First observation of the proton emitter $^{116}_{57}La_{59}$: Evidence for strong neutron-proton pairing from proton separation energies and emission probabilities

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The discovery of the new proton emitter $^{116}_{57}La_{59}$, 23 neutrons away from the only stable La isotope, $^{139}_{57}La_{82}$, is reported. The ^{116}La nuclei were synthesised in the fusion-evaporation reaction $^{58}Ni(^{64}Zn, p4n)^{116}La$ and identified via their proton radioactivity using the MARA mass spectrometer and the silicon detectors placed at its focal plane. Comparisons of the measured proton energy ($E_p = 718\pm9$ keV) and half-life ($T_{1/2} = 50\pm22$ ms) with values calculated using the Universal Decay Law approach indicate that the proton is emitted with an orbital angular momentum $l = 2$ and that its emission probability is enhanced relative to its closest, less exotic, odd-even lanthanum isotope ($^{117}_{57}La_{60}$) while the proton-emission Q-value is lower. We propose this to be a signature for the presence of strong neutron-proton pair correlations in this exotic, neutron deficient system. The observations of γ decays from isomeric states in ^{116}La and ^{117}La are also reported.

Introduction. Nucleonic pair correlations, also commonly called "pairing", play an important role in the structure of atomic nuclei. Well-known manifestations of the nuclear pairing effect, which is similar to condensed-matter physics phenomena such as superconductivity and superfluidity, are the odd-even staggering of nuclear binding energies [1], seniority symmetry [2–4] in the low-lying spectra of spherical even-even nuclei, and the reduced moments of inertia and backbending effect [5, 6] in rotating deformed nuclei. The first fundamental theoretical description of pairing in condensed matter physics focused on the properties of systems composed of large numbers of fermions, such as the electrons in superconductors (Bardeen-Cooper-Schrieffer (BCS) theory [7, 8]). The same formalism can be applied to atomic nuclei if the limited number of nucleons that can be bound in such systems is properly taken into account. A basic feature of BCS theory is that it treats systems with identical fermions moving in time-reversed orbitals as correlated pairs with opposite spins ($J = 0$), so-called Cooper pairs. Mean-field models of atomic nuclei based on the BCS approach, such as Hartree-Fock-Bogoliubov theory [9], therefore treat the neutron and proton pairing fields separately. These pairing fields give rise to the nuclear odd-even mass differences for neutrons and protons independently and are called isospin $T = 1$ or isovector pairing. However, the unique coexistence of two distinct fermionic systems (neutrons and protons) in the nucleus may produce additional pairing modes not found elsewhere in Nature. In particular in nuclei with equal or nearly equal neutron and proton numbers ($N \approx Z$) enhanced correlations arise between neutrons and protons that occupy orbitals with the same quantum numbers. The normal isovector pairing mode based on like-particle neutron-neutron (nn) and proton-proton (pp) Cooper pairs can be generalized to include neutrons and protons which may then also form isospin $T = 1$, angular momentum $J = 0$ np pairs. Of special interest is the long-standing question of the possible presence of a new and structurally different np pairing mode termed isoscalar pairing [10–19] predicted to be built from isospin $T = 0$, $J > 0$ np pair correlations. Many theoretical calculations predict that isoscalar pairing may only manifest itself clearly in the heaviest, most exotic neutron deficient nuclei with $A > 80$, for a review, see Ref. [20]. Calculations using isospin-generalized BCS and HFB equations including pp, nn, np ($T = 1$), and np ($T = 0$) Cooper pairs indicated that there may exist a second-order quantum phase transition in the ground states of $N = Z$ nuclei from $T = 1$ pairing below mass 80 to a predominantly $T = 0$ pairing phase above mass 90, with the intermediate mass 80-90 region showing a co-existence of $T = 0$ and $T = 1$ pairing modes [21]. There are even predictions for a dominantly $T = 0$ ground-state pairing condensate in $N \sim Z$ nuclei around mass 130 [22].

Proton radioactivity, i.e. spontaneous and direct proton emission, is a rare nuclear decay mode observed for the ground-states or isomeric states of some extremely neutron deficient nuclides (around 30 are known to date). Ground-state proton radioactivity was first observed by Hofmann *et al.* for ^{151}Lu [23] and by Klepper *et al.* for ^{147}Tm [24]. The first observation of proton radioactivity in the region of neutron deficient nuclei above tin, was made for ^{109}I and ^{113}Cs by Gillitzer *et al.* [25]. Proton emission from odd-odd nuclides may reveal effects of the residual proton-neutron interactions between the odd valence neutron and proton [26]. It has previously been observed for a few such cases that the proton-decay Q value, Q_p, of the odd-odd nucleus is lower than that of its less-exotic neighboring odd-even isotope. This was first inferred by Page *et al.* for the ^{108,109}I pair [27] and recently confirmed by direct measurement of proton emission from ^{108}I by Auranen *et al.* [28]. Another example is the ^{112,113}Cs pair [29]. Mass models fail consistently in reproducing this behavior. This might be attributed to the attractive residual force between the odd valence neutron and proton in the odd-odd systems [27, 31] but the nature of this neutron-proton interaction is until now unknown.

In this Letter, we present the discovery of the extremely neutron deficient, $T_Z = 1$, isotope $^{116}_{57}$La$_{59}$ via its radioactive proton decay and the observation of γ-ray transitions from low-lying microsecond isomers in ^{116,117}La. We propose that the observed differences in measured proton decay Q values and proton formation probabilities between neighboring isotopes provide a new signature and evidence for strong neutron-proton pairing in these exotic systems.

Experimental details and Results. The experiment was performed at the Accelerator Laboratory of the University of Jyväskylä using the ^{58}Ni(^{64}Zn, $p3n$)^{117}La and ^{58}Ni(^{64}Zn, $p4n$)^{116}La fusion-evaporation reactions. A ^{64}Zn beam was accelerated by the K130 cyclotron and let to impinge upon an isotopically enriched (99.8%) metallic target foil of ^{58}Ni with an areal density of 750 μg cm^{-2}. During the main production run (39 h of irradiation time) the beam kinetic energy was 330 MeV with an average intensity of 4.8 pnA. The charged-particle detector array JYTube (Jyväskylä-York Tube) [32] was arranged at the target position to detect evaporated charged particles emitted in the reactions. The fusion residues recoiled out of the target, and were transmitted and separated according to their mass to electric charge state ratio, A/q, to the focal plane detector system of the vacuum-mode recoil separator MARA (Mass Analysing Recoil Apparatus) [33]. The electric and magnetic dipole fields of MARA were set to accept fusion residues with mass $A = 117$ and (nominally) charge state $q = 30.5^+$ for the central "reference" trajectory and to accept recoils in four charge states from 29^+ to 32^+. At the MARA focal plane, recoils were passed through a position-sensitive multiwire proportional counter before being implanted into a double-sided silicon strip detector (DSSD) [32, 33]. The DSSD was 300 μm thick, with an active area of 128 mm $\times$ 48 mm, and was electrically segmented into 72 vertical strips in the y plane and 192 horizontal strips in the x plane, giving a total of 13824 quasipixels. A second layer of silicon with a thickness of 500 μm was located directly behind the DSSD to veto punch-through events due to uncorrelated high-energy light charged particles and to detect β particles escaping after leaving a partial energy signal in the DSSD. Five clover Ge detectors were placed surrounding the focal plane of MARA in a close-packed geometry and used for the measurement of delayed γ rays following recoil implantations and/or charged particle decays detected in the DSSD. All detector signals were time stamped using a global 100 MHz clock and recorded independently by the triggerless data acquisition system [34]. The data were analyzed online and offline using the GRAIN software package [35].

The energy calibration of the DSSD was accomplished using a standard mixed-isotope (^{241}Am, ^{244}Cm, ^{239}Pu) α radioactive source with its three main α-energy peaks, as well as in-beam data for the known proton decay of ^{109}I [37], its α-decaying daughter ^{108}Te as well as for the α-emitters ^{109}Te and ^{110}I [38]. The ^{109,110}I and ^{108,109}Te ions were produced in reactions using a higher beam energy of 370 MeV in a brief run after the main experiment, see the corresponding characteristic proton and alpha decay peaks in Figure 1 (a).

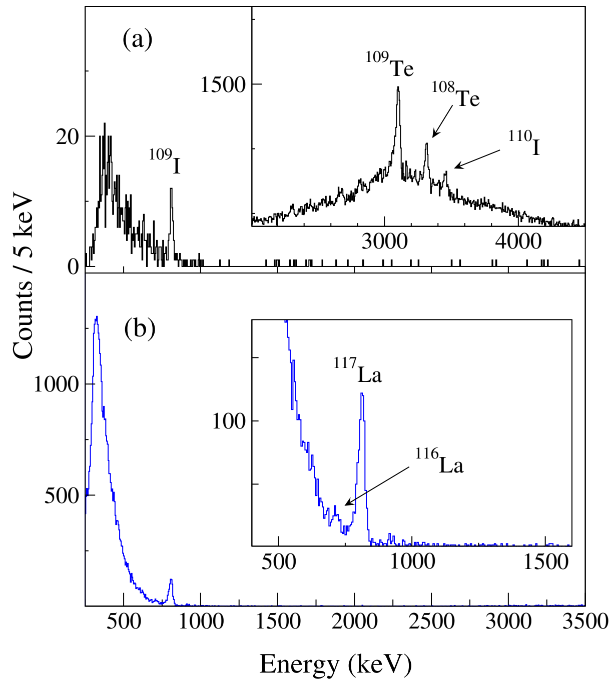

FIG. 1. DSSD energy spectra for punch-through-vetoed decay events. (a) Energy spectrum registered in the DSSD at the beam energy of 370 MeV for decay events ocurring within 300 μs of an ion implantation into the same DSSD quasipixel. The proton-decay peak of ^{109}I is indicated. The inset shows α-decay peaks from ground-state decays of ^{110}I and ^{108,109}Te obtained by applying a recoil-decay correlation time of 3 s. (b) Energy spectrum registered in the DSSD at the beam energy 330 MeV for decay events occurring within 100 ms after an ion implantation. The previously known proton-decay peak of ^{117}La is indicated. The inset shows the enlarged low-energy region. See text for details.

Figure 1 (b) shows the decay energy spectra recorded in the DSSD at the beam energy of 330 MeV. The corresponding events were punch-through vetoed and with the requirement that the decay occurred within 100 ms after the implantation of a recoil in the same DSSD quasipixel as well as with the requirement that the multiplicity of evaporated charged particles detected in JYTube was 0 or 1. The high background in the low-energy region is mainly from β decays of strongly populated nuclides closer to the stability line. The peak at 808(5) keV corresponds to the proton decay of ^{117}La and its half-life was measured to be

$T_{1/2} = 21.6(31)$ ms, in agreement with the previous measurements of the ground-state proton decay of ^{117}La [39–41]. The yield of $\sim$ 680 counts in this peak corresponds to a production cross section of $\sim$ 120 nb, assuming a MARA transmission efficiency of 35% for mass 117.

The low-energy region of the spectrum in Figure 1 (b) is enlarged and shown in the inset. A small peak at 718(9) keV is clearly visible above the β background with a statistics of 40(14) counts, which we assign to the ground-state proton decay of ^{116}La. These proton-decay events could be further enhanced relative to the β-decay background by requiring that they occurred in delayed coincidence with γ rays at the focal plane of MARA as discussed below.

Recoil-γ-decay event chains were investigated in order to search for isomeric γ-ray transitions in ^{117}La and the newly discovered isotope ^{116}La. The γ-ray energy spectrum recorded by the clover detectors sourrounding the MARA focal plane for events registered within 8 μs of recoil implantation and gated by ^{117}La protons (Figure 2(a)), reveals a peak with 9 counts at 192 keV. Based on a maximum likelihood analysis [42], a lifetime $\tau = 3.9^{+1.9}_{-0.9}$ μs ($T_{1/2} = 2.7^{+1.3}_{-0.7}$ μs) was determined for the corresponding isomeric level. Using Weisskopf estimates, we assign the multipolarity of the 192 keV transition to be of magnetic quadrupole (M2) character. This would correspond to a lifetime of 4.6 μs which is in fair agreement with the measured lifetime of the new observed isomeric state for ^{117}La. In contrast, the lifetime for a state depopulated entirely by an E3 transition of the same energy would be around four orders of magnitude longer, while the estimated lifetime of an E2 transition would be more than two orders of magnitude shorter (equivalent to about one tenth of the flight time through the MARA separator). The isomeric state is most likely situated at 192 keV excitation energy since there are no significant other peaks present in the spectrum of Figure 2(a), apart from La X-rays which are expected due to internal conversion of the M2 transition. A tentative spin-parity $7/2^-$ is consequently assigned to the 192 keV level, based on the previous assignment of the ground state as $(3/2^+)$ [39, 41]. The intensity of the observed 192 keV transition indicates an isomeric population ratio of the order of 30%. This would be in agreement with the observation of prompt γ rays from ^{117}La by Liu *et al.* [41] if a significant fraction of those γ rays emanate from a rotational cascade feeding the isomer. Evidence for an isomeric M2 transition at an energy of 182 keV was also observed to be correlated with the proton decay of ^{116}La, as shown in Figure 2(b). Its half-life was determined to be $T_{1/2} = 2.0^{+2.8}_{-0.8}$ μs.

Figure 3 (a) displays the energy spectrum for decay events occurring within 100 ms of a recoil implantation in the DSSD and in delayed coincidence with recoil-decay-correlated isomeric γ rays. In this way, the dominant β background in the DSSD spectra could be greatly reduced and the new proton peak assigned to ^{116}La becomes more pronounced. The inset shows a two-dimensional spectrum of decay energy versus ion mass number, where only ions with charge state 29^+ were selected. For this charge state the recoil implantation rate was substantially lower with a correspondingly lower burden of random correlations between the implantation of fusion residues and β rays. The upper part of the inset to Fig. 3 (a) shows the measured mass distributions for recoils with mass 116 and 117 in the same charge state. The distributions drawn in red and blue correspond to the strongly populated nuclides ^{116}I and ^{117}Xe, respectively, which are obtained by gating on their characteristic isomeric γ rays. In the two-dimensional histogram of energy vs mass the high-energy group corresponds to the ground-state proton decays of ^{117}La, while the low-energy distribution corresponds to β-decay events. The five events in the middle group coincide with the distribution for ions with mass 116 and with the proton line of the new isotope ^{116}La. Panel (b) shows decays within 3 s of a recoil implantation. Using the logarithmic binning method described by Schmidt *et al.* [42], a two-component function was used to fit the lifetime of the decay events within the shaded region of Figure 3 (b). A half-life of $T_{1/2} = 50(22)$ ms was deduced for the proton decay of ^{116}La.

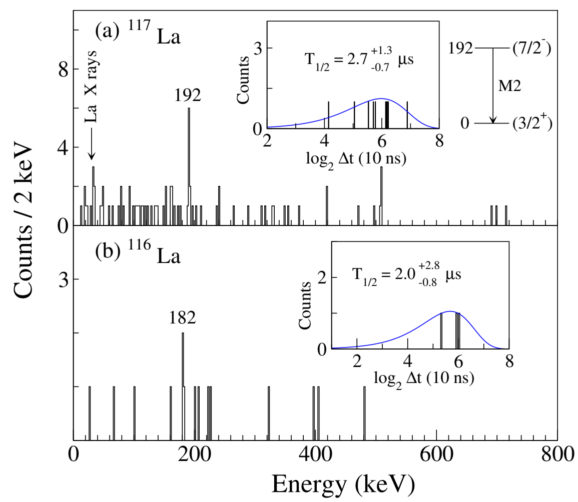

FIG. 2. (a) Selection of γ-ray energies detected within 8 μs of a recoil implantation into the DSSD and followed by a proton decay from ^{117}La in the same quasipixel. The inset shows the time distribution of 192-keV γ-rays selected in this way. The fit of the lifetime using the maximum likelihood method [42] is indicated by the blue curve. The proposed corresponding level scheme obtained for ^{117}La is indicated to the right. (b) Same as (a) tagged by ^{116}La proton decays and for 182-keV γ-rays.

Discussion. The ground-state and low-lying yrast configurations of the neutron deficient lanthanum isotopes are predicted to be moderately quadrupole-deformed ($\beta_2 \approx 0.3$) [43] and based on valence protons occupying Nilsson [44] orbits from the near-degenerate $2d_{5/2}$ and $1g_{7/2}$ sub-shells. Levels based on configurations with $g_{9/2}/h_{11/2}$ spherical parentage that extrude/intrude with increasing deformation may also come close to the Fermi level. The detailed balance determining which configuration actually forms the ground state may provide important input for

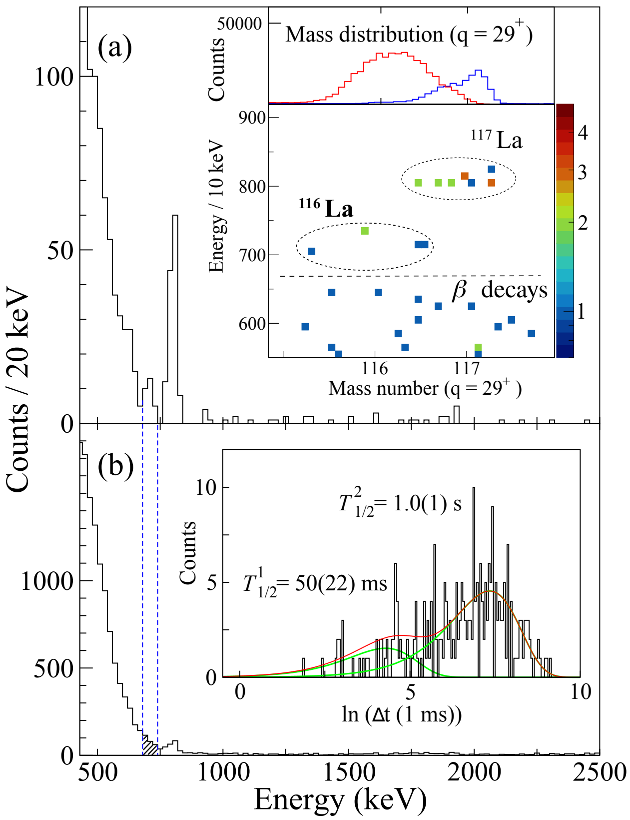

FIG. 3. Projected decay energy spectrum measured in the DSSD for events correlated with a prior detection of a recoil-γ event. (a) The decay events were required to occur within 100 ms after recoil implantation into the same DSSD quasipixel. The lower inset shows the two-dimensional distribution of ion mass vs decay energy for ions in charge state $q = 29^+$. The upper inset shows the mass distributions of ^{116}I and ^{117}Xe ions from the same experiment measured in MARA drawn in red and blue, respectively. (b) The decay events were measured within 3 s of a recoil implantation. The blue dashed lines are drawn to illustrate the location of the proton peak of ^{116}La, as indicated by the shaded area. The inset shows the logarithmic time spectrum of the decay events in the shaded region, fitted using a two-component function (red). The green curves show each fitted component. The distribution of higher ln(Δt) values corresponds to the random-correlated β-ray background with its effective fitted half-life.

stringent tests of effective nucleon-nucleon interactions used in current nuclear models. This situation also provides the necessary requirements for nuclear isomerism created by yrast spin traps [45, 46]. The relevant Nilsson configurations near the Fermi level have been discussed extensively in the literature for neighboring isotopes closer to the line of beta stability, see e.g. the recent work on ^{119}Cs [47]. In ^{121}La, the most neutron deficient lanthanum isotope for which excited states have been identified to date, two rotational bands have been observed and assigned to be built on the $9/2^+[404]$ and $1/2^-[550]$ Nilsson orbitals [48].

In theoretical studies of the proton emission rate from ^{117}La the adiabatic approach suggested ground-state spin-parity $J^\pi = 3/2^+$ [49], while non-adiabatic calculations proposed $J^\pi = 3/2^+$ or $J^\pi = 3/2^-$ [50, 51]. The $J^\pi = 3/2^+$ assignment corresponds to the $3/2^+[422]$ Nilsson orbital [47] of mainly $2d_{5/2}$ parentage and hence emission of predominantly $l = 2\hbar$ protons. On the other hand, $J^\pi = 3/2^-$ would correspond to the $3/2^-[541]$ $1h_{11/2}$ intruder configuration [47] and, consequently, $l = 5\hbar$ proton emission which would be expected to be subject to additional strong hindrance due to the larger centrifugal barrier. For the proton emitter ^{121}Pr, it was proposed [52] that the $J^\pi = 7/2^-$ member of such a configuration could form the ground state as a result of a strong Coriolis interaction, i.e. a kinematic coupling of the angular momenta of the odd valence proton and the core. Even if this state is unlikely to form the ground state of ^{117}La it may well produce a low-lying excited state resulting in a spin-trap isomer. The 192-keV M2 transition found in this work is therefore assigned to populate the $3/2^+$ ground state from this $7/2^-$ state as indicated in Figure 2. For ^{116}La, the spin-parity of its ground-state is considerably more difficult to assess due to the numerous possibilities to couple the low-lying proton and neutron configurations. Therefore, even though the 182-keV M2 isomeric transition is most likely of similar character, it was not placed into a tentative level scheme.

The Universal Decay Law is a convenient, model independent microscopic approach to quantum tunneling theory which can be applied to all forms of ground-state to ground-state radioactive decays involving the emission of protons and heavier charged particles [53, 54]. The expression for the half-life can be written as

$$T_{1/2} = \frac{\hbar \ln 2}{\Gamma_l} = \frac{\ln 2}{\upsilon} \left| \frac{H_l^+(\chi,\rho)}{RF_l(R)} \right|^2 \quad (1)$$

where l the angular momentum carried by the emitted particle and υ is its outgoing velocity. The distance parameter, R, denotes the point where the radial wave function describing the proton in the internal region of the nucleus is matched with its outgoing wave function. H_l^+ is the Coulomb-Hankel function which can be well approximated by the Wentzel-Kramers-Brillouin (WKB) value [55]. The formation probability, $|RF_l(R)|^2$, that can be extracted from Eq. 1 can provide a more precise evaluation of the influence of the nuclear structure on the proton-decay, especially in the case of a deformed nucleus [54].

Figure 4 shows the $|RF_l(R)|^2$ values as extracted from the experimental half-lives and Q values from the ground-state proton-decays observed to date in odd-Z elements between $Z = 53$ and 83. In the present analysis it was not possible to accurately measure the β-decay branching ratio for the ground-state decay of ^{116}La. Based on the theoretical calculations by Möller *et al.*, which predicted a partial β-decay half-life of 124 ms for ^{116}La [56], the proton-decay branching ratio is estimated to be 60(18)% and, consequently, the partial proton-decay half-life is deduced as $T_{1/2,p} = 84^{+86}_{-50}$ ms. The formation probability for ^{116}La is furthermore calculated under two alternative assumptions, that the orbital angular momentum carried by the emitted proton is $l_p = 2\hbar$ or $l_p = 4\hbar$, while $l_p = 0$, $l_p = 1\hbar$ and $l_p = 3\hbar$ have been excluded based on the lack of proton orbitals with s, p or f parentage close to the

Fermi level. It is clearly seen from Figure 4 that a reasonable value for the formation probability in the proton decay of ^{116}La can only be obtained when $l = 2\hbar$. This firmly assigns the proton component of the ground-state wave function of ^{116}La to be of predominantly $d_{5/2}$ parentage, which is consistent with the results for ^{117}La derived in the present work as well as the neighboring proton-emitters $^{108,109}_{53}$I, $^{112,113}_{55}$Cs, $^{121}_{59}$Pr, and $^{130,131}_{63}$Eu [37].

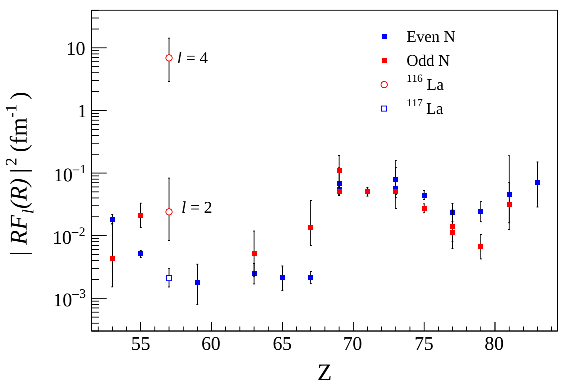

FIG. 4. Proton formation probabilities for the ground-state decays in the odd-Z elements between $Z = 53$ and 83 as a function of the proton number Z. The Q_p values and half-lives for ^{116,117}La are from the results obtained in the present work. The remaining experimental data used for this plot are taken from Ref. [37] as well as from the recently reported results on ^{108}I [28] and ^{183}Bi [57]. Experimental uncertainties in the half-lives and Q-values have been taken into account when calculating the error bars of the formation probabilities. The formation probability, $|RF_l(R)|^2$, has been calculated for ^{117}La using the proton orbital angular momentum $l_p = 2\hbar$ and for ^{116}La using two alternative values, $l_p = 2\hbar$ and $l_p = 4\hbar$. For ^{116}La, the partial proton-decay half-life was calculated based on the theoretically predicted β-decay partial half-life of 124 ms [56]. Information on the values of l_p, Q_p and half-lives used for the calculation as well as the derived formation probabilities for other proton-emitters can be found in Table II of Ref. [58].

It can be observed that the proton formation probabilities shown in Figure 4, are divided into two main regions at $Z = 69$, where $|RF_l(R)|^2$ reaches the maximum value for the nucleus $^{144}_{69}$Tm. Focusing first on the even-N nuclides in the region with $Z \geq 69$ they have consistently the largest $|RF_l(R)|^2$ values with a dip appearing around $_{77}$Ir - $_{79}$Au. The behavior is associated with the nuclear deformation of these proton emitters which are predicted to be spherical or weakly-deformed in this region [43] and the ground-state proton wave function to be dominated by a single-particle orbit [59]. Consequently, the formation probability of this region is generally large, and the dip in $|RF_l(R)|^2$ around the iridium and gold proton emitters is likely related to the transitional onset of weakly-deformed shapes [43]. For the proton emitters with $Z < 69$ the $|RF_l(R)|^2$ values are consistently around one order of magnitude smaller and it was noted that this may be expected in this deformed region [55] for which the decay primarily involves only the low- l components of the deformed ground-state configuration [59–61]. However, the deformation effect on the formation probability seems to saturate already at moderate deformations since the $|RF_l(R)|^2$ values exhibit a nearly constant trend also beyond the strongly deformed $_{63}$Eu - $_{67}$Ho rare-earth nuclei. For the weakly deformed $_{53}$I - $_{55}$Cs nuclei the $|RF_l(R)|^2$ values again start increasing as a function of decreasing Z, approaching the spherical shapes close to the doubly-magic, self-conjugate nucleus $^{100}_{50}$Sn$_{50}$.

Turning now to the odd-odd isotopes, a striking effect is visible in Figure 4, namely that three of the odd-N isotopes in the deformed region; $^{112}_{55}$Cs, $^{116}_{57}$La, and $^{140}_{67}$Ho have significantly larger $|RF_l(R)|^2$ values than the closest, less exotic, even-N isotope. This implies that the presence of the odd valence neutron induces a facilitating effect on the proton emission in these odd-odd systems compared with the neighboring isotope where all neutrons are paired. At the same time, the differences between the ground-state to ground-state proton-decay Q values of the odd-N proton-emitters and their less exotic nearest even-N isotopes, $\Delta Q_p^{oe} = Q_p^N - Q_p^{N+1}(N \text{ odd})$, are negative: $\Delta Q_p^{oe} = -153(8)$ keV for $^{112,113}_{55}$Cs, -90(10) keV for $^{116,117}_{57}$La, and -83(11) keV for $^{140,141}_{67}$Ho. We also note that while $\Delta Q_p^{oe} = -222(14)$ keV for $^{108,109}_{53}$I the difference between the corresponding $|RF_l(R)|^2$ values is within the experimental uncertainties and therefore inconclusive.

The enhanced $|RF_l(R)|^2$ values for these odd-odd proton emitters and the corresponding negative ΔQ_p^{oe} values for $^{112,113}_{55}$Cs, $^{116,117}_{57}$La, and $^{140,141}_{67}$Ho isotopic pairs stand out by themselves. The *coincidence* of these quantities is even more remarkable and seems to require an explanation beyond the current understanding of proton radioactivity.

We have considered various mechanisms that might explain the observed effect and find that a possible scenario involves neutron-proton pairing effects, which may be enhanced in extremely neutron deficient nuclei for which neutron and proton numbers are approximately equal and neutrons and protons near the Fermi level therefore move in similar orbits. The properties of odd-odd nuclides are of particular interest in this sense since like-particle pair correlations are expected to be blocked by the odd valence particles.

How could then an enhanced np pairing strength contribute to these effects? As discussed above, a sufficient number of valence neutrons and protons moving in identical orbits may produce an np pairing condensate that could increase the binding of exotic, extremely neutron deficient odd-odd systems compared with those situated further down in the valley of β stability. But such an effect could also be a result of a strong attractive residual np interaction as discussed in earlier work [29]. The enhancement of the $|RF_l(R)|^2$ values for the odd-odd proton emitters, which indicate that the odd neutron facilitates the emission of the proton instead of binding it stronger, seems, however, at first glance contradictory to a situation with an increased binding of the same due to a strong residual np interaction between the two valence nucleons. On the other hand, for a large np pair gap of the isovector type one may expect properties similar to "normal" isovector pairing of the like-particle type. Such pair correlations have commonly been assumed to become especially important at the nuclear surface, but there are few rigorous treatments of this difficult problem. In a study of the spatial distribution of isovector pairing strength within the HFB approach, Sandulescu *et al.* [62] calculated the neutron pairing density for the chain of even-even tin isotopes between doubly-magic $^{100}_{50}\mathrm{Sn}_{50}$ to $^{132}_{50}\mathrm{Sn}_{82}$. While the probability density for d-wave particles, which are relevant for the considered proton emitters, was predicted to be at a minimum at the nuclear surface, the pairing density was found to be sustained throughout the nucleus and with a clear enhancement at the surface. In a subsequent study, Vigezzi *et al.* [63] investigated the spatial distribution of the pairing density in ^{120}Sn using a bare nucleon-nucleon potential approach as well as with a pairing interaction induced by the exchange of collective vibrations. The resulting pairing density was found to be strongly peaked on the nuclear surface.

We propose that a mechanism by which proton emission in an odd-odd nucleus could be enhanced is via an isovector np pairing condensate and its modifying effects on the valence proton wave function. This might result in a more surface-peaked proton density distribution, thereby facilitating the tunneling process through the Coulomb and centrifugal barriers. A dynamic enhancement of the proton emission probability involving np pairs scattering into proton continuum states could also play a role in such a process. The fact that the effect is not observed in the region of near-spherical or weakly deformed nuclei is consistent with a smaller pairing gap in these nuclei while the deformed region with its higher level density and larger pairing gap, as well as closer proximity to the $N = Z$ line seems more favorable. Whether the observed effect could also be explained by np pairing of the isoscalar type is a possibility that requires additional theoretical investigation. These observations highlight the importance of proton emission as a probe of fundamental nuclear structure theory and illustrate the need for pushing the experimental boundaries of proton emission measurements further.

Conclusions. The extremely neutron deficient isotope ^{116}La has been discovered via its ground-state proton emission ($E_p = 718(9)$ keV, $T_{1/2}$ =50(22) ms). The proton decay of ^{117}La has also been remeasured ($E_p = 808(5)$ keV, $T_{1/2} = 21.6(31)$ ms). Isomeric transitions of M2 character belonging to ^{117}La and ^{116}La were observed with energies and corresponding half-lives ($E_\gamma = 192$ keV, $T_{1/2} = 1.8^{+0.6}_{-0.4}$ μs) and ($E_\gamma = 182$ keV, $T_{1/2} = 2.0^{+2.8}_{-0.8}$ μs), respectively. An enhanced proton emission probability for ^{116}La as well as for a few other odd-odd proton emitters compared with their neighboring, less-exotic, odd-even isotopes is observed and found to coincide with negative ΔQ_p^{oe} values in the same cases. This unexpected effect is proposed as a manifestation of strong neutron-proton pairing of the isovector type in these systems.

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[1] W. Heisenberg, Z. Phys. **78**, 156 (1932).
[2] A. de Shalit and I. Talmi, *Nuclear Shell Theory* (Academic Press, New York, 1963).
[3] I. Talmi, *Simple Models of Complex Nuclei* (Harwood Academic Press, Switzerland, 1993).
[4] D. Rowe and G. Rosensteel, Phys. Rev. Lett. **87**, 172501 (2001).
[5] H. R. A. Johnson and J. Sztarkier, Phys. Lett. B **34**, 605 (1971).
[6] F. Stephens and R. Simon, Nucl. Phys. A **183**, 257 (1972).
[7] J. Bardeen, L. N. Cooper, and J. R. Schrieffer, Phys. Rev. **106**, 162 (1957).
[8] J. Bardeen, L. N. Cooper, and J. R. Schrieffer, Phys. Rev. **108**, 1175 (1957).
[9] J. Dobaczewski, H. Flocard and J. Treiner, Nucl. Phys. A **422**, 103 (1984).
[10] J. Engel, K. Langanke and P. Vogel, Phys. Lett. B **389**, 211 (1996).
[11] J. Engel, S. Pittel, M. Stoitsov, P. Vogel, and J. Dukelsky, Phys. Rev. C **55**, 1781 (1997).
[12] O. Civitarese, M. Reboiro, and P. Vogel, Phys. Rev. C **56**, 1840 (1997).
[13] A. Goodman, Adv. Nucl. Phys. **11**, 263 (1979).
[14] W. Satuła and R. Wyss, Phys. Rev. Lett. **87**, 052504 (2001).
[15] G. Martínez-Pinedo and K. Langanke and P. Vogel, Nucl. Phys. A **651**, 379 (1999).
[16] D. D. Warner, M. Bentley, and P. Van Isacker, Nature Phys. **2**, 311 (2006).
[17] B. Cederwall *et al.*, Nature (London) **469**, 68 (2011).
[18] B. Cederwall *et al.*, Phys. Rev. Lett. **124**, 062501 (2020).
[19] X. Liu *et al.*, Phys. Rev. C **104**, L021302 (2021).
[20] S. Frauendorf, A.O. Macchiavelli, Part. Nucl. Phys. **78**, 24 (2014).
[21] A. L. Goodman, Phys. Rev. C **60**, 014311 (1999).
[22] A. Gezerlis, G. F. Bertsch, Y.L. Luo, Phys. Rev. Lett. **106**, 252502 (2011).
[23] S. Hofmann, W. Reisdorf, G. Münzenberg, F.P. Heßberger, J.R.H. Schneider, P. Armbruster, Z. Phys. A **305**, 111 (1982).
[24] O. Klepper, T. Batsch, S. Hofmann, R. Kirchner, W. Kurcewicz, W. Reisdorf, E. Roeckl, D. Schardt, G. Nyman, Z. Phys. A **305**, 125 (1982).
[25] A. Gillitzer, T. Faestermann, K. Hartel, P. Kienle, E. Nolte, Z. Phys. A **326**, 107 (1987).
[26] P.J. Woods, C.N. Davids, Annu. Rev. Nucl. Part. Sci. **47**, 541 (1997).
[27] R. D. Page *et al.*, Z. Phys. A **338**, 295 (1991).
[28] K. Auranen *et al.*, Phys. Lett. B **792**, 187 (2019).

[29] R.D. Page *et al.*, Phys. Rev. Lett. **72**, 1798 (1994).
[30] Suzuki *et al.*, Phys. Rev. Lett. **119**, 192503 (2017).
[31] M. Patial, P. Arumugam, A. K. Jain, E. Maglione, L. S. Ferreira, Phys. Rev. C **88**,, 054302 (2013).
[32] J. Uusitalo, J. Saren, J. Partanen, J. Hilton, Acta Physica Polonica B **50**, 319 (2019).
[33] J. Sarén *et al.*, Nucl. Instr. Meth. Phys. Res. A **266**, 4196 (2008).
[34] I. H. Lazarus *et al.*, IEEE Trans. Nucl. Sci. **48**, 567 (2001).
[35] P. Rahkila *et al.*, Nucl. Instrum. Methods Phys. Res. Sect. A **595**, 637 (2008).
[36] E. S. Paul *et al.*, Phys. Rev. C **51**, 78 (1995).
[37] B. Blank and M. J. G. Borge, Prog. Part. Nucl. Phys. **60**, 403 (2008).
[38] F. Heine, T. Faestermann, A. Gillitzer, J. Homolka, M. Köpf *et al.*, Z. Phys. A, **340**, 225 (1991).
[39] F. Soramel *et al.*, Phys. Rev. C **63**, 031304(R) (2001).
[40] H. Mahmud *et al.*, Phys. Rev. C **64**, 031303(R) (2001).
[41] Z. Liu *et al.*, Phys. Lett. B **702**, 24 (2011).
[42] K. Schmidt, C.-C. Sahm, K. Pielenz, and H.-G. Clerc, Z. Phys. A **316**, 19 (1984).
[43] P. Möller, A. J. Sierk, T. Ichikawa, and H. Sagawa, At. Data Nucl. Data Tables **109–110**, 1 (2016).
[44] S. G. Nilsson Mat.-Fys. Medd., **29**, 16 (1955).
[45] A. K. Jain, B. Maheshwari, S. Garg, M. Patial, B. Singh, Nucl. Data Sheets **128**, 1 (2015).
[46] H. L. Liu, F. R. Xu, S. W. Xu, R. Wyss, and P. M. Walker, Phys. Rev. C **76**, 034313 (2007).
[47] K. K. Zheng *et al.*, Phys. Rev. C **104**, 044305 (2021)
[48] B. Cederwall *et al.*, Z. Phys. A **338**, 463 (1991).
[49] E. Maglione *et al.*, Phys. Rev. **59**, R589 (1999).
[50] B. Barmore *et al.*, Phys. Rev. **62**, 054315 (2000).
[51] H. Esbensen and C.N. Davids, Phys. Rev. C **63**, 014315 (2001).
[52] M. C. Lopes, E. Maglione, and L. S. Ferreira, Phys. Lett. B **673**, 15 (2009).
[53] C. Qi, F. R. Xu, R. J. Liotta, and R. Wyss, Phys. Rev. Lett. **103**, 072501 (2009).
[54] D. S. Delion, *Theory of Particle and Cluster Emission* (Springer-Verlag, Berlin, 2010).
[55] D. S. Delion, R. J. Liotta, and R. Wyss, Phys. Rev. Lett. **96**, 072501 (2006).
[56] P.Möller, M. R. Mumpower, T. Kawano, W. D. Myers, At. Data Nucl. Data Tables **125**, 1 (2019).
[57] D.T. Doherty *et al.*, Phys. Rev. Lett. **127**, 202501 (2021).
[58] C. Qi, D. S. Delion, R. J. Liotta, and R. Wyss, Phys. Rev. C **85**, 011303(R) (2012).
[59] C. Qi, R. Liotta, and R. Wyss, Prog. Part. Nucl. Phys. **105**, 214 (2019).
[60] E. Maglione, L. S. Ferreira, and R. Liotta, Phys. Rev. Lett. **81**, 538 (1998).
[61] E. Maglione, L. S. Ferreira, and R. Liotta, Phys. Rev. C **59**, R589 (1999).
[62] N. Sandulescu, P. Schuck, X. Viñas, Phys. Rev. C **71**, 054303 (2005).
[63] E. Vigezzi *et al.*, AIP Conference Proceedings **1120**, 92 (2009).