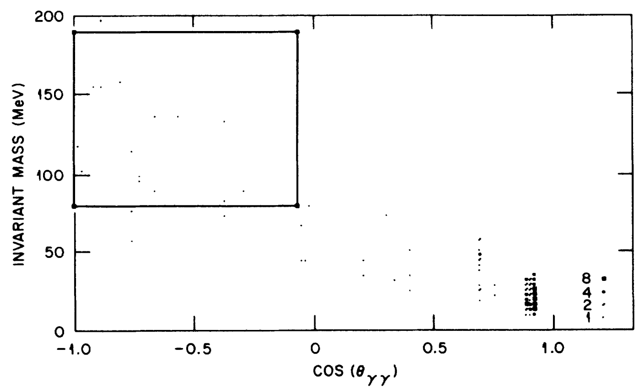

FIG. 2. Same as Fig. 1 but excluding cosmic-ray events by the requirement that the arrival times of the two-detector events be within 1.25 ns of each other.

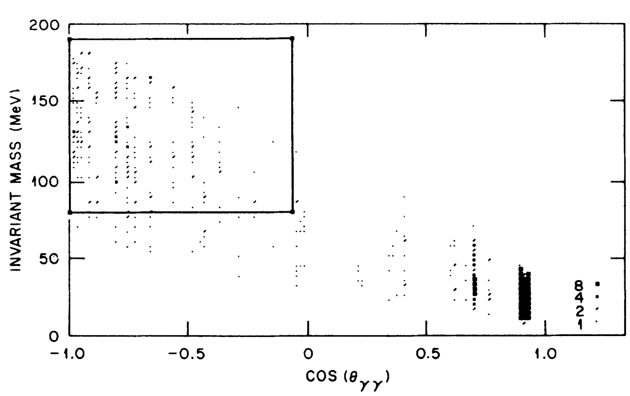

FIG. 1. Two-dimensional map in invariant mass vs correlation angle for all correlated two-detector events within ± 100 ns of a fission trigger. The outline marks the region expected for π^0 decay events associated with the spontaneous fission of ^{252}Cf. The limits correspond to invariant masses from 85 to 185 MeV and correlation angles from 92° to 180°.

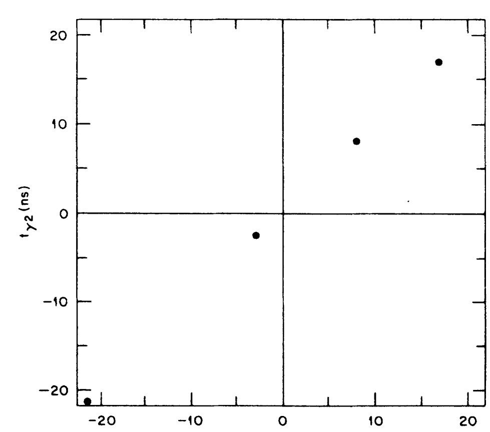

FIG. 3. Time distribution of correlated ($\Delta t < 1.25$ ns) two-detector events relative to the occurrence of the fission trigger event. No candidate events remain within the prompt time window of ± 2 ns about $t = 0$.

spin spectrometer for high energy events, we then required that the time difference between pulses in two associated photon detectors be less than 1.25 ns. This criterion provided an effective bias against cosmic rays since the transit time for particles with $v = c$ across the spin spectrometer, and passing thru its center, corresponds to a mean arrival time difference of 1.78 ns between the two opposite NaI(Tl) detectors. For the events excluded by the application of this criterion, we found that the detector in the upper hemisphere of the spin spectrometer was triggered first, as expected for cosmic rays. The events in the two-dimensional map of invariant mass versus correlation angle meeting the equal-time requirement are shown in Fig. 2. Clearly, the spin spectrometer works quite effectively in this "self-shielding" mode as we are able to exclude most cosmic-ray events within the generous time window of the fission trigger event (± 100 ns). In Fig. 3, we show the time distribution of these remaining events (candidates for pion-accompanied fission) relative to the time of the fission trigger. These events are distributed in a random fashion. When the criterion of "prompt coincidence" with the fission trigger (± 2 ns) is applied, we have no candidate events, that is, zero events which could be taken for π^0 decay events occurring in the center of the spin spectrometer and associated with a fission decay of ^{252}Cf.

Applying the interval distribution of Poisson statistics, we deduce that the spontaneous fission decay associated with π^0 emission is $< 1 \times 10^{-9}$ of the normal spontaneous fission decay of ^{252}Cf at the 90% confidence level ($< 1.5 \times 10^{-9}$ at the 95% confidence level). It might be possible with the technique used here to reduce this upper limit by a factor of about 100, for example, by increasing the source strength by a factor of 5 and running 20 times longer. To reduce the limit still further, a more effective veto of cosmic rays, for example, with a high efficiency anticoincidence shield, would be required. This is not a simple matter for a device as large as the spin spectrometer, but the effort and the expense could be justified if there were sufficient theoretical interest and a realistic prediction of the expected branching ratio as based on a plausible dynamic model.

Note added in proof. A pion branching ratio, relative to fission, of $< 5 \times 10^{-9}$ has recently been reported [C. Cerruti *et al.*, Z. Phys. A **329**, 383 (1988)].

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