Ground State Proton Radioactivity from ^{121}Pr: When Was This Exotic Nuclear Decay Mode First Discovered?

A. P. Robinson,[1] P. J. Woods,[1] D. Seweryniak,[2] C. N. Davids,[2] M. P. Carpenter,[2] A. A. Hecht,[3] D. Peterson,[2] S. Sinha,[2] W. B. Walters,[3] and S. Zhu[2]

[1]*School of Physics, The University of Edinburgh, Edinburgh EH9 3JZ, United Kingdom*
[2]*Argonne National Laboratory, Argonne, Illinois, 60439 USA*
[3]*University of Maryland, College Park, Maryland, 20742 USA*

(Received 12 April 2005; published 12 July 2005)

Ground-state proton radioactivity has been identified from ^{121}Pr. A transition with a proton energy of $E_p = 882(10)$ keV [$Q_p = 900(10)$ keV] and half-life $t_{1/2} = 10^{+6}_{-3}$ ms has been observed and is assigned to the decay of a highly prolate deformed $3/2^+$ or $3/2^-$ Nilsson state. The present result is found to be incompatible with a previously reported observation of ground-state proton radioactivity from ^{121}Pr, which would have represented the discovery of this phenomenon.

DOI: 10.1103/PhysRevLett.95.032502 PACS numbers: 21.10.Tg, 23.50.+z, 27.60.+j

In 1970 two groups independently reported evidence for the then new phenomenon of nuclear proton radioactivity [1,2], the decay of a nucleus by the emission of a single constituent proton. In the ensuing decades, proton radioactivity has proved to be a uniquely valuable tool for studying the structural properties of large numbers of nuclei beyond the proton drip line [3,4]. The experiment performed at Harwell by Jackson *et al.* [1] reported the serendipitous discovery of proton radioactivity from a high spin isomer in ^{53}Co, the ground state being proton bound. This was promptly confirmed by Cerny *et al.* in work performed at Berkeley [5]. Meanwhile, at Dubna, evidence was being reported of a 0.7–1.0 MeV proton decay produced by the fusion reaction ^{32}S + ^{96}Ru [2]. It was initially speculated the activity was from ^{122,123}Pr [2]. Subsequent experiments by this group [6] using two different techniques resulted in the report of a "proton emitter of 0.83(5) MeV with a half-life ~1 s," hypothesized as being from the ground state of a light praseodymium isotope, with ^{121}Pr being specifically proposed. The paper concluded "additional experiments with a method of higher sensitivity are needed." Over three decades later, with the issue still unresolved, we have set out to identify ground-state proton radioactivity from ^{121}Pr to test this historic claim.

Ironically, ^{121}Pr is one of the most remote proton emitters in the region of the proton drip line between $Z = 51$–83. It can be accessed by the $1p6n$ heavy ion fusion evaporation channel using stable beam-target combinations. This has been achieved only once before when the $1p6n$ channel was successfully used to identify proton decay from ^{135}Tb, with a cross section of ~3 nb [7]. Generally speaking, the cross section for producing proton emitters rapidly decreases with increasing neutron evaporation for $1pxn$ channels. For comparison, $1p2n$ evaporation channels typically have $\sigma \sim 50$ μb, and $1p4n$ channels typically have $\sigma \sim 200$ nb [3]. The Dubna experiments used the ^{32}S + ^{96}Ru fusion reaction, whereas here we chose to feed the same compound nucleus using the ^{36}Ar + ^{92}Mo reaction since the ATLAS accelerator facility at Argonne can produce intense beams of ^{36}Ar ions, and ^{92}Mo is a very stable target material, with high isotopic purity.

A 240 MeV beam of ^{36}Ar was used to bombard a 0.7 mg cm^{-2} thick ^{92}Mo target, giving a center-of-target excitation energy ~110 MeV to optimize production of ^{121}Pr via the $1p6n$ evaporation channel. The experiment was run for approximately 6 days with an average beam current ~25 pnA. The Argonne Fragment Mass Analyzer (FMA) [8] was set to transmit $A = 121$ recoils with charge states $q = 22$ and 23 through two slits in front of the focal plane detectors. A position-sensitive parallel grid avalanche counter located at the focal plane provided A/q, time of arrival, and energy-loss signals for the recoiling nuclei. After passing through this detector the recoils were implanted into a 60 μm thick 80×80 double sided strip detector (DSSD) [7] situated behind the focal plane of the FMA. The DSSD was instrumented in parallel with semi-Gaussian shaping amplifiers and delay-line amplifiers to allow half-lives down to ~1 μs to be measured [9]. A 0.3 mm thick, large area, silicon detector was placed directly behind the DSSD to partially veto background from β-delayed protons.

Figure 1(a) shows the energy spectrum for decay events in the DSSD occurring within 30 ms of an $A = 121$ recoil being implanted into the same quasipixel. For comparison, Fig. 1(b) shows decays occurring within 250 ms of implantation. This latter spectrum is fairly featureless in the energy region of interest, exhibiting a broad distribution of events characteristic of partial energy deposition from β-delayed proton decays. Likely $A = 121$ sources for this activity are the proton-rich even Z nuclei, ^{121}Ce and ^{121}Ba. In Fig. 1(a) there is clear evidence for a peak around 880 keV. There is no equivalent structure in either the rest

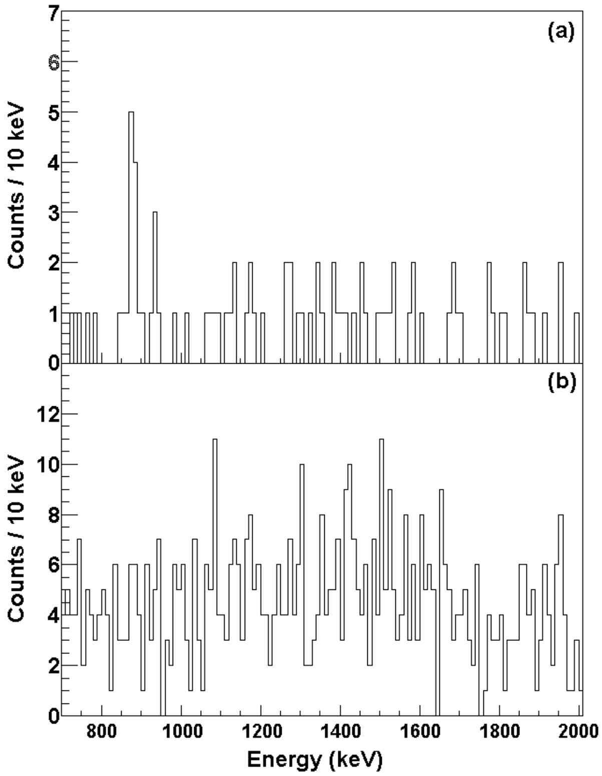

FIG. 1. Decays in the DSSD produced using a 240 MeV ^{36}Ar beam to bombard a ^{92}Mo target, within (a) 30 ms of implantation of an $A = 121$ residue into a DSSD quasipixel, showing the ground-state proton decay peak of ^{121}Pr, and (b) within 250 ms of implantation of an $A = 121$ residue, showing the structure of the background from long-lived β-delayed proton emitters.

of Fig. 1(a) or more importantly in Fig. 1(b), where the background is relatively low in this energy region. The counts in the peak correspond to a cross section of $\sim$300 pb (within a factor of $\sim$2 uncertainty), assuming a combined FMA and DSSD efficiency of $\sim$8%. The half-life was determined to be 10^{+6}_{-3} ms using the method of maximum likelihood. The low decay energy and short half-life are incompatible with α radioactivity, so we conclude the decay line represents proton radioactivity. This being the case, we obtain a proton decay energy $E_p = 882(10)$ keV [$Q_p = 900(10)$ keV], using a calibration from the known proton emitter, ^{147}Tm [$E_p = 1051(3)$ keV] [10].

The Möller-Nix mass model [11] and the Liran-Zeldes mass formula [12] are known to predict proton decay Q values well in this region of the proton drip line. Both approaches predict that the neighboring proton-rich $A = 121$ isobars ^{121}Ce and ^{121}La are proton bound, making it very unlikely that these nuclei are the source of the proton radioactivity. The Möller-Nix mass model gives a Q-value prediction of 521 keV for ^{121}Pr, while the Liran-Zeldes formula predicts a value of 1180 keV (the latest mass tables of Audi *et al.* [13] quote the experimental value reported by the Dubna group in [6]). The present Q value of 900(10) keV therefore lies between the two theoretical predictions, and is consistent with proton decay from the ground state of ^{121}Pr.

Assuming a spherical $g_{7/2}$ proton configuration for ^{121}Pr leads to a calculated half-life unphysically longer than that observed, and can be ruled out as a possibility. The Möller-Nix mass model [11] predicts a large quadrupole deformation $\beta_2 = 0.31$ with a $3/2^-$ proton configuration for the ground state of ^{121}Pr. The $3/2^+[422]$ and $3/2^-[541]$ deformed Nilsson proton orbitals are expected to be located near the Fermi surface, and are approximately degenerate. In the neighboring highly deformed proton emitter, ^{117}La, the decay rate was compatible with either configuration [14]. Calculations of the decay rate of ^{121}Pr as a deformed proton emitter were carried out in the adiabatic limit, using the Green's function technique of Esbensen and Davids [15]. For the present case a $\approx$100% proton branch is assumed since the half-life is much shorter than the predicted β^+ decay half-life of $\sim$300 ms [16]. The spectroscopic factor $u^2 = 0.51$ was obtained from a BCS calculation. The half-life calculations for $K = 3/2^+$ and $3/2^-$ are compared to the experimental half-life in Fig. 2. Not shown are calculated half-lives for the nearby, but more tightly bound $1/2^+[420]$ and $9/2^+[404]$ orbitals, which are at least an order of magnitude too short or too long, respectively. The decay rate for ^{121}Pr is again found to be consistent with a highly prolate deformed $3/2^+$ or $3/2^-$ ground-state configuration. There is very tentative evidence of a second weaker peak in Fig. 1(a) at an energy of $\sim$930 keV that could possibly be due to proton decay of an isomeric state; however, more data would be essential to infer this. In summary, the data clearly point to the observation of ground-state proton decay from a highly deformed Nilsson configuration, entirely consistent with previous proton decay measurements and theoretical ex-

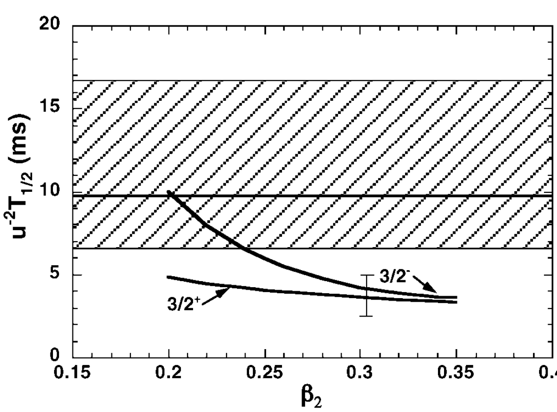

FIG. 2. Proton half-life vs quadrupole deformation β_2 for ^{121}Pr. The hatched area encloses the experimental measurement. Calculations are shown for the $3/2^+[422]$ and $3/2^-[541]$ proton Nilsson configurations for a prolate shape, β_4 scaled by the equation $\beta_4 = 0.26\beta_2$, and a spectroscopic factor $u^2 = 0.51$. A generic error bar on the calculated half-life due to uncertainties in the proton energy is shown for the $3/2^+[422]$ orbital.

pectations for nuclei in this region. The question remains: is the ground-state proton decay of ^{121}Pr reported here compatible with that reported by the Dubna Group? In order to address this crucial issue, we first review below the Dubna experiments.

In Refs. [2,6] by the Dubna Group, a gas-filled mass separator was used to separate the recoiling nuclei of interest and deposit them in the central volume enclosed by a telescope consisting of three cylindrical coaxial proportional counters. The inner counter provided ΔE information, while the second counter measured the residual energy E. The first two counters were connected in a coincidence circuit, and the third counter provided a veto for particles which were not stopped in the second counter. The minimum decay time observable with this method was 0.3 ms [2,6]. In Ref. [2] direct ("soft") proton decay events were reported as being produced using a ^{32}S beam energy of 190 MeV on a ^{96}Ru target, corresponding to an excitation energy $\sim$90 MeV in the compound nucleus, ^{128}Nd. These events decreased in yield when the beam energy was increased to 210 MeV. The decays were in the energy range of 0.7–1.0 MeV with a half-life range of 0.2–2 s. This initial work was followed up by a more detailed study using the gas-filled separator [6] in which a higher ^{32}S beam energy of 240 MeV was used, corresponding to a maximum compound nucleus excitation energy $\sim$128 MeV. In this instance, two separate proton decay groups were reported, one with a proton centroid energy of 0.8 MeV (no error quoted) and a half-life of 0.6(3) s, and a second group with a centroid energy of 1.2(1) MeV and a half-life of 2.1(6) s. The number of events was found to decrease when the beam energy was decreased below 240 MeV. The Dubna Group concluded the activity was sufficiently long-lived to attempt an alternative, slower method to verify their results, which was also reported in [6]. A helium gas jet system based on the method of Macfarlane and Griffioen [17] was used to collect the recoiling nuclei on a rotating disk, which subsequently placed the collected activity under a detector telescope. The disk was divided into 8 sections. When one section of the disk was facing the detector telescope, the activity was collected on the neighboring section. The disk was rotated by 45° every 3 s, and the time of rotation was $\sim$15 ms [6]. Using this approach, a single peak of proton decay events with $E_p = 830(50)$ keV and $t_{1/2} = 1400(800)$ ms was reported to be produced with $\sigma \sim 100$ nb [6]. This group of events was associated with the events around 0.8 MeV seen using the gas-filled separator system in Ref. [6], but no evidence was found for the higher energy group. The main source of background in all the Dubna experiments was from β-delayed proton emitters produced as fusion reaction products [2,6]. Reference [6] concludes "the appearance of a group of protons with energy $\sim$0.83 MeV cannot be attributed to background effects" and that "the results do not contradict the assumption that this is the ^{121}Pr nucleus produced in a reaction in which a proton and six neutrons are emitted," which "can be connected with proton decay from the ground state."

The results of the present work give agreement with the energy of the protons, but crucially the lifetime measured here is far too short to be compatible with the Dubna values, and, in particular, the present activity would not have been observable using the slower helium jet technique in which a peak is reported in [6]. Furthermore, the cross section of $\sim$100 nb [6] for the $1p6n$ channel is unrealistically high, both in relation to the present work, and in comparison to all presently known systematics for $1p6n$ channels in this region of nuclei. It is perhaps noteworthy that, at the time of the first Dubna experiments, there was an expectation that the $1p6n$ cross section would be in excess of 1 μb [2], whereas current systematics suggest a typical value of $\sim$1 nb consistent with the present experimental result of $\sim$300 pb. We therefore conclude that ground-state proton radioactivity from ^{121}Pr was not observed in the Dubna experiments, but was observed for the first time in the present work.

One might then ask, what did the Dubna Group observe? A simple, plausible explanation would be an artifact associated with background from β-delayed proton emission. Ground-state proton decay from heavier Pr isotopes can certainly be ruled out as being incompatible with the present result, since the proton energies would be much lower. Proton decay from a possible isomer in heavier Pr isotopes would seem incompatible with the observation that the proton yields were found to decrease with beam energies below 240 MeV [6], well above the expected excitation function maximum for the production of such isotopes. Such an explanation would be possible for the lower energy group of events observed at lower ^{32}S beam energies in the first Dubna experiment [2], but is inconsistent with these events being associated with the low energy events reported in the later experiments [6]. Reference [6] "does not exclude the possibility" of the 0.83(5) MeV group being from "a light isotope of lanthanum"; however, results now reported for the ground-state proton decay of ^{117}La [14] are incompatible with this result, again due to the relatively short half-life (26 ms).

In summary, we report the observation of ground-state proton radioactivity from ^{121}Pr, and find its decay rate consistent with emission from a highly deformed prolate nuclear shape. This leaves Pm as the only odd-Z element between $Z = 51$–83 for which proton radioactivity has not been reported. Our present result for the proton energy, when compared with mass models and proton energies for other neighboring elements, indicates ^{125}Pm is very likely the heaviest Pm isotope for which proton decay is the dominant ground-state branch. This is consistent with the nonobservation of proton decay from ^{126}Pm [18]. Our result makes it extremely unlikely that ground-state proton radioactivity was observed in the early pioneering Dubna experiments, which were nonetheless remarkable for their time. We therefore conclude that ground-state proton radioactivity was first identified at GSI in 1981 for the isotope ^{151}Lu by Hofmann and collaborators [19].

[1] K. P. Jackson, C. U. Cardinal, H. C. Evans, N. A. Jelley, and J. Cerny, Phys. Lett. **33B**, 281 (1970).
[2] V. A. Karnaukhov, D. D. Bogdanov, and L. A. Petrov, in *Proceedings of the International Conference on the Properties of Nuclei far from the Region of Beta-Stability, Leysin, Switzerland, 1970* (CERN Report No. 70-30, p. 457).
[3] P. J. Woods and C. N. Davids, Annu. Rev. Nucl. Part. Sci. **47**, 541 (1997).
[4] See, for example, *Proceedings of the 2nd International Symposium on Proton-Emitting Nuclei, Legnaro, Italy*, AIP Conf. Proc. No. 681, edited by E. Maglione and F. Soramel (AIP Press, New York, 2003).
[5] J. Cerny, J. E. Esterl, R. A. Gough, and R. G. Sextro, Phys. Lett. **33B**, 284 (1970).
[6] D. D. Bogdanov, V. P. Bochin, V. A. Karnaukhov, and L. A. Petrov, Sov. J. Nucl. Phys. **16**, 491 (1973).
[7] P. J. Woods, P. Munro, D. Seweryniak, C. N. Davids, T. Davinson, A. Heinz, H. Mahmud, F. Sarazin, J. Shergur, W. B. Walters, and A. Woehr, Phys. Rev. C **69**, R051302 (2004).
[8] C. N. Davids, B. B. Back, K. Bindra, D. J. Henderson, W. Kutschera, T. Lauritsen, Y. Nagame, P. Sugathan, A. V. Ramayya, and W. B. Walters, Nucl. Instrum. Methods Phys. Res., Sect. B **70**, 358 (1992).
[9] A. P. Robinson, C. N. Davids, G. Mukherjee, D. Seweryniak, S. Sinha, P. Wilt, and P. J. Woods, Phys. Rev. C **68**, 054301 (2003).
[10] P. J. Sellin, P. J. Woods, T. Davinson, N. J. Davis, K. Livingston, R. D. Page, A. C. Shotter, S. Hofmann, and A. N. James, Phys. Rev. C **47**, 1933 (1993).
[11] P. Möller, J. R. Nix, W. D. Myers, and W. J. Swiatecki, At. Data Nucl. Data Tables **59**, 185 (1995).
[12] S. Liran and N. Zeldes, At. Data Nucl. Data Tables **17**, 431 (1976).
[13] G. Audi, O. Bersillon, J. Blachot, and A. H. Wapstra, Nucl. Phys. **A729**, 337 (2003).
[14] H. Mahmud, C. N. Davids, P. J. Woods, T. Davinson, A. Heinz, G. L. Poli, J. J. Ressler, K. Schmidt, D. Seweryniak, M. B. Smith, A. A. Sonzongi, J. Uusitalo, and W. B. Walters, Phys. Rev. C **64**, R031303 (2001).
[15] H. Esbensen and C. N. Davids, Phys. Rev. C **63**, 014315 (2001).
[16] P. Möller, J. R. Nix, and K.-L. Kratz, At. Data Nucl. Data Tables **66**, 131 (1997).
[17] R. D. Macfarlane and R. D. Griffioen, Nucl. Instrum. Methods **24**, 461 (1963).
[18] H. Mahmud, Ph.D. thesis, University of Edinburgh (unpublished).
[19] S. Hofmann *et al.*, *Proceedings of the 4th International Conference on Nuclei far from Stability*, Helsingør, Denmark, edited by P. G. Hansen and O. B. Nielsen (CERN Report No. 81-09, p. 190).