EUROPEAN ORGANIZATION FOR NUCLEAR RESEARCH

Letter of Intent to the ISOLDE and Neutron Time-of-Flight Committee

Preparation for the study of the transitional nucleus ^{191}Po with high-resolution laser spectroscopy at CRIS

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T.E. Cocolios[1], K.T. Flanagan[2], A.N. Andreyev[3], C. Babcock[2,4], J. Billowes[2], I. Budincevic[5], B. Cheal[2], V.N. Fedosseev[4], R. Garcia Ruiz[5], M. Huyse[5], K.M. Lynch[1,2], B.A. Marsh[4], G. Neyens[5], J. Papuga[5], T.J. Procter[2], S. Roethe[4,6], M.D. Seliverstov[3], H.H. Stroke[7], P. Van Duppen[5], K.D.A. Wendt[6], P.M. Walker[8]

[1] *ISOLDE, CERN, Geneva, Switzerland*
[2] *University of Manchester, Manchester, United Kingdom*
[3] *University of the West of Scotland, Paisley, United Kingdom*
[4] *EN Dept, CERN, Geneva, Switzerland*
[5] *Katholieke Universiteit Leuven, Leuven, Belgium*
[6] *Johannes Gutenberg Universität, Mainz, Germany*
[7] *New York University, New York, NY, USA*
[8] *University of Surrey, Surrey, United Kingdom*

Spokesperson: Thomas Elias Cocolios Thomas.Elias.Cocolios@cern.ch
Contact person: Kieran Flanagan Kieran.Flanagan@cern.ch

Abstract:
Recent results from the IS456 experiment at ISOLDE on the changes in the mean-square charge radii of the neutron-deficient polonium isotopes using the in-source resonant laser spectroscopy technique with the RILIS has revealed that $^{191}_{84}$Po is a key nucleus in the understanding of shape coexistence around $^{186}_{82}$Pb$_{104}$. Higher resolution is however required in order to determine the spin of the ground state ($T_{1/2} = 22$ ms) and of the long-lived high-spin isomer ($T_{1/2} = 93$ ms). Ground-state properties related to shape (changes in the mean-square charge radii, electric quadrupole moments) and to nuclear configuration (magnetic dipole moment) are also essential. We therefore propose to study ^{191}Po with the CRIS setup at ISOLDE. The limited production rates do not allow for this measurement to be performed with the existing facility; beam extraction developments and an upgrade of the incident proton beam energy to 2.0 GeV are necessary to improve the production rates. As a step toward this experiment at CRIS, we propose to do on-line preliminary work with ^{193,195,196,204}Po to ascertain the feasibility of this study.

Requested shifts: 5 shifts

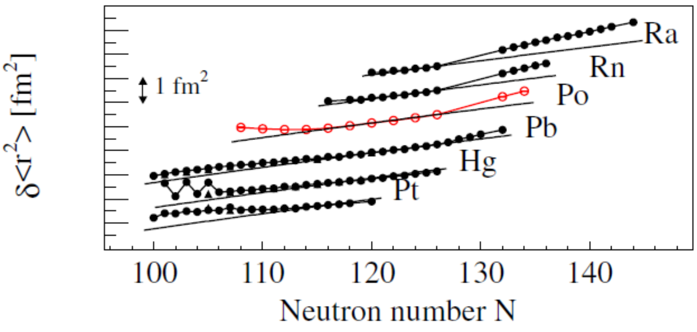

Figure 1: Changes in the mean-square charge radii of the even-Z isotopes around $Z = 82$. The solid lines represent the prediction of the spherical droplet model. For polonium, only the even-A isotopes are shown.

1 Physics Motivation

The region of nuclei around $Z = 82$ and $N = 104$ is characterized by the occurrence of different shapes at low energy [1, 2]. The most dramatic case is that of ^{186}Pb where the ground state and the first two excited states are believed to be of three different shapes [3]. The richness of the nuclear structure of the neighboring isotopes makes this region an ideal ground to study nuclear models and has attracted considerable interest in the recent years, both experimentally and theoretically [4–21]. In a shell-model approach, shape coexistence arises from the scattering of pairs of protons across the $Z = 82$ shell gap [1, 2]. With already two valence protons outside the $Z = 82$ shell closure, the polonium isotopes ($Z = 84$) have received a particular attention [6].
In a recent effort at ISOLDE, the ground-state properties of the neutron-deficient polonium isotopes have been studied with the ISOLDE Resonant Ionization Laser Ion Source (RILIS) [20–23]. It allowed the extraction of the changes in the mean-square charge radii of the isotopes $^{192-211,216,218}$Po, and of the magnetic dipole moment of the ground state and long-lived isomers in the odd-A isotopes. The changes in the mean-square charge radii revealed a gradual departure from the prediction of the spherical droplet from $N \leq 114$ in even-N isotopes, as seen in Fig. 1 [21]. This departure is very different from that of the mirror case mercury ($Z = 80$), with a pair of proton holes in the $Z = 82$ shell closure: only odd-A mercury isotopes in the vicinity of $N = 104$ display a major departure from the spherical droplet model [24].
The odd-A polonium isotopes are still under scrutiny. Preliminary results on the changes in the mean-square charge radii are consistent with the even-A isotopes picture and do not display any dramatic odd-even staggering, as in platinum ($Z = 78$) [25] or mercury. Their magnetic dipole moments are also consistent with the assigned configurations, though there are some departures from the similar configurations in lead and mercury. A publication on those isotopes is under preparation [22].
The most exotic isotope which was studied in this series is ^{191}Po. With a low-spin ground state and a high-spin isomer, this isotope is generally described similarly to the heavier odd-A polonium isotopes $^{193-199}$Po. ^{191}Po has also been studied in fusion-evaporation

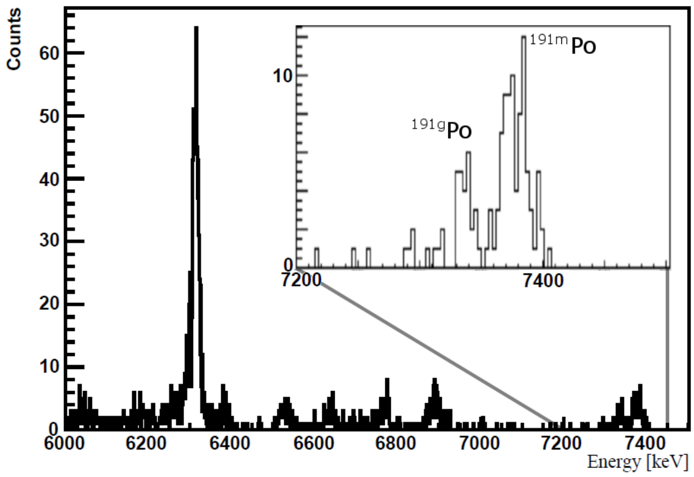

Figure 2: α decay of ^{191}Po observed with the Windmill detector setup at LA1 in the course of experiment IS456.

experiments at the RITU spectrometer of the University of Jyväskylä (Finland) [9]. The spin assignment for each isomer was made on the basis of the spin of the ground state and long-lived isomer in ^{187}Pb, of the measured hindrance factors in the α decay of ^{191}Po, and of the conversion coefficients in the subsequent γ-ray emissions.
In the recent ISOLDE study, both the low-spin ground state ($T_{1/2}$ = 22 ms) and the high-spin isomer ($T_{1/2}$ = 93 ms) have been observed, as shown in Fig. 2. In the earlier study of the production of the polonium beams with the ISOLDE RILIS [26], it has been observed that the high-spin isomer of the odd-A isotopes is systematically produced with higher intensity than the ground state. This is attributed to the large angular momentum transfer in the spallation reaction. The excited nuclei subsequently decay preferably via yrast levels, which eventually lead to the high-spin isomer.
One should also note that due to the slow release of the polonium isotopes, only a fraction of the produced atoms may be released from the target, depending upon the half life of the isotope. If this effect is accounted for, a systematic ratio of 1 : 5 between the low-spin ground state and high-spin isomer remains in ^{193,195,197}Po. If one considers similarly the ^{191}Po isotope, a ratio 5 : 1 is observed, in strong discrepancy with respect to the heavier isotopes. The low-spin ground state should not have been observed at all in experiment IS456, considering its short half life and the reduced cross section for its production. The overabundance of the ground state in the beam cannot be explained by the tuning of the laser frequency, as both hyperfine structures have been scanned across and covered similarly (see Fig. 3).
Furthermore, the scan of the hyperfine structure of the low-spin ground state, though of limited statistics, has revealed a structure very dissimilar to that of the heavier odd-A isotopes, as shown in Fig. 3. These changes could have several origins, from deformation to difference in spin. It appears then that this isotope is a turning point in the evolution of the structure of the neutron-deficient polonium isotopes. The observation of a low-lying prolate band in ^{190}Po [27], in contrast to the weakly oblate bands observed in the heavier

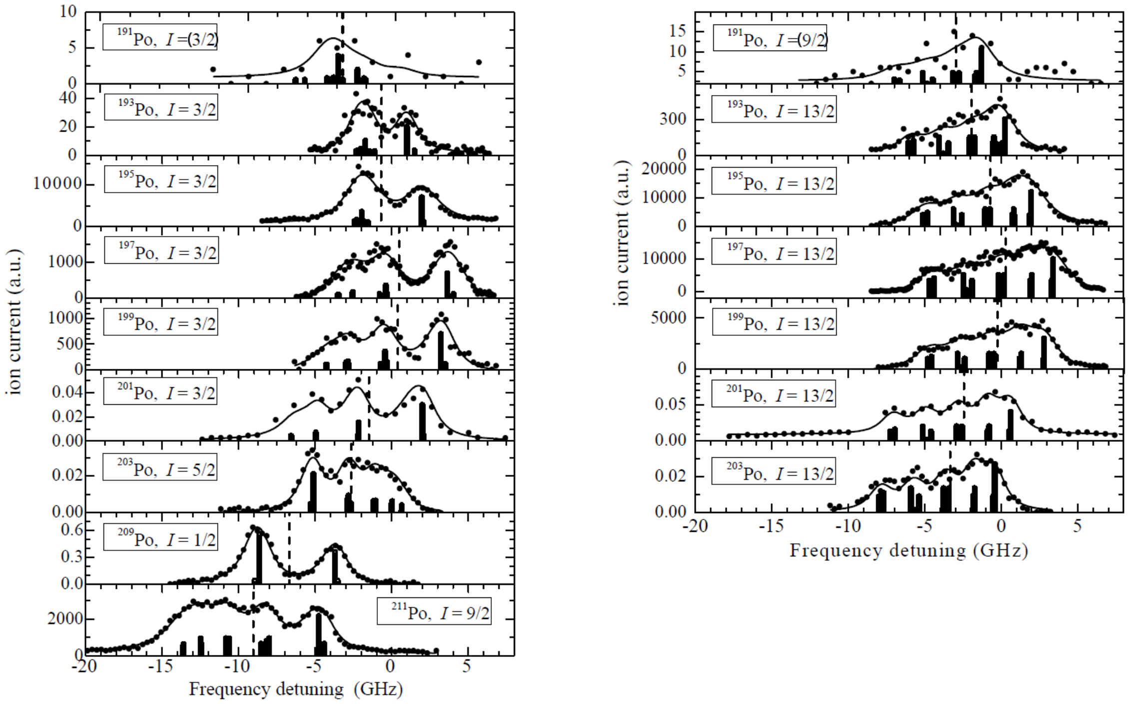

Figure 3: Hyperfine structure scan of the 843 nm transition in polonium in experiment IS456. The solid line represents the best fit for the spin indicated; the histogram represents the distribution of the hyperfine components. Left: low-spin state. Right: high-spin state.

polonium isotopes, further motivates the transitional nature of ^{191}Po.
The lighter isotopes $^{186-190}$Po have been studied in fusion-evaporation reactions at the SHIP velocity filter in GSI, where the α decay pattern of the odd-A isotopes ^{187,189}Po shows a departure from the trend of the even-A isotopes $^{186-200}$Po [28]. This has been interpreted as a sign of deformation in the ground state, also predicted by Beyond Mean Field calculations [21, 29, 30].
By studying the ground-state and isomer properties of the isotope ^{191}Po, we shall determine the spin, configuration, and shape of those two states which lay at a cornerstone of this region of shape coexistence.

2 Experimental technique and requirements

ISOLDE is only facility worldwide where the requested beam may be produced with the required beam quality for laser spectroscopy.

2.1 Beam production and delivery

The ^{191}Po atoms are produced in the proton-induced spallation of a UC$_x$ target at ISOLDE. Using the RILIS with the lasers set in narrowband mode in the course of ex-

Isotope	$T_{1/2}$	RILIS / Surface			Hot plasma		
		Prod [ion·s^{-1}]	in 160 ms [ion·s^{-1}]	Cont	Prod [ion·s^{-1}]	in 160 ms [ion·s^{-1}]	Cont
^{191g}Po	22 ms	0.1	0.1		1	1	
^{191m}Po	93 ms	0.3	0.3		3	3	
^{191m}Bi	150 ms	0.1	0.1	3 : 1	10^2	10^2	1 : 33
^{191}Bi	12 s	10	2	1 : 6	10^4	$1.5 \cdot 10^3$	$1 : 5 \cdot 10^2$
^{191}Pb	2 min	100	15	1 : 50	10^7	$1.5 \cdot 10^6$	$1 : 5 \cdot 10^5$
^{191}Tl	5 min	$5 \cdot 10^5$	$8 \cdot 10^4$	$1 : 2.7 \cdot 10^5$	-	-	-
^{191}Hg	50 min	-	-	-	10^6	$1.5 \cdot 10^5$	$1 : 5 \cdot 10^4$

Table 1: Summary of the current beam composition at mass $A = 191$ using the RILIS for polonium or a hot-plasma ion source. For each source, the production rate is given, the expected beam for the first 160 ms after the proton impact, and the ratio to the beam of interest (^{191m}Po). The production rates for ^{191}Po are extrapolated based on the observed activity during the narrowband study of experiment IS456. The ratio of ^{191}Po with respect to each contaminant in the beam, labeled 'Cont' in the table, are 4 times better for the low-spin ground state than those quoted here.

periment IS456, a rate of 0.03 ion·s^{-1} has been observed in resonance for ^{191m}Po and 0.01 ion·s^{-1} for ^{191g}Po. A factor 10 increase is expected for broadband operation.
Surface-ionized isobaric ^{191}Bi was observed in the α-decay spectrum. Surface-ionized isobaric ^{191}Pb and ^{191}Tl should also be present, the latter representing the most important contaminant, with a rate of $\sim 10^5$ ion·s^{-1} [1].
The polonium atoms may also be ionized in a hot-plasma ion source. The ionization efficiency is expected to be higher by an additional order of magnitude ($\times 10$). The contaminations will however be much more enhanced, especially with ^{191}Pb at a rate of 10^7 ion·s^{-1}, and ^{191}Hg at a rate of 10^6 ion·s^{-1}.
The ion beam is accelerated with a 50 kV potential and separated from other masses in the high resolution separator (HRS) and cooled and bunched in the ISOLDE cooler & buncher (ISCOOL). Due to the short half life of the ground state ^{191g}Po ($T_{1/2} = 22$ ms), only the first two bunches after proton impact on target can be utilized, assuming a bunch release rate of 50 Hz, while up to eight bunches may be utilized in the case of the isomer ^{191m}Po ($T_{1/2} = 93$ ms).
At such proximity to the proton pulse, most of the short-lived ^{191m}Bi will also be released, but all other longer-lived contaminants will be suppressed. The beam contamination is then reduced accordingly by a factor 1 : 25 for ^{191g}Po or 1 : 6 for ^{191m}Po. The beam purity is summarized in Table 1.

2.2 CRIS

The beam is sent to the CRIS setup. It is first neutralized in an alkali-vapor charge-exchange cell (CEC). The neutralization efficiency for polonium is unknown and will be

[1] yields of ^{191}Tl with RILIS are $5 \cdot 10^7$ ion·s^{-1}, with a selectivity of ~ 200.

investigated.

After the CEC, the atom beam keeps drifting along the beam line while the surviving ions are deflected away with an electrostatic potential. A set of three laser beams is then overlapped with the atom beam to resonantly excite the beam from its ground state, as shown in Fig. 5. Two tunable lasers have to be used to produce the 255.8 nm and 843 nm transitions, the former requiring frequency tripling from 767 nm. The final non-resonant step is provided by a frequency-doubled Nd:YAG laser. All three laser systems match existing CRIS proposals, on copper (IS531, 244 nm and 249 nm) and francium (IS471, 423 nm from frequency-doubled 846 nm).

The reionized beam is deflected via an electrostatic potential to a counting station. The ion beam impinges on a copper dynode biased at -500 V. The secondary electrons emitted upon impact are accelerated towards a multichannel plate detector (MCP) to be recorded. The beam can alternatively be sent to an ultra-thin carbon foil of the CRIS decay spectroscopy station (DSS), which is surrounded by two silicon detectors to measure the energy of α particles or to count β particles. The energy resolution of the α particle detection is similar to that shown in Fig. 2 and allows an unambiguous identification of either isomer.

The hyperfine structure of the isotope is scanned by Doppler tuning the frequency of the second laser at 843 nm in the rest frame of the atoms by adjusting their velocity with a potential applied at the entrance of the CEC. The resolution of the CRIS technique is not limited by the Doppler broadening of the high-temperature ion source, unlike in the case of in-source laser spectroscopy [31]. The high temperature is responsible for a large energy distribution in the ion source; however, upon acceleration, this distribution translates to a small velocity spread as $\delta E \propto v \cdot \delta v$. A resolving power as low as 100 MHz can be achieved, with which it will be possible to resolve the hyperfine structure components. These were left unresolved in the IS456 data as the linewidth was ~ 3 GHz (Fig. 3). This improved resolution will allow the extraction of spins, accurate isotope shifts, magnetic dipole moments, and electric quadrupole moments [31].

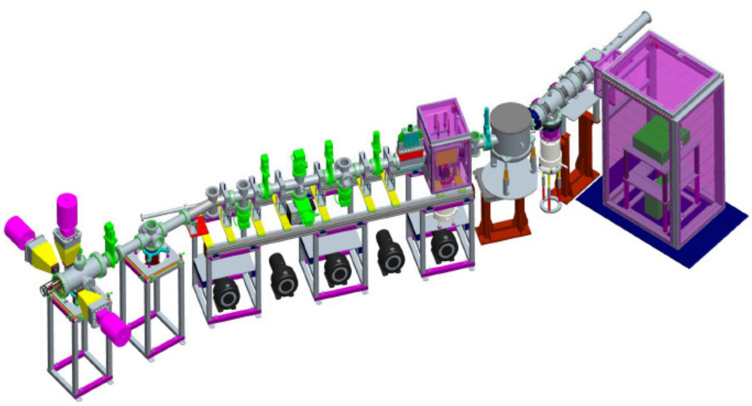

Figure 4: Layout of the CRIS setup at ISOLDE.

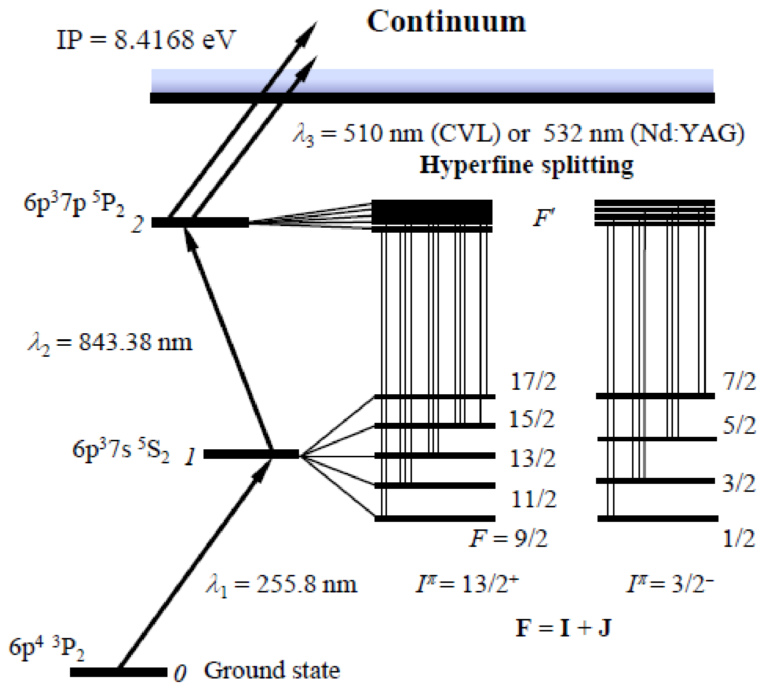

Figure 5: Laser ionization scheme of polonium.

2.3 Feasibility

2.3.1 2.0 GeV PSBooster

Based on the production rates and contamination levels presented in Table 1, the cleaner conditions of the RILIS are preferred, even if the absolute beam intensity is smaller.
In the case of ^{191m}Po, assuming an efficiency of the CRIS technique of 1 : 30 as per the original proposal of experiment IS471, the current beam intensity would yield a rate of 0.01 count per second at the experiment. The background rate arises from non-resonant collisional re-ionization of atoms in the beam line. At a pressure of 10^{-9} mbar, the current typical pressure of the CRIS beam line, an occurrence of $1 : 10^5$ is observed. A count rate of 1 count per second can then be expected. 10^4 s are then necessary to have a hint of an effect at the highest point of the hyperfine structure. For a scan with 100 points, a total of 10^6 s is then required, which represents 278 days. These conditions make this experiment impossible at a facility such as ISOLDE.
It is possible to further improve the vacuum of the CRIS setup, as has been demonstrated by the WITCH collaboration, with the use of getter material in the beam line. Such an improvement is foreseen in the near future (2014) and the re-ionization rate would then be improved to $1 : 10^6$.
In order to improve the experimental conditions, higher beam intensity would also be beneficial. The RILIS is already very efficient and there cannot be dramatic improvements on the ionization efficiency without extensive online developments to search for auto-ionizing states or Rydberg series.

The use of a fresh target can surely be beneficial. The production rates for ^{191}Po from experiment IS456 were acquired after a few days of irradiation. The target had by then started to degrade, increasing further the release time of polonium. The shorter-lived isotopes are the most affected by those effects.
It would also be beneficial to improve the release properties of polonium from the target. Studies are ongoing for a new generation of UC_x targets with overall improved release properties. Alternative release mechanisms may also be explored. For example, the extraction of molecular compounds could be favorable. In the case of the polonium atom, the PoO_2 molecule is a promising candidate. The presence of oxygen in the target prevent however the use of UC_x as target material and ThO should be used instead. The release properties depend also from the interaction of the atom of interest with the target assembly material. Polonium has a high reactivity to tantalum; the exposed tantalum elements should therefore be lined with rhenium foils. All the aforementioned developments are currently undertaken by the ISOLDE Target Group in the frame of other research proposals and letters of intent. It is however difficult at this stage to quantify the improvement on the production rates of ^{191}Po.
Finally, the spallation cross section may also be improved upon, by increasing the energy of the primary proton beam to 2.0 GeV. A factor 10 improvement in beam intensity is foreseen for the isotopes in this region. This applies to all elements listed in Table 1.
By combining these two improvements (reduced re-ionization rates and increased beam intensity), a count rate of at least 0.1 count per second for the isotope of interest can be reached at CRIS, compared to 1 count per second for the background rate. This would translate into a 3-hour long scan, which is reasonable for such an exotic study. We therefore motivate the upgrade to the CERN driver facility to provide ISOLDE with higher energy (2.0 GeV) in the near future.

2.3.2 Preliminary tests at CRIS

The study of the polonium isotopes at CRIS is conditional on many factors. Due to the radioactive nature of its isotopes, the polonium atom has not been extensively studied. The atomic spectroscopy has only been performed within a lamp [32], or for 2 transitions with laser spectroscopy at off-line atomic beam laser spectroscopy facility [33] and at ISOLDE [20, 21, 26]. There is no experience on the neutralization of a polonium beam with use of an alkali vapor. Though no laser development is required for the ionization of polonium, it is the first element for which a 3-step ionization scheme will be tested at CRIS. Altogether, we request **2 shifts** to demonstrate the possibility of ionizing polonium at CRIS, using any beam of polonium. Even-A isotopes with no hyperfine structure are best suited; the best compromise between production rates and radio-protection concerns are ^{196,204}Po.
The proposed study relies also on the possibility to resolve the complex hyperfine structure of the odd-A isotopes and to separate the low-spin ground state from the high-spin isomer. It is therefore essential to test the technique on the isotopes ^{193,195}Po, nearest to the isotope of interest. For that purpose, we request **3 shifts**.
Beam properties are presented in Table 2. Contamination is not an issue for any of those beams.

Isotope	$T_{1/2}$ [s]	Prod [ions$\cdot\mu C^{-1}$]
^{193g}Po	0.45	70
^{193m}Po	0.24	100
^{195g}Po	4.64	25000
^{195m}Po	1.92	55000
^{196}Po	5.8	480000
^{204}Po	12708	11000000

Table 2: Production rates of the polonium beams of interest for the preliminary tests [26]. The improved power of the Nd:YAG RILIS upgrade results in higher production rates than those presented in this table.

3 Shifts and developments requests

Summary of requested shifts: In order to demonstrate the possibility of ultimately performing the studies of ^{191}Po, we request **5 shifts** at the CRIS beam line with ^{193,195,196,204}Po, using a standard UC_x target and the RILIS.
Developments: In order to have sufficient beam intensity, a full proposal will be coupled to an increase in the production rates, expected to be achieved, amongst other developments, with an upgrade to 2.0 GeV of the proton beam energy from the PSBooster.

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Appendix

DESCRIPTION OF THE PROPOSED EXPERIMENT
The experimental setup comprises: (*CRIS and its decay spectroscopy station DSS*)

Part of the	Availability	Design and manufacturing
CRIS	☒ Existing	☒ To be used without any modification

HAZARDS GENERATED BY THE EXPERIMENT Hazards named in the document relevant for the fixed CRIS installation.