Early onset of deformation in the neutron-deficient polonium isotopes

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Abstract. In-source laser spectroscopy has been performed at CERN-ISOLDE with the RILIS laser ion source on $^{191-204,206,208-211,216,218}$Po. New information on the β decay of ^{199}Po were extracted in the process, challenging previous results. Large-scale atomic calculations were performed to extract the changes in the mean-square charge radius $\delta\langle r^2 \rangle$ from the isotope shifts. The $\delta\langle r^2 \rangle$ for the even-A isotopes reveal a large deviation from the spherical droplet model for $N < 116$.

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1. Introduction

In the vicinity of $Z = 82$, the interplay between microscopic and macroscopic effects gives rise to shape coexistence at low energy [1, 2, 3, 4], with the most extreme case of ^{186}Pb where the first three nuclear states are 0^+ states with different deformations [5]. The extent of the mixing between those shapes can be determined by investigating the evolution of the shape of the nucleus in its ground state through the study of the changes in the mean-square charge radius $\delta\langle r^2\rangle$ along isotopic chains.

The knowledge on the $\delta\langle r^2\rangle$ of the even-Z isotopes with $Z \leq 82$ and $N \sim 104$ is extensive [6, 7, 8, 9, 10, 11, 12], but the data are lacking for the more exotic nuclei beyond the $Z = 82$ shell closure. In a recent series of experiments, the $\delta\langle r^2\rangle$ of the neutron-deficient isotopes $_{84}$Po have thus been investigated at the CERN-ISOLDE facility using the resonant ionisation laser ion source (RILIS). In this contribution, we report on the findings of these experiments with a particular emphasis on new decay spectroscopy data that were extracted for ^{199}Po .

2. Experimental technique

The polonium isotopes were produced at the CERN-ISOLDE facility [13] via the proton-induced spallation (1.4 GeV, 1.4 μA on average) of ^{238}U (50 g·cm^{-2}). The reaction products diffused out of the target held at high temperature (~ 2000°C) to an ion source cavity where they were resonantly ionised with the RILIS [14, 15] using a three-step atomic excitation [16]. Due to the high temperature of the ion source, isobaric thallium and francium isotopes were also surface ionised.

The polonium beam was accelerated to an energy of 50 keV and mass separated in a dipole magnet. Beams of $^{191-198,211,216,218}$Po were implanted in one of ten thin carbon foils (20 μg·cm^{-2}) in the Windmill setup [17]. The implantation position was surrounded by one or two silicon detectors to measure the energy of the α particles emitted in the decay of the polonium isotopes and of its daughters. For ^{195}Po, two high-purity germanium detectors were added to additionally measure the energy of γ rays.

Beams of $^{199-204}$Po were implanted in the aluminized mylar tape of the ISOLDE tape station. The β particles emitted in the decay were monitored with a plastic scintillator. The energy of the subsequent γ rays was measured with a high-purity germanium detector. Beams of $^{206,208-210}$Po were monitored using a Faraday cup.

3. Extraction of the atomic and nuclear spectra

The atomic resonances were acquired by scanning the frequency of the transition at 843.38 nm [16] while monitoring the ionisation rate of the isotopes of interest. For the longer-lived isotopes $^{206,208-210}$Po with the highest beam intensity, the ion beam current was measured. For the shorter-lived isotopes, the particles emitted by the nuclear decay were counted. For the even-A isotopes and for ^{211}Po, a single isomer was observed, and counting the α ($^{192-198,211,216,218}$Po) or β ($^{200-204}$Po) particles was sufficient. In the case of the remaining odd-A isotopes, it was necessary to rely on the characteristic energy of the α particles ($^{193-197}$Po) or γ rays ($^{199-203}$Po) emitted in the decay of each isomer. In the analysis of those decay spectra, new information were uncovered, as already reported for ^{195}Po [18]. In this contribution, we report on the γ-ray spectra observed in the β decay of ^{199}Po to ^{199}Bi.

The level structure of ^{199}Bi was studied at CERN-ISOLDE in the β decay of ^{199}Po following the α decay of ^{203}Rn and in the α decay of ^{203}At [19], at the joint institute for nuclear research (JINR), Dubna (Russia), following the fusion-evaporation reactions of a ^{10}Be beam on a ^{197}Au target [20], and at the university isotope separator at Oak Ridge (UNISOR) facility, Oak Ridge (TN, USA), in the fusion-evaporation of ^{14}N on natural Ir [21]. In the latter work, the discrepancies between the three approaches have been thoroughly highlighted.
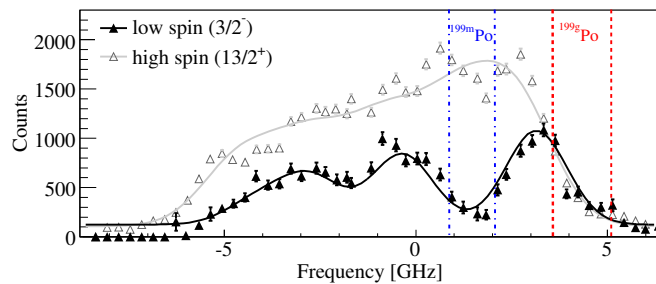
Figure 1. Hyperfine spectra of the low- (full black triangles) and high- (open gray triangles) spin isomers of ^{199}Po following the 246 keV and 1002 keV γ-ray transitions, respectively. The red dashed and blue dash-dotted lines delimit the regions of the scans that have been selected to generate the purified spectra shown in figure 2.
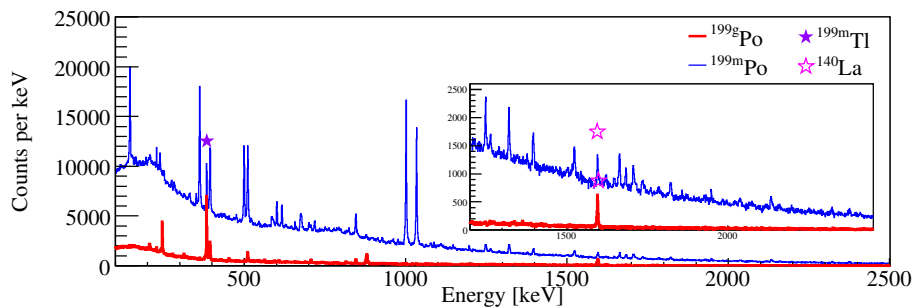

Figure 2. Purified γ-ray spectra made from a linear combination of the raw spectra generated from the regions highlighted in figure 1. The thick red line shows a pure ^{199g}Po spectrum while the thin blue line shows a pure ^{199m}Po spectrum. Contamination from ^{140}La (implanted the week prior to the experiment) and ^{199m}Tl are marked with stars.

The UNISOR study [21] is the first to produce a mass-separated beam of ^{199}Po to study the β decay independently from other polonium isotopes or from other decay modes (α decay of ^{203}At in ISOLDE [19]). The reaction used at UNISOR populates more strongly the high-spin isomer ^{199m}Po while the α decay of ^{203}Rn in ISOLDE populates in contrary more the low-spin ground state ^{199g}Po. Based on those observations, the parent of the different lines was proposed.

The hyperfine spectra for ^{199g}Po and ^{199m}Po were extracted in our experiment by following the intensity of the 246 keV and 1002 keV γ-ray transitions, respectively (see figure 1). By selecting a specific frequency range, it is possible to enhance the production of one of the two isomers over the other, as demonstrated with ^{70}Cu [22, 23] and ^{185}Pb [24]. The selected ranges for ^{199}Po are shown in figure 1. While it is possible to produce a clean beam of ^{199m}Po, it is more difficult for ^{199g}Po as its hyperfine structure overlaps completely with that of ^{199m}Po. It is however possible to produce a pure spectrum of either ^{199g}Po or ^{199m}Po by a linear combination of the spectra produced in the two ranges. Those purified γ-ray energy spectra are shown in figure 2. Many transitions appear to be of pure origin, like that at 880 keV (^{199g}Po) or that at 1034 keV (^{199m}Po), as suggested in [21]. There are however discrepancies, as seen, for example, for the 846 keV transition, present in both γ-ray energy spectra. The lists of observed γ rays for ^{199g,m}Po and their relative intensities are given in tables 1 and 2.

Table 1. List of γ-ray transitions in ^{199g}Po and their intensities in this work and the previous studies [19, 20, 21].

This work		UNISOR [21]		JINR [20]		ISOLDE [19]	
E_γ [keV]	I_γ	E_γ [keV]	I_γ	E_γ [keV]	I_γ		
206.83(9)	17.9(14)	206.7	18			206.6	18.9
245.88(2)	100	246.0	100	246.0	100	245.9	100
278.76(17)	6.7(6)						
313.07(11)	13.4(11)						
393.95(3)	94.2(72)	394.2					
452.69(17)	17.6(15)	452.5	14.2				
526.89(19)	10.9(10)	527.0	9.6				
563.47(23)	18.2(71)						
607.42(13)	19.5(16)						
707.10(8)	28.9(23)						
815.52(20)	24.1(20)	815.3	9.6				
845.82(7)	55.6(43)	845.7	82.8			845.8	63
879.49(5)	184(14)	880.2				880.4	
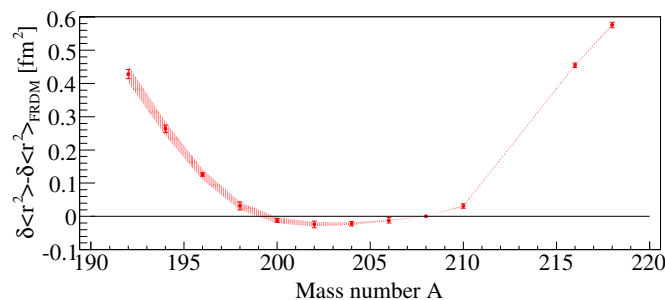
Figure 3. Difference between the even-A polonium $\delta\langle r^2 \rangle$ and the predicted value by the spherical finite range droplet model [25, 26]. The error bars represent the statistical uncertainties while the shaded area represents the systematic uncertainty arising from the atomic calculations.

4. Analysis of the atomic spectra

The atomic spectra observed in this work reveal the hyperfine structure, as seen in figure 1, and the isotope shift between any two isotopes. Those experimental spectra can then be related to the nuclear observables of interest, namely the magnetic dipole and electric quadrupole moments for the hyperfine structure, and the $\delta\langle r^2 \rangle$ for the isotope shift [27]. For the latter, the relation involves two atomic parameters, denoted M and F in reference [27]. These two parameters cannot be evaluated experimentally as the radioactive nature of the polonium isotopes hinders experimental atomic studies. Instead, we performed large-scale atomic calculations, as described in references [28, 29]. The accuracy of those calculations could be tested against an experimental modified King plot, which relates the M and F parameters of two different atomic transitions. In this work, we compared our measurements with the 843.38 nm transition to previous measurements with the 255.8 nm transition [30]. The discrepancy between the calculations and the experimental data is used to determine the systematic uncertainty resulting from the

Table 2. List of γ-ray transitions in ^{199m}Po and their intensities in this work and the previous studies [19, 20, 21].

This work		UNISOR [21]		JINR [20]		ISOLDE [19]	
E_γ [keV]	I_γ	E_γ [keV]	I_γ	E_γ [keV]	I_γ		
145.74(2)	9.9(5)	145.6	19.4			145.8	16.3
227.91(15)	1.2(1)			229.1	10.2		
239.12(7)	2.1(1)	239.3	7.5				
351.65(10)	1.6(1)						
361.88(1)	24.5(13)	361.9	36.6	361.6	47	361.6	27
383.50(3)	12.4(6)						
394.13(2)	14.7(8)	394.2					
499.47(2)	21.7(11)	499.7	21.9	499.8	42.3		
601.32(4)	7.4(4)	601.2	10.9				
616.90(4)	7.7(4)	616.4	1.3				
675, 73(7)	11.7(6)						
717.94(8)	4.4(2)	717.8	6.1				
824.56(17)	2.8(2)	825.2	3.6				
845.69(5)	11.3(6)	845.7	19.8			845.8	35
1001.74(1)	100	1001.7	100	1002.0	100	1002.0	100
1034.00(1)	102.7(53)	1033.8	83	1034.4	100	1034.0	117
1077.86(26)	3.2(2)						
1147.33(16)	3.5(2)						
1197.19(13)	2.7(2)	1197.5	4.2				
1248.43(7)	7.8(4)	1248.4	8.7				
1263.64(25)	2.6(2)	1262.8	3.2				
1321.00(8)	9.8(5)	1320.1	10.0				
1396.22(11)	7.0(4)	1395.9	7.3				
1523.66(12)	6.8(4)	1523.6	6.8				
1621.9(4)	2.3(1)						
1647.38(33)	1.4(1)						
1663.57(9)	6.6(3)	1663.4	7.5				
1683.47(15)	4.4(2)	1683.2	6.3				
1707.19(16)	7.8(4)	1706.2	6.5				
1735.3(4)	3.4(2)						
1822.07(21)	5.3(3)	1822.1	4.0				
1857.19(19)	0.8(1)						
1948.53(15)	1.8(1)	1949.4	3.3				
2037.6(5)	2.4(1)	2036.7	1.7				
2133.92(21)	3.9(2)	2133.1	3.4				
2322.42(34)	1.6(1)						
2376.01(55)	2.1(1)						

use of the calculated atomic factors. Finally, the $\delta\langle r^2\rangle$ of the even-A polonium isotopes are extracted and shown in figure 3.

The $\delta\langle r^2\rangle$ depart dramatically from the predictions of the spherical finite range droplet model (FRDM) [25, 26] for $N \leq 114$. The interpretation of this departure is discussed in reference [29] in terms of beyond mean field calculations [31, 32] as resulting from the very soft nature of those isotopes, rather than from static deformation. The different behaviour with respect to the mercury isotopes, where only odd-A isotopes depart from the spherical trend [7], suggests an involvement of the high-j orbitals occupied by the protons. Further studies are planned at current and future facilities to investigate this claim, such as at the collinear resonant ionisation spectroscopy (CRIS) setup at CERN-ISOLDE [33, 34].

Acknowledgments

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