Blurring the Boundaries: Decays of Multiparticle Isomers at the Proton Drip Line

R. J. Carroll,[1] R. D. Page,[1] D. T. Joss,[1] J. Uusitalo,[2] I. G. Darby,[1] K. Andgren,[3] B. Cederwall,[3] S. Eeckhaudt,[2] T. Grahn,[2] C. Gray-Jones,[1] P. T. Greenlees,[2] B. Hadinia,[3] P. M. Jones,[2] R. Julin,[2] S. Juutinen,[2] M. Leino,[2] A.-P. Leppänen,[2] M. Nyman,[2] D. O'Donnell,[1,4] J. Pakarinen,[2] P. Rahkila,[2] M. Sandzelius,[2] J. Sarén,[2] C. Scholey,[2] D. Seweryniak,[5] and J. Simpson[4]

[1]*Department of Physics, Oliver Lodge Laboratory, University of Liverpool, Liverpool L69 7ZE, United Kingdom*
[2]*Department of Physics, University of Jyväskylä, P.O. Box 35, FI-40014 Jyväskylä, Finland*
[3]*Department of Physics, Royal Institute of Technology, SE-10691 Stockholm, Sweden*
[4]*STFC, Daresbury Laboratory, Daresbury, Warrington WA4 4AD, United Kingdom*
[5]*Argonne National Laboratory, Physics Division, Argonne, Illinois 60439, USA*
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A multiparticle spin-trap isomer has been discovered in the proton-unbound nucleus $^{158}_{73}Ta_{85}$. The isomer mainly decays by γ-ray emission with a half-life of 6.1(1) μs. Analysis of the γ-ray data shows that the isomer lies 2668 keV above the known 9^+ state and has a spin $10\hbar$ higher and negative parity. This 19^- isomer also has an 8644(11) keV, 1.4(2)% α-decay branch that populates the 9^+ state in ^{154}Lu. No proton-decay branch from the isomer was identified, despite the isomer being unbound to proton emission by 3261(14) keV. This remarkable stability against proton emission is compared with theoretical predictions, and the implications for the extent of observable nuclides are considered.

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Identifying the full extent of the nuclear landscape is a long-standing issue of fundamental importance in nuclear physics. For odd-Z nuclei, proton emission determines whether a given nuclide will be too short lived to be isolated and studied. In light elements, the lifetimes for proton emission quickly become extremely short with increasing neutron deficiency, so their decays can be studied close to a production target [1]. For heavier odd-Z elements, the larger Coulomb barrier means that lifetimes drop more gradually and can become significantly longer than the typical flight time of $\sim$1 μs through a recoil separator. The centrifugal barrier also has a strong effect, increasing lifetimes for larger orbital angular momentum changes Δl. Consequently, experimental studies of odd-Z nuclei have identified over 30 proton-emitting nuclei, with examples for most elements from iodine ($Z = 53$) to bismuth ($Z = 83$), all having proton-decay Q values $Q_p < 2$ MeV and $\Delta l \leq 5\hbar$ [2]. These studies reveal how half-lives of low-lying states generally decrease with increasing neutron deficiency (see Fig. 1), dropping more rapidly when proton or α-particle emission dominates.

The occurrence of high-spin isomers lying at excitation energies well above 1 MeV could blur the boundary of heavy, proton-rich nuclei that are observable experimentally ($t_{1/2} \geq 1$ μs). Multiparticle spin-trap isomers occur in nuclei with limited numbers of valence nucleons in orbitals with large angular momentum quantum numbers [4]. Isomers provide unique opportunities to observe proton and α decays with larger Δl values than simpler low-lying states. Although the decays of isomers may have greater Q values than those of ground states, which would shorten lifetimes, large Δl values inhibit decays so that isomers can have significantly longer half-lives. For example, in ^{212}Po the (18^+) isomer at 2.9 MeV α decays to the 0^+ ground state of ^{208}Pb with a half-life of 45 s, whereas its 0^+ ground state has a half-life of only 0.3 μs [5].

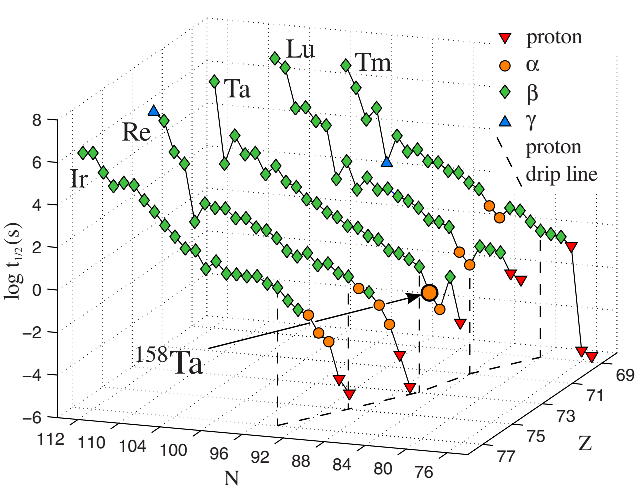

FIG. 1 (color online). Measured half-lives of longest-living states of neutron-deficient Tm, Lu, Ta, Re, and Ir isotopes. Main decay modes are indicated by symbols given in the key. The solid lines connect successive data points from Ref. [3] for each element. The dashed lines indicate the proton drip line, beyond which proton emission from nuclear ground states is energetically possible ($Q_p > 0$). No states are known in nuclei beyond the lightest isotopes shown.

In this Letter, we report the discovery of a high-lying spin-trap isomeric state in the proton-unbound nuclide ^{158}Ta, which has three valence neutrons outside an $N = 82$ core. The isomer decays by both α-particle and γ-ray emission and is a rare example of an α decay involving a large spin change. Although the isomer is highly unbound to proton emission, no proton-decay branch was observed, providing an opportunity to understand the effects of such isomers on the proton-rich boundary of the nuclear landscape.

The experiment was performed at the University of Jyväskylä Accelerator Laboratory. The ^{158}Ta nuclei were produced in fusion-evaporation reactions induced by 255 MeV ^{58}Ni ions bombarding an isotopically enriched, self-supporting ^{102}Pd target foil of thickness $\sim 1\ \mathrm{mg\,cm^{-2}}$. An average beam intensity of 4.3 particle nA was delivered for 139 h. Prompt γ rays were measured by using the JUROGAM array, which comprised 43 Compton-suppressed Ge detectors. The ^{158}Ta ions recoiled out of the target and were transported within 0.5 μs by using the gas-filled separator RITU [6,7] to the GREAT spectrometer [8] situated at its focal plane. The ions passed through a multiwire proportional counter and were implanted into the adjacently mounted double-sided silicon strip detectors (DSSDs). Each DSSD had an active area of 60 mm $\times$ 40 mm and was 300 μm thick. The strips on their front and back surfaces were orthogonal, and the strip pitch of 1 mm on both faces provided 4800 independent pixels. X rays and γ rays emitted during decay processes in the DSSDs were measured by using a planar and a Clover Ge detector. All detector signals were passed to the triggerless data acquisition system [9], where they were time stamped with a precision of 10 ns. The data were analyzed by using the GRAIN [10] and RADWARE [11] software packages.

In total, 7.6×10^8 evaporation residues were implanted into the DSSDs at an average rate of 1.5 kHz. The most abundant nuclides were ^{157}Lu and ^{157}Hf, which were produced with cross sections of $\sim$5 mb and $\sim$2 mb via the $3p$ and $2p1n$ evaporation channels, respectively. Prompt γ-ray transitions at the target position and delayed γ rays at the focal plane were assigned as feeding specific α-decaying states in the $1p1n$ evaporation residue ^{158}Ta by using standard tagging techniques [12,13]. Two levels were previously known in ^{158}Ta, both of which decay by α-particle emission [14,15]: the 2^- ground state [$E_\alpha = 5968(5)$ keV, $t_{1/2} = 46(4)$ ms] and a 9^+ state at an excitation energy of 141(9) keV [$E_\alpha = 6046(4)$ keV, $t_{1/2} = 35(1)$ ms]. The spin assignments were proposed on the basis of empirical coupling rules for odd-odd nuclei [15]. A total of 3.5×10^5 α decays of the 9^+ state in ^{158}Ta were recorded, corresponding to a cross section of $\sim$20 μb. Figure 2(a) shows the energy spectrum of prompt γ rays measured in JUROGAM correlated with these α decays. The corresponding spectrum of delayed γ rays measured in the Clover detector is shown in Fig. 2(b). These delayed γ-ray transitions have a common

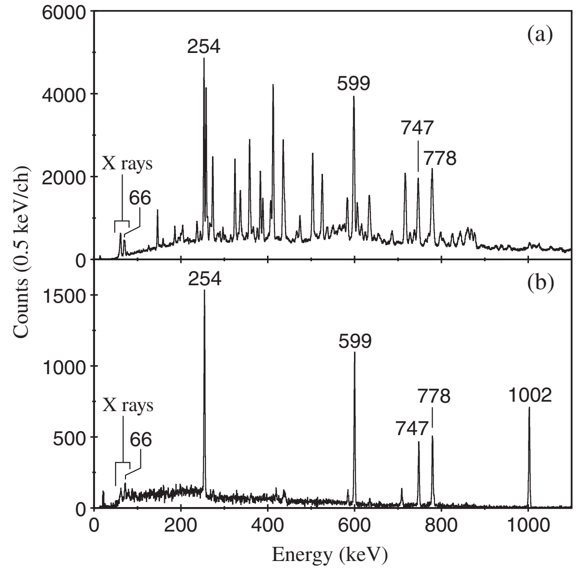

FIG. 2. Energy spectra of γ rays associated with ions implanted into the DSSDs and followed by 6046 keV α decays of ^{158}Ta in the same pixel within 175 ms. (a) Prompt γ rays measured by using JUROGAM. (b) Delayed γ rays measured by using the GREAT Clover detector within 30 μs of the implantation of a correlated ^{158}Ta ion. Transitions included in the partial level scheme are labeled with their energies in keV.

half-life of 6.1(1) μs, indicating that they are part of the decay path from a single isomer. Analysis of γ-ray coincidences resulted in the partial level scheme presented in Fig. 3. For example, 66 keV γ rays measured in the planar detector were observed in coincidence with γ rays at 254, 599, 747, and 1002 keV in the Clover detector but not 778 keV.

The isomeric state lies 2668 keV above the 9^+ state and decays mainly via the 1002 keV transition, which is clearly

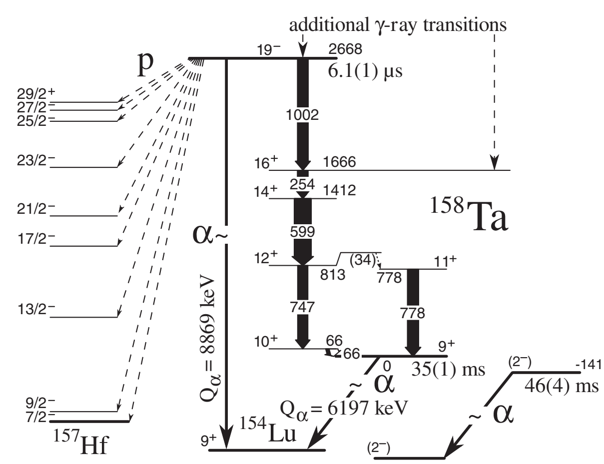

FIG. 3. Partial level scheme of ^{158}Ta with level energies in keV indicated relative to the 9^+ state. The dashed arrows indicate possible proton-decay branches from the 19^- isomer to excited states in ^{157}Hf [16]. The 34 keV transition was inferred from γ-ray coincidences.

visible in Fig. 2(b) but absent from the prompt spectrum of Fig. 2(a). Its lifetime is consistent with an $E3$ multipolarity assignment for the depopulating transition with a reduced transition strength of $B(E3) = 0.101(4)$ Weisskopf units. The angular distributions of the 254, 599, 747, and 778 keV γ rays measured by using JUROGAM are consistent with those of known stretched $E2$ transitions measured during the same experiment. These $E2$ assignments and the $E3$ assignment for the 1002 keV transition are compatible with the measured intensities, after allowing for internal conversion. The cascade of $E2$ transitions is assigned as connecting the multiplet of states from 16^+ to 10^+ with a $\pi h^n_{11/2}\nu f^2_{7/2}h_{9/2}$ configuration that is observed in odd-odd $N = 85$ isotones from $^{150}_{65}$Tb to $^{160}_{75}$Re [17]. Intensity and internal conversion arguments also require the multipolarity of the 66 keV transition to be $M1$ [18,19], supporting the proposed spin assignment for the 9^+ state [15]. The isomer has a spin $10\hbar$ higher and negative parity. A possible structure for this isomer is $[\pi h^{-3}_{11/2}\nu f_{7/2}h_{9/2}i_{13/2}]19^-$, analogous to that proposed in ^{152}Ho, which decays by $E3$ γ-ray emission with $B(E3) = 0.92(3)$ W.u. [20].

An α decay from the 19^- isomer in ^{158}Ta to the 9^+ state in ^{154}Lu was also observed [see Fig. 4(a)] with a yield of 1600 counts. The assignment of this $E_\alpha = 8644(11)$ keV activity is based on the following observations: Its half-life is 6.4(4) μs, which agrees with the value deduced from the isomer γ decays; the Q_α value agrees within uncertainties with the combination of the Q_α value of the 9^+ state and the excitation energy of the 19^- isomer; and the same prompt γ-ray transitions as those feeding the 19^- isomer are observed when tagging separately on the α-decay and γ-decay branches of the isomer. Comparing the yields of the 8644 and 6048 keV α-decay peaks associated with these prompt γ rays populating the 19^- isomer allowed an α-decay branching ratio of 1.4(2)% to be deduced. This in turn leads to a partial α-decay half-life of 440(70) μs. Allowing for the spin and parity change gives a reduced α-decay width of 14(2) keV [21], corresponding to a factor of 5 hindrance relative to the α decay of the ground state of ^{212}Po. This reflects structural changes involved in the α decay and compares with hindrance factors of $\sim$20 for α decays of the nearby $N = 84$ spin-trap isomers, which are visible in Fig. 4(a) [14]. The partial half-lives of all these isomers near $N = 82$ are shorter than those of lower-lying states, unlike spin-trap isomers near ^{208}Pb, so their gain in Q_α value outweighs the hindrance caused by the increased centrifugal barrier.

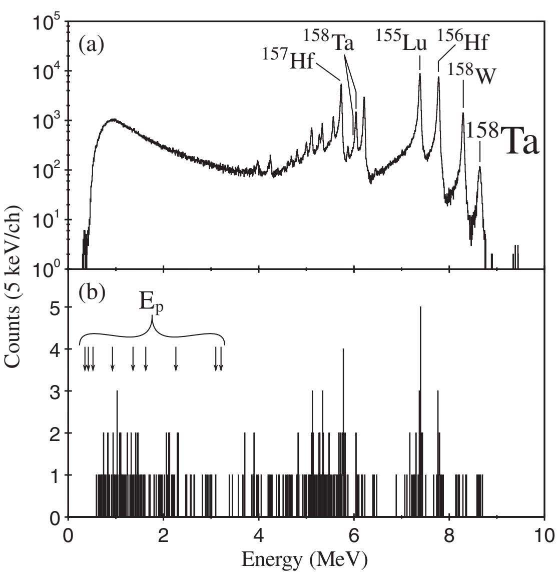

FIG. 4. (a) Energy spectrum of all decays occurring within 30 μs of an ion implantation into the same DSSD pixel. The continuum below 4 MeV is α particles escaping from the DSSD without depositing their full energy. (b) Energy spectrum of decays from (a) that are followed within 575 ms by the α decay of ^{157}Hf, the daughter of ^{158}Ta proton decays. Vestiges of the strongest α peaks in (a) appear through random correlations. The arrows indicate the energies E_p expected for proton-decay lines.

Proton emission is more strongly affected by large spin changes than α decay, because for protons the Coulomb barrier is smaller and the centrifugal barrier is larger for a given spin change, owing to the smaller reduced mass. As with α decays of isomers, the gain in Q_p value is at the expense of an increase in the centrifugal contribution to the barrier through which the particle has to tunnel and has a strong effect on the partial proton-decay half-life. Whether there is an overall increase or decrease in the half-life of an isomer depends on the delicate balance between these two competing effects.

There are $\sim$120 nuclei that are known to have high-spin ($>9\hbar$) isomers at excitation energies above 1.5 MeV and half-lives > 1.5 μs [3]. Including the ^{158}Ta isomer discovered in this work, isomers in only 13 of these nuclei are proton unbound, two of which decay by direct proton emission. The first observation of proton radioactivity was from a $19/2^-$ isomer in $^{53}_{27}$Co, lying at an excitation energy of 3.2 MeV [22–24]. The isomer mainly undergoes β decay, with a $\sim$1.5% proton-decay branch proceeding via $\Delta l = 9\hbar$ emission to the ground state of ^{52}Fe. This remains the highest known Δl value for any proton decay [2]. More recently, direct proton emission has been observed from a (21^+) isomer in $^{94}_{47}$Ag [25,26]. This 6.7 MeV isomer decays to at least one excited state in ^{93}Pd with a branching ratio of a few percent. The spins and parities of the states involved are uncertain, but the confirmed transition is thought to populate an excited state through $\Delta l = 4\hbar$ emission. In both ^{53}Co and ^{94}Ag, the extra Q_p value available from the excitation energy of the isomer is sufficient for proton emission to occur, even though their ground states are bound and therefore lie within the proton drip line [27].

Of the remaining cases, five are nuclides beyond the proton drip line [27], from a small region just above the $N = 82$ shell closure: $^{153,154,155}_{71}$Lu and $^{157,158}_{73}$Ta. Although these nuclei have no states that are bound, proton emission has been observed only from the ground state of ^{157}Ta [28]. The present data were searched for a proton-decay branch from the ^{158}Ta isomer. From the known Q_p value of the 9^+ state of 594(14) keV [15], a Q_p value of 3261(14) keV can be deduced for the 19^- isomer. Any proton decay of this isomer would be followed by the known α decay of ^{157}Hf [$E_\alpha = 5729(4)$ keV, $t_{1/2} = 115(1)$ ms] [14]. Figure 4(b) shows the events in Fig. 4(a) that are followed by an α decay of ^{157}Hf within 575 ms. There is no statistically significant peak at any of the energies expected for decays populating known excited states in ^{157}Hf (see Fig. 3) [16], giving a 95% confidence level upper limit of 0.008% for any proton-decay branch [29].

The excitation energies, spins, and parities of yrast states that could be populated by proton emission from the new isomer in ^{158}Ta and the multiparticle isomers in ^{157}Ta and ^{153,154,155}Lu are known [16,30–33]. Partial half-lives for proton-decay branches from the multiparticle isomers in ^{155}Lu and ^{157,158}Ta estimated by using a simple formula that reproduces measured proton-decay half-lives [34] are presented in Fig. 5. This formula is based on the systematics of proton-decay half-lives, after taking into account the effects of the Coulomb barrier, Δl, and the Q_p value. Although the direct proton decay of the 19^- isomer in ^{158}Ta to the $7/2^-$ state in ^{157}Hf would have a much longer partial half-life than the decays of low-lying states, the most favoured proton-decay branches to excited states in ^{157}Hf would be faster, which is a similar situation to that observed in ^{94}Ag [25,26]. However, the proton decays would not compete with the measured γ- and α-decay branches, as is observed experimentally. In the case of ^{155}Lu, proton emission from the isomer would be faster than from lower-lying states, which are only just unbound, whereas for ^{157}Ta all proton-decay paths from the isomer have longer partial half-lives than low-lying states. In ^{153}Lu, the $\pi s_{1/2}$ isomer at 80 keV would have the shortest partial half-life, with those of the isomer decays and the $\pi h_{11/2}$ ground state being comparable. The excitation energy of the ^{154}Lu isomer is not known [32].

The 19^- state in ^{158}Ta is an important example of the stability against proton emission that can be achieved in highly unbound spin-trap isomers. Lifetimes and decay modes are extremely sensitive to decay energies and spin changes, raising the possibility of high-spin multiparticle isomers in other nuclei beyond the proton drip line with much longer half-lives than their lower-lying, low-spin states.

In nuclei away from closed shells, high-spin isomers are also found in regions of axially symmetric deformed nuclei through the formation of "K traps." Decays requiring large changes of the quantum number K, the projection of the nucleus' total angular momentum onto its symmetry axis, are strongly hindered [4,35]. The proton drip line passes through shell closures, near which multiparticle spin-trap isomers can be formed, and regions of nuclear deformation, where K isomers can occur. Multiparticle isomers could

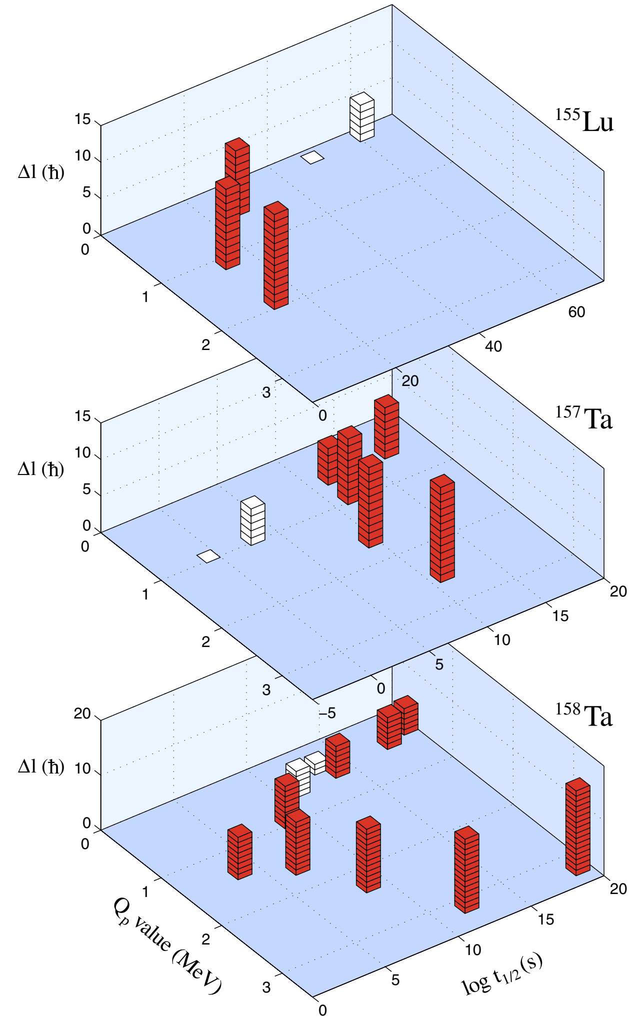

FIG. 5 (color online). Partial half-lives for proton emission from states in ^{155}Lu, ^{157}Ta, and ^{158}Ta calculated by using the method of Ref. [34]. Values for emission from low-lying states are indicated in white, while those for multiparticle spin-trap isomers are shown in red. Spins and excitation energies of states in the daughter nuclei are taken from Refs. [16,30,33].

therefore blur the boundaries of the nuclear landscape by providing the last observable nuclear states beyond the proton drip line. Calculations suggest that multiparticle isomers in superheavy nuclei [36] and neutron-rich nuclei [37,38] may also have enhanced stability. Decays of long-lived multiparticle isomers beyond the expected boundaries of the nuclear landscape therefore remain an intriguing possibility for future experiments and an important challenge for models of atomic nuclei.

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[1] A. M. Rogers *et al.*, Phys. Rev. Lett. **106**, 252503 (2011).
[2] B. Blank and M. J. G. Borge, Prog. Part. Nucl. Phys. **60**, 403 (2008).
[3] National Nuclear Data Center Evaluated Nuclear Structure Data File, http://www.nndc.bnl.gov/ensdf/.
[4] P. M. Walker and G. D. Dracoulis, Nature (London) **399**, 35 (1999).
[5] A. R. Poletti, G. D. Dracoulis, A. P. Byrne, and A. E. Stuchberry, Nucl. Phys. **A473**, 595 (1987).
[6] M. Leino *et al.*, Nucl. Instrum. Methods Phys. Res., Sect. B **126**, 320 (1997).
[7] J. Uusitalo *et al.*, Nucl. Instrum. Methods Phys. Res., Sect. B **204**, 638 (2003).
[8] R. D. Page *et al.*, Nucl. Instrum. Methods Phys. Res., Sect. B **204**, 634 (2003).
[9] I. H. Lazarus *et al.*, IEEE Trans. Nucl. Sci. **48**, 567 (2001).
[10] P. Rahkila, Nucl. Instrum. Methods Phys. Res., Sect. A, **595**, 637 (2008).
[11] D. C. Radford, Nucl. Instrum. Methods Phys. Res., Sect. A **361**, 297 (1995).
[12] K.-H. Schmidt, R. S. Simon, J.-G. Keller, F. P. Hessberger, G. Münzenberg, B. Quint, H.-G. Clerc, W. Schwab, U. Gollerthan, and C.-C. Sahm, Phys. Lett. **168B**, 39 (1986).
[13] E. S. Paul *et al.*, Phys. Rev. C **51**, 78 (1995).
[14] R. D. Page, P. J. Woods, R. A. Cunningham, T. Davinson, N. J. Davis, A. N. James, K. Livingston, P. J. Sellin, and A. C. Shotter, Phys. Rev. C **53**, 660 (1996).
[15] C. N. Davids *et al.*, Phys. Rev. C **55**, 2255 (1997).
[16] A. F. Saad *et al.*, Z. Phys. A **351**, 247 (1995).
[17] P. J. Sapple *et al.*, Phys. Rev. C **84**, 054303 (2011).
[18] T. Kibédi, T. W. Burrows, M. B. Trzhaskovskaya, P. M. Davidson, and C. W. Nestor, Jr., Nucl. Instrum. Methods Phys. Res., Sect. A **589**, 202 (2008).
[19] A. N. Andreyev, P. A. Butler, R. D. Page, D. E. Appelbe, G. D. Jones, D. T. Joss, R. D. Herzberg, P. H. Regan, J. Simpson, and R. Wadsworth, Nucl. Instrum. Methods Phys. Res., Sect. A **533**, 422 (2004).
[20] S. André, C. Foin, D. Santos, D. Barnéoud, J. Genevey, Ch. Vieu, J. S. Dionisio, M. Pautrat, C. Schück, and Z. Meliani, Nucl. Phys. **A575**, 155 (1994).
[21] J. O. Rasmussen, Phys. Rev. **113**, 1593 (1959).
[22] K. P. Jackson, C. U. Cardinal, H. C. Evans, N. A. Jelley, and J. Cerny, Phys. Lett. **33B**, 281 (1970).
[23] J. Cerny, J. E. Esterl, R. A. Gough, and R. G. Sextro, Phys. Lett. **33B**, 284 (1970).
[24] J. Cerny, R. A. Gough, R. G. Sextro, and J. E. Esterl, Nucl. Phys. **A188**, 666 (1972).
[25] I. Mukha *et al.*, Phys. Rev. Lett. **95**, 022501 (2005).
[26] J. Cerny, D. M. Moltz, D. W. Lee, K. Peräjärvi, B. R. Barquest, L. E. Grossman, W. Jeong, and C. C. Jewett, Phys. Rev. Lett. **103**, 152502 (2009).
[27] M. Wang, G. Audi, A. H. Wapstra, F. G. Kondev, M. MacCormick, X. Xu, and B. Pfeiffer, Chin. Phys. C **36**, 1603 (2012).
[28] R. J. Irvine *et al.*, Phys. Rev. C **55**, R1621 (1997).
[29] G. J. Feldman and R. D. Cousins, Phys. Rev. D **57**, 3873 (1998).
[30] D. Seweryniak *et al.*, Phys. Rev. C **71**, 054319 (2005).
[31] D. Nisius *et al.*, Phys. Rev. C **52**, 1355 (1995).
[32] J. H. McNeill, A. A. Chishti, P. J. Daly, W. Gelletly, M. A. C. Hotchkis, M. Piiparinen, B. J. Varley, P. J. Woods, and J. Blomqvist, Z. Phys. A **344**, 369 (1993).
[33] C. T. Zhang *et al.*, Z. Phys. A **345**, 327 (1993).
[34] D. S. Delion, R. J. Liotta, and R. Wyss, Phys. Rev. Lett. **96**, 072501 (2006).
[35] P. M. Walker and G. D. Dracoulis, Hyperfine Interact. **135**, 83 (2001).
[36] F. R. Xu, E. G. Zhao, R. Wyss, and P. M. Walker, Phys. Rev. Lett. **92**, 252501 (2004).
[37] L. K. Peker, E. I. Volmyansky, V. E. Bunakov, and S. G. Ogloblin, Phys. Lett. **36B**, 547 (1971).
[38] V. P. Bugrov, V. E. Bunakov, S. G. Kadmenskiĩ, and V. I. Furman, Sov. J. Nucl. Phys. **42**, 34 (1985).