This is the accepted manuscript made available via CHORUS. The article has been published as:

β-Delayed Neutron Spectroscopy Using Trapped Radioactive Ions
Phys. Rev. Lett. 110, 092501 — Published 26 February 2013
DOI: 10.1103/PhysRevLett.110.092501
$\beta$-delayed neutron spectroscopy using trapped radioactive ions

R. M. Yee,1, 2 N. D. Scielzo,1 P. F. Bertone,3 F. Buchinger,4 S. Caldwell,3, 5 J. A. Clark,3 C. M. Deibel,3, 6 J. Fallis,3, 7 J. P. Greene,3 S. Gulick,4 D. Lasca,3, 8 A. F. Levand,3 G. Li,3, 4 E. B. Norman,2 M. Petretti,1 G. Savard,3, 5 R. E. Segel,8 K. S. Sharma,3, 7 M. G. Sternberg,3, 5 J. Van Schelt,3, 5 and B. J. Zabrisky3

1 Physical and Life Sciences Directorate, Lawrence Livermore National Laboratory, Livermore, California 94550, USA
2 Department of Nuclear Engineering, University of California, Berkeley, California 94720, USA
3 Physics Division, Argonne National Laboratory, Argonne, Illinois 60439, USA
4 Department of Physics, McGill University, Montréal, Québec H3A 2T8, Canada
5 Department of Physics, University of Chicago, Chicago, Illinois 60637, USA
6 Joint Institute for Nuclear Astrophysics, Michigan State University, East Lansing, Michigan 48824, USA
7 Department of Physics and Astronomy, University of Manitoba, Winnipeg, Manitoba R3T 2N2, Canada
8 Department of Physics and Astronomy, Northwestern University, Evanston, Illinois 60208, USA

(Dated: January 23, 2013)

A novel technique for $\beta$-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 $\gamma$ 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 $\beta$-delayed neutron energy spectrum and branching ratio were determined by detecting the $\beta^-$ 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.

PACS numbers: 29.30.Hs, 26.30.Hj

The $\beta^-$ decay of neutron-rich nuclei often populate excited states in daughter nuclei, and when these states are above the neutron-binding energy they can de-excite by $\gamma$-ray or neutron emission, with the latter process identified as $\beta$-delayed neutron ($\beta$n) emission. The properties of $\beta$n 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 exists. High quality data also has the potential to help determine neutron-capture rates [18, 19] for neutron-rich isotopes needed to understand the non-equilibrium phase of r-process nucleosynthesis [20–22] and to support the stockpile stewardship mission [23, 24].

Historically, $\beta$n 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 $\beta$n 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 $\beta$n 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 $\approx 1$ 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 $\beta$-recoil ion coincidence measurements. A similar approach can be applied to perform $\beta$n spectroscopy from the $\beta$-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 max-
imum (FWHM) and total intrinsic detection efficiencies of $\gtrsim 60\%$ are achievable. Backgrounds from scattered neutrons and $\gamma$ 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 $\beta n$ 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 $\beta$ decay, (2) $\beta$-delayed $\gamma$ rays emitted by the isotope being studied, and (3) $\beta$ singles.

A proof-of-principle experiment was conducted by studying a standard, well-known $\beta n$ 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 $V_{RF} \cos(2\pi ft)$ with $V_{RF} = 200$ V and $f = 264$ kHz. The stability condition of the trap [39] was chosen such that $\gtrsim 2^+\text{ ions (and therefore all } \beta^-\text{ decay daughters) were not confined.}$

A plastic scintillator $\Delta E$-$E$ telescope and metal-anode chevron micro-channel plate (MCP) were used for $\beta$ and recoil-ion detection, respectively, as shown in Fig. 1, and characterized in detail in Ref. [40]. The total detection efficiency for $\beta$-recoil ion coincidences was $\approx 0.05\%$. The $\Delta E$ detector, which only has a small ($= 1\%$) detection efficiency for $\gamma$ rays and neutrons, was used to identify $\beta$ particles in coincidence with recoil ions. The telescope was separated from the vacuum by a beryllium window providing a 150-keV threshold for $\beta$ detection. Gamma rays were detected using 80% and 140% relative-efficiency high-purity germanium detectors (HPGe).

Ions were captured in the BPT every 5 s, accumulated for 145 s, then ejected towards 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 ($I_\gamma = 3.5 \pm 0.4\%$) $\gamma$ rays emitted following $^{137}$I $\beta$ decay [41] and the 455-keV ($I_\gamma = 31 \pm 3\%$) $\gamma$ ray emitted following $^{137}$Xe $\beta$ decay [34] in coincidence with $\beta$ particles. The ratio of $^{137}$I to $^{137}$Xe was consistent with the independent yields (IV) from $^{252}$Cf fission [42]. Although no known $\gamma$ ray from $^{137}$Te $\beta$ 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 IV (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 $\beta^-$ 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 $< 160$ eV unless a neutron is emitted. Emission of a neutron with energy $E_n$, following $\beta$ decay yields $^{136}$Xe ions with recoil energies of $E_{m_n}/m_{130}$, where $m_n$ and $m_{130}$ are the masses of the neutron and $^{136}$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 $530 \pm 50$ keV [34], yielding an average $\beta n$ 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 $\beta$-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 [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 reach 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 $\beta^-$ decay [34].

The measured TOF difference between the $\beta^-$ and the recoil-ion is shown in Fig. 2. Scattering of $\beta$ particles or $\gamma$ rays between the $\beta$ telescope and MCP detector can produce a prompt coincidence that determines $t = 0$. The $\beta n$ recoils have TOFs $> 0.44 \mu$s. The broad peak at TOFs $\gtrsim 2 \mu$s corresponds to recoil ions following the $\beta^-$ decay of $^{137}$I, $^{137}$Te, and $^{137}$Xe. Spurious peaks at very short times incompatible with recoil-ion TOFs resulted from electrical pick-up observed by both $\beta$ and recoil-ion detectors.
For decays of trapped $^{137}$I, $P_n = \frac{N_{\beta\gamma}}{N_{\beta}}$ where $N_{\beta\gamma}$ is the total number of decays resulting in $^{136}$Xe recoil ions, and $N_{\beta}$ is the total number of $\beta$ decays. $N_{\beta\gamma}$ is determined from $\int_{x_{\beta\gamma}}^{x_{\beta}} n_{x_{\beta\gamma}} \, dx_{\beta\gamma}$, where $n_{x_{\beta\gamma}}$ is the number of recoil ions observed in the time window 0.44–1.38 $\mu$s (corresponding to 200–2000 keV neutrons). $\varepsilon_{\beta\gamma}$ is the $\beta$ detection efficiency for these events, $\Omega_{137}$ is the fraction of $^{136}$Xe ions that hit the MCP detector active area, $\varepsilon_{\beta}$ is the $^{136}$Xe intrinsic recoil-ion detection efficiency, and $f = 92.5\pm2.5\%$ is the fraction of the $\beta n$ 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 $\beta n$ spectrum was not contaminated with events from recoil ions from $\beta$ decay to the ground state or $\gamma$ ray emitting states. $N_{\beta\gamma}$ was determined three ways by measuring recoil ions ($N_{\beta\gamma}^0$), $\beta$-delayed $\gamma$ rays ($N_{\beta\gamma}^\gamma$), and $\beta$ singles ($N_{\beta\gamma}^\beta$).

$N_{\beta\gamma}$ is given by $\frac{n_{\beta\gamma}}{\varepsilon_{\beta}}$, where $n_{\beta\gamma}$ is the number of $\beta$-delayed $\gamma$ rays emitted in the remainder of the decays is anticipated to be an effect smaller than the $\sim10\%$ experimental uncertainty of this work.

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

The values of $\Omega_{137}$ and $\Omega_{136}$ 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 $\beta$ particle hits the $\Delta E$ detector. Recoil ions with $\gtrsim 500$ 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 $\gtrsim 4.2 \mu$s can give rise to TOF events as short as $\approx 3.2 \mu$s. With the $\beta$-recoil ion detectors at right angles, the recoil-ion detection efficiency is only mildly dependent on the details of the $\beta$-decay kinematics. The sensitivity to the RF amplitude, ion cloud size, and details of the $\beta$ decay were folded into the uncertainty of the recoil-ion detection efficiency.

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 $\Omega_{137}$, which is sensi-
The βn-energy spectrum, shown in Figure 3, was reconstructed from the velocity of $^{139}$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 mono-energetic neutrons from 200 keV 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 is subtracted. The $^{137}$Te βn energy spectrum is not known, but is expected to contribute only ≈ 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 (≈ 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 nucleus following the β decay of trapped $^{137}$I. The coincident detection efficiency can be increased to ≈ 2% (a factor of ≈ 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/sec. Results from multiple detector angle combinations can determine any anisotropies that could arise from forbidden transitions.

The neutron-energy resolution can be improved to ≈ 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 ≈ 50 ms can be studied. In addition to fission fragments, this approach can be used to study a variety of isotopes produced at ISOL or fragmentation facilities where 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.

We thank P. A. Vetter for lending the β and MCP detectors, C. J. Lister for assistance with the HPGe detectors, and S. G. Prussin for fruitful discussions on βn spectroscopy. This work was supported by U.S. DOE under Contracts DE-AC02-06CH11357 (ANL), DE-AC52-07NA27344 (LLNL), DE-FG02-98ER41086 (Northwestern U.), NSERC, Canada, under Application No. 216974 and the Department of Homeland Security. This material is based upon work supported by the National Science Foundation Graduate Research Fellowship under Grant No. DGE-0668477. R. M. Yee acknowledges support

<table>
<thead>
<tr>
<th>Method</th>
<th>$P_n$ (%)</th>
</tr>
</thead>
<tbody>
<tr>
<td>(1) Low-energy recoil ions</td>
<td>6.80 ± 0.88</td>
</tr>
<tr>
<td>(2) β-delayed γ rays</td>
<td>6.88 ± 1.05</td>
</tr>
<tr>
<td>(3) β singles</td>
<td>6.95 ± 0.76</td>
</tr>
<tr>
<td>2011 IAEA evaluation</td>
<td>7.33 ± 0.38</td>
</tr>
</tbody>
</table>

**TABLE I. Summary of $^{137}$I βn branching ratios.**
from the Lawrence Scholar Program at LLNL and the Berkeley Nuclear Research Center.