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Spin-trap isomers in deformed, odd-odd nuclei in the light rare-earth region near $N = 98$

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Masses of neutron-rich, odd-odd Pm, Eu, and Tb nuclei near $N = 98$ were measured using the Canadian Penning Trap mass spectrometer at the Californium Rare Isotope Breeder Upgrade (CARIBU) facility. High-resolution mass measurements yielded the discovery of spin-trap isomers at $N = 97$ in ^{162}Tb, and in the $N = 99$ isotones of ^{160}Pm and ^{164}Tb. Furthermore, no evidence of long-lived isomers were observed at $N = 95$ in ^{158}Eu, at $N = 97$ in ^{158}Pm, nor at $N = 101$ in ^{164}Eu and ^{166}Tb. These experimental observations are compared to results from multiquasiparticle blocking calculations.

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In axially symmetric deformed nuclei the projection of the total angular momentum of individual nucleons onto the symmetry axis Ω is approximately a good quantum number. By breaking pair(s) of protons (π) or neutrons (ν), or both, high-K ($K = \sum \Omega_i$) multiquasiparticle states can be found at relatively low excitation energies, which are often long-lived since the depopulating transitions proceed between levels with significantly different K values [1–3].

A somewhat different mechanism can be attributed for the formation of low-lying isomers in deformed odd-odd nuclei, where for a given two-quasiparticle configuration, two states can be formed due to the effect of residual proton-neutron interactions, one with $K_{>}=\Omega_\pi + \Omega_\nu$ and the other with $K_{<}=|\Omega_\pi - \Omega_\nu|$. Which of the two states will be lowest in energy depends on the intrinsic spin projection of the unpaired proton and neutron, and the ordering is governed by the so-called Gallagher-Moszkowski rule [4]. When orbitals with large Ω are near both the neutron and proton Fermi surfaces, the $K_<$ and $K_>$ states can have a large spin difference, and, as a consequence, the connecting γ-ray transition can be significantly hindered, thus leading to the formation of long-lived, spin-trap isomers. A feature of such isomers is the low-multiplicity of the depopulating γ-ray cascade, which coupled with the long lifetimes ($T_{1/2} > 100$ ms), makes time-correlated, γ-ray spectroscopy studies an impractical method for their discovery and characterization. However, high-resolution Penning trap mass measurements in conjunction with β-decay spectroscopy studies have proven to be a powerful tool for the discovery of isomers and the elucidation of their properties as recently demonstrated by Hartley *et al.* [5].

The observation of isomers and their characterization can provide valuable information on single-particle excitation energies and on the spin-dependent residual interactions between unpaired nucleons, which are an essential input for understanding the structure of deformed, odd-odd nuclei [6,7]. This is particularly relevant for neutron-rich nuclei in the deformed, light rare-earth region, given their involvement in the formation of the elemental abundance peak near A $\approx$ 165 by the rapid neutron-capture process (see, for example, Ref. [8] and references therein), where knowledge is limited owing to the paucity of experimental nuclear data.

In this work, we report on new high-resolution mass studies of neutron-rich, odd-odd Pm, Eu, and Tb nuclei in the light rare-earth region with the intention of characterizing the presence of long-lived isomerism near $N = 98$. Long-lived isomers were discovered in ^{160}Pm and ^{162,164}Tb and their excitation energies were determined from the measured masses. These isomers are interpreted as spin traps and the revelation of their structure is aided by predictions made from multiquasiparticle blocking calculations using a deformed Woods-Saxon potential and the Lipkin-Nogami prescription

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of the pairing correlations. The observation of isomers in the $N = 99$ isotones, but not in the $N = 101$ cases provide evidence for deficiencies in the theoretical models to correctly predict the ordering of single-particle levels in deformed nuclei above $N = 98$.

The mass measurements were conducted using the Canadian Penning Trap mass spectrometer (CPT) [9] located in the low-energy experimental area of the Californium Rare Isotope Breeder Upgrade (CARIBU) facility [10] at Argonne National Laboratory. Ions of neutron-rich rare-earth nuclei — originating from the spontaneous fission of a ≈ 1 Ci ^{252}Cf source held inside a large-volume gas catcher — were extracted at a charge state of 2^+, filtered by A/q, and collected in a radio-frequency (rf) quadrupole cooler/buncher before being injected into a multireflection time-of-flight mass separator (MR-TOF) [11] for further beam purification. Highly mass-resolved beams ($R = m/\Delta m > 100\,000$) were then delivered to the CPT where the phase-imaging ion-cyclotron-resonance (PI-ICR) technique [12] was used to directly measure the cyclotron frequencies (ν_c) of trapped ions. Masses were then determined by measuring the ν_c of a reference ion with a similar A/q and a previously well-established mass, here one of $C_6H_6^+$ or $^{84}Kr^+$.

In PI-ICR, ν_c is determined by measuring the accumulated phase advance (ϕ_{adv}) of a trapped ion's orbital motion inside the Penning trap over a period of time where no rf excitations are applied — referred to here as the *accumulation time* (t_{acc}). The cyclotron frequency and ionic mass (m_{ion}) are related to these quantities by

$$\nu_c = \frac{qB}{2\pi m_{ion}} = \frac{\phi_{adv}}{2\pi t_{acc}}, \quad (1)$$

where q is the ion's charge and B is the magnetic field strength at the center of the Penning trap. Further details of the PI-ICR mass measurement procedure at the CPT and a discussion of this method's systematic effects can be found in Refs. [13,14]. One of the main advantages of PI-ICR over the previously employed time-of-flight ion-cyclotron-resonance (TOF-ICR) technique is a $\approx$20-fold increase in achievable mass resolution (up to $R \approx 25\,000\,000$). This improvement not only facilitates accurate discrimination from nearby isobaric contamination, but also makes PI-ICR the ideal tool to search for low-lying, long-lived isomeric states. This has already been demonstrated by the CPT for the cases of ^{146}La, ^{163}Gd [13], and ^{160,162}Eu [5]. Building upon the success of these results, the masses of several neutron-rich odd-odd Pm, Eu, and Tb nuclei were measured and the results are summarized in Table I.

The general measurement procedure used in this work was to first identify the isotope of interest and neighboring isobars by scanning the MR-TOF mass-selection gate and measuring the ν_c of the transmitted ions using short accumulation times (typically $t_{acc} < 50$ ms). After a positive identification, beam purification in the MR-TOF was optimized and several more ν_c measurements with gradually increasing accumulation times (up to ≈ 700 ms) were performed. As the Penning trap resolving power increases, the ν_c of any newly appearing beam inhabitants were measured and either identified as molecular contamination or labeled as candidate isomeric states. The nature of the molecular contamination typical to CARIBU beams of light rare-earth nuclei is well understood from recent CPT campaigns of high-resolution mass measurements in this region [14]. A bloated majority of these contaminants are molecular combinations of neutron-rich fission fragments with simple hydrocarbons or water. Confirmation of such a molecule then involves the identification of an isotopic analog molecule in neighboring mass number beams with a population fraction in line with the predicted fission yields of the radioactive component. Finally, a precision ν_c measurement of the isotope of interest was conducted according to the procedure outlined in Ref. [13]. Sample ν_c measurement results for ^{156,158,160}Pm, ^{158,160,162,164}Eu, and ^{162,164,166}Tb are shown in Fig. 1. Isomeric states in ^{156,160}Pm, ^{160,162}Eu, and ^{162,164}Tb are evident from the resolved double-peaked structure within such a narrow ν_c domain. The relative size of the two peaks is indicative of the approximate population fractions of each state found in each respective beam.

To demonstrate the accuracy of measuring small excitation energies with the CPT using the PI-ICR technique, the case of ^{156}Pm was chosen as it is easily produced at CARIBU and has a known long-lived isomer [15]. Previously, the mass of ^{156}Pm was measured by the CPT using TOF-ICR [16], but due to the inherently limited mass resolution the isomeric state was not observed. With the PI-ICR technique this isomer was trivially resolved and an excitation energy of 152.2(27) keV was determined, in agreement with the known value of 150.3(1) keV [15]. Also reported in Ref. [16] were the first mass results of ^{158,160}Eu and ^{158}Pm, which were measured to a precision of ≈ 20 keV. In the present work, a search for isomers was performed in the cases of ^{158}Eu and ^{158}Pm but none were observed. However, from this process the masses of both nuclei were again determined and agreement with the previous CPT data set [16] was attained, while the mass uncertainties have been improved tenfold. Recently, isomeric states in ^{160,162}Eu were discovered from PI-ICR mass measurements with the CPT and were independently confirmed through β-decay spectroscopy measurements [5], together with earlier studies on ^{162}Eu [17]. We also discovered an isomer in ^{162}Tb ($N = 97$) with an excitation energy of 285.5(32) keV. Owing to the large excitation energy of this isomer, a relatively short t_{acc} of just 125 ms was used to adequately resolve the two states. Consequently, the peaks in the ^{162}Tb panel of Fig. 1 are somewhat broader than the others due to the reduced resolving power. The only previous mass information for ^{162}Tb was from β-decay endpoint measurements [18–20] culminating in a mass value that lies between the two states found here.

Masses of ^{158,160}Pm, ^{162}Eu, and ^{164}Tb were also recently measured at JYFLTRAP using the TOF-ICR technique [21]. In comparison to the present results, agreement is only found for the mass of ^{158}Pm. However, in contrast to the present work, no isomers were previously reported in ^{160}Pm, ^{162}Eu, or ^{164}Tb — presumably due to the lower sensitivity and mass resolution achievable with the TOF-ICR approach. In fact, the JYFLTRAP mass values for these three nuclei are found to lie between these determined for the ground and isomeric states in the present work, which is indicative that unresolved admixtures of two states were reported in Ref. [21]. The β-decay half-lives of ^{160}Pm, ^{162}Eu, and ^{166}Tb were also

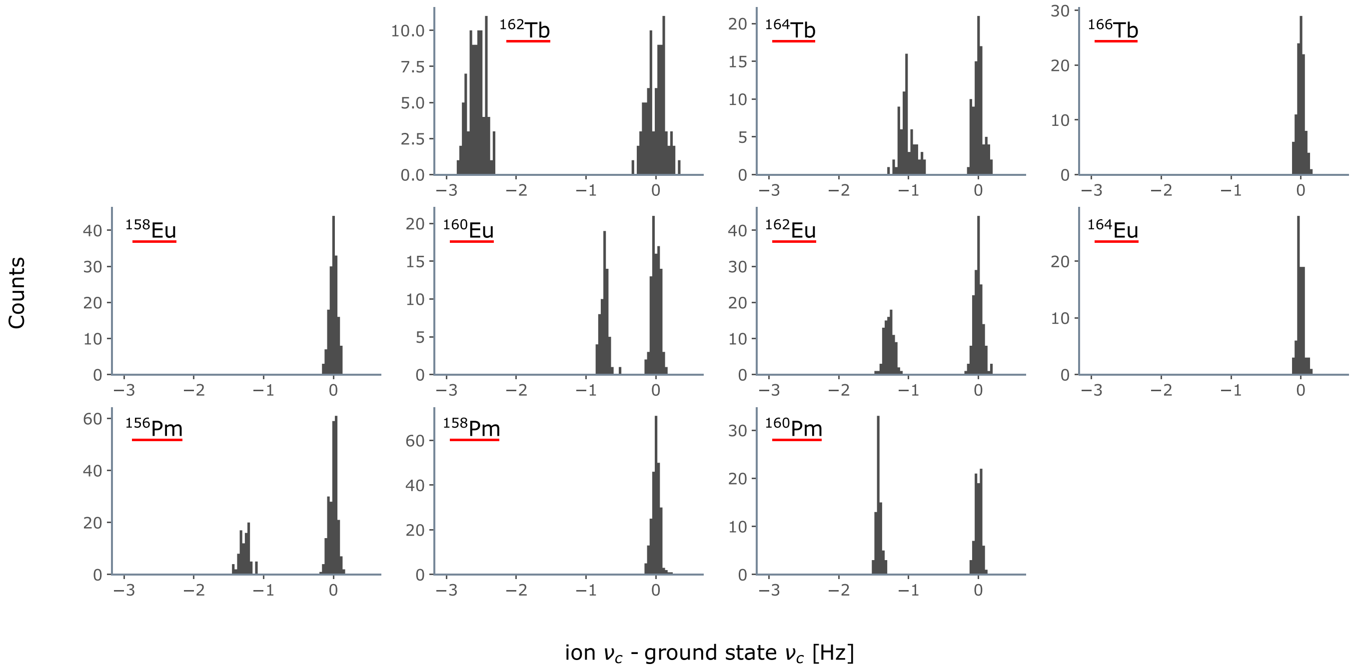

FIG. 1. Sample measurements of ν_c for each nuclide discussed in this work. Each panel is a histogram of calculated cyclotron frequencies for every ion detected in a single PI-ICR measurement relative to the average ν_c of the ground state within a 3-Hz window, corresponding to $\approx$ 350 keV in this mass region. Spectra with two peaks indicate the presence of both a ground state and a long-lived isomer.

measured at the RIKEN facility [22], but no long-lived isomers were reported in these nuclei. Because of limited statistics, only beta spectra were used in the analysis of these data sets and, therefore, they were not able to distinguish between the ground-state and isomer decays. More recently, another set of rare-earth masses were reported by JYFLTRAP [23] including a remeasurement of ^{162}Eu utilizing PI-ICR where they now find agreement with the CPT result from Ref. [5].

In the $N = 101$ cases of ^{164}Eu and ^{166}Tb, cyclotron frequencies were measured with accumulation times ranging between 230 and 245 ms. No isomeric states were observed in these two nuclei and the masses are in good agreement with the new values reported in Ref. [23].

To understand the structure of the observed states, multi-quasiparticle blocking calculations, similar to those reported in Ref. [5], were carried out. The energies of single-particle levels were initially taken from the Woods-Saxon potential with "universal" parameters [24] and deformation parameters β_2, β_4, and β_6 from Ref. [25], but their ordering was modified to approximately reproduce the experimental one-quasiparticle states in the neighboring odd-A nuclei. The most striking difference between the theoretical and empirical sets of single-particle levels is the inversion of the $\nu 1/2[521]$ and $\nu 7/2[633]$ orbitals at $N = 99$, and the $\nu 3/2[521]$ and $\nu 5/2[642]$ orbitals at $N = 95$. The pairing correlations were treated using the Lipkin-Nogami prescription [26] with fixed strengths of $G_\pi = 23.5/A$ MeV and $G_\nu = 15.5/A$ MeV and included the effect of blocking. The predicted favored configurations, corresponding quantum numbers, and excitation energies for the ground and isomeric states in the nuclei under study are given in Table I. The excitation energies were derived following the prescription of Ref. [27] and the Gallagher-Moszkowski splitting energies of Ref. [28].

In the present work, the observed isomers in the odd-odd Pm, Eu, and Tb isotopes can be interpreted as spin traps and associated with the $K_>$ and $K_<$ states within the same configuration. Specifically, their structure can be understood as a coupling of the proton $\pi 5/2[532]$ $(Z = 61)$, $\pi 5/2[413]$ $(Z = 63)$, or $\pi 3/2[411]$ $(Z = 65)$ orbital with the neutron $\nu 3/2[521]$ $(N = 95)$, $\nu 5/2[523]$ $(N = 97)$, $\nu 7/2[633]$ $(N = 99)$, or $\nu 1/2[521]$ $(N = 101)$, as schematically shown in Fig. 2.

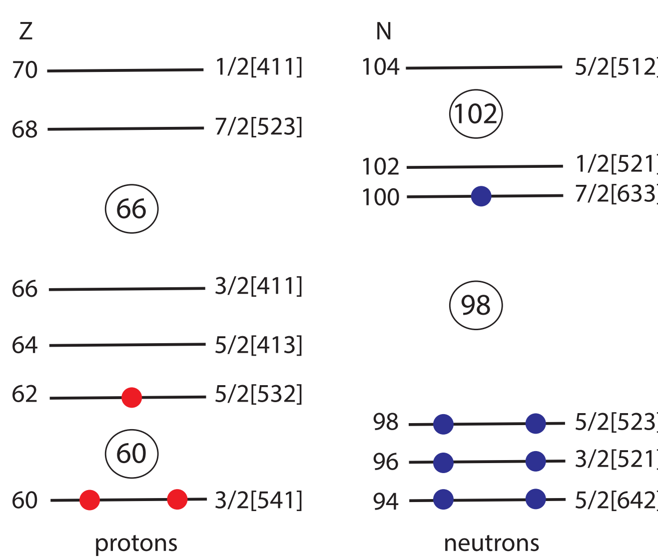

FIG. 2. Empirical single-particle levels near the proton and neutron Fermi surface for ^{160}Pm.

TABLE I. Cyclotron frequency ratios ($\nu_c^{\rm ref}/\nu_c$) and mass excesses (ME) for the ground and isomeric states measured by the CPT. The excitation energies (E_x) for the observed isomers are given alongside the predicted K values, spins, and parities (K^π, I^π), configurations, and excitation energies.

		Measured			Predicted		
Nuclide	Ref. Ion	$\nu_c^{\rm ref}/\nu_c$	ME [keV]	E_x [keV]	K^π, I^π	Configuration	E_x [keV]
N = 95							
^{156}Pm^{2+}	$C_6H_6^+$	0.998 957 107 (16)	−64168.5 (23)	0.0	4^+, 4^+	$\pi5/2[532]\otimes\nu3/2[521]$	0
^{156m}Pm^{2+}	$C_6H_6^+$	0.998 958 154 (10)	−64016.3 (15)	152.2 (27)	1^+, 1^+	$\pi5/2[532]\otimes\nu3/2[521]$	214
^{158}Eu^{2+}	$C_6H_6^+$	1.011 748 662 (14)	−67270.6 (20)	0.0	1^-, 1^-	$\pi5/2[413]\otimes\nu3/2[521]$	0
^{158m}Eu^{2+}					4^-, 4^-	$\pi5/2[413]\otimes\nu3/2[521]$	110
N=97							
^{158}Pm^{2+}	$C_6H_6^+$	1.011 804 811 2 (69)	−59106.6 (10)	0.0	0^+, $(0^+, 1^+)$[b]	$\pi5/2[532]\otimes\nu5/2[523]$	0
^{158m}Pm^{2+}					5^+, 5^+	$\pi5/2[532]\otimes\nu5/2[523]$	110
^{160}Eu^{2+}	^{84}Kr$^+$	0.952 978 970 2 (58)[a]	−63493.4 (9)	0.0	5^-, 5^-	$\pi5/2[413]\otimes\nu5/2[523]$	0
^{160m}Eu^{2+}	^{84}Kr$^+$	0.952 979 565 4 (51)[a]	−63400.4 (8)	93.0 (12)	0^-, (1^-)[b]	$\pi5/2[413]\otimes\nu5/2[523]$	114
^{162}Tb^{2+}	$C_6H_6^+$	1.037 384 012 (14)	−65879.4 (20)	0.0	1^-, 1^-	$\pi3/2[411]\otimes\nu5/2[523]$	0
^{162m}Tb^{2+}	$C_6H_6^+$	1.037 385 975 (17)	−65593.9 (25)	285.5 (32)	4^-, 4^-	$\pi3/2[411]\otimes\nu5/2[523]$	140
N=99							
^{160}Pm^{2+}	^{84}Kr$^+$	0.953 046 770(13)	−52894.6 (20)	0.0	6^-, 6^-	$\pi5/2[532]\otimes\nu7/2[633]$	0
^{160m}Pm^{2+}	^{84}Kr$^+$	0.953 047 989 (70)	−52704 (11)	191 (11)	1^-, 1^-	$\pi5/2[532]\otimes\nu7/2[633]$	142
^{162}Eu^{2+}	^{84}Kr$^+$	0.964 926 876 6 (97)[a]	−58723.9 (15)	0.0	1^+, 1^+	$\pi5/2[413]\otimes\nu7/2[633]$	0
^{162m}Eu^{2+}	^{84}Kr$^+$	0.964 927 901 (12)[a]	−58563.7 (19)	160.2 (24)	6^+, 6^+	$\pi5/2[413]\otimes\nu7/2[633]$	128
^{164}Tb^{2+}	^{84}Kr$^+$	0.976 822 644 (12)	−62105.0 (19)	0.0	5^+, 5^+	$\pi3/2[411]\otimes\nu7/2[633]$	0
^{164m}Tb^{2+}	^{84}Kr$^+$	0.976 823 571 (77)	−61960 (12)	145 (12)	2^+, 2^+	$\pi3/2[411]\otimes\nu7/2[633]$	170
N=101							
^{164}Eu^{2+}	^{84}Kr$^+$	0.976 879 399 (16)	−53232.8 (25)	0.0	3^-, 3^-	$\pi5/2[413]\otimes\nu1/2[521]$	0
^{166}Tb^{2+}	^{84}Kr$^+$	0.988 767 521 (10)	−57808.9 (16)	0.0	1^-, 1^-	$\pi3/2[411]\otimes\nu1/2[521]$	0

[a]From Ref. [5].
[b]Spin affected by the Newby term [33].

It should be noted that the Woods-Saxon and Nilsson modified oscillator potentials with "universal" parameters [24,29], as well as the folded-Yukawa model [30], failed to reproduce the correct ordering of single-particle neutron states above $N = 98$. This was recently discussed in Refs. [5,31,32], where it was pointed out that the ordering of the $\nu7/2[633]$ and $\nu1/2[521]$ orbitals need to be reversed. In the frame of the projected shell model, a change in the spin-orbit interaction was introduced in Ref. [32] to explain the ordering of neutron single-particle levels near $N = 98$. The present studies also confirm such an inversion, which is manifested by the observation of isomers in the ^{160}Pm, ^{162}Eu, and ^{164}Tb ($N = 99$) isotones, but not in the ^{164}Eu and ^{166}Tb ($N = 101$) ones. Since the low-Ω, $\nu1/2[521]$ orbital is established at $N = 101$ (see Fig. 2), its coupling to the $\pi5/2[413]$ (^{164}Eu) and $\pi3/2[411]$ (^{166}Tb) proton orbitals would lead to $K_> =$ 3- and 2- and $K_< =$ 2- and 1- states, respectively. The spin differences are small and, hence, no spin-trap isomers are expected.

A puzzling feature of the present data, which is not understood at present, is the nonobservation of long-lived isomers in ^{158}Eu ($N = 95$) and ^{158}Pm ($N = 97$) where the $\pi5/2[413]\otimes\nu3/2[521]$, $\pi5/2[532]\otimes\nu5/2[523]$ configurations, respectively, are predicted to be lowest in energy. While a γ-ray decaying isomeric state has previously been reported in ^{158}Pm, only a lower bound of $T_{1/2} > 16\,\mu$s [34,35] could be established. If the true half-life of this state is less than ≈ 100 ms it may have been too short-lived to resolve from the observed ^{158}Pm ground state in our data set.

In summary, we discussed a recent campaign of mass measurements conducted with the CPT mass spectrometer in which the long-lived isomeric landscape of neutron-rich odd-odd nuclei in the light rare-earth region near A $\approx$ 165 was thoroughly combed for the first time. Owing to the high mass-resolving power offered by the PI-ICR technique, spin-trap isomers was discovered in ^{162}Tb ($N = 97$) and in the $N = 99$ isotones of ^{160}Pm and ^{164}Tb, and can be dismissed in the cases of ^{164}Eu and ^{166}Tb at $N = 101$. Modification of the single-particle level ordering, predicted by several theoretical models, is needed to explain the existence of isomers in the $N = 99$ isotones and their absence in the $N = 101$ ones. Detailed decay spectroscopy studies in this region of the nuclear chart would be valuable to further probe the properties of the isomers discovered here and to explore the lack of observed isomerism in ^{158}Eu and ^{158}Pm.

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