



High-spin states in ^{212}Po above the α -decaying (18^+) isomer

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ABSTRACT

The nucleus ^{212}Po has been produced through the fragmentation of a ^{238}U primary beam at 1 GeV/nucleon at GSI, separated with the FRagment Separator, FRS, and studied via isomer γ -decay spectroscopy with the RISING setup. Two delayed previously unknown γ rays have been observed. One has been attributed to the E3 decay of a 21^- isomeric state feeding the α -emitting 45-s (18^+) high-spin isomer. The other γ -ray line has been assigned to the decay of a higher-lying 23^+ metastable state. These are the first observations of high-spin states above the ^{212}Po (18^+) isomer, by virtue of the selectivity obtained via ion-by-ion identification of ^{238}U fragmentation products. Comparison with shell-model calculations points to shortfalls in the nuclear interactions involving high- j proton and neutron orbitals, to which the region around $Z \sim 100$ is sensitive.

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Being the heaviest doubly-magic nucleus discovered so far, ^{208}Pb is a cornerstone of the Segrè chart. The structure of the surrounding nuclei showcases textbook “shell-model” states, coming from nucleon couplings outside the closed core [1,2]. In this

region, the ^{212}Po isotope occupies a unique place, offering the possibility to study the coupling of protons and neutrons forming its low-lying yrast structure [3–5] and the appearance of low-energy levels of unnatural parity decaying by E1 transitions. The nature of these levels, shell-model multiplets or α -cluster states, is still under debate [6–8]. In particular, the α -decaying 2922 keV level, with tentative spin-parity assignment (18^+) [5,9], is a state at the crossroads of the single-particle and α -cluster structure. This paper reports the first observation of excited states above the (18^+) isomer. This was possible by exploiting the potential of fragmentation of ^{238}U beams at relativistic energies in combination with ion-by-ion fragment identification.

A wealth of information on the nuclear interaction in this region has been achieved by measuring excited levels coming from the coupling of the valence nucleons or holes near ^{208}Pb [10–12]. Nuclear models for the region $Z \approx 100$ are sensitive to nucleon-nucleon interaction of the high- j proton $\pi i_{13/2}$ and neutron $\nu j_{15/2}$ shells which are filled in high-spin states in nuclei close to ^{208}Pb [13]. These states have an *a priori* pure wave function easy to calculate within the shell model. On the other hand, concerning a possible mixing of octupole E3 single-particle excitations with the ^{208}Pb collective 3^- state, the availability of high j -orbitals in the valence space, like $\nu j_{15/2}$ and $\pi i_{13/2}$, and the limited number of nucleons make ^{212}Po an interesting testing ground for the understanding of such mixing.

Several predictions were made for the existence of high-spin negative-parity states above the (18^+) isomer in ^{212}Po , on the basis of already identified states in the ^{213}At isotope and simple shell-model considerations [14]. The authors observed that in ^{212}Po a 21^- state should lie a few hundred keV above the 2922 keV long-lived, α -decaying, (18^+) isomer. It could mix with configurations involving the coupling to the ^{208}Pb 3^- state. This 21^- state, with only an E3 decay path open to the (18^+) isomer, should then be itself an isomer in the μs range, based on the systematics in the region. Experimental confirmation of such a prediction has been missing, mainly because of the difficulty of identifying isomers decaying by γ -ray emission to the long-living (18^+) state in typical fusion-evaporation reactions, like those performed with the Euroball array [6], or transfer reactions [3,5]. High-energy fragmentation reactions (~ 1 GeV/nucleon) can provide an alternative, with substantially lower production cross-section but very high selectivity in both mass and atomic number of the resulting fragments. In this work, we employed this technique to study ^{212}Po .

Neutron-rich isotopes beyond the $N = 126$ shell closure were produced by fragmentation of a 1 GeV/nucleon ^{238}U primary beam impinging on a 2.5 g/cm² Be target. The ^{238}U beam was delivered by the UNILAC-SIS18 accelerator with an intensity of about $1.5 \cdot 10^9$ ions/spill, with an extraction time of ~ 1 s followed by ~ 2 s of beam-off period. A 223 mg/cm² Nb stripper was mounted

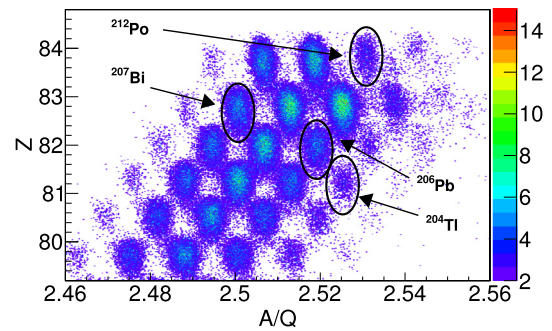


Fig. 1. Experimental isotope identification matrix; some isotopes of interest are encircled and labelled.

after the target to maximise the number of fully-stripped ions. Reaction products were then separated and selected according to their magnetic rigidity ($B\rho$) with the double-stage magnetic spectrometer FRS [15]. Ions of interest were unambiguously identified on an event by event basis with the experimental setup that is described in detail in Refs. [16–23]. This set-up has been shown to achieve sufficient discrimination in both Z and A/Q ratio, with $Q = Z$ for fully stripped nuclei, that contamination between neighbouring nuclei can be considered negligible. At such high Z , this fully stripped ion identification is a unique capability at GSI. The discrimination plot obtained with this analysis is shown in Fig. 1.

Fully identified ions were then slowed down in a thick Al-degrader followed by a plastic stopper. The implantation depth in the stopper was the same for all fragments with same Z and A/Z thanks to the monochromatic energy structure of the secondary beam. The implantation plane was surrounded by the RISING γ spectrometer, which consisted of 105 germanium crystals arranged in 15 clusters with 7 crystals each [24–26]: the full-energy γ -ray peak detection efficiency of the array was measured to be 15% at 662 keV, but the presence of the stopper lowered it to 13% at the same energy. The coupling of the γ spectrometer with the stopper enabled the performance of isomer spectroscopy with a time correlation window of up to 100 μs between ion implantation and γ -ray detection. The prompt flash produced during the implantation of heavy ions in the stopper affects isomer spectroscopy studies, requiring the search of a γ -ray event in a time window starting approximately 0.1 μs after the implantation. Especially at low energies (100–200 keV), due to the HPGe-detectors' time resolution and time-walk effect, this limits the sensitivity for isomers with lifetimes $\lesssim 100$ ns.

In Fig. 2 (a) the γ spectrum is shown, taken in delayed coincidence with about 3700 fully-stripped ^{212}Po ions obtained by selecting a time window of $\Delta t = 0.1 - 11.5$ μs after implantation. The spectrum is made by requiring a valid event in only one crystal

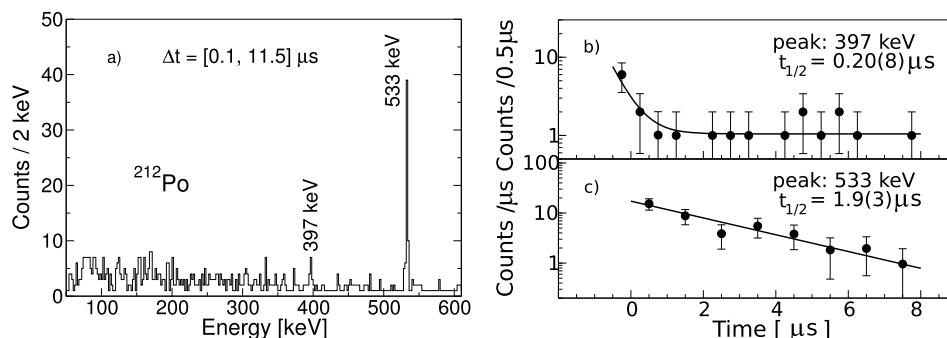


Fig. 2. (a) Delayed γ -ray spectrum of ^{212}Po , showing the two lines assigned to the depopulation of the states above the (18^+) isomer of this isotope. The spectrum has been produced selecting a time window of $\Delta t = [0.1, 11.5]$ μs after the implantation. Both lines attributed to ^{212}Po present an exponential time distribution, displayed in panel b) for the 397 keV line (where also a constant background term is added) and in panel c) for the 533 keV line, along with their estimated half-lives.

Table 1

Values of transition strengths (in W.u.) deduced from the lifetimes of the states decaying by the 533 and 397 keV lines.

$B(\sigma L)$ [W.u.]	533 keV	397 keV
$B(E1)$	$7(1) \cdot 10^{-10}$	$1.5(9) \cdot 10^{-8}$
$B(E2)$	$9(1) \cdot 10^{-5}$	$4(1) \cdot 10^{-3}$
$B(E3)$	18(3)	$1.2(5) \cdot 10^3$
$B(M1)$	$3.9(7) \cdot 10^{-8}$	$8(3) \cdot 10^{-7}$
$B(M2)$	$5(1) \cdot 10^{-3}$	0.16(6)

per cluster, to reduce the high-multiplicity background events [27]. Two γ -ray peaks can be seen: one at 397 keV, with an area of 14(6) counts, and one at 533 keV, with 57(12) counts. The statistical uncertainty of each transition energy is ± 1 keV. The time distribution of each identified transition (after background subtraction) has been χ^2 fitted with an exponential decay plus a constant background term, as shown in Fig. 2(b) and 2(c), yielding a half-life of $t_{1/2} = 0.20(8)$ μ s for the 397 keV line and of $t_{1/2} = 1.9(3)$ μ s for the 533 keV one. These lines have not been observed in ^{212}Po previously [28]. The spectrum has been carefully checked but no other peaks were found. Add-back treatment was also performed, without the requirement of in-cluster multiplicity equal to one, but yielded no substantial increase in statistics, but higher background events at low energies. Coincidence γ - γ spectra have been built, requiring two events in a time window of $\Delta t_{\text{coinc}} = 5$ μ s. No clear coincidence events between the two lines were registered, with one event amidst rather numerous seemingly random coincidences. With the given statistics, if the two lines belong to the same cascade, the probability that the number of observed coincidences is zero or one at most, is estimated to be $28_{-11}^{+20}\%$ and $62_{-18}^{+21}\%$, respectively, according to Poisson distribution. Such values are non negligible, and do not allow the rejection of the possibility of a sequential emission of the two γ rays. In such a case, the difference in lifetime unambiguously places the state depopulated by the 397 keV transition above the level decaying by the 533 keV one. The absence of any other coincidences between the 397 and the 533 keV transitions and other known lines of ^{212}Po strongly suggests the population of levels above the 2922 keV (18^+) α -decaying isomer. The newly observed levels therefore are placed at 3455 keV and 3852 keV, assuming unique decay chain, or 3455 and 3319 keV in case of two parallel decay paths.

From the measured half-lives the strength of the two transitions can be deduced: the results obtained assuming different multiplicities can be found in Table 1.

The deduced strengths favour an E3 assignment to the 533 keV transition, showing the typical enhancement due to the coupling to the ^{208}Pb 3^- core vibration. In the region, an almost identical strength value is found in two other known E3 transitions in the polonium isotopic chain, namely the $16^+ \rightarrow 13^-$ in ^{210}Po and the $(43/2^+) \rightarrow (37/2^-)$ in ^{211}Po . Both decays were studied with the present data set as well, yielding a strength value $B(E3) = 18(1)$ and $17(1)$ W.u. for ^{210}Po and ^{211}Po , respectively. These can be compared with the literature values of $B(E3) = 19(1)$ [29] and $17(5)$ [30] W.u.

Regarding the 397 keV line, strength values seem only compatible with an M2 assignment or suppressed E1 or E2. All other assignments would imply an uncharacteristic strength value for the region. The most probable multiplicities therefore limit the maximum spin value of the populated states to $16 < J < 23$.

Previous studies with fragmentation reactions connected the spin value of a γ -decaying isomer to its isomeric ratio (IR), via the so called ‘‘sharp cut-off model’’ [31]. In this work, following the same approach, the experimental IR is computed to assess the spin value of the two isomeric states observed. To do so, hypotheses

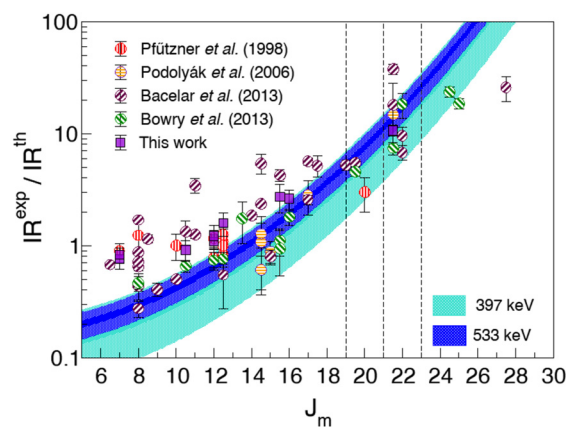


Fig. 3. Isomeric ratios (IR^{exp}) computed for different isomers and different isotopes with the data set of this experiment, normalized to the value predicted by the sharp cut-off model (IR^{th}), compared with results from Refs. [31,35–37]. The IR of the 397 (turquoise) and 533 keV (blue) lines of ^{212}Po are represented with shaded areas spanning along all spin values; vertical lines corresponding to spin $J = 19, 21$, and 23 are added to guide the eye.

Table 2

Isomeric ratio values (IR^{exp}) calculated from this dataset, compared to their predicted value according to the sharp cut-off model (IR^{th} , rounded to the nearest integer) [31].

Isotope	J^π	$\alpha^\dagger (\cdot 10^{-2})$	IR^{exp} (%)	IR^{th} (%)	$\text{IR}^{\text{exp}}/\text{IR}^{\text{th}}$
^{204}Tl	(12^-)	3.95(6)	33(5)	31	1.1(2)
	$(7)^+$	3.65(6)	50(10)	66	0.8(1)
$^{205}\text{Pb}^{\dagger\dagger}$	$25/2^-$	1.075(15)	<32	27	<1.2
^{206}Pb	12^+	0.774(11)	35(9)	29	0.8(3)
	7^-	8.92(13)	53(8)	64	0.8(1)
^{207}Bi	$21/2^+$	13.76(20)	34(7)	37	0.9(2)
^{210}Po	16^+	4.36(7)	22(4)	9	2.6(5)
^{211}Po	$(43/2^+)$	10.44 (15)	12(1)	1	11(1)
	$(31/2^-)$	4.61(7)	25(7)	9	2.7(8)

\dagger Taken from Brlcc [32].

$\dagger\dagger$ The intensity of the 27.7 keV γ -ray transition that depopulates this isomer has not been measured with accuracy, but has been estimated to be <87% by Poletti et al. [34], which is the value adopted.

and formulas of Pfützner et al. [31] have been adopted, drawing the internal conversion coefficients from Brlcc [32] (assuming null mixing ratio coefficient for both transitions), γ -ray efficiency from source measurements, time-of-flight, and kinematic factors from measurements and LISE++ [33] calculations. The IR for the 397 keV transition is $\text{IR}^{\text{exp}}=10(5)\%$ for E1, E2, E3 and M1 multipolarity, and $13(7)\%$ assuming M2 character. Regarding the 533 keV transition, its IR does not vary strongly changing multipolarity or assuming feeding from the 397 keV line or not. It ranges from $14(3)\%$ to $16(3)\%$. Note that since the 397 keV transition originates from a state with relatively short lifetime compared to that of the 533 keV transition, the determined IR for the 533 keV line already includes the feeding through the higher lying isomeric state.

IRs computed for other known long-living states in the region are compared to values taken from other fragmentation experiments [31,35–37] in Fig. 3; experimental values (IR^{exp}) are expressed in terms of their ratio to their sharp cut-off prediction (IR^{th}). Results obtained in this work (presented in Table 2) are fairly compatible with the values of Pfützner et al. [31] and Bacelar et al. [37]. Data also clearly show an almost exponential increase of the $\text{IR}^{\text{exp}}/\text{IR}^{\text{th}}$ ratio with the spin of the decaying isomeric state, which has been already observed by the same authors. A thorough discussion on this trend and the physics involved can be found in Ref. [35,38] and references therein. The shaded areas (representing

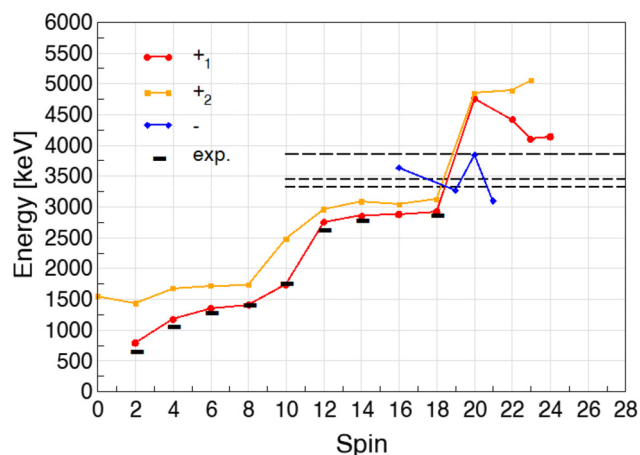


Fig. 4. Partial level scheme of ^{212}Po . The energy levels obtained by our shell-model calculations using the Kuo-Herling interaction (red circles, orange squares and blue diamonds) are compared with the experimental level energies (black bars) from previous work [6]. The three black dashed lines represent the possible placing of the two lines observed in this work: middle one for the 533 keV line, uppermost and lowermost for the 397 keV one, in the two hypotheses of sequential and parallel decay.

the IR of the 397 keV, in turquoise, and 533 keV lines, in blue) fall below, by a factor of $2 \div 5$, the majority of other experimental data for spin values lower than $J=14$, further reassuring that the newly observed states observed in this work lie indeed above the (18^+) α -decaying isomer.

The aforementioned experimental results can be compared to shell-model calculations. Dracoulis et al. [14] predicted a $21^- - 19^- - 18_2^+ - 23^+ - 24^+$ sequence above the (18^+) α -decaying state, noticing that the 21^- and the 23^+ states should be long-lived, with a lifetime of hundreds of microseconds and a few nanoseconds respectively. While the predicted energy of the two γ transitions depopulating the two isomeric states are different from those of the peaks observed in this work, their lifetime predictions, allowing energy rescaling, match nicely the measured half-lives of the two states, strongly suggesting a 21^- and 23^+ assignment.

This implies nonetheless that the two transitions form a cascade, which can be supported with 28% probability from experimental data, considering the coincidence event random. To consider other alternatives, shell-model calculations for this isotope were performed, employing the Kuo-Herling (KH) interaction [39] and using the shell-model codes ANTOINE and NATHAN [40–42].

The available single-particle orbits spanned from $Z=82$ to $Z=126$ in the proton space, encompassing the $h_{9/2}$, $f_{7/2}$, $f_{5/2}$, $p_{3/2}$, $p_{1/2}$ and $i_{13/2}$ shells, and from $N=126$ to $N=184$ for the neutron configuration, and thus the $g_{9/2}$, $d_{5/2}$, $g_{7/2}$, $s_{1/2}$, $d_{3/2}$, $i_{11/2}$ and $j_{15/2}$ shells, without truncation. The calculations reproduce all level energies very well up to the (18^+) α -decaying isomeric state, with a mean square deviation of 55 keV; they also reproduce the $16^+/18^+$ inversion [5,14], predicting the two states almost degenerate and close to the experimental energy of the α -decaying isomer. Above 3 MeV excitation energy, our calculations predict the $21^-/19^-/23^+/24^+$ sequence in line with the suggestions discussed earlier [14] (Fig. 4). Shell-model calculations were also performed by Warburton [43], with the KH interaction - results are compatible with ours.

The 18^+ spin-parity is built from a $\nu[g_{9/2}i_{11/2}]_{10^+}$ neutron configuration and two major proton configurations. The dominant component of the wave function (60%) has both protons in the $h_{9/2}$ shell, while the other component (38%) sees one proton in the $h_{9/2}$ and the other proton in the $f_{7/2}$ single-particle orbitals. Regarding the 21^- state, it is built from a rather pure (82%) wave function coupling the $\pi[h_{9/2}i_{13/2}]_{11^-}$ proton configuration to the

$\nu[g_{9/2}i_{11/2}]_{10^+}$ neutron configuration. This level is calculated at 3098 keV, 180 keV above the 2922 keV isomer and fairly close to the calculation of Dracoulis et al. [14]. Above the 21^- level, the 19^- (3263 keV), 23^+ (4104 keV) and 24^+ (4135 keV) states are predicted, with similarly pure configurations, coupling the leading $\pi[h_{9/2}i_{13/2}]_{11^-}$ partition with the $\nu[g_{9/2}^2]_{8^+}$, $\nu[g_{9/2}j_{15/2}]_{12^-}$ and $\nu[i_{11/2}j_{15/2}]_{13^-}$ neutron configurations, respectively. It has to be noticed that configurations containing the $\pi i_{13/2}$ and $\nu j_{15/2}$ orbitals may couple to the ^{208}Pb 3^- state: following notation of Ref. [44], such states should be written with a tilde, e.g., $\pi \tilde{i}_{13/2}$, denoting with this an admixture of the pure $\pi i_{13/2}$ orbital with the $\pi f_{7/2} \otimes 3^-$ and $\pi h_{9/2} \otimes 3^-$ coupled configurations. E2 and E3 transitions have also been calculated in order to determine the most likely candidates for the two observed states. Standard effective charge values were used for the E2 decays, while for the E3 transitions a proton effective charge value of $2.5e$ was used [45,46], while the neutron effective charge value was kept fixed at $0.5e$. No E1 or M2 transitions were calculated, since no value in this region is established in literature.

None of the calculated E2 strengths is consistent with the experimental numbers listed in Table 1. The $21^- \rightarrow 18^+$ E3 decay has a strength of 13 W.u. which is compatible with the value deduced for the 533 keV transition and in line with the typical strength of mixed $\pi i_{13/2} \rightarrow h_{9/2}$ decays in this region [45]. A change in the neutron effective charge produced no significant variation in the computed strength value of this decay.

Regarding the 397-keV line, the lack of a clear coincidence between the two lines does not allow for a firm assignment. However the assignment of this transition to the 16^+ , 19^- , $20^- \rightarrow 18^+$ transitions would be either in contrast with observed lifetime or energy. In fact, those states are either predicted too high in energy (more than 0.5 MeV than the observed), or would open much faster decay paths from the 21^- state (analogous to observed transitions in neighbouring isotopes) that would make the level non-isomeric, even if they could not be observed during this experiment. Similar arguments can be used to rule out a 19^- , 20^- or $22^+ \rightarrow 21^-$ decay assignment. Possibilities that cannot be ruled out completely are also $17^+ \rightarrow 18^+$ and $20^- \rightarrow 21^-$ assignment, which have no observed analogues in the neighbouring isotopes. The most straightforward assignment is the M2 $23^+ \rightarrow 21^-$ decay. This assignment is consistent with the deduced strength (according to both systematics, Ref. [47] and references therein, and shell-model calculations performed by Dracoulis et al. [14]), does not require the presence of un-observed γ rays. Other high-spin states are predicted too distant in energy, or require transitions of nature which have no observed match in any of the neighbouring isotopes. Furthermore, this assignment would match nicely with the prediction of a $47/2^-$ state in ^{211}Po about 400 keV higher than the $43/2^+$ [30]. The observation of such level would strengthen our arguments. Moreover, it has been already noticed [46] that shell-model calculations employing the Kuo-Herling interaction, while extraordinarily successful at low spins, are somewhat less reliable when reproducing the high-spin spectrum of isotopes in this region, with a deviation of ~ 200 -300 keV.

We have tried to reproduce the energy of the observed states by altering the single-particle energy (SPE) values of the $i_{13/2}$ and $j_{15/2}$. We managed to reproduce the energy of the 21^- state by increasing by 0.5 MeV the $i_{13/2}$ SPE. This modification is numerically similar to the ^{208}Pb 3^- state induced depression of the estimation of the $i_{13/2}$ SPE (410 keV) calculated by Hamamoto [48]. We were not able to reproduce the 23^+ level energy, however, as it seems to require a decrease of the estimation of the $j_{15/2}$ SPE, probably due to the fact that its neutron configuration $\nu[g_{9/2}j_{15/2}]_{12^-}$ does not couple to the octupole vibration of the core due to Pauli blocking. The 21^- level energy seems, in our calculations, to be insensitive to the placing of the $j_{15/2}$ orbital. These results suggest that the

Kuo-Herling interaction, despite its excellent predictive power at low spins across a large region of the nuclear chart, presents shortfalls in the description of high-spin structures. A more thorough discussion on the subject is beyond the scope of this paper, but this preliminary investigation calls for extensive large shell-model calculations to understand how this configuration-dependent 3^- coupling can be reproduced in a shell-model framework.

In summary, decay γ -ray spectroscopy of relativistic ^{238}U fragmentation reaction products allowed to study the high-spin structure of ^{212}Po , searching for predicted μs isomers above the α -decaying long-lived (18^+) isomer. Two γ -ray transitions have been observed for the first time and attributed as follows: the 533 keV ($21^- \rightarrow 18^+$) E3 transition and the 397 keV ($23^+ \rightarrow 21^-$) M2 transition. The half-lives of the two decaying states have been measured and the transition strengths deduced. From the experimental point of view, now that key γ -ray transitions above the isomer are established, it should be possible to exploit data from other reactions, with higher cross sections, and use these energies and half-lives to extend further the ^{212}Po level structure.

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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