

Emergence of an island of extreme nuclear isomerism at high excitation near ^{208}Pb



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ABSTRACT

Metastable states with $T_{1/2} = 8(2)$ ms in ^{205}Bi and $T_{1/2} = 0.22(2)$ ms in ^{204}Pb , with ≈ 8 MeV excitation energy and angular momentum $\geq 22 \hbar$, have been established. These represent, by up to two orders of magnitude, the longest-lived nuclear states above an excitation energy of 7 MeV, ever identified in the nuclear chart. Additionally, the half-life of the 10.17 MeV state in ^{206}Bi has been determined to be 0.027(2) ms, the next highest value in this highly excited regime. These observations indicate the emergence of an island of extreme nuclear isomerism arising from core-excited configurations at high excitation in the vicinity of the doubly closed-shell nucleus ^{208}Pb . These results are expected to provide discriminating tests of the effective interactions used in current large-scale shell-model calculations.

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Metastable states in atomic nuclei, also referred to as isomers, represent the manifestation of the associated wave functions being pure and quite distinct from those of other levels in their vicinity. Consequently, transition rates for decay of these states are orders of magnitude lower than those of nearby levels. The exploration of a variety of nuclear isomers, whose decay may be hindered by the required large change in angular momentum, or in its projection on the symmetry axis of a deformed nucleus, or in its shape, or a considerable difference in the configurations of initial and final levels, leads to crucial insights which further the understanding of the strongly-interacting, nuclear many-body system. Specifically, isomeric properties play a major role in refining effective interactions for shell-model calculations of near-spherical nuclei. Detailed descriptions are available in recent reviews [1–4]. In some instances, the degree of hindrance of the decay may be quite extreme, leading to isomeric half-lives which are larger by many orders of magnitude in comparison to those of other simi-

lar states. Some examples of such extreme nuclear isomerism are the: (a) “spin isomer” in ^{180}Ta ($Z=73$) with $T_{1/2} > 7.1 \times 10^{15}$ y [5], (b) “K isomer” in ^{178}Hf ($Z=72$) with $T_{1/2} = 31(1)$ y [6], (c) “shape isomer” in ^{242}Am ($Z=95$) with $T_{1/2} = 14(1)$ ms [7]. All of these isomers lie at relatively low (< 2.5 MeV) excitation energy. In the region around the doubly closed-shell nucleus ^{208}Pb , a notable isomer at relatively low excitation is the α -decaying 2.93-MeV state with $T_{1/2} = 45.1(6)$ s in ^{212}Po [8]. With an increase in excitation energy, a trend of decreasing half-lives is evident. In lighter nuclei, with fewer valence nucleons and lower level density, in some instances, longer-lived states may result, such as the 45.9(6)-s level at 6.958 MeV, with total angular momentum (henceforth, referred to as spin), $I = 12 \hbar$, in ^{52}Fe ($Z=26$) [9], and the β -decaying (21^+), 0.40(4)-s state at 6.67 MeV in ^{94}Ag [10]. The longest-lived isomers at very high excitation (> 7 MeV), known prior to this work, are the 8.533-MeV state in ^{212}Fr ($Z=87$) with $T_{1/2} = 23.6(21)$ μs [11], and the 8.095-MeV level in ^{213}Fr with $T_{1/2} = 3.1(2)$ μs [12]. In fact, the isomer in ^{212}Fr had been characterized as an “outstanding example” of a spin trap in near-spherical nuclei, and given its long half-life and high excitation energy, had been termed as an “extreme isomer” [2]. It had also been recognized then that “more extreme isomers might exist in heavy nuclei” [2]. It should be noted though that no specific predictions were made. The isomers

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reported in the present work have been discovered two decades later, despite the large body of recent work on isomers in the $A = 200$ – 215 region by several groups worldwide [13]. The presence of such long-lived states in the >7 – 8 MeV excitation range, which decay by γ -ray emission, is noteworthy.

Surveys of isomers across the nuclear chart [13,14] reveal that many long-lived states at high excitation are known to exist in heavy nuclei, which lie near the line of β -stability. As a result, these isomers are difficult to access experimentally, since compound nuclear fusion-evaporation reactions, which can populate levels at the highest excitation, favor the production of isotopes deficient in neutrons. Inelastic excitation and multi-nucleon transfer reactions, on the other hand, can be used to access nuclei near the line of stability or even on the neutron-rich side. However, the cross sections and highest spin attainable are limited in comparison with fusion-evaporation products. With the sensitivity provided by large γ -ray detector arrays, and pulsed beams from accelerators with a range of timing options, exploring isomers at high excitation near the line of β -stability becomes feasible. An impressive example is the recent study of the heaviest known doubly closed-shell nucleus ^{208}Pb ($Z=82$, $N=126$), wherein states up to spin $30 \hbar$ and excitation energy, $E_x = 16.4$ MeV, have been established [15]. The focus of the present work is the region near ^{208}Pb , where the availability of numerous orbitals with high values of intrinsic angular momentum, e.g., $h_{11/2}$ proton (π) and $i_{13/2}$ neutron (ν) holes, and $\pi h_{9/2}$, $\pi i_{13/2}$ and $\nu g_{9/2}$ particles, leads to conditions conducive for the realization of long-lived states at very high excitation. Recent work, including that of this collaboration [16–18], has revealed the presence of several states with half-lives up to hundreds of microseconds at intermediate excitation, but the $23.6(21)$ - μs isomer in ^{212}Fr at 8.533 MeV was thus far the longest-lived state above 7 MeV [11]. The present work describes newly identified metastable states in ^{205}Bi ($Z = 83$) and ^{204}Pb whose half-lives are about two orders of magnitude larger than other isomers in a similar excitation range. Additionally, the half-life of the 10.17 -MeV state in ^{206}Bi , which was previously reported to be $> 2 \mu\text{s}$ [19], has been measured and found to be slightly longer than that of the ^{212}Fr isomer. These newly-identified metastable states have a different character as compared to the so-called “spin isomers” in the Rn-Fr-Ra region, as will be described below.

The work described in this letter involves the population of highly-excited levels in isotopes of Pb, Bi and other elements through multi-nucleon transfer reactions with heavy, highly-energetic projectiles: (a) 1450 -MeV ^{209}Bi and (b) 1430 -MeV ^{207}Pb beams, incident on a 50 mg/cm^2 Au target. The Gammasphere detector array [20], which at the time consisted of 100 Compton-suppressed high-purity germanium detectors, was used to record coincident γ rays emitted within $\approx 1 \mu\text{s}$ of each other. Details regarding the experiment and data analysis are presented elsewhere [21]. In previous reports on ^{205}Bi and ^{204}Pb , levels up to 6.7 MeV and 8.1 MeV, respectively, had been identified utilizing α particles as projectiles incident on ^{205}Tl and ^{204}Hg targets [22,23]. These experiments were focused on the decays of short-lived levels, and a number of transitions up to $I \approx 20 \hbar$ were placed in the respective level schemes. The focus of the present work was on identifying metastable states and establishing their half-lives and decay paths.

Pulsed beams from the ATLAS accelerator at Argonne National Laboratory were used in different beam-sweeping intervals: successive bursts separated by ≈ 825 ns up to 8 s, enabled a search for and identification of isomers with half-lives in the microseconds, milliseconds and seconds time ranges. The data were collected in “beam-off” periods of $800 \mu\text{s}$, 3 ms, 3 s and 8 s, i.e., the pulsed beam was deflected away from the target for these durations. The coincidence window was $\approx 1 \mu\text{s}$. When beam pulses were separated by 825 ns, three- or higher-fold coincidence data were collected. For larger time periods, during the “beam-off”

periods of $800 \mu\text{s}$ and above, two- and higher-fold data were recorded. The data were sorted offline into histograms of two, three and four dimensions involving energy and time parameters, and subsequently analyzed using the RADWARE and TSCAN suite of programs [24,25]. Some examples of the histograms used for the data analysis are listed here: (a) two-, three- and four-dimensional symmetric, γ -energy histograms for establishing the excited level structures; (b) time-gated, triple- γ energy coincidence histograms to establish long half-lives; (c) energy-energy-time difference histograms for determining half-lives $T_{1/2} < 1 \mu\text{s}$; (d) prompt-delayed, two- and three-dimensional γ -energy histograms for identifying coincidence events across isomeric states with $T_{1/2} < 1 \mu\text{s}$; (e) angle-sorted, γ -energy, asymmetric matrices to determine transition multipolarities using the method of directional angular correlations from oriented states (DCO) [26]. The so-called “prompt” and “delayed” coincidence events corresponded to the detection of at least three γ rays within ± 40 ns and 50 – 650 ns of the trigger, respectively, when the beam pulses were separated by 825 ns.

A total of thirty new γ rays have been placed in the level schemes of ^{204}Pb and ^{205}Bi from the present work. However, only the transitions crucial for establishing the spin and parity quantum numbers of the isomeric levels are discussed. A paper describing the detailed level schemes for ^{204}Pb and ^{205}Bi deduced from this work is being prepared [27]. The newly established γ rays, along with previously reported ones, are displayed in Fig. 1. The γ -ray spectra illustrated in Figs. 1 and 2 represent three-fold delayed coincidence events. In Figs. 1(a) and 1(b), the summed coincidence spectra obtained with two simultaneous gates on all combinations of pairs of transitions in the previously known cascades in: (a) ^{204}Pb : 1006 – 325 – 618 – 167 – 1046 – 316 – 433 keV spanning $I = 9$ to $19 \hbar$, and (b) ^{205}Bi : 881 – 286 – 697 – 641 – 600 – 516 keV spanning $I = 9/2$ to $31/2 \hbar$, are presented. The quality of the data with fewer gating transitions, specifically the three decay branches from the isomer in ^{204}Pb , are displayed in Fig. 2.

The level scheme of ^{204}Pb has been extended up to a new long-lived isomer at $E_x = 8349$ keV, the half-life of which has been determined to be $0.22(2)$ ms [Fig. 3(a)], by inspecting the time distribution of the summed coincidence counts in the previously known 1006 – 325 – 618 – 167 – 1046 – 316 – 433 keV cascade spanning $I = 9$ – $19 \hbar$ [23], when the beam was incident on the target for $200 \mu\text{s}$, and data were collected during a 800 - μs beam-off period. In the case of ^{205}Bi , an inspection of the 800 - μs and 3 -ms beam-off data indicated that the half-life of the isomer was greater than these periods. Therefore, data from the next higher available pulsing period (3 -s beam-off) were scrutinized. The time distribution of the summed coincidence counts in the previously known 641 – 600 – 516 -keV cascade [Fig. 1(b)] spanning a spin range $25/2$ to $31/2 \hbar$ [12] indicate a half-life of $8(2)$ ms for the metastable state in ^{205}Bi [Fig. 3(b)]. To determine the half-life of the 10.17 -MeV level in ^{206}Bi , previously reported to be $> 2 \mu\text{s}$ [19], the time distribution of the summed coincidence counts of the most intense γ rays in the cascade between the 10.17 and 1.045 MeV levels was inspected in the 800 - μs beam-off data, leading to a half-life of $0.027(2)$ ms, as indicated in Fig. 3(c). A comparison of the above half-lives with those of other isomeric levels above an excitation energy of 7 MeV [14], and with $T_{1/2} > 1 \mu\text{s}$, across the nuclear chart is displayed in Fig. 4, where it is evident that the data points for ^{204}Pb and ^{205}Bi are outliers compared to those previously identified.

The excitation energy and spin-parity quantum numbers for the newly-identified isomer in ^{204}Pb were established as $E_x = 8349$ keV and $I^\pi = (22^+)$. The 481 -keV γ ray in ^{204}Pb (Fig. 1) is not observed in the so-called “prompt” data, which are recorded within a few tens of nanoseconds of the beam being incident on the target, but is clearly visible in the data collected during the “beam-off” periods. Therefore, it is attributed to the direct deexcitation of

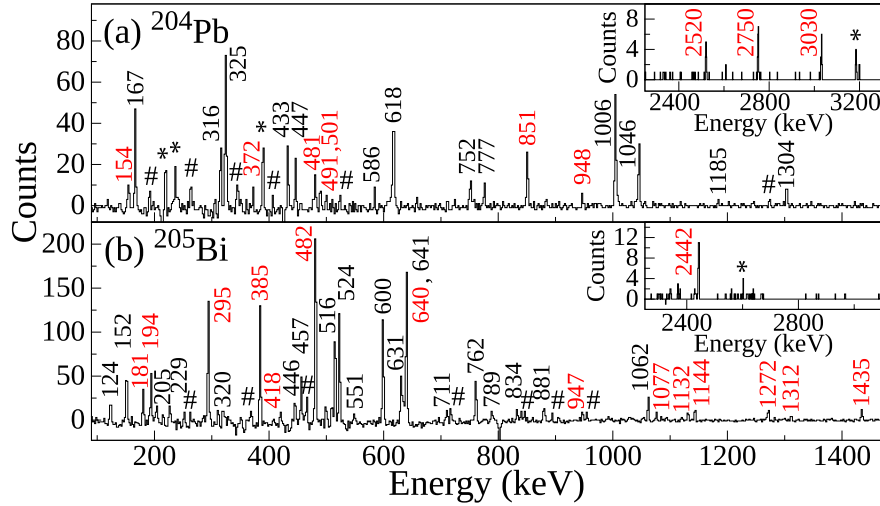


Fig. 1. Coincidence spectra illustrating γ rays observed in the deexcitation of the $T_{1/2} = 0.22(2)$ ms and $8(2)$ ms isomers in (a) ^{204}Pb and (b) ^{205}Bi , respectively. Transitions with energy > 2 MeV are displayed in the insets. Asterisks and hash marks designate unplaced and contaminant γ rays, respectively. The ones in black font were established earlier [22,23], while those in red have been identified from the present work.

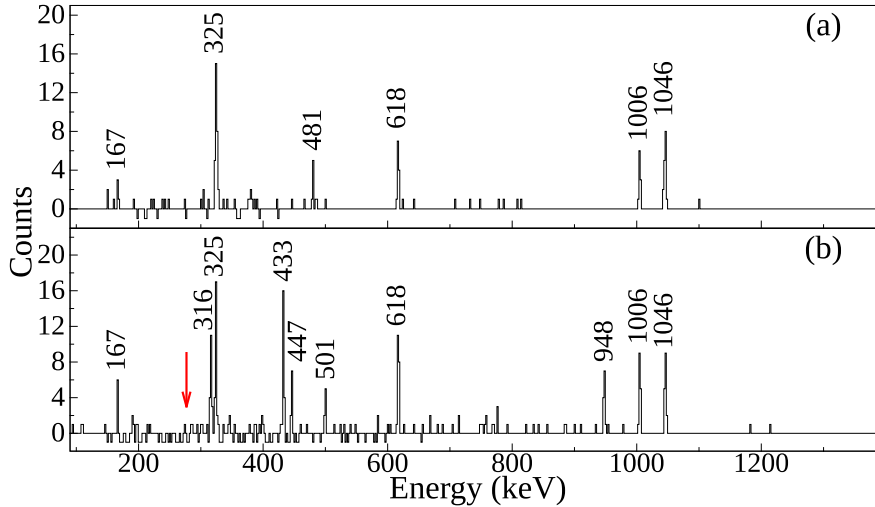


Fig. 2. Summed coincidence delayed spectra illustrating the decay of the isomer in ^{204}Pb : (a) the 2520-keV transition in coincidence with the previously established 1006-325-618-1046 keV cascade ([23]), (b) the 1304-keV γ ray in coincidence with the 1006-325-618-1046-316-433 cascade identified earlier. It is evident that, in the former instance, the 481-keV γ ray is the only newly identified one in addition to the gating transition at 2520 keV. In the latter instance, only the 501- and 948-keV transitions are newly established. The arrow indicates the absence, in the delayed data, of the 277-keV, $E_x = 8126$ keV to 7849 keV γ ray observed in the prompt data from the previous work. This γ ray is clearly visible in the present prompt data, but not in the delayed spectrum. The 481-, 501- and 948-keV peaks are evident only in the delayed spectra. The present analysis, with the observed coincidence relationships, and the energy sums for the above three parallel decay paths, unambiguously establish the excitation energy of the isomer in ^{204}Pb as 8349 keV.

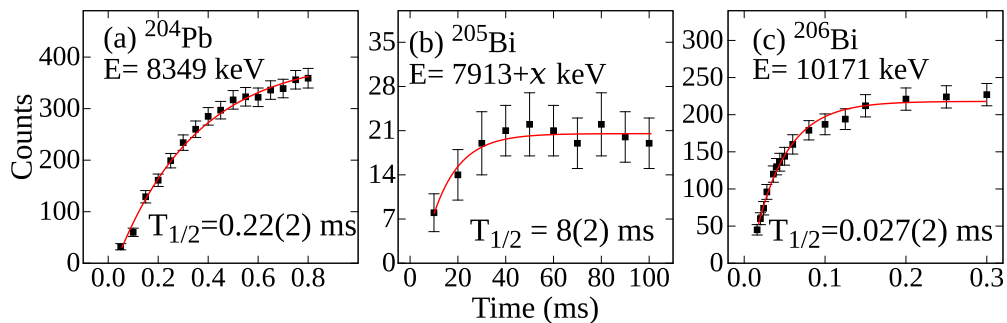


Fig. 3. Time distributions for the decay of isomers with excitation energy ≥ 8 MeV in: (a) ^{204}Pb , (b) ^{205}Bi , and (c) ^{206}Bi . The integral counts, along with the half-lives inferred from the fits, are displayed.

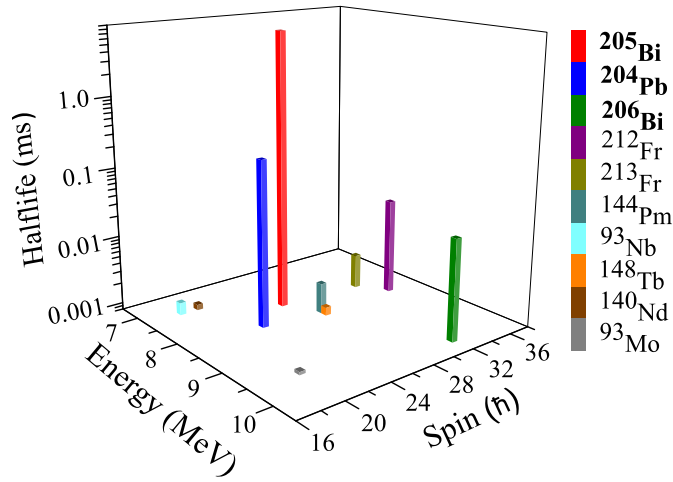


Fig. 4. Long-lived states ($T_{1/2} > 1 \mu\text{s}$) above an excitation energy of 7 MeV in nuclides across the nuclear chart. Note that the half-lives are displayed on a logarithmic scale. The large difference between the half-lives of the isomers in ^{205}Bi and ^{204}Pb established from this work, and those in other nuclei, is evident.

the 8349-keV isomer. The 481- and 2520-keV γ rays, which are newly identified in the present work, are found to be in cascade, with the latter directly feeding the previously established 16^+ , 5348-keV level with a proposed $\nu(i_{13/2}^{-2}, f_{5/2}^{-1}, p_{3/2}^{-1})$ configuration in ^{204}Pb [23]. The 2520-keV transition most likely has $E3$ character based on a calculation following the prescription in previous work [28–31], described below, leading to an $I^\pi = (19^-)$ assignment for the $E_x = 7868$ -keV initial level. The expected transition energy for the $E3$ excitation built on the four-nucleon-hole, 16^+ state can be estimated as follows. The unperturbed energy of the $E3$ excitation in ^{208}Pb would be 2615 keV. On account of the coupling to configurations involving multiple nucleons, energy shifts would result from the two $i_{13/2}$ neutrons and the two low- j neutrons. The final energy can be expressed as the sum of energy shifts corresponding to the individual constituents of such a configuration, which, in this case, turns out to be 2483 keV, in fair agreement with the experimentally observed 2520-keV value, thus validating its $E3$ assignment. To determine the multipolarity of the 481-keV transition feeding the level deexcited by the 2520-keV γ ray, intensity balance considerations have been used, for which a detailed procedure may be found in our earlier work [16–18]: either $E3$ or $M1$ character is inferred from the experimental value of 0.14(3) for the total conversion coefficient, due to its similarity with the corresponding theoretical values (0.111 and 0.119, respectively) from BRICC [32]. Based on typical transition rates expected for γ rays with different multiplicities, $M1$ character appears unlikely. An $E3$ character for the 481-keV transition would imply spin-parity quantum numbers (22^+) for the isomeric level. This would be consistent with similar isomeric transitions in $^{212,213}\text{Fr}$, ^{206}Bi [11,12,19], and many other nuclei in the vicinity of ^{208}Pb . It may be noted that there are no long-lived isomers in this region which decay via $M1$ transitions.

In ^{205}Bi , it was only possible to constrain the spin-parity of the isomer based on various experimental and theoretical considerations, but a firm assignment, comparable to the ^{204}Pb case, was not possible. All γ rays assigned to ^{205}Bi [Fig. 1(b)] are visible in the “prompt” data ruling out the possibility that any of these directly deexcite the isomer. The 8-ms isomer, therefore, most likely deexcites through one or more unobserved low-energy transitions with large conversion coefficients, accounting for their absence in the spectra. Similar considerations, as those outlined above for ^{204}Pb , have been employed in the case of ^{205}Bi , where the 2442–482–295 keV cascade feeds the previously established 4696-keV, $37/2^-$

level [22]. From the present work, the energy of this level is inferred to be about 1 keV lower i.e., 4695 keV. The 2442-keV γ ray feeds this level, and a similar calculation as in ^{204}Pb leads to the inference of its $E3$ character. Therefore, the level at 7136 keV deexcited by the 2442-keV γ ray is assigned $I^\pi = (43/2^+)$. Intensity balance considerations imply $M1$ character for the 295-keV transition. The 482-keV γ ray may have either $M1$ or $E3$ character, with the latter being excluded by the prompt decay of the associated level. The 2442–482–295 keV cascade is therefore assigned $E3$ – $M1$ – $M1$ multipolarity, with the topmost level at 7913 keV having possible $I^\pi = (45/2^+)$ or $(47/2^+)$. The 8-ms isomer in ^{205}Bi probably decays to this level through an unobserved transition. It may be noted that there is another level at 7971 keV in ^{205}Bi which has a prompt decay, and is fed by the decay of the isomer through an unobserved transition. It is not clear from the data whether the isomer in ^{205}Bi has two decay paths: one each to the levels at 7913 and 7971 keV, or only one decay to the 7971-keV state, which then deexcites to the 7913-keV level. All of the above considerations will be described in our detailed future paper [27]. Regarding the excitation energy of the 8-ms isomer in ^{205}Bi , a value 7913+x keV is listed, where $x < 150$ keV. This upper limit has been estimated taking into account the statistics in the data, inferred transition probabilities, theoretical conversion coefficients of the expected $E3$ or $M2$ transitions [32], and the efficiency of the detector array. A transition of 150 keV would have theoretical total conversion coefficients of 18.9 and 18.8 for $M2$ and $E3$ multiplicities, respectively. The available statistics in the data and the efficiency of the array at 150 keV would imply 2–3 counts for such a γ ray, with a background of about 1 count. The resultant peak may or may not be discernible in the spectra, therefore the upper limit of 150 keV. Of course, it is quite possible that the actual energy or energies of the transitions deexciting the isomer is significantly lower, which is why $x < 150$ keV represents an upper limit only. We have performed additional calculations towards a more sophisticated estimate which will be described in our future paper [27]. In view of the measured half-life and the corresponding inferred transition rates for different multiplicities, and based on the results of shell-model calculations performed (described below), a $(51/2^-)$ spin-parity assignment appears quite probable for the isomer implying a decay through low-energy (< 150 keV) $E3$ and/or $M2$ transitions.

Prior to this work, metastable states reported in this region beyond $E_x = 7$ MeV, with the highest half-lives, were in the Rn–Fr–Ra isotopes [1,2,11,12]. The ones identified in Tl–Pb–Bi isotopes prior to this work were primarily at lower excitation [28–31,33], except the states at very high excitation in ^{208}Pb [15]. The primary differences in the nature of the isomers in the above two regions are evident from an inspection of: (i) excitation energy (E_x) as a function of $I(I+1)$, where I is the spin, and (ii) reduced $E3$ transition probabilities $[B(E3)]$ for the decay of these isomers. As demonstrated in Fig. 5(a), the $I^\pi = (22^+)$ isomeric state in ^{204}Pb follows the trajectory of the lowest-energy levels at lower spin, as is also the case for others in neighboring nuclei. However, the $I^\pi = (34^+)$ isomeric level in ^{212}Fr [Fig. 5(b)] is found to lie distinctly lower in energy at the given spin in relation to the trajectory defined by the lower-lying states, attesting to its nature as a “spin-trap” isomer, similar to the situation in neighboring isotopes of Rn, Fr and Ra. Another striking difference is evident in the $B(E3)$ values for the decay of the isomers in these two regions [Figs. 5(c) and 5(d)]. In the Tl–Pb–Bi region, including the isomers newly identified from this work, the $B(E3)$ values are found to be in the vicinity of those expected from Weisskopf single-particle transition rates [Weisskopf units (W.u.)], or to be significantly lower. For instance, the $B(E3)$ value for the 481-keV transition in ^{204}Pb is determined to be 0.098(11) W.u. In the Rn–Fr–Ra region, an increase ranging from 20–40 units in comparison to the single-particle tran-

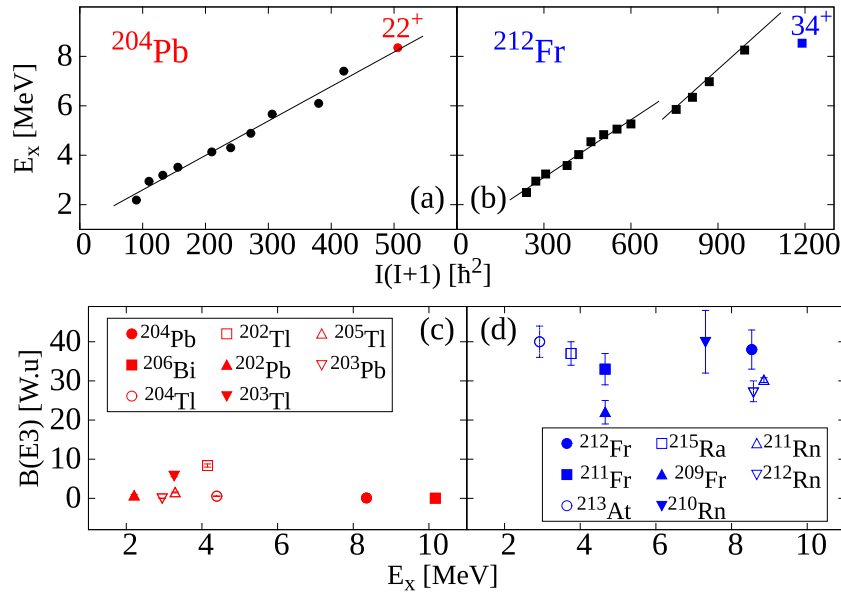


Fig. 5. (a), (b) Locations of isomers in ^{204}Pb and ^{212}Fr in the excitation energy (E_x) - angular momentum (I) plane. The dashed lines are intended to guide the eye. (c), (d) Reduced $E3$ transition probabilities $[B(E3)]$ for isomer decays in the Tl-Pb-Bi and Rn-Fr-Ra region, respectively. The evidently different nature of these two types of isomers is discussed in the text.

sition rates is visible in the $B(E3)$ values. This is attributed to transitions involving a change $\Delta j = \Delta l = 3$ between states [1]. The relatively lower half-lives in this region can be explained in terms of the resultant enhancement in the $E3$ transition probabilities. In the Tl-Pb-Bi region though, no such enhancement is visible, and for the isomers reported in the present work, the half-lives are considerably larger. It is relevant to mention here that $E3$ decays from the 23^+ and 28^- isomers at 11.4 MeV and 13.7 MeV in ^{208}Pb have been observed to be enhanced by factors of 32 and 56 W.u., respectively [15], similar to the $15/2^- \rightarrow 9/2^+$ transition in ^{209}Pb which has a strength of 26(7) W.u. This enhancement is similar to that observed in the Rn-Fr-Ra region and may be attributed to the very high excitation in ^{208}Pb which makes it possible to sample the $j_{15/2}$ neutron orbital, resulting in transitions of the $\nu j_{15/2} \rightarrow \nu g_{9/2}$ type. At low spin, in ^{209}Pb , the $15/2^-$ level is 1.42 MeV above the $9/2^+$ ground state, with these levels resulting from the occupation of the $j_{15/2}$ and $g_{9/2}$ orbitals, respectively, therefore the half-life of the $15/2^-$ level is only 1.36(30) ns [34]. It is possible that long-lived states at high excitation may also be realized in Po ($Z = 84$) and At ($Z = 85$) isotopes with $N < 126$, and neutron-rich ones with $Z < 82$, which would become accessible with rare-isotope beams.

It may be noted that the isomers in the Rn-Fr-Ra region were populated through fusion-evaporation reactions, as compared to multi-nucleon transfer reactions in the present work. In the latter case, quality spectroscopic data at high excitation are relatively more difficult to obtain, therefore long-lived isomers in this energy range in the Tl-Pb-Bi region around the line of stability remained undiscovered until now. It is noteworthy that the isomers in the Tl-Pb-Bi region involve either hole-hole or particle-hole excitations, and in the Rn-Fr-Ra case particle-particle configurations, comprising nucleons in high- j orbitals.

To aid in the understanding of the experimental data, shell-model calculations have been performed for ^{204}Pb and ^{205}Bi using the KHH7B effective interaction in the model space $Z = 58-114$ and $N = 100-164$ around doubly-magic ^{208}Pb using the OXBASH code [35]. The model space includes the proton orbitals $1d_{5/2}$, $0h_{11/2}$, $1d_{3/2}$ and $2s_{1/2}$ below $Z = 82$, and the $0h_{9/2}$, $1f_{7/2}$, and $0i_{13/2}$ ones above, and the neutron orbitals $0i_{13/2}$, $2p_{3/2}$, $1f_{5/2}$, and $2p_{1/2}$ below $N = 126$ and the $1g_{9/2}$, $0i_{11/2}$, and $0j_{15/2}$ ones above it. For the KHH7B effective interaction, the cross-shell two-

body matrix elements (TBMEs) are taken from the G-matrix potential (H7B) [36], while the proton-neutron hole-hole and particle-particle TBMEs are from the Kuo-Herling interaction [37], with modifications included later [38]. For ^{204}Pb , two sets of calculations were performed with $t = 0$ and $t = 1$, where $t = 0$ represents no excitation across the $Z = 82$ and $N = 126$ shell gaps. In the $t = 1$ case, the calculations involve the excitation of one nucleon across the $Z = 82$ and $N = 126$ shell gaps. Mixing between $t = 0$ and core-excited configurations is blocked in these calculations. For ^{205}Bi , the $1d_{5/2}$, $0h_{11/2}$, $1d_{3/2}$ and $2s_{1/2}$ proton orbitals are completely filled, with the unpaired proton occupying either the $0h_{9/2}$, $1f_{7/2}$ or $0i_{13/2}$ orbitals. The 22 valence neutrons have been allowed to occupy only the $0i_{13/2}$, $2p_{3/2}$, $1f_{5/2}$ and $2p_{1/2}$ orbitals below $N = 126$. Results of shell-model calculations using the KHH7B interaction for other nuclei in this region have been recently reported [16,39–41].

In ^{204}Pb , the shell-model calculations indicate the presence of multiple $t = 1$ states with spin-parity quantum numbers 19^- and 20^- , and excitation energy $\approx 7-8$ MeV. A 20^+ state with a $\nu i_{13/2}^{-4}$ configuration is also expected to lie in this vicinity. It is quite unlikely that any of these states are candidates for the isomer at 8349 keV since, in that case, the 481-keV deexciting transition (Fig. 1) would be of dipole character and would be inconsistent with the 0.22(2) ms half-life. The calculations indicate that states with spin $> 20 \hbar$ can arise in either of two ways viz., the $\pi(h_{11/2}^{-1}, h_{9/2}) \otimes \nu(i_{13/2}^{-2})$ and the $\pi(h_{11/2}^{-1}, h_{9/2}) \otimes \nu(f_{5/2}^{-1}, p_{1/2}^{-1}, i_{13/2}^{-2})$ configurations. While both these configurations can lead to the 22^+ level calculated to be at 8085 keV, the amplitude for the 4-quasiparticle state is found to be 14.2%, while for the one with six quasiparticles it is 56.1%. Though the excitation energies are reasonably reproduced in most cases, the shell-model calculations do not give a good account of the measured transition probabilities.

In ^{205}Bi , levels with spin-parity quantum numbers $43/2^-$, $45/2^+$, $45/2^-$, $47/2^+$, $47/2^-$ and $51/2^-$ are calculated to lie in the region between 6.5-7.7 MeV. It is quite unlikely that the isomer at an excitation energy of 7913+x keV has $I \leq 47/2 \hbar$ since, in that case, relatively fast $E2$ or $M2$ transitions of several hundred keV would deexcite to levels with spin up to $43/2 \hbar$, inconsistent with the measured 8-ms half-life. While the shell-model calculations indicate that multiple $51/2^-$ states are possible, only one of

these, with the $\pi i_{13/2} \otimes \nu(f_{5/2}^{-1}, i_{13/2}^{-3})$ configuration, is low enough in energy to be consistent with the experimental value. Four other $51/2^-$ states, all involving the $\nu g_{9/2}$ orbital, are possible but are calculated to lie above 9 MeV, and are therefore unlikely to be candidates for the isomeric configuration. Further, all other levels such as the $53/2^+$ state with the $\pi i_{13/2} \otimes \nu(i_{13/2}^{-4})$ configuration are also unlikely since they lie above 9 MeV. Therefore, based on the expectation from the shell-model calculations, the only candidate for the isomer would be the $51/2^-$ state with the $\pi i_{13/2} \otimes \nu(f_{5/2}^{-1}, i_{13/2}^{-3})$ configuration.

In the previous work on ^{206}Bi [19], configuration assignments for the isomers with $I^\pi = (28^-)$ and (31^+) , and half-lives of 155 ns and 0.027 ms, respectively, had not been proposed. The (28^-) isomer likely results from the $\pi i_{13/2} \otimes \nu[i_{13/2}^{-3}, (p_{1/2}^{-1}, g_{9/2})]_{43/2^-}$ configuration, with the (26^+) state to which it decays having a $\pi h_{9/2} \otimes \nu[i_{13/2}^{-3}, (p_{1/2}^{-1}, g_{9/2})]_{43/2^-}$ one. Thus, the isomerism would be associated with the $\pi i_{13/2} \rightarrow \pi h_{9/2}$ transition with a strength $B(M2) = 0.028(3)$ W.u., consistent with hindered $M2$ decays in this region. There are two possibilities for the configuration of the (31^+) isomer: either a neutron core excitation, $\pi i_{13/2} \otimes \nu[i_{13/2}^{-3}, (p_{1/2}^{-1}, j_{15/2})]_{49/2^+}$, or a proton core excitation, $\pi[i_{13/2}, (h_{11/2}^{-1}, h_{9/2})] \otimes \nu(i_{13/2}^{-3})$. The measured half-life leads to $B(E3) = 0.0276(21)$ W.u., consequently the latter scenario (proton core excitation) seems more likely, since in the former instance, the $\nu j_{15/2} \rightarrow \nu g_{9/2}$, $\Delta j = \Delta l = 3$, $E3$ transition would be expected to have a transition probability $B(E3) > 20$ W.u., far from the value deduced from the present data.

The new discoveries from the present work have direct implications for understanding the structure of nuclei in the vicinity of the heaviest, doubly-magic nucleus, ^{208}Pb . The identification of isomers at high excitation with complex core-excited configurations, where the excitation energy and spin-parity are established firmly from experiment, challenges large-scale shell-model calculations using available interactions. Additionally, with a precise knowledge of the half-lives of the isomers and the decay paths, tests of the predicted electromagnetic properties are also feasible. Further, long-lived isomeric states in general, and particularly in the present cases, tend to have relatively pure configurations, and are, therefore, more suitable to discriminate between different theoretical predictions. As stated earlier, the character of the isomers at high spin in the Tl-Pb-Bi region is different from that of the long-lived states previously established in the Rn-Fr-Ra one (which are yrast spin-traps, and have enhanced $E3$ transition rates). Therefore, different considerations have to be factored in their respective descriptions. At present, the commonly used effective interactions in this region are the: (a) KHHE ($Z = 58-82$, $N = 82-126$), (b) KHH7B ($Z = 58-114$, $N = 100-164$), (c) KHPE ($Z = 82-126$, $N = 126-184$), (d) V_{low-k} ($Z = 82-126$, $N = 126-184$), and (e) KHM3Y ($Z = 50-126$, $N = 82-184$). It is recognized that, while these interactions have satisfactory predictive power at low to intermediate excitation, there are major limitations in the description of high-spin phenomena, as for example, outlined in the previous work on ^{206}Bi [19]. The present results, viz., the excitation energy, spin-parity and electromagnetic properties of the three longest-lived metastable states at high excitation observed thus far in the periodic chart constitute an important contribution which should improve our understanding considerably.

To summarize, metastable states have been established in ^{205}Bi , ^{204}Pb and ^{206}Bi , with half-lives of 8(2) ms, 0.22(2) ms and 0.027(2) ms, respectively, constituting the three highest values of half-lives above an excitation energy of 7 MeV across the nuclear chart. This suggests that nuclear isomerism built on core-excited configurations is quite robust even under such extreme conditions. The emergence of a new frontier of metastable states in nuclei with $T_{1/2} \gg 1 \mu\text{s}$ at high excitation, with the hindrance attributable

to the difference in the configurations of isomeric states and the levels to which they decay, close to the line of β -stability in the region around ^{208}Pb , is indicated. These results constitute a fertile testing ground for large-scale shell-model calculations, along with available effective interactions. With focused experiments to identify more such instances, using multi-nucleon transfer reactions with highly-energetic, heavy-ion beams and suitably long pulsing periods, coupled with the sensitive detection of γ rays using large detector arrays, which are being planned by this collaboration, similar long-lived states are expected to be found in neighboring nuclei, thus redefining one extreme of isomerism.

Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

Data availability

Data will be made available on request.

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