# EUROPEAN ORGANIZATION FOR NUCLEAR RESEARCH Addendum to IS456 Study of polonium isotopes ground state properties by simultaneous atomic- and nuclear-spectroscopy

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April 21, 2008

#### Abstract

The study of the changes in the mean-square charge radius along the neutrondeficient polonium (Z = 84) isotopic chain with the RILIS has been very successful. Data on <sup>193-200,202,204</sup>Po has been collected. Large deviation from the spherical droplet model have been observed for the most neutron-deficient isotopes. We propose to complete the systematics on the odd-A isotopes with the measurement of the hyperfine structure and isotope shifts of <sup>201,203</sup>Po, to reduce our systematic uncertainty having a greater overlap with previous data set by measuring the isotope shifts of <sup>208,209,210</sup>Po and finally to establish whether the shell effects beyond N = 126 are similar to those observed in lead and bismuth by measuring the isotope shifts of <sup>211,211m,212m,216,217,218</sup>Po. We also outline the required developments for a faster release of <sup>192</sup>Po, necessary to complete the original proposal. Altogether we require **24 shifts**.

# Introduction

We propose to study the changes in the mean-square charge radii and the moments of the ground states along the polonium (Z = 84) isotopic chain to directly observe the effect of different shapes, coexisiting at low-excitation energy in the neutron-deficient polonium isotopes.

Thanks to large optical shifts around Z = 82 (of the order of 1 GHz per two mass units) and hyperfine coefficients (of the same order of magnitude), it is possible to use the RILIS to study those ground- (and isomer-) state properties.

In this addendum to experiment IS456, we briefly recall the physics motivation for the study of the change in the mean-square charge radius of polonium. We then report on the measurement of August 2007 and the status of the analysis. We finally present the case of the neutron-rich  $^{211,211m,212m,216,217,218}$ Po, that of the intermediate  $^{201,203,208,209,210}$ Po and outline the required developments to complete the study of the neutron-deficient  $^{192}$ Po.

#### **Physics** interest



Figure 1: Changes in the mean-square charge radius as a function of the neutron number N for Pt [1], Hg [2], Pb [3, 4, 5, 6, 7] and Po [8] before the start of experiment IS456. The points represent the experimental data while the lines are the spherical droplet model predictions. Changes are relative to a reference isotope circled for each element.

Shape coexistence at low excitation energy in nuclei is a phenomenon for which interest has been continuously growing on both the experimental and theoretical fronts [9, 10, 11, 12, 13]. The region around the neutron mid-shell N = 104 and closed proton shell Z = 82is especially prolific. The platinum isotopes (Z = 78) ground states show a transition from the weakly deformed oblate shape at A < 176 and A > 188 to a strongly prolate configuration for  $176 \leq A \leq 188$  [1, 14, 15]. This whole region is thus rich in shape coexistance. The effects on the changes in the mean-square charge radii can be seen on Fig. 1. Similarly the mercury isotopes (Z = 80) around N = 104 show shape coexistence and shape transition and, for the lightest isotopes ( $N \leq 106$ ), a large odd-even staggering in the charge radii as well as a similarly large difference between the charge radii of the ground state and the isomers [2].

More recently, the mean-square charge radii of the neutron-deficient <sup>182-190</sup>Pb isotopes have been studied at ISOLDE by the same collaboration in experiment IS407 using the same technique proposed here. This study across the mid-shell N = 104 along the Z = 82 closed shell revealed that charge distribution of the lead nuclei have only little deviation from the spherical droplet model prediction. This corresponds to a weak mixing only, despite the proximity of a 0<sup>+</sup> excited state for the lightest nuclei. A comparison with Beyond Mean Field calculations [16] and IBM-type predictions [17] shows that this deviation can be explained by the sensitivity of the mean-square charge radius to small changes in the pairing gap and/or the weak mixing in its wavefunction [6]. The success of this experiment was two-fold. First, the technique proved to be working down to the very short-lived <sup>182</sup>Pb isotope (T<sub>1/2</sub>=55 ms), with a count rate as low as a few ions per second. Secondly, the results obtained challenged the models beyond what was originally expected and triggered discussion toward a better understanding of the Beyond Mean-Field model and the Interacting Boson Model. [6]

The polonium isotopes (Z = 84) are important for understanding the transition across the proton shell closure of Z = 82. Although extensive studies have been performed on the neutron-deficient isotopes [18, 19, 20, 21, 22, 23, 24, 25], the measurement of the charge radii was limited to the longest lived isotopes <sup>200,202,204–210</sup>Po [8]. The status of the charge radii around Z = 82 before the start of IS456 is summarised in Fig. 1.

The recent  $\alpha$ -,  $\beta$ - and  $\gamma$ -spectroscopy investigations on neutron-deficient polonium isotopes down to A = 190 have indeed highlighted ground state shape coexistence. Several  $\alpha$ - and  $\beta$ -decay experiments have concluded that the ground state has a spherical shape near the closed neutron shell N = 126. The excited oblate deformed states become lower in energy toward the neutron-deficient region [18, 20, 23] and a strong mixture between the two configurations is observed for <sup>194</sup>Po [19]. Recent work even concluded on <sup>194</sup>Po being mostly oblate deformed [26]. It is established that <sup>192</sup>Po has an oblate deformed state for main component in the ground state, as observed by in-beam  $\gamma$ -spectroscopy [21]. In <sup>191</sup>Po, it was shown from the large hindrance in the  $\alpha$ -decay that the 13/2<sup>+</sup> isomer is deformed and possesses an important intruder component [22]. In <sup>190</sup>Po (mother nucleus of <sup>186</sup>Pb where triple shape coexistence was first observed [11]) a low-lying prolate band has been identified [24]. Finally, a recent study of <sup>186–188</sup>Po  $\alpha$ -decay suggests that the ground state of those isotopes is in a deformed prolate state [25]. Those observations should be reflected on the charge radii similarly as in the case of mercury and platinum, although the crossing of the proton gap makes systematic predictions considerably difficult.

On the neutron-rich side, the lead isotopes are the only even-Z isotopes in this region (Z = 78 - 84) measured beyond N = 126 [3]. The changes in mean-square charge radius at N = 126 show a kink (Fig. 1) that can not be reproduced by most nuclear models. So far, only Relativistic Mean Field calculations achieve to reproduce this shell effect [27, 28]. It would be interesting to search for a similar behaviour in the next even-Z isotopes (Po - Z = 84) and verify the similarity to the intermediate odd-Z case (Bi - Z = 83), which displays a behaviour comparable to that of lead [29].



Figure 2: Optical spectra for the even-A isotopes <sup>194,196,198,200,202,204</sup>Po. The fitted line is a simple Gaussian profile; a detailed analysis taking into account the total line shape is ongoing.

#### Experimental run from August 2008

The even-A isotopes <sup>194,196,198,200,202,204</sup>Po and odd-A isotopes <sup>193,195,197,199</sup>Po, the latter with a low spin ground state and a high spin isomer, were successfully produced and studied with the RILIS using one of the schemes recently developed at ISOLDE [30]. <sup>193–199</sup>Po were identified through their characteristic  $\alpha$ -decay at GLM, <sup>202</sup>Po through its  $\beta$ -decay and <sup>199–200,204</sup>Po through the  $\gamma$  radiation following their  $\beta$ -decay at the ISOLDE yield station. The optical profile of each of those 14 isotopes and isomers have been extracted and preliminary results are already available [31].

The optical spectra of the even-A polonium isotopes (Fig. 2) yields directly the isotope shift between any two isotopes. Since all have a 0<sup>+</sup> ground state spin, their optical spectra are made of a single peak. As a reference isotope, the  $\beta$ -decaying <sup>202</sup>Po is measured simultaneously with  $\alpha$ -decaying isotopes.



Figure 3: a. Optical spectrum of the low spin ground state of <sup>197</sup>Po. b. Optical spectrum of the high spin isomer of <sup>197</sup>Po.

The complicated structure of the odd-A isotopes, as shown for example in the case of <sup>197</sup>Po in Fig. 3, required a careful analysis to extract the hyperfine parameters related to the magnetic dipole and the electric quadrupole moments, before the isotope shift could be determined. An analysis of the  $\chi^2$  distribution created by varying the hyperfine parameters, allowed for the determination of the isotope shifts between those isotopes and <sup>202</sup>Po.

The isotope shifts extracted are shown in Fig. 4. The evolution of the even-A isotopes shows a departure from the linear trend beyond A = 198 while the odd-even staggering reverses beyond A = 196. Hard conclusions can however only be drawn from a comparison over the whole isotopic chain once the mean-square charge radii are extracted from those isotope shifts.

In the case of the odd-A polonium isotopes, the systematics on the magnetic dipole and electric quadrupole moments could also be extracted for the low and high spin states. The moments for the first two isotopes where the isomerism from the neutron in the  $i_{13/2}$ orbital is observed, namely <sup>201,203</sup>Po, are unfortunately not yet available to observe the complete systematic.

Thanks to an overlap between the isotopes studied in this work and that of Kowalewska et al. [8], we can compare the isotope shifts for two different optical transitions studied to determine the ratio of the their electronic F-factor, necessary to extract the change in the mean-square charge radius from the isotope shift. This technique, developed by



Figure 4: Isotope shifts between all the measured polonium isotopes. The reference is  $^{202}$ Po.



Figure 5: King plot between the data from [8] and IS456.

King [32], compares modified isotope shifts, where the different mass contributions are normalised and the values line up (provided there are no second order effects in the transitions involved). Fig. 5 shows the current King plot for IS456. The line should go through the origin and is determined with only two points, yielding a ratio of F-factors of -0.48(5). This is the largest source of uncertainty in the determination of the change in the mean-square charge radius, displayed in Fig. 6. For determination of the electronic F-factors for both spectroscopic transitions, calculations based on multiconfigurational wave functions are under investigation by S. Fritzsche.

Although the analysis is ongoing, it is already possible to conclude that this technique is sensitive enough and well suited for extracting structure information above Z = 82.



Figure 6: Mean-square charge radii of the Pb and Po isotopic chain.

# Further study of the polonium isotopic chain

We propose first to complete the study of the systematics of the moments and isotope shifts in the odd-A polonium isotopes towards the neutron-deficient side. The current knowledge indeed excludes <sup>201,203</sup>Po where the long-lived isomerism between low-spin state and high-spin state from the  $i_{13/2}$  orbital begins. Those anchor points are necessary to complete the systematic study of the moments as the  $i_{13/2}$  neutron shell is depopulated. The decay branches and half-lives are very well suited to measure  $\gamma$  decay lines as was done for <sup>199</sup>Po.

The current accuracy on the change in the mean-square charge radius is limited by the knowledge of the electronic F-factor. By measuring more isotope shifts overlapping with the previous data set from Kowalewska et al. [8] one could extend the King plot to have a better relative measurement of that parameter and have a good comparison with the atomic calculations. For that purpose, we propose to measure  $^{208,209,210}$ Po for which the optical spectra are simple (no quadrupole contribution to the hyperfine structure). Those long-lived isotopes have to be measured in a Faraday cup and beam contamination, especially from the isobaric  $^{208-210}$ Fr isotopes, has to be be suppressed. In recent tests at ISOLDE, high suppression of francium contaminants has been achieved using a quartz transfer line [33].

In order to observe the kink across the N = 126 shell-closure, we also propose to measure the isotope shift in the neutron-rich isotopes  $^{211,211m,212m,216,217,218}$ Po. By comparing the evolution of the polonium isotopic chain to that of lead as was done for the lighter isotopes in [8] or for the bismuth isotopes in [29], it is possible to estimate the order of magnitude of the isotope shift to be about 30 GHz between  $^{202}$ Po and  $^{216}$ Po and more as we go further from N = 126. This isotope shift is still within the scanning capabilities of the RILIS.

Finally, it would be very interesting to complete the study of the neutron-deficient isotopes from the original proposal. The study of  $^{192}$ Po was impossible in the course of

2007 as radiation protection required the proton beam on the GPS target to be limited to 1  $\mu$ A. Moreover, the release of polonium revealed to be quite slow, resulting in high decay losses. Altogether <sup>192</sup>Po was not observed during the run. Although higher proton intensity is now available, improving the release from the target-ion-source system is necessary to measure this isotope.

Isotope	Yield	Half-life	Counts	Isomer	Yield	Half-life	Counts
	$[\mu C^{-1}]$	$[\mathbf{s}]$	per step		$[\mu C^{-1}]$	$[\mathbf{s}]$	per step
<sup>192</sup> Po	$1.6 \cdot 10^{0}$	0.0332	14				
<sup>201</sup> Po	$1.6 \cdot 10^{6}$	918	60000	<sup>201</sup> <i>m</i> Po	$1.25 \cdot 10^{7}$	534	60000
<sup>203</sup> Po	$1.3 \cdot 10^{7}$	2160	11000	<sup>203m</sup> Po	$2.1 \cdot 10^{6}$	45	65000
<sup>208</sup> Po	$2 \cdot 10^{6}$	$9 \cdot 10^{7}$	FC				
<sup>209</sup> Po	$1.2 \cdot 10^{6}$	$3\cdot 10^9$	FC				
<sup>210</sup> Po	$5 \cdot 10^5$	$1.2 \cdot 10^{7}$	FC				
<sup>211</sup> Po	$6.2 \cdot 10^2$	0.516	8521	$^{211m}$ Po	$5 \cdot 10^5$	25.2	300000
				<sup>212m</sup> Po	$4 \cdot 10^{5}$	45.1	150000
<sup>216</sup> Po	$1.2 \cdot 10^{0}$	0.15	10				
<sup>217</sup> Po	$2.2 \cdot 10^{1}$	1.53	160				
<sup>218</sup> Po	$6.6 \cdot 10^{2}$	183	700				

# Yields and measurement feasibility

Table 1: Estimated yields from ABRABLA [34, 35] convoluted to the release and half-lives of the polonium isotopes of interest with a UC<sub>x</sub> target (density 50 g·cm<sup>-2</sup>) and estimated number of counts with optimal cycling time (1 super cycle implantation and 0, 1 or 2 super cycles decay); the longest-lived isotopes shall be measured in a Faraday cup (FC). The left column is for the ground states while the right column is for the isomeric states.

#### Yields

The yields of all the isotopes discussed previously differ greatly one from the other. The half-lives are also factoring in differently. As the yields of none of those isotopes have been measured with the RILIS, they are evaluated from ABRABLA calculations where the release and half-lives are convoluted as done in [30]. The yields and estimated number of decays at the different stations are given in Table 1.

Note for  $^{209-210}$ Po that the decay of short-lived precursor isotopes in the target can increase the yields by a factor of 30 [36]. The estimated yields are thus very conservative.

#### Contamination and limits

In the study of the  ${}^{201,203,208-210}$ Po isotopes, the highest contamination that can be expected is that of the isobaric francium. This contamination can be however suppressed using a quartz transfer line down to  $10^6 \text{ ions} \cdot \text{s}^{-1}$  [37, 33]. The next major isobaric contamination is that of the thallium isotopes which have yields of the same order of magnitude

than those of the laser ionised polonium isotopes [38]. In the case of a Faraday cup measurement, as proposed for  $^{208-210}$ Po, this would be visible as a constant base line but it would not put the measurement in jeopardy. In the case of of  $\gamma$  radiation detection, the separation from this isobaric contamination is guaranteed.

The study of the neutron-rich  $^{211-212,216-218}$ Po is very sensitive to the contamination from the isobaric francium nuclei as well as that from neighbouring masses. During the on-line measurement of August 2007 at the GLM beam line, contamination of  $^{213}$ Fr was found in the  $\alpha$ -decay spectrum of mass A = 216. This study would thus require the use of a quartz transfer line and the HRS mass separator. The use of HRS does not allow for simultaneous measurement of a reference isotope and will therefore extend the measuring time.  $^{216-218}$ Po can also be studied in the release bunching mode [39] thanks to the short half-life of the isobaric francium. No other contamination is then expected and the experiment is feasible, despite the low estimated yields comparable to those of  $^{193-194}$ Po.

The study of the very short-lived <sup>192</sup>Po is however very unlikely to be feasible in the current conditions. Although the estimated number of decays is higher than that of <sup>216</sup>Po and despite improvements in our detection efficiency (tripling of the  $\alpha$  detection solid angle), the yield estimate has a high uncertainty as the half-life of this isotope (0.033 s) is out of the range of previously measured isotopes [30]. The yield could be lower by an order of magnitude. The development of a target-ion source system with faster release is necessary to overcome the decay losses of such an isotope before it can be ionised.

### Request for beam-time

In-order to complete the study of shape coexistence in the neutron-deficient polonium isotopes and to directly observe the shell effects beyond N = 126, it is necessary to

- 1. complete the study of the moments and the change in the mean-square charge radii of  $^{201,203}$ Po,
- 2. increase the overlap with the previous data set by measuring the isotope shift with  $^{208,209,210}$ Po,
- 3. measure the isotope shifts of the neutron-rich  $^{211,211m,212m,216,217,218}$ Po.

All those isotopes are produced using a  $UC_X$  target and while we require a quartz transfer line and the use of HRS for some isotopes, it is important to note that it does not prevent the other isotopes from being measured. All isotopes can therefore be measured over the same run in the same conditions.

Based on the estimated yields and number of decays as well as on the experience from the measurement of August 2007 and the extra time required to measure the reference isotope apart from the measurement of interest, we request the following:

0. RILIS setup and optimisation		3 shifts
1. isotope shift and hyperfine structure	<sup>201,203</sup> Po	4 shifts
2. isotope shift	<sup>208,209,210</sup> Po	3 shifts
3. isotope shift	$^{211,211m,212m,216,217,218}$ Po	14 shifts
Total number of requested shifts	24 shifts	

#### Request for target developments

As <sup>192</sup>Po is a keyu isotope in the study of the neutron-deficient polonium isotopes, it is of great importance to assert its shape from the measurement of its isotope shift. As the yields are extremely limited, only the technique of resonant laser ionisation spectroscopy, used in experiment IS456, is suited to extract this information. Due to the slow release of polonium and the short half-life of this isotope, large decay losses occur before the ionisation can take place. We therefore request the development for faster release of polonium in order to later submit an addendum to finalise the study of the neutrondeficient polonium isotopic chain.

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