Discovery of ^{229}Rn and the Structure of the Heaviest Rn and Ra Isotopes from Penning-Trap Mass Measurements

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The masses of the neutron-rich radon isotopes $^{223-229}$Rn have been determined for the first time, using the ISOLTRAP setup at CERN ISOLDE. In addition, this experiment marks the first discovery of a new nuclide, ^{229}Rn, by Penning-trap mass measurement. The new, high-accuracy data allow a fine examination of the mass surface, via the valence-nucleon interaction δV_{pn}. The results reveal intriguing behavior, possibly reflecting either a $N = 134$ subshell closure or an octupolar deformation in this region.

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The nuclear binding energy reflects the net effect of all aspects of the underlying fundamental forces [1]. Its evolution as a function of proton or neutron number provides information of great importance for nuclear structure. Joining the most basic nuclear characteristics of size and weight is that of shape, usually quantified by deformation, when a nucleus is considered not to be spherical.

As the capacious valence orbitals of very heavy nuclides begin to fill with nucleons, increasing varieties of deformation become possible. It is well known [2] that one of these degrees of freedom, of importance in the light actinide nuclei, is that of octupole correlations and octupole deformations. Möller *et al.* [3] show octupole contributions to the binding energy and note that the strongest contribution is centered at ^{222}Rn. The role of octupole deformations, and therefore the study of their impact, is important in several respects. The existence of the actinides and the heaviest nuclei is specifically due to quantum effects in the underlying single particle levels which also manifest in associated collective correlations. The measurement of new masses in this region of octupole correlations therefore provides a significant constraint on future microscopic models, for example, in approaches exploiting density functional theory which is widely used in many areas of many-body physics [4]. Further, octupole correlations are known to greatly enhance the sensitivity to nuclear electric dipole moments: understanding the relations between masses and such correlations is therefore of importance in the wider context of tests of fundamental symmetries. Finally, another important impact of the study of masses in this region is to be found in the development of microscopic mass models for nucleosynthesis, which are particularly vulnerable in heavy nuclei.

In this Letter we report on seven new masses of neutron-rich radon nuclei including the new isotope ^{229}Rn, which marks the first discovery of a nuclide using a Penning-trap mass spectrometer. The results allow the extraction of important new values for the valence proton-neutron interaction in the $A \sim 222$ mass region, confirming and significantly extending a unique anomaly in these interaction strengths that may indeed be related to octupole degrees of freedom in this region.

Penning traps are used extensively [5] to provide accurate mass data of radionuclides with half-lives as short as only a few milliseconds and production yields of a few hundred ions per second [6]. The high resolving power has allowed the separation of isomeric states [7] and even the detection of a new isomer, in the case of ^{65}Fe [8].

The measurements reported in this Letter were performed at the double Penning-trap mass spectrometer ISOLTRAP [9] located at the on-line isotope separator facility ISOLDE at CERN. The radon nuclides were produced by spallation of a 50-g/cm^2 thick UC$_X$ target by pulses of up to 3×10^{13} protons at an energy of 1.4 GeV from CERN's Proton Synchrotron Booster accelerator. The nuclear reaction products diffused from the hot target through a water-cooled transfer line into a high-efficiency arc-discharge ion source, designed via a new approach

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TABLE I. Frequency ratios $r = \nu_{c,\mathrm{ref}}/\nu_c$, relative mass uncertainties $\delta m/m$, and mass excesses of the measured radon isotopes Δ and their literature values Δ_{lit} [15]. Extrapolated mass excess values are marked with *. The reference ion was $^{133}Cs^+$ with $m(^{133}\mathrm{Cs}) = 132.905\,451\,932(23)$ u [15].

A	$r = \nu_{c,\mathrm{ref}}/\nu_c$	$\delta m/m$	Δ (keV)	Δ_{lit} (keV)
220	1.655 400 519(80)	5×10^{-8}	10 614(10)	10 613.4(2.2)
223	1.678 052 066(83)	5×10^{-8}	20 396(10)	20 300*(300*)
224	1.685 592 713(121)	7×10^{-8}	22 435(15)	22 440*(300*)
225	1.693 150 170(174)	10×10^{-8}	26 555(22)	26 490*(300*)
226	1.700 691 984(129)	8×10^{-8}	28 739(16)	28 770*(400*)
227	1.708 249 568(145)	8×10^{-8}	32 875(18)	32 980*(420*)
228	1.715 792 924(180)	11×10^{-8}	35 249(22)	35 380*(410*)
229	1.723 350 324(106)	6×10^{-8}	39 362(13)	

[10]. Off-line studies showed that the ionization efficiencies for noble gases are 5–20 times larger than with the standard ISOLDE plasma ion source (MK7 FEBIAD [11]), and reach 60% for radon (estimated through extrapolation from the He-Xe noble-gas series).

The singly charged ions were accelerated to 30 keV and separated with the high-resolution mass separator with a resolving power of the order of 4000 for a first suppression of any residual isobaric contaminants. The resulting ion beam was injected into the recently installed radio frequency quadrupole cooler ISCOOL [12] (working in transmission mode) to improve the ion-beam emittance. The ions were stopped and bunched in the ISOLTRAP cooler and buncher in order to prepare them for capture into the following two Penning traps. The first, cylindrical trap was used for cooling and isobaric cleaning. The high-precision mass measurements were carried out in the second, hyperboloidal precision Penning trap. Here, the cyclotron frequency $\nu_c = qB/(2\pi m)$ of the ion was measured via the time-of-flight (TOF) ion-cyclotron-resonance detection technique [13], where q and m are the charge and the mass of the ion, respectively, and B the magnetic field strength. In order to calibrate the unknown magnetic field at the time of the measurement, the cyclotron frequency of ^{133}Cs, having a well-known mass, was measured immediately before and after that of the ion of interest. The resolving power, given by the product of the excitation time t_{exc} and the cyclotron frequency ν_c, was $\sim 4 \times 10^5$.

For $^{220,223-228}$Rn, all resonances were taken with an excitation time of 1.2 s. For ^{229}Rn a total of four resonances with excitation times of 100 ms, 600 ms, and twice 1.2 s were recorded. The resonances were fitted to the theoretical curve [13] in order to extract the cyclotron frequency. The data analysis followed the procedure described in [14]. In addition to the systematic uncertainty of 8×10^{-9}, a relative mass-dependent uncertainty of $1.6 \times 10^{-10}(m - m_{\mathrm{ref}})/u$ was added quadratically to the uncertainty of the mean of the measured frequency ratios.

Although the cold transfer line allows only gaseous species to reach the ion source, the TOF ion-cyclotron resonances were carefully checked for the presence of any contaminations, that might shift the center frequency, by a count-rate-class analysis [14]. No anomalies were detected.

The experimental results are given in Table I, which shows for each investigated radon isotope the frequency ratio of the reference ion $^{133}Cs^+$ compared to the ion of interest, the relative uncertainties, the derived mass excesses, and the literature values. The well-known mass of ^{220}Rn [16] was measured as a cross-check and compares very well with [15].

^{229}Rn has never been observed, thus only estimates for the half-life and mass excess were available. The first confirmation that the new isotope ^{229}Rn was produced at ISOLDE came from a measurement of the half-life at the ISOLDE spectroscopy station, shown in Fig. 1. This measurement gives a half-life of $12^{+1.2}_{-1.3}$ s for a nuclide with

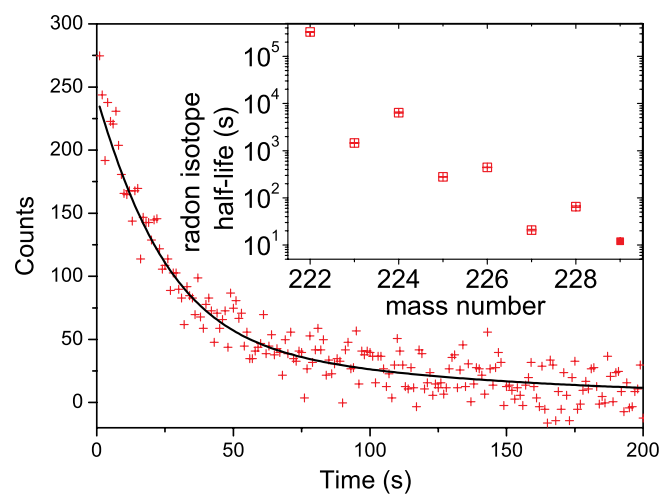

FIG. 1 (color online). Beta-decay curve of ^{229}Rn. Data from 10 separate acquisitions were added and background was subtracted. The fit includes ^{229}Rn with an estimated half-life $t_{1/2}$ (^{229}Rn) of 12 s along with its daughter ^{229}Fr (50.2 s) and granddaughter ^{229}Ra (4.0 m). Inset: Radon half-lives with $t_{1/2}$ (^{229}Rn) nicely following the trend.

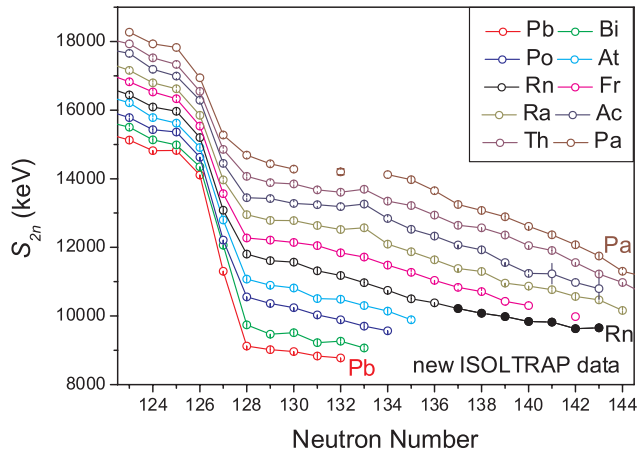

FIG. 2 (color online). Two-neutron separation energies (S_{2n}) for Pb-Pa isotopes with $N = 123$–144 [15]. New radon data are represented by filled circles. A deviation from the parallel decrease of S_{2n} in neighboring chains is visible.

mass number 229 then delivered to ISOLTRAP. The fit accounted for beta counts from the decays of the daughter isotope ^{229}Fr and granddaughter ^{229}Ra (both known). When shown together with the known half-lives of the lighter radon isotopes (see inset of Fig. 1), the trend is convincing. When the new S_{2n} values are plotted (as a function of N) alongside neighboring isotopic chains (see Fig. 2) striking continuity is visible that indicates that the chain of Rn isotopes has been measured. Though the S_{2n} of ^{229}Fr could fall in the same place, we can exclude this nuclide since we looked for its well-known mass, over 3.5 MeV away from the one we measured, and easily within range of the resolving power of the precision trap. Also the yield attributed to ^{229}Rn follows the normal linear trend (note the logarithmic scale), as visible in Fig. 3 which

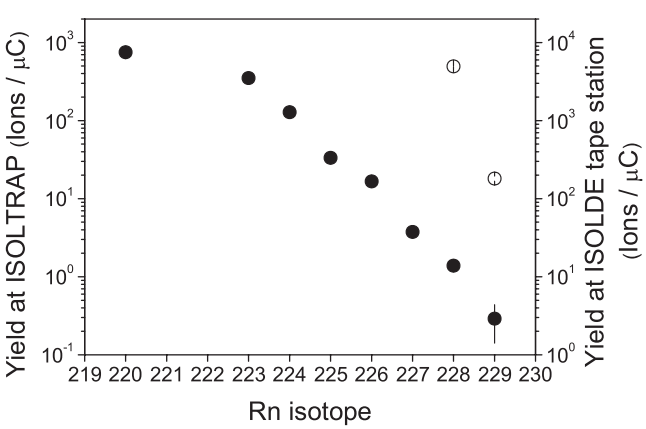

FIG. 3. Yields of the different radon isotopes. The closed circles show the yield measured in the ISOLTRAP precision trap during our measurements. The open circles represent the yield obtained at the ISOLDE tape station. The comparison for ^{228}Rn gives the ISOLTRAP efficiency of about 0.2%, which is in the same range as that for ^{229}Rn.

shows yields for the different radon isotopes in our precision trap. For $^{228-229}$Rn these relative yields are comparable to those determined from the ISOLDE tape station but differ slightly since the measurements were performed at different times during the run. Target conditions are known to vary significantly as the target ages. Furthermore, the data were carefully analyzed in order to exclude any other atomic or molecular isobars. (i) Resonances of possible single-ion contaminants, like ^{229}Fr, are too far away from our measured frequency. (ii) We have verified all possible molecular contaminants with up to three different atoms constituting the molecule in the range of ± 2 Hz, which is 4 times the FWHM of the ^{229}Rn resonance and is much larger than the resonance uncertainty of 0.02 Hz. In addition, none of them is likely to be produced at ISOLDE. (iii) The high amplitude ("TOF effect") of the TOF resonance, comparable with amplitudes for the other radon isotopes, shows that the $A = 229$ beam was very clean after our purification trap.

Atomic masses give nuclear binding and separation energies, and various combinations (double differences) of binding energies provide empirical filters that isolate specific nucleonic interactions [17]. In particular, the δV_{pn} values give the average interaction between the last protons and the last neutrons in even-even and even Z–odd N nuclei [18,19]:

$$\delta V_{pn}^{ee}(Z,N) = \tfrac{1}{4}[(B_{Z,N} - B_{Z,N-2}) - (B_{Z-2,N} - B_{Z-2,N-2})],$$
$$\delta V_{pn}^{eo}(Z,N) = \tfrac{1}{2}[(B_{Z,N} - B_{Z,N-1}) - (B_{Z-2,N} - B_{Z-2,N-1})],$$

where $B_{Z,N}$ is the binding energy. Thus the present measurements of the masses of $^{223-229}$Rn allows us to investigate the critical valence p-n interaction which affects many aspects of nuclear structure such as the single particle energies, magic numbers, collectivity, the onset of deformation, and the geometrical shapes in atomic nuclei [20–22].

Empirical δV_{pn} values in this region are shown in Fig. 4. The peak for ^{208}Pb is well understood in terms of the high spatial overlaps of protons and neutrons filling orbits just below their respective closed shells. This figure, however, shows a second striking feature: the Ra isotopes around $N = 134$ exhibit a sharp peak, similar in magnitude to that in lead. This was already described in [23] as the *Ra puzzle*, and it was speculated that if the mass data are correct, the p-n interactions might sense a softness to octupole deformation that is known in this region. However, the Ra peak is so strong that it is important to first confirm it experimentally.

Although our result for ^{220}Rn has somewhat larger uncertainty (10 keV) than previous results, it clearly confirms the large value of δV_{pn} for ^{223}Ra in Fig. 4 (top), removing any suspicion that it was due to an incorrectly measured Rn mass. The peak near $N \sim 135$ is the most extreme excursion of an individual δV_{pn} value from local trends (except

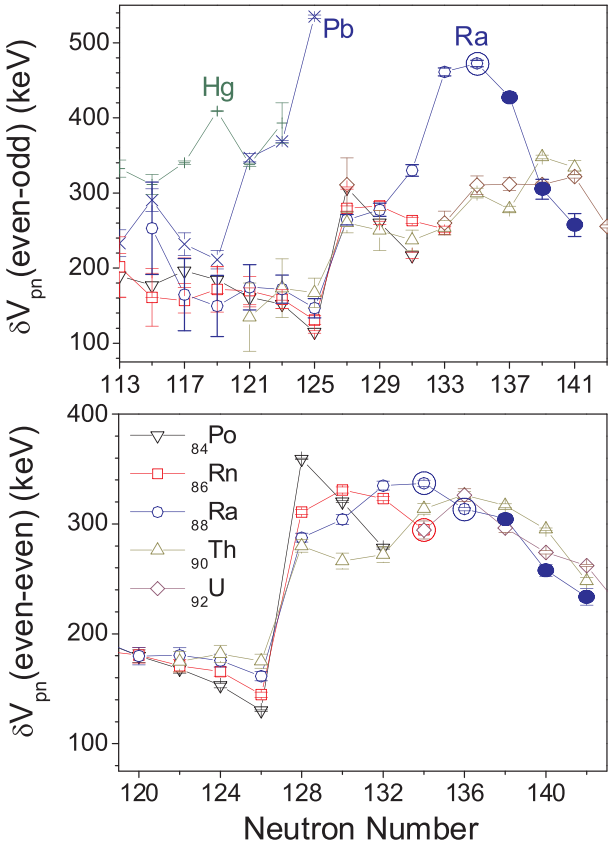

FIG. 4 (color online). Top: Empirical δV_{pn} (even-odd) values for the ^{208}Pb region (from Ref. [23]), with the addition of the new Ra δV_{pn} values (solid circles). The δV_{pn} values involving ^{220}Rn, whose mass is confirmed in the present study, are encircled. Bottom: As top but for even-even nuclei.

for doubly magic nuclei and $N = Z$ nuclei whose origins lie in single particle overlaps and the Wigner force, respectively). Even more importantly, the new results significantly extend, and, indeed, complete, the mapping of a striking anomaly revealing that this point is not an isolated singularity at $N = 135$ but rather part of a very well developed peak in δV_{pn} (even-odd) which terminates at $N = 139$ and which is unique in the nuclear chart. That it appears in a region that is also characterized by the best known example of octupole correlations and of a possible neutron subshell effect at $N = 134$ [2] makes it intriguing and important. It is particularly unexpected that an anomaly of this magnitude would occur in a region of collective structure, where smooth behavior is anticipated due to many-particle correlations. An understanding of structure in this region, including octupole deformations, must account for this anomaly.

In contrast, the δV_{pn} (even-even) values for Ra are consistent with the other nuclei in this region as seen in Fig. 4 (bottom). One interesting feature is that the δV_{pn} values for neighboring elements have similar behavior but are systematically shifted to the right for each successive Z. A similar pattern is visible in the rare-earth-metal region [24] except that here it appears quite early in the p-n major shells while, in the rare-earth-metal region, it is near mid-shell. Whether this behavior is coincidental, or reflects a similar microscopic origin, remains to be studied.

In summary, we have directly determined the masses of $^{223-229}$Rn by precision mass spectrometry for the first time. We have also identified for the first time ^{229}Rn and measured its half-life. These new mass measurements provide significant extensions of known masses in this region, which are important for understanding the binding of the heavy nuclei, which have an impact on nucleosynthesis in the actinide region, and which provide new constraints on modern models of structure and of the mass surface. Before our study, the Ra anomaly in δV_{pn} was restricted to two δV_{pn} values, and therefore there was some doubt about its existence. The present mass measurements prove that it does exist and that it is part of a systematic deviation from the general trend of δV_{pn} values which constitutes the most significant anomaly in p-n interaction strengths anywhere in the nuclear chart (except for $N = Z$ nuclei)—and therefore, the most significant anomaly in collective nuclei. Our results extend the data to show the full behavior of this anomaly. As this occurs in a region of octupole correlations, these results provide a new signature of such correlations, and a new aspect of their influence on nuclear binding.

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