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Revealing the Nature of yrast States in Neutron-Rich Polonium Isotopes

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Polonium isotopes having two protons above the shell closure at Z = 82 show a wide variety of lowlying, high-spin isomeric states across the whole chain. The structure of neutron-deficient isotopes up to ²¹⁰Po (N = 126) is well established as they are easily produced through various methods. However, there is not much information available for the neutron-rich counterparts for which only selective techniques can be used for their production. We report on the first fast-timing measurements of yrast states up to the 8⁺ level in ^{214,216,218}Po isotopes produced in the β^- decay of ^{214,216,218}Bi at ISOLDE, CERN. In particular, our new half-life value of 607(14) ps for the 8_1^+ state in ²¹⁴Po is nearly 20 times shorter than the value available in the literature and comparable with the newly measured half-lives of 409(16) and 628(25) ps for the corresponding 8_1^+ states in ^{216,218}Po, respectively. The measured $B(E2; 8_1^+ \rightarrow 6_1^+)$ transition probability values follow an increasing trend relative to isotope mass, reaching a maximum for ²¹⁶Po. The increase contradicts the previous claims of isomerism for the 8⁺ yrast states in neutron-rich ²¹⁴Po and beyond. Together with the other measured yrast transitions, the B(E2) values provide a crucial test of the different theoretical approaches describing the underlying configurations of the yrast band. The new

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experimental results are compared to shell-model calculations using the KHPE and H208 effective interactions and their pairing-modified versions, showing an increase in configuration mixing when moving toward the heavier isotopes.

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The atomic nucleus, much like an atom itself, is composed of discrete particles, protons and neutrons, which fill energy levels according to quantum principles. In many ways, it behaves as a collection of individual nucleons in specific quantum states. Nuclear isomers are excited metastable states with half-lives of the order of nanoseconds and longer. The isomerism is usually caused by large differences between initial and final states in physical properties, such as total angular momentum, its projection on the symmetry axis (K-isomers), shape, seniority, or the low energy difference between the states [1,2]. By determining the decay energy, branching ratio of de-exciting transitions, and half-life of the state in question, one can deduce the reduced transition probability (a measure of the decay strength usually expressed in Weisskopf units or W.u. [3]) and compare it to the single-particle estimate (which assumes that the decay involves a single nucleon). The ratio of the experimental and single-particle values defines a hindrance (or enhancement) factor, which is related to the underlying decay mechanism or the collective nature of a transition. The occurrence of isomeric states in the vicinity of doubly magic shell closures represents one of the important benchmarks for testing the predictive power of the nuclear shell model using different residual nucleon-nucleon interactions [4].

In the region around the doubly magic ${}^{208}_{82}\text{Pb}_{126}$, lowenergy nuclear structure is often dominated by a relatively high-*j*, single-particle orbital (with $j \ge 7/2$). Seniority (ν) represents the number of particles that are not paired (i.e., they are not coupled to angular momentum J = 0) and can be regarded as a good quantum number. Seniority isomers arise due to the selection rules associated with the conservation of ν and are encountered in semimagic nuclei because electric quadrupole (E2) transitions between $\nu = 2$ states (e.g., $8^+_1 \rightarrow 6^+_1$) are small when the valence shell is close to half filled [5].

Polonium isotopes, having two protons in the $h_{9/2}$ orbital above the Z = 82 shell closure, are especially suitable for testing the seniority scheme across the long chain of isotopes, also crossing the N = 126 shell closure. A large bulk of data exists in the literature on excited states and reduced transition probabilities B(E2) in neutron-deficient ^{198–208}Po isotopes. These nuclei can be studied through fusion–evaporation reactions with heavy ions [6,7], Coulomb excitation experiments [8,9], or β^+ /EC (electron capture) decay [10]. These studies generally agree that the structure of the yrast states (defined as the states of lowest energy for a given angular momentum), including the 8^+ seniority isomers, in the neutron-deficient polonium isotopes is dominated by the two-proton configuration $\pi(h_{9/2})^2$ coupled to the quadrupole vibrations of the underlying even-even ²⁰⁸Pb core [11–13].

For isotopes heavier than ²¹⁰Po, the high-*j* neutron orbital $\nu(g_{9/2})$ starts to be filled. It is therefore expected to compete with the $\pi(h_{9/2})^2$ in determining the configuration of 0^+-8^+ yrast states. The experimental studies of neutron-rich nuclei with Z > 82 and N > 126 are more challenging, as only very specific production techniques can be employed. One of the methods to populate excited states in such nuclei is through multinucleon transfer reactions such as ¹⁸O + ²⁰⁸Pb, used to study ^{210,212,214}Po [14–17].

The previous nuclear structure study of ²¹⁴Po [15] reported its yrast 8⁺ state half-life of 13(1) ns and $B(E2; 8_1^+ \rightarrow 6_1^+) = 0.54(4)$ W.u., underlining its isomeric character. Based on the resemblance between the excitation energies of yrast states in ^{214,216,218}Po, the authors proposed that similar isomers might exist in ^{216,218}Po, their main excitation mechanism being one-neutron-pair breaking.

To reach isotopes beyond ²¹⁴Po, high-energy fragmentation or spallation in direct or inverse kinematics are the only methods of choice [18,19]. In the past, by using proton-induced spallation in ²³²Th and ²³⁸U at ISOLDE, CERN, yrast states in ^{216,218}Po up to 8⁺ have been populated through the β^- decay of ^{216,218}Bi [20,21]; however, half-life measurements of excited states were not performed.

In this Letter, we report on the first fast-timing measurements of the yrast 2^+ , 4^+ , 6^+ , and 8^+ states in 214,216,218 Po populated in the β^- decay of 214,216,218 Bi at ISOLDE, CERN. The present results strongly disagree with the previous literature value of the $T_{1/2}(8_1^+)$ in 214 Po [15]. The extracted B(E2) values are compared to state-of-the-art shell-model calculations in order to understand the underlying structure of the yrast bands of 214,216,218 Po leading to a comprehensive characterization of the single-particle excitations in the "northeast" region of 208 Pb in the nuclear chart. We note that the detailed discussion of the experimental setup, decay schemes, and newly identified states in 214,216 Po from the same experiment has been recently reported for the β^- decay of 214 Bi [22] and 216 Bi [23]. The 214,216,218 Bi isotopes were produced at ISOLDE,

The ^{214,216,218}Bi isotopes were produced at ISOLDE, CERN [24] by bombarding a 50-g/cm²-thick UC_x target with the 1.4 GeV proton beam delivered by the Proton Synchrotron Booster. The bismuth atoms released from the target were selectively ionized by the Resonance Ionization



FIG. 1. β -gated γ -ray spectra recorded by the HPGe (black) and LaBr₃(Ce) (red) detectors following the β^- decay of ²¹⁴Bi (a), ²¹⁶Bi (b), and ²¹⁸Bi (c). The yrast transitions in ^{214,216,218}Po are labeled. γ rays from the β^- decays of ^{84–88}Br were also identified, originating from Ba^{84–88}Br⁺ radioactive molecules produced in the target and matching mass 214, 216, or 218. The most intense contaminant visible in the region of interest is marked with the * symbol in panel (c) and originates from the β^- decay of ⁸⁷Kr.

Laser Ion Source [25], accelerated to 50 keV and massseparated using the ISOLDE High Resolution Separator. The bismuth ions were implanted on an aluminized Mylar[®] tape at the center of the ISOLDE Decay Station (IDS) detection setup [26] equipped with a fast EJ232 plastic scintillator used as a β detector, four HPGe Clover detectors for the detection of γ rays in the daughter nuclei, and two small-volume (30.5 cm³) conic LaBr₃(Ce) detectors. This detection setup enabled the measurement of nuclear half-lives using the $\beta \gamma(t)$, $\gamma \gamma(t)$, and $\beta \gamma \gamma(t)$ fast-timing method [27,28], a well-established technique at IDS employing a similar setup as described in Refs. [29–32]. It covers half-lives between 10 ps and 100 ns by using fasttiming detectors such as LaBr₃(Ce) and plastic scintillators in coincidence with HPGe detectors. More details about the calibration procedure of the detectors is provided in Supplemental Material [33].

Energy spectra recorded by the HPGe and LaBr₃(Ce) detectors in coincidence with β^- events are shown in Fig. 1. The production yields of ^{214,216,218}Bi ions were measured to be >2 × 10⁴ (limited in order to reduce the count rate in the detectors), 1.5 × 10³, and 2 × 10² ions/µC, considering an average proton current of 1.2, 1.6, and 1.8 µA and a total running time of 3, 6.5, and 13 h, respectively.

The half-lives of the 8_1^+ states in ^{214,216,218}Po were extracted through the $\beta\gamma\gamma(t)$ method by gating on β particles in the plastic scintillator as the START signal and the $8_1^+ \rightarrow 6_1^+ \gamma$ rays in the LaBr₃(Ce) detectors as the STOP signal for the time-to-amplitude converters. An extra gating condition was required on γ rays in the HPGe detectors originating from any of the three transitions below the 6_1^+ levels $(6_1^+ \rightarrow 4_1^+, 4_1^+ \rightarrow 2_1^+, \text{ and } 2_1^+ \rightarrow 0_1^+)$



FIG. 2. (a)–(c) Background-subtracted time-difference distributions between β particles recorded in the fast plastic scintillator and γ rays directly depopulating the yrast 8⁺ states in ^{214,216,218}Po, extracted from β - γ_{La} - γ_{Ge} events. (d)–(l) Delayed (black) and antidelayed (red) background-subtracted time distributions between γ rays directly feeding and de-exciting the yrast 2⁺, 4⁺, and 6⁺ states in ^{214,216,218}Po, extracted from γ_{La} - γ_{La} events. The half-lives, reported in Table I, were measured using the decay-slope method [(a)–(c)] and the centroid-shift method [(d)–(1)] [27,28]. The longer-lived component (tail) in the ²¹⁴Po 6⁺ spectrum (d) originates from the 8⁺ half-life due to an overlap in the LaBr3(Ce) energy spectra between the feeding (240 keV) and de-exciting (245 keV) transitions. However, it was excluded when evaluating the centroid shift of the distribution.

in order to further reduce the background of resulting timedifference distributions, shown in Fig. 2.

The possible delayed contributions from high-lying, long-lived levels feeding the 8_1^+ states were investigated by HPGe gating on the γ -ray transitions from above. In the case of ^{214,216}Po, we report half-lives of 73(7) and 155(14) ps for the 8_2^+ 1824.5- keV [22] and 1802.6-keV [20] levels, respectively. For ²¹⁸Po, no long-lived states feeding the 8_1^+ level were identified, either because of the lack of statistics or their absence. The LaBr₃(Ce) detectors cannot resolve the transitions feeding (240 keV) and deexciting (244 keV) the yrast 8^+ state in ²¹⁴Po [see Fig. 1(a)]; however, the two contributions were taken into account for

TABLE I. Experimental (Expt.) $T_{1/2}$ and B(E2) values in ^{214–218}Po nuclei (Nucl.) measured in the present Letter and Ref. [15] (bold), compared to calculated B(E2)s using various effective interactions: H208, KHPE, and their pairing-modified versions.

			$B[E2; J^{\pi} \to (J-2)^{\pi}]$ (W.u.)				
Nucl.	J^{π}	$T_{1/2}$ (ps) Expt.	Expt.	H208	H208 -m	KHPE	KHPE -m
²¹⁴ Po	$2^+_1 \\ 4^+_1 \\ 6^+_1 \\ 8^+_1$	13(5) 35(5) 118(5) 607(14) 13(1) ns	7(3) 18(3) 16(1) 11.3(3) 0.54(4) [15]	13.6 16.9 11.4 1.2	14.3 18.5 14.7 9.2	12.3 13.3 5.9 0.1	13.2 16.5 12.6 6.6
²¹⁶ Po	$\begin{array}{c} 2^+_1 \\ 4^+_1 \\ 6^+_1 \\ 8^+_1 \end{array}$	11(5) 21(5) 31(5) 409(16)	13(6) 26(6) 37(6) 24(1)	18.1 25.2 18.8 16.2	18.9 27.1 25.0 17.8	17.7 22.1 9.8 3.2	18.7 25.2 23.2 15.5
²¹⁸ Po	$2^+_1 \\ 4^+_1 \\ 6^+_1 \\ 8^+_1$	<15 <15 20(8) 628(25)	>13 >33 40(16) 7.8(3)	19.2 29.9 28.3 8.5	20.1 31.5 35.0 16.2	14.8 21.5 2.8 1.0	15.3 22.9 3.2 0.003

the half-life analysis. The values of the half-lives reported in Table I are corrected for the contributions mentioned above and result from the convolution fit between a Gaussian (prompt response of the fast-timing detectors), an exponential decay, and a flat background.

The time distributions for the decays of 2_1^+ , 4_1^+ , and 6_1^+ states in ^{214,216,218}Po are shown in Figs. 2(d)–2(l). The deduced half-lives are reported in Table I and have been measured through the $\gamma\gamma(t)$ method by extracting the timing information from the LaBr₃(Ce) detectors for γ rays directly populating and depopulating each level.

In the case of ²¹⁴Po, a significant discrepancy is observed between the newly measured half-life value of $T_{1/2}(8_1^+) =$ 607(14) ps, and the previous one of 13(1) ns [15]. The latter was measured using the slope fitting of timedifference distributions from HPGe detectors, known for having a much slower and less accurate time response [40] compared to the LaBr₃(Ce) detectors [29].

To link the newly measured data to the underlying nuclear structure, two effective interactions were used in the present Letter for the calculation of low-energy levels and *E*2 transition rates: (i) the recently developed H208 effective interaction [4,41] already used in the interpretation of 212,214,216 Po new experimental data [22,23,42,43]; and (ii) the well-established Kuo-Herling interaction as modified by Warburton and Brown [44], denoted as KHPE. More details regarding the calculations are provided in Supplemental Material [33].

Additional modifications of the two interactions, denoted by H208-m and KHPE-m, were implemented within this Letter in order to reconcile calculated and experimental B(E2) values but maintain the good agreement for other observables using the original interactions. Specifically, the diagonal and off-diagonal $1h_{9/2}$ protonpairing matrix elements were reduced by 100 keV in H208-m. In the case of the KHPE-m interaction, only a larger reduction of 200 keV produced a similar improvement for ^{210–216}Po. However, in ²¹⁸Po, the 8_1^+ state excitation energy becomes lower than the 6_1^+ one, and, therefore, the reduction was limited to 100 keV in the case of ²¹⁸Po. The $1h_{9/2}0i_{13/2}$ matrix elements were not modified, as they had an insignificant effect on the studied *E*2 transitions.

The calculated and experimental excitation energies of the $0^+ - 8^+$ yrast levels in ^{210–218}Po isotopes are shown in Fig. 3. The comparison indicates that the H208 interaction is in better agreement with the adopted experimental data [45–48] than KHPE, except for ²¹⁰Po [49]. A common shortcoming is a compression by about 200 keV observed for the 8^+_1 state of ²¹⁸Po, which we attribute to the adopted truncation for that nucleus.

In order to investigate in detail the wave-function structure of the yrast states, Table II in Supplemental Material [33] reports their main components (with $\geq 10\%$ contribution), where we note the following points: (i) all the yrast 2⁺-8⁺ states in ²¹⁰Po (N = 126) have nearly pure-proton $1h_{9/2}$ contribution; and (ii) for N > 126 the contribution from the pure $1h_{9/2}$ diminishes, and the composition is more fragmented. The neutron $1g_{9/2}$ state starts to play a strong role in all the yrast states, e.g., nearly 54% in 0⁺-6⁺ states in ²¹²Po, and with a further admixture of $1f_{7/2}$ proton for the 8_1^+ state. An even more fragmented wave function can be seen for ²¹⁴⁻²¹⁸Po, with the added contribution from the $0i_{11/2}$ neutron orbital.

The calculated $B[E2; J^{\pi} \rightarrow (J-2)^{\pi}]$ reduced transition probabilities interconnecting the yrast 0^+-8^+ states are compared to our new results and to available data from the literature [15,16,45,49,50] in Fig. 4, Table I here, and Table I in Supplemental Material [33]. The results of the



FIG. 3. The calculated 0^+-8^+ yrast energy levels of eveneven $^{210-218}$ Po isotopes using H208, H208-m, KHPE, and KHPE-m interactions, compared to the available experimental data [15,45–49].



FIG. 4. $B[E2; J^{\pi} \rightarrow (J-2)^{\pi}]$ values for transitions between ^{214,216,218}Po yrast states measured in the present Letter (full circles with uncertainties) and from adopted experimental values [45,49] or the most recent experimental results [15,16,50]. The comparison with shell-model calculations using the H208 interaction, initial and pairing-modified versions (colored lines), is presented. Lower limits for the experimental B(E2) values are indicated with an arrow pointing up. All the values are reported in Table I.

H208 and KHPE calculations are close to each other, with the exception of ²¹⁸Po, where different truncations are implemented. Overall, they are within error bars with the new measurements, with few exceptions mentioned below.

We first note the overestimation of the $B(E2, 2_1^+ \rightarrow 0_1^+)$ values in ^{210,212,214}Po isotopes by a factor of two. However, the calculated $B(E2; 4_1^+ \rightarrow 2_1^+)$ and $B(E2; 6_1^+ \rightarrow 4_1^+)$ values are very well reproduced by the shell-model calculations, shown in Table I, which suggests that the correct structure of the 2_1^+ -state wave function was taken into account. This $B(E2, 2_1^+ \rightarrow 0_1^+)$ discrepancy was already explained in Refs. [44,51] by the absence of the contribution from particle-hole core excitations in the 0_1^+ wave function, and, later, the α -cluster model was proposed to address this issue [50].

The most striking discrepancy is observed for the $B(E2; 8^+_1 \rightarrow 6^+_1)$ values in ²¹⁴Po, where our newly measured value is at least 10 times larger than the original shell-model-calculated one, questioning the isomeric nature of the 8^+_1 state in ²¹⁴Po and excluding any hindrance originating from seniority selection rules. This represents the first experimental indication of an increased fragmentation of the yrast states' wave functions. To understand the origin of this inconsistency between theory and our new measurements, the modified effective interactions were employed. As a consequence, there was a remarkable increase of the calculated $B(E2; 8^+_1 \rightarrow 6^+_1)$ value in ²¹⁴Po, from 1.2 to 9.2 W.u. in H208-m and from 0.1 to



FIG. 5. The wave-function components of ²¹⁴Po states calculated in seniority scheme where T(x, y) represents the number of neutron (x) or proton (y) pairs being broken. The initial and pairing-modified versions of the H208 effective interaction are employed within the NATHAN code.

6.6 W.u. in KHPE-m, close to the measured value of 11.3(3) W.u., as shown in Fig. 4 and Table I.

To provide a complementary insight into the effect of the aforementioned pairing modifications on the wave functions' structure of the lowest states in ²¹⁴Po, the same calculations are performed in the seniority (ν) scheme using the *j*-coupled NATHAN code [52]. The results obtained using H208 and H208-m are displayed in Fig. 5, where T(x, y) represents the number of neutron (x) or proton (y) pairs being broken [e.g., the 0^+_1 -state wave function is dominated by a 66% contribution from T(0,0), which corresponds to $\nu = 0$, and the 2^+_1 by 45% of T(1,0) corresponding to $\nu = 2$]. No important change between the two versions can be observed in the seniority structure of the $0^+_1, 2^+_1, 4^+_1$, and 6^+_1 states. The effect of reducing the $1h_{9/2}$ proton-pairing matrix elements is apparent on the structure of the two lowest 8^+ states. In H208, the main wave function component of the 8^+_1 state was characterized by seniority $\nu = 2$ (one neutron pair broken). The 8^+_2 state was initially predicted very close in energy to the 8^+_1 state, connected by a strong E2 transition to the 6_1^+ state and characterized by a mixing of $\nu = 2, 4$, and 6. Reducing the pairing in H208-m leads to an inversion of the $8^+_{1,2}$ states, and the $\pi(1h_{9/2}1f_{7/2})$ state comes below the $\nu g_{9/2}^4$ 8⁺ one, resulting in a good agreement between experiment and calculations. In this regard, it is important to notice that similar discrepancies between calculated and experimental B(E2) values have been found before in ¹³⁶Sn [53] and Pb isotopes [54]. All these cases share a common point that the inclusion of the effective three-body forces via the adjustment of the two-body matrix elements improves the agreement with the experimental data [55].

The increasing trend of the experimental $B[E2; J^{\pi} \rightarrow (J-2)^{\pi}]$ values for transitions from the J = 2, 4, 6, and 8 states, shown in Fig. 4, was predicted theoretically in neutron-rich Po isotopes up to A = 216, with the exception of a dip in ²¹⁴Po, for the $8^+_1 \rightarrow 6^+_1$ transition (see Figs. 6–9 from Ref. [4]). The trend was explained through the increase of the collectivity and quadrupole correlations with respect to the neutron number, leading to strong electromagnetic strengths.

The sudden decrease of the experimental $B(E2; 8_1^+ \rightarrow 6_1^+)$ for ²¹⁸Po is also reproduced theoretically, despite the adopted truncation in this nucleus. This can be attributed to the contribution from $i_{13/2}$ and $j_{15/2}$ proton and neutron orbitals in the structure of the 8_1^+ and 6_1^+ states. A strong transition probability of 27 W.u. is calculated when excluding the aforementioned orbitals. Their inclusion determines a reduction of quadrupole correlations due to pairing, leading to the $B(E2; 8_1^+ \rightarrow 6_1^+)$ decrease observed in ²¹⁸Po.

In conclusion, the B(E2) transition probabilities of yrast states up to the 8⁺ level in ^{214,216,218}Po isotopes populated in the $\hat{\beta}^-$ decay of ^{214,216,218}Bi are established for the first time through fast-timing measurements at the ISOLDE Decay Station. The comparison with shell-model calculations using the standard versions of the H208 and KHPE effective interactions shows reasonable agreement for the lower-lying transitions. A significant disagreement was observed in the case of the $8^+_1 \rightarrow 6^+_1$ transitions, especially for ²¹⁴Po where a much higher transition probability was measured (B(E2) = 11.3(3) W.u. instead of 0.54(4) W.u. [15]), disproving its previously claimed isomeric character. Similarly, based on the large B(E2) values, the 8^+_1 states in ^{216–218}Po should not be considered isomeric in the context of the seniority scheme as there is no characteristic hindrance observed in their decays. This latter observation is indicative of an increased fragmentation of the yrast states' wave functions. An improved agreement between theory and the present measurement was achieved after reducing the pairing strength of the interactions, effectively inverting the lowest predicted 8^+ states and confirming the two-proton configuration $\pi(1h_{9/2}1f_{7/2})$ of the yrast 8^+ state dominated by quadrupole correlations. The new findings can serve as a guideline for future analysis, providing a clear indication of the nature of 8^+_1 states in neutron-rich polonium isotopes. Additionally, they serve as an extremely valuable input for the different theoretical approaches and constitute a stringent test for the effective interactions.

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