β-Delayed Neutron Spectroscopy Using Trapped Radioactive Ions

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A novel technique for β-delayed neutron spectroscopy has been demonstrated using trapped ions. The neutron-energy spectrum is reconstructed by measuring the time of flight of the nuclear recoil following neutron emission, thereby avoiding all the challenges associated with neutron detection, such as backgrounds from scattered neutrons and γ rays and complicated detector-response functions. $^{137}I^+$ ions delivered from a ^{252}Cf source were confined in a linear Paul trap surrounded by radiation detectors, and the β-delayed neutron-energy spectrum and branching ratio were determined by detecting the β^- and recoil ions in coincidence. Systematic effects were explored by determining the branching ratio three ways. Improvements to achieve higher detection efficiency, better energy resolution, and a lower neutron-energy threshold are proposed.

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The β^- decay of neutron-rich nuclei often populates excited states in daughter nuclei, and when these states are above the neutron-binding energy they can de-excite by γ-ray or neutron emission, with the latter process identified as β-delayed neutron (βn) emission. The properties of βn emission are important to both the pure and applied nuclear physics communities [1]. Neutron-emission branching ratios are needed to determine how short-lived neutron-rich isotopes synthesized in the astrophysical r process decay back to stability [2–7]. Both neutron branching ratios and energy spectra are required for nuclear reactor kinetics calculations for reactor safety studies [1,8–10] and are important for future Generation IV reactor designs [11]. Delayed-neutron measurements aid in the understanding of the nuclear structure of neutron-rich nuclei [12–15] and are needed to improve nuclear-structure models [16] and empirical predictions [17] used to determine the properties of nuclei for which no data exist. High quality data also have the potential to help determine neutron-capture rates [18,19] for neutron-rich isotopes needed to understand the nonequilibrium phase of r-process nucleosynthesis [20–22] and to support the stockpile stewardship mission [23,24].

Historically, βn detection has dealt with significant experimental compromises, namely, the detection of neutrons with high efficiency or modest energy resolution, but not both. Furthermore, much of the βn data have large uncertainties [25,26], and recent measurements have revealed discrepancies as large as factors of 2–4 [2,12], warranting further experimental investigation.

In this work, a new technique is demonstrated which circumvents the challenges associated with neutron detection by instead studying the nuclear recoil. In concert with facilities discussed in Ref. [27] to provide the requisite intense ion beams, improvements to βn measurements can be made.

Ion traps have revolutionized mass spectrometry and have the potential to do so for decay spectroscopy as well. These devices can confine cooled radioactive ions to a $\approx 1\ \mathrm{mm}^3$ volume in vacuum, where they decay nearly at rest. The emitted radiation emerges from the trap with negligible scattering, and therefore the nuclear recoil can be studied. Recent measurements using atom traps [28–30] and ion traps [31] have inferred the neutrino momentum from β-recoil-ion coincidence measurements. A similar approach can be applied to perform βn spectroscopy from the β-recoil-ion coincidence time of flight (TOF). Here, neutron emission leads to high-energy recoils having short TOFs, with the lower-energy recoil imparted by the leptons being a small perturbation to the measurement. As conservation of momentum allows for the reconstruction of one unobserved particle, the study of two-neutron emission is also possible but requires the detection of the additional neutron by other means.

The recoil-ion technique offers several promising advantages over conventional neutron-detection techniques. It yields TOF spectra with a near-Gaussian response, avoiding the spectral unfolding techniques typically required to extract the neutron-energy spectrum from a complicated detector response [32,33]. Neutron-energy resolutions approaching 3% full width at half-maximum (FWHM) and total intrinsic detection efficiencies of $\gtrsim 60\%$ are achievable. Backgrounds from scattered neutrons and γ rays, a challenge in traditional neutron detection, are avoided entirely because they have significantly shorter TOFs than the nuclear recoils. To verify the control of systematic effects, the βn branching ratio (P_n) can be obtained by comparing the higher-energy recoil ions characteristic of neutron emission to (1) the lower-energy recoil ions following β decay, (2) β-delayed γ rays emitted by the isotope being studied, and (3) β singles.

A proof-of-principle experiment was conducted by studying a standard, well-known βn precursor, ^{137}I ($t_{1/2} = 24.5 \pm 0.2$ s [34], $Q_\beta = 6027 \pm 8$ keV [35], $P_n = 7.33 \pm 0.38\%$ [11]). Fission fragments from a $\approx$ 1-mCi ^{252}Cf spontaneous fission source were thermalized in a large-volume gas catcher [36], extracted, then bunched and further cooled using a radio-frequency (rf) quadrupole ion guide [37]. Only singly charged ions with a mass of 137 u were selected using a timed deflection pulse and a He buffer gas-filled Penning trap [38] and delivered to the Beta-decay Paul Trap (BPT), an open-geometry linear Paul trap described in Ref. [39]. The trap was operated with time-varying voltages of the form $V_{\mathrm{rf}} \cos(2\pi f t)$ with $V_{\mathrm{rf}} = 200$ V and $f = 264$ kHz. The stability condition of the trap [39] was chosen such that $\geq 2+$ ions (and therefore all β^--decay daughters) were not confined.

A plastic scintillator ΔE-E telescope and metal-anode chevron microchannel plate (MCP) were used for β and recoil-ion detection, respectively, as shown in Fig. 1, and characterized in detail in Ref. [40]. The total detection efficiency for β-recoil-ion coincidences was $\approx 0.05\%$. The ΔE detector, which only has a small ($\approx 1\%$) detection efficiency for γ rays and neutrons, was used to identify β particles in coincidence with recoil ions. The telescope was separated from the vacuum by a beryllium window

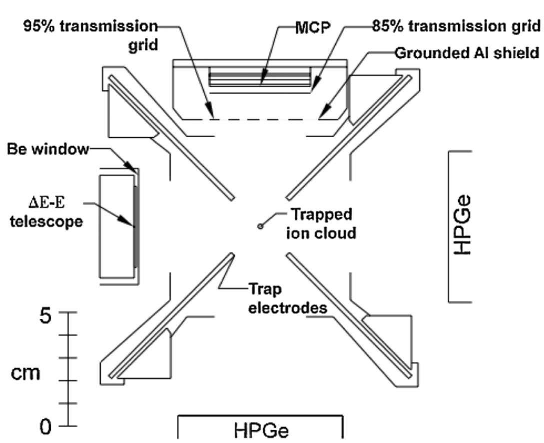

FIG. 1. End-on view of the BPT and detectors.

providing a 150-keV threshold for β detection. Gamma rays were detected using 80% and 140% relative-efficiency high-purity germanium (HPGe) detectors.

Ions were captured in the BPT every 5 s, accumulated for 145 s, and then ejected toward a diagnostic silicon detector that monitored the contents of the trap. The trap was left empty for a period of 5, 25, or 40 s at the end of each cycle to assess backgrounds. Trap contents were also monitored by detecting a single peak from the 1218-keV ($I_\gamma = 12.8 \pm 1.3\%$) and 1220-keV ($I_\gamma = 3.5 \pm 0.4\%$) γ rays emitted following ^{137}I β decay [41] and the 455-keV ($I_\gamma = 31 \pm 3\%$) γ ray emitted following ^{137}Xe β decay [34] in coincidence with β particles. The ratio of ^{137}I to ^{137}Xe was consistent with the independent yields from ^{252}Cf fission [42]. Although no known γ ray from ^{137}Te β decay ($t_{1/2} = 2.49 \pm 0.05$ s, $P_n = 2.99 \pm 0.16\%$ [34]) was observed, an amount consistent with the independent yield (after correction for decay losses during the ion preparation) was assumed to also be present. The buildup of activity in the trap was consistent with the ^{137}I $t_{1/2}$, implying a trap storage $t_{1/2}$ of >220 s, and therefore $\gtrsim 93\%$ of the ^{137}I decays in the trap. A trapping efficiency of $\gtrsim 60\%$ was achieved for ions entering the BPT.

The β^- decay of ^{137}I yields ^{137}Xe ($t_{1/2} = 229.08 \pm 0.78$ s, $S_n = 4025.56 \pm 0.10$ keV [35]) ions with recoil energies <170 eV unless a neutron is emitted. Emission of a neutron with energy E_n following β decay yields ^{136}Xe ions with recoil energies of $\frac{E_n m_n}{m_{136}}$, where m_n and m_{136} are the masses of the neutron and the ^{136}Xe ion, respectively. As E_n can extend to 1987 ± 8 keV [35,43], ^{136}Xe ions are expected to have energies up to 14.6 keV. However, the average neutron energy is 530 ± 50 keV [34], yielding an average βn-recoil-ion energy closer to 3.9 keV. Most of the daughter ions emerging from the trap are expected to have charge state 2+, as β-decay studies have shown that $\approx 80\%$ of the daughter ions retain all the orbital electrons and the emission of a neutron is expected to result only in limited additional ionization (see Ref. [40] and references therein). Simulations indicate that, for recoil ions characteristic of neutron emission, the differences between charge states 2+ through 5+ can be neglected, as they have a $<1\%$ effect on both the fraction of ions that reaches the MCP detector and the TOF. Internal conversion could result in ions having higher charge states [44], but no significant conversion has been observed in ^{137}I β^- decay [34].

The measured TOF difference between the β^- and the recoil ion is shown in Fig. 2. Scattering of β particles or γ rays between the β telescope and MCP detector produces a prompt coincidence that determines $t = 0$. The βn recoils have TOFs > 0.44 μs. The broad peak at TOFs $\gtrsim 2$ μs corresponds to recoil ions following the β^- decay of ^{137}I, ^{137}Te, and ^{137}Xe. Spurious peaks at very short times incompatible with recoil-ion TOFs resulted from the electrical pickup observed by both β^- and recoil-ion detectors.

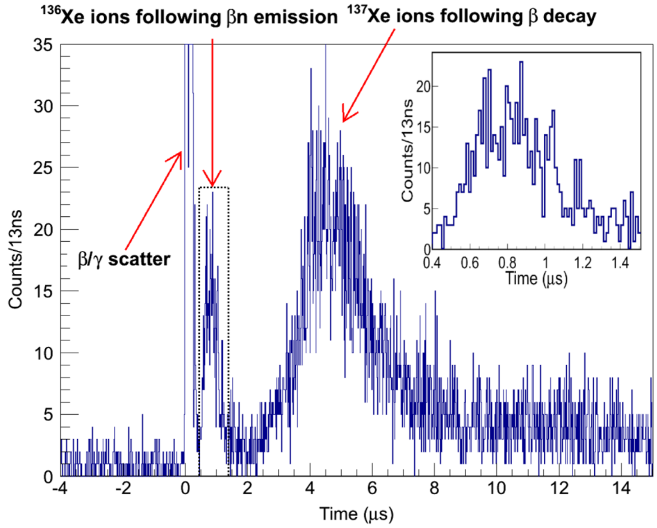

FIG. 2 (color online). Recoil-ion TOF spectrum collected with a 30 ion/s $^{137}I^+$ beam. The TOF spectrum of the ^{136}Xe recoil ions from βn emission, highlighted by the dotted box, is shown in the inset.

For decays of trapped $^{137}I^+$, $P_n = \frac{N_{\beta 136}}{N_\beta}$, where $N_{\beta 136}$ is the total number of decays resulting in ^{136}Xe recoil ions and N_β is the total number of β decays. $N_{\beta 136}$ is determined from $\frac{n_{\beta 136}}{f \varepsilon_{\beta 136} \Omega_{136} \varepsilon_{136}}$, where $n_{\beta 136}$ is the number of recoil ions observed in the time window 0.44–1.38 μs (corresponding to 200–2000-keV neutrons), $\varepsilon_{\beta 136}$ is the β detection efficiency for these events, Ω_{136} is the fraction of ^{136}Xe ions that hits the MCP detector active area, ε_{136} is the ^{136}Xe intrinsic recoil-ion detection efficiency, and $f = 92.5 \pm 2.5\%$ is the fraction of the βn spectrum expected to fall in this energy window based on previous studies of ^{137}I [45–48]. The 200-keV neutron-detection threshold, limited only by the larger than necessary electric fields for this ion trap, was conservatively selected to ensure that the βn spectrum was not contaminated with events from recoil ions from β decay to the ground state or γ-ray emitting states. N_β was determined three ways by measuring recoil ions (N_β^r), β-delayed γ rays (N_β^γ), and β singles (N_β^β).

N_β^r is given by $\frac{n_{\beta 137}}{\varepsilon_{\beta 137} \Omega_{137} \varepsilon_{137} (1 - P_n)}$ where $n_{\beta 137}$ is the number of ^{137}Xe recoil ions observed, $\varepsilon_{\beta 137}$ is the β detection efficiency for these events, Ω_{137} is the fraction of ^{137}Xe ions that hit the MCP detector active area, and ε_{137} is the ^{137}Xe intrinsic recoil-ion detection efficiency. Corrections were applied for the recoil ions expected from the β decay of $^{137}Xe^+$ and $^{137}Te^+$ ions in the trap and the expected number of βn recoils in this time window.

N_β^γ is determined from $\frac{n_{\beta\gamma}}{\varepsilon_\gamma I_\gamma \varepsilon_{\beta 137}}$, where $n_{\beta\gamma}$ is the number of β-γ coincidences from β-delayed γ rays at 1218 and 1220 keV from ^{137}I decay, ε_γ is the γ-ray detection efficiency, and I_γ is the absolute γ-ray intensity.

N_β^β is given by $\frac{n_\beta}{\varepsilon_{\beta 137}}$, where n_β is the number of observed β's from trapped ^{137}I, accounting for backgrounds from ^{137}Te and ^{137}Xe β decay, untrapped ^{137}I β decay, and radiation from the room.

The intrinsic detection efficiency of an MCP approaches the open-area ratio for ions with $\gtrsim 2$ keV of kinetic energy [49–52], regardless of species or charge state [53,54]. Here, the recoil ions are all charge state 2+ or higher and strike the MCP well above the detection threshold with kinetic energies >5.3 keV, ensured by the −2.65 kV potential applied to the MCP face. Therefore, ε_{136} is assigned the open-area ratio value of 0.60 ± 0.03 and $\frac{\varepsilon_{136}}{\varepsilon_{137}} \approx 1$.

Detailed Monte Carlo simulations were developed to interpret the results. A β-decay code adapted from Ref. [30] was used to generate β^- and recoil-ion spectra for ^{137}I, ^{137}Xe, and ^{137}Te decays. Although the ^{137}I β decay to the ground state ($45.2 \pm 0.5\%$ of the total [34]) and many of the hundreds of transitions to excited states at energies below S_n are likely first forbidden, there are essentially no data to determine potential deviations from allowed spectra and the complicated decay scheme is likely incomplete. For these decays, the lepton momenta were generated from an allowed distribution and γ-ray cascades were approximated as consisting of one or two isotropically emitted γ rays. For decays to excited states above the neutron-separation energy of ^{137}Xe, prior experimental results [47] are consistent with calculations based on the gross theory of β decay [55] that indicate that, for ^{137}I, $\approx$ 75%–80% are expected to be allowed. For these allowed decays, no correlation between the β particle and neutron momentum is expected. Any potential anisotropic neutron emission in the remainder of the decays is anticipated to be an effect smaller than the ~10% experimental uncertainty of this work.

The ratio $\frac{\varepsilon_{\beta 137}}{\varepsilon_{\beta 136}}$ was determined to be 1.24 ± 0.02 (as decays to the excited states that can lead to βn emission yield lower-energy β particles) by propagating β^- particles through a detailed model of the BPT in GEANT4 [56]. The uncertainty is based on the reliability of the GEANT4 model, the β detection threshold, and the ^{137}I decay scheme [34].

The values of Ω_{137} and Ω_{136} were determined to be $1.39 \pm 0.10\%$ and $2.96 \pm 0.04\%$, respectively, by propagating the recoil ions through the electric fields of the trap using SimIon 3D version 8.0 [57] for decays where the β particle hits the ΔE detector. Recoil ions with $\lesssim 500$ eV of energy are especially susceptible to perturbations by the rf fields. Unperturbed $^{137}Xe^{2+}$ ions that would otherwise have a drift time of $\gtrsim 4.2$ μs can give rise to TOF events as short as ≈ 3.2 μs. With the β and recoil-ion detectors at right angles, the recoil-ion detection efficiency is only mildly dependent on the details of the β-decay kinematics. The sensitivity to the rf amplitude, ion cloud size, and details of the β decay were folded into the uncertainty of the recoil-ion detection efficiency.

TABLE I. Summary of the ^{137}I βn branching ratios.

Method	P_n (%)
(1) Low-energy recoil ions	6.80 ± 0.88
(2) β-delayed γ rays	6.88 ± 1.05
(3) β singles	6.95 ± 0.76
2011 IAEA evaluation [11]	7.33 ± 0.38

The P_n values determined from the three approaches all share the measurement of the high-energy recoil ions and are summarized in Table I. The largest source of uncertainty in method (1) is from Ω_{137}, which is sensitive to the details of the β decay and the electric field in the trap. The largest uncertainty in method (2) is the 10% uncertainty in the γ-ray intensity [41], while the P_n measured in method (3) is limited by the $\approx$ 6% statistical uncertainty in $n_{\beta 136}$.

The βn-energy spectrum, shown in Fig. 3, was reconstructed from the velocity of ^{136}Xe recoil ions using conservation of momentum. As the recoil ions from neutron emission are only minimally perturbed by the electric fields, the velocity can be determined simply from the average distance to the MCP and the TOF. The broadened TOF response from the recoil imparted by the leptons and the impact of the rf fields was determined for recoil ions from monoenergetic neutrons from 200 to 1500 keV using SimIon. The measured recoil-ion TOF spectrum was corrected for the β detection efficiency determined from the GEANT4 simulations, after the flat background from accidentals was subtracted. The ^{137}Te βn-energy spectrum is not known but is expected to contribute only $\approx$ 3% of the total βn counts. In Fig. 3, the βn-energy spectrum of ^{137}I determined here is in excellent agreement with the results of Refs. [45–48], if convoluted with the energy resolution ($\approx$ 10%) of this measurement.

A novel method for studying β-delayed neutron-energy spectra and branching ratios has been demonstrated by measuring the large momentum kick imparted to the

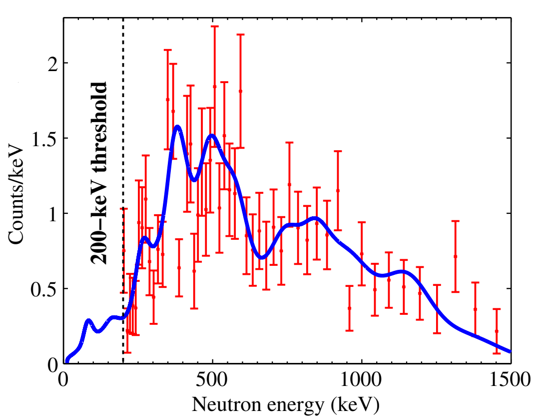

FIG. 3 (color online). Comparison of the βn-energy spectrum for ^{137}I measured here with a known spectrum from Ref. [47] that has been convoluted with the energy resolution currently obtained from the recoil ions (shown by the solid line).

nucleus following the β decay of trapped ^{137}I. The coincident detection efficiency can be increased to $\approx$ 2% (a factor of $\approx$ 40 larger than in this work) by placing more β and MCP detectors in the available space around the trap. This would allow measurements to be performed on ion beams as weak as 0.1–1 ion/s. Results from multiple detector angle combinations can determine any anisotropies that could arise from forbidden transitions.

The neutron-energy resolution can be improved to $\approx$ 3% FWHM by better determining the ion trajectories using position-sensitive MCPs. The impact of the rf electric fields, which impart some energy to the low-energy recoil ions, can be reduced by bringing the electrodes closer to the trap center so that a smaller voltage can generate the same trapping potential. With less perturbation to the recoil ions from β decay to the ground state and γ-ray emitting states, these ions will have longer TOFs. For most βn precursors, nearly background-free measurements of the neutron spectrum can be performed to energies as low as 25–50 keV, ultimately limited at an energy where the neutron and lepton recoils are comparable.

Significantly higher statistics can be collected at a fission-fragment beam facility such as the Californium Rare Ion Breeder Upgrade (CARIBU) [58], where isotopes with half-lives as short as $\approx$ 50 ms can be studied. In addition to fission fragments, this approach can be used to study a variety of isotopes produced at isotope separator on-line or fragmentation facilities where a stopped beam infrastructure is available. The presence of contaminant isobars in the trapped ion sample can be avoided by using a high-resolution double-focusing magnetic spectrometer, purifier Penning trap, or reflectron to ensure that only the desired isotope is delivered to the βn spectroscopy trap. This experimental approach can also be adapted for use with laser traps for the elements that can be efficiently collected and confined [59], where isobaric and even isomeric purity is guaranteed and the electric field can be tailored to the needs of the measurement. By implementing the aforementioned improvements, the technique will be capable of collecting βn spectra with high efficiency, excellent energy resolution, and low neutron-energy thresholds, while avoiding many of the complications and limitations of existing methods.

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