EUROPEAN ORGANIZATION FOR NUCLEAR RESEARCH

Proposal to the ISOLDE and Neutron Time-of-Flight Committee

Laser & decay spectroscopy and mass spectrometry of neutron-rich mercury isotopes south-east of ^{208}Pb

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A. Algora[1], B. Andel[2], A. N. Andreyev[3], S. Antalic[2], D. Balabanski[4] M. Benhatchi[5], J. Benito[6], C. Bernerd[7], K. Blaum[8], J. A. Briz[6], R. B. Cakirli[9], K. Chrysalidis[7], T. Cocolios[10], J. G. Cubiss[3], T. Day Goodacre[11], V. N. Fedosseev[7], L. M. Fraile[6], L. P. Gaffney[12], G. Georgiev[5], P. F. Giesel[13], K. Gladnishki[14], R. Heinke[7], Y. Hirayama[15], A. Illana[6], D. Kocheva[14], U. Köster[16], D. Lange[8], R. Lica[17], Yu. A. Litvinov[18], D. Lunney[5], B. A. Marsh[7], A. McFarlane[3], J. Mišt[2], A. Morales[1], M. Mukai[19], S. Naimi[5], L. Nies[7], T. Niwase[15], J. Ojala[12], B. Olaizola[20], C. Page[3], R. D. Page[12], J. Pakarinen[21], Z. Podolyak[22], G. Rainovski[14], M. Rosenbusch[15], S. Rothe[7], P. Schury[15], Ch. Schweiger[8], L. Schweikhard[13], S. Sharma[23], A. Sitarčík[2], P. Srivastava[23], K. Stoychev[7], M. Stryjczyk[21], P. Van Duppen[10], Y. Watanabe[15], J. Wilson[3], Z. Yue[3,7] + IDS collaboration

[1]IFC, CSIC, Valencia, Spain; [2]Comenius University in Bratislava, Slovakia; [3]University of York, UK; [4]ELI-NP, Bucharest, Romania; [5]CNRS/Université Paris-Saclay, France; [6]Universidad Complutense de Madrid, Spain; [7]CERN, Geneva, Switzerland; [8]Max-Planck-Institute for Nuclear Physics, Germany; [9]Department of Physics, Istanbul University, Turkey; [10]IKS, KU Leuven, Belgium; [11]University of Manchester, UK; [12]University of Liverpool, UK; [13]Universität Greifswald, Germany; [14]Sophia University, Bulgaria; [15]WNSC, IPNS, KEK, Japan; [16]ILL, Grenoble, France; [17]IFIN-HH, Romania; [18]GSI, Germany; [19]RIKEN, Japan; [20]IEM-CSIC, Madrid, Spain; [21]JYFL, Jyvaskyla, Finland; [22]University of Surrey, UK; [23]Department of Physics, IIT, Roorkee.

Spokesperson: J. G. Cubiss [james.cubiss@york.ac.uk], D. Lange [daniel.lange@cern.ch], A. N. Andreyev [andrei.andreyev@york.ac.uk], U. Köster [koester@ill.fr]
Contact person: Ch. Schweiger [christoph.schweiger@cern.ch], Z. Yue [zixuan.yue@york.ac.uk]

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Abstract: Following the successful completion of LOI244 in 2023 (development of neutron-rich mercury beams with a UC_x+quartz line target [1]), we propose to study the properties of the neutron-rich isotopes $^{209-212}$Hg through a combination of decay and laser spectroscopy, as well as mass spectrometry. The nuclei south-east of ^{208}Pb are of key interest for nuclear structure and nuclear astrophysics studies. However, the difficulty in accessing them in the laboratory means there is a scarcity of data. Hence, new measurements of β-decay properties, masses, charge radii and magnetic dipole moments provide critical tests for models that attempt to describe this region of the nuclear chart.

Summary of requested shifts: 21 shifts

1 Introduction

Mercury isotopes with $A \leq 208$ have been successfully studied in our previous works at ISOLDE using a molten lead target [2]-[5], which cannot be used to study heavier isotopes as ^{208}Pb does not contain enough neutrons. Therefore, the only way to produce more neutron-rich mercury nuclides with proton-induced reactions is with a uranium (or thorium) target. However, such target and ion source systems produce isobaric contamination from surface-ionised francium, which have extremely high in-target production yields for A=209-212 (10^8-10^9 atoms/μC). The very high intensity of this contamination has prevented previous experimental access to this mass region. Hence, despite their importance for nuclear structure and astrophysics, the properties of the neutron-rich mercury isotopes are poorly known, with the limited available data collected in experiments at other facilities (see following section for details).

In 2023, our LOI244 [1] run measured the yields and purity of the neutron-rich $^{207\text{-}211}$Hg beams using a UC_x+quartz line target. Our tests successfully demonstrated sufficiently high production yields and a strong enough suppression of the francium contamination by the quartz line for dedicated studies of these neutron-rich mercury isotopes. An example of the β-tagged γ-ray spectrum collected for ^{210}Hg at the ISOLDE Decay Station (IDS) is shown in Figure 1, with RILIS ionising lasers turned on (black) and off (red). Strong transitions following the ^{210}Tl→^{210}Pb β decay are seen in the lasers on spectrum, along with several new transitions assigned to the ^{210}Hg→^{210}Tl decay (new γ-ray transitions and thallium x-rays were identified for the decays of ^{209,211}Hg also). The absence of these transitions from the lasers off spectrum demonstrates that no surface ionised thallium was produced, therefore all the observed activity originates from laser-ionised mercury precursors. Similarly, no transitions from the decay of ^{210}Fr are observed, due to a combination of the suppression from the quartz line, the IDS tape movement, and the β-tagging technique.

During the yield measurements, cumulative singles γ rates of >0.5 Hz were recorded at IDS for transitions belonging to the decays of $^{209\text{-}211}$Hg, using just 4 HPGe clover detectors. These rates are sufficient to perform precision decay and decay-tagged laser spectroscopy, along with mass spectrometry studies of these nuclei during a dedicated beamtime.

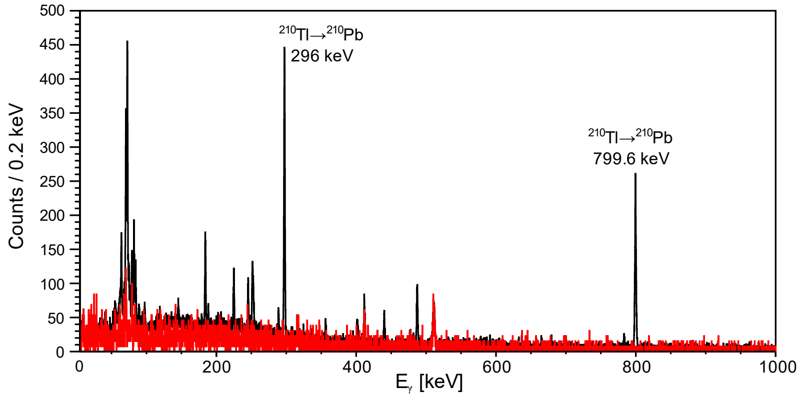

Figure 1 - β-tagged γ energy spectrum for A=210 taken during LOI244 [1] with lasers on (black) and off (red). The two spectra are normalised to run time, data with lasers on and off were recorded in 7.3 and 1 hours, respectively. The known γ-ray transitions from the ^{210}Tl→^{210}Pb β decay are labelled. Note that neither of these transitions are present in the lasers off spectrum, indicating that there is no surface ionized thallium contamination.

2 Previously known data and motivation for new study of neutron-rich Hg

β-decay properties

The nuclei in the region south-east of doubly-magic ^{208}Pb (see Figure 2) are important for constraining astrophysical nucleosynthesis models [6]. Critically, astrophysical network calculations rely on input of basic nuclear properties, such as masses, half-lives ($T_{1/2}$), decay modes, and the probability of β-delayed neutron emission. The mercury isotopes with N≥126 have valence particle configurations which allow first-forbidden β decays to directly compete with allowed Fermi and Gamow-Teller transitions (see for example the ^{208}Hg→^{208}Tl decay [7]), which can significantly alter their lifetimes relative to expectations [8]-[10]. Furthermore, information on the isotopes in this region provides stringent tests for large-scale shell model calculations [10]-[12], which have been used to calculate properties such as masses, $T_{1/2}$, log*ft* values, level schemes, and magnetic moments. However, there are presently little experimental data available (see Figure 2) to compare the models to due to the difficulty in accessing this region in the laboratory, with the available data coming from studies employing relativistic fragmentation, or deep-inelastic reactions [13]-[19].

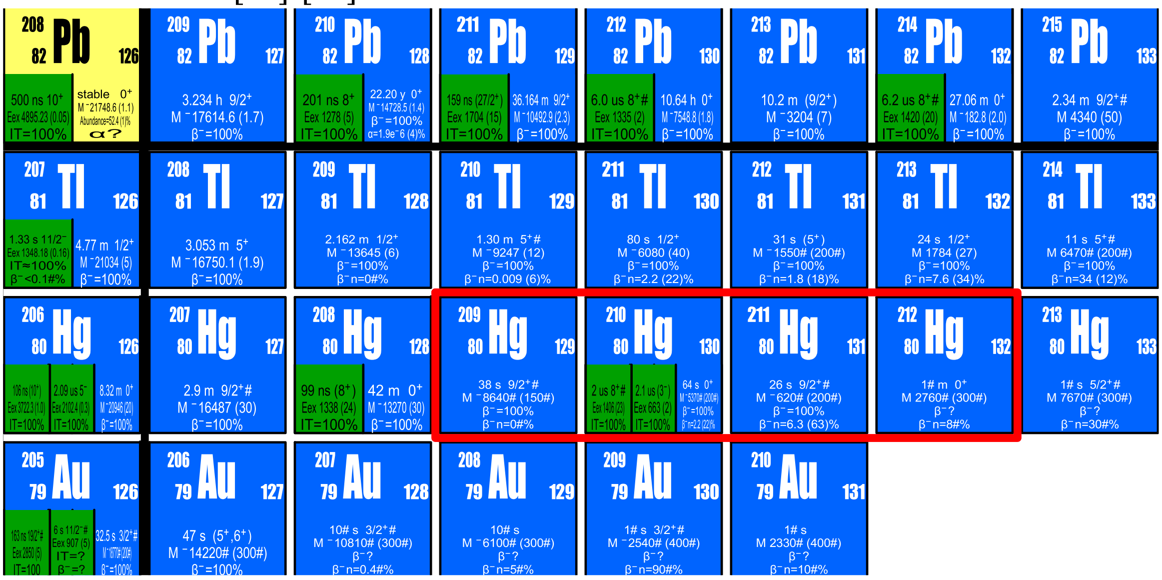

Figure 2 – Chart of nuclides for isotopes with Z≤82, N≥126, with the $^{209\text{-}212}$Hg isotopes we propose to study highlighted by the red rectangle. Data taken from the NUBASE2020 evaluation [20]. The $T_{1/2}$ and masses labelled with '#' are not measured but estimated from systematic trends.

Previously, values of $T_{1/2}(^{208}\text{Hg}) = 41^{+5}_{-4}$ min and $T_{1/2}(^{209}\text{Hg}) = 35^{+9}_{-6}$ s were measured after production in multi-nucleon transfer reactions and chemical separation, at the Heavy Ion Research Facility Lanzhou (HIRFL) [13],[14]. Later fragmentation studies at relativistic energies with the FRS at GSI [15]-[17], with statistics of the order of just a few tens to hundreds of counts for $^{208\text{-}211}$Hg [17], found $T_{1/2}(^{208}\text{Hg}) = (132.2 \pm 50.0)$ s and $T_{1/2}(^{209}\text{Hg}) = (6.3 \pm 1.1)$ s. This disagreement between the two values for ^{208}Hg was partially resolved when $T_{1/2}(^{208}\text{Hg})$ = 135(10) s was re-measured at ISOLDE [7], which is consistent with, but more precise than the value of the GSI study [17].

However, the remaining large discrepancy in the lifetime of ^{209}Hg suggests that its β-decay should be restudied. Indeed, our preliminary data from LOI244 indicate that the decay scheme from the HIRFL study [14] is incorrect. Instead, our data contains several new γ-ray transitions, along with some that were observed following the decay of a 95 ns, J^{π}=(17/2^{+}) isomer in ^{209}Tl, produced in relativistic fragmentation reactions at GSI [18]. However, dedicated measurements are required to collect sufficient statistics for constructing a reliable decay scheme.

There are presently no data for the β-decay schemes of ^{210,211}Hg. However, our preliminary results have identified several new γ-ray transitions in coincidence with thallium x-rays, following the decays of ^{210,211}Hg (see Figure 1). Our observed β-γ coincidence rates for these isotopes are sufficient for the first β-decay schemes for ^{210,211}Hg to be constructed.

Furthermore, all collected decay data for $^{209\text{-}211}$Hg will be used to place constraints on β-delayed neutron emission probabilities. This will be done by observing whether γ transitions belonging to $^{A\text{-}1}$Tl/Pb are present in data collected at mass setting A.

Currently, results for subsets of the properties discussed above from large-scale shell model computations are available up to ^{211}Hg and ^{213}Tl. New, dedicated calculations are planned for the data sets we will collect.

Atomic masses

Presently, the measured masses for mercury isotopes only extend to A=208 [21]. The masses of nuclei near ^{208}Pb can be used to probe the interaction strength between the last proton and neutron, reflected by δV_{pn} which is extracted from nuclear binding energies B:

$$\delta V_{pn}(Z,N) = 1/4[\{B(Z,N) - B(Z,N-2)\} - \{B(Z-2,N) - B(Z-2,N-2)\}],$$

for even-even nuclei [22] (similar δV_{pn} formulas for odd-A nuclei can be found in [23]). The proton-neutron interaction is an essential ingredient when discussing configuration mixing, the onset of collectivity in nuclei and nuclear deformation, as well as the nuclear symmetry energy. Indeed, as the proton-neutron interaction is short range, δV_{pn} is predicted to increase when the last protons and neutrons occupy orbitals with strong spatial overlaps. In the regions surrounding ^{208}Pb shown in Figure 3(a), a "cross" pattern in δV_{pn} values is expected, whereby δV_{pn} is large for isotopes in quadrants II and III, and small for I and IV.

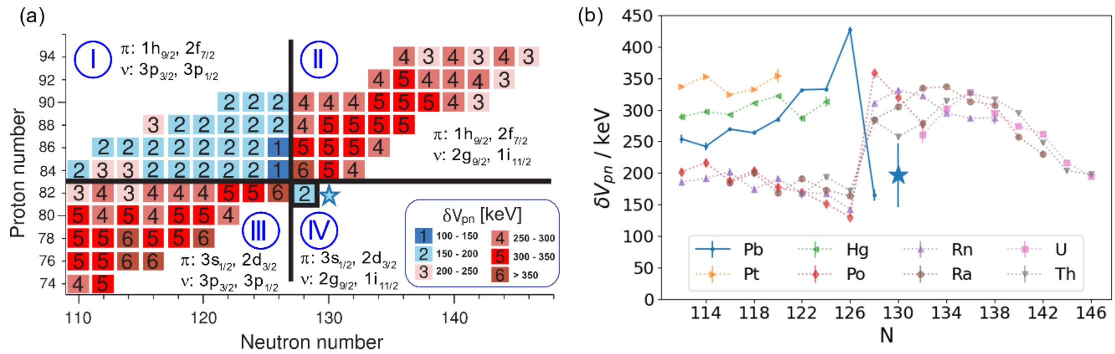

Figure 3 – (a) Values for δV_{pn} as a function of Z and N (adapted from [21]), and (b) δV_{pn} values for different isotopic chains as a function of N with mass values extracted from AME2020 [24] (adapted from [25]). In (a), the quadrants are labelled with the orbitals the last protons and neutrons are expected to occupy in the spherical nuclei near Z=82 and N=126. The star in both panels indicates $\delta V_{pn}(^{212}\mathrm{Pb})$ which will be extracted in our measurements, with the error bars in (b) representing the uncertainty in the estimation from systematics given in AME2020.

Currently, many experimental masses are known for isotopes in sectors I, II and III, with the data following the expected pattern. However, only one data point for ^{210}Pb exists for region IV [see Figure 3(a)]. This result comes from a mass measurement of ^{208}Hg, made at GSI using the storage ring ESR [21]. Additional data in this so-far unexplored region is sorely needed to test the prediction of a low δV_{pn} value in these isotopes. Masses of mercury isotopes are needed to extend the δV_{pn} values along the lead chain.

In the early 2000s, the weakening of the lead shell gap in neutron-deficient isotopes was observed through mass measurements, though energies of the first 2^+ states contradicted this [26]. Later, the reduction of the lead shell-gap energy was explained by an interplay of deformation energies in even-even mercury and polonium nuclei, the masses of which are used in the extraction of the lead shell-gap. It is also possible that neutron-rich mercury isotopes are deformed. Our mass measurements will be a sensitive probe to detect a possible onset of such deformation.

Furthermore, it is presently unfeasible to directly reach r-process nuclides in these heavy nuclei. Therefore, it is of utmost importance to constrain nuclear mass models, which show deviations of several MeV in this region. We note that mass precisions of the order of 50 keV are needed for nuclear astrophysics applications [27],[28]. In this context, we will provide new data in the vicinity of magic nuclei, which are particularly important in model adjustments.

Our data from LOI244 taken with the ISOLTRAP multi-reflection time-of-flight mass spectrometer (MR-ToF MS) show the feasibility of measuring the masses of at least ^{209,210}Hg (see Figure 4). A total of ~150 and ~300 ions were detected for ^{209,210}Hg, respectively. Due to the long buncher loading time chosen for the yield and contamination study in these tests, high counts per experimental cycle were observed. However, the isobaric contamination from lead-polonium induces space-charge effects [29] on the less abundant mercury isotopes, systematically shifting the ToF of the latter.

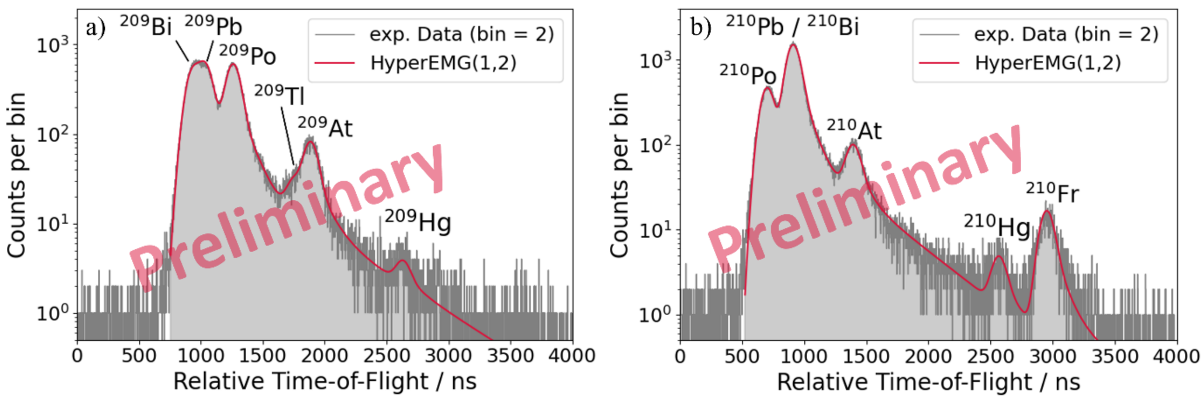

Figure 4 – Time-of-Flight spectra for (a) A = 209 and (b) A = 210 collected with the ISOLTRAP MR-ToF MS during the yield and contamination study of LOI244. The mass determination of ^{209,210}Hg is complicated by systematic ToF-shifts from space-charge effects induced by more abundant isobaric contamination. Increasing the statistics with lower ion loads (by selecting shorter buncher loading times) will allow precision mass measurements of ^{209,210}Hg. Note that the observed contaminants are long-lived, and therefore do not affect the proposed decay and laser studies. The spectra confirm the strong suppression of the dominant francium contamination, which has in-target production yields of 10^8-10^9 atoms/μC.

New and improved measurements with lower ion loads are required to reduce and characterise this shift, and accurately determine the masses of the mercury isotopes of interest.

Charge radii and magnetic dipole moments

Magnetic dipole moments (μ) and changes in mean-squared charge radii ($\delta\langle r^2\rangle$) extracted from hyperfine structure (hfs) and isotope shift (IS) measurements can be used to probe how ground state configurations and deformations evolve with changing neutron number. Our results for the neutron-deficient mercury isotopes [2],[3] concluded the story on shape staggering in mercury isotopes near N=104, whilst our studies of the neutron rich ^{207,208}Hg [4],[5] were the first confirmation of the "kink" in charge radii when crossing N=126, below Z=82. Extending these measurements above ^{208}Hg is one of our goals.

A consistent description of the observed kinks in the gradient of experimental charge radii, and differences in μ from the Schmidt values of nuclei close to shell closures remains a challenge for nuclear structure theory, with many competing approaches on the market (see for example [4],[5],[30]-[37]). As μ values are dominated by the configurations of valence particles/holes, they provide additional, critical constraints on the models that attempt to describe the structures in this region.

3 Proposed program for the neutron-rich Hg isotopes

Our results from LOI244 using the quartz transfer line have shown that the neutron-rich $^{209-211}$Hg isotopes can be produced with intensities high enough to study their properties through a combination of mass spectrometry, decay, and laser spectroscopy. The proposed studies are summarised in Table 1.

Table 1 - Summary of proposed measurements for the different isotopes. Solid circles indicate measurements that can definitely be made successfully, hollow circles for challenging ones that will be attempted. Yield estimates are based on the preliminary decay data, using the known transition intensities from the thallium to lead β-decays.

		Measurement		
Isotope	**Yield estimate [ions/μC]**	**β-γ decay**	**Mass**	**IS + hfs**
^{209}Hg	4(1)	●	●	●
^{210}Hg	4(1)	●	●	●
^{211}Hg	0.5(1)	●	○	○
^{212}Hg	n/a	○		

Mercury beams will be produced using the standard, three-step RILIS laser ionisation scheme used in [1]-[5]. The extracted ions will be mass separated using the ISOLDE separator and sent to the measurement devices discussed below. Half-life and decay studies will be performed at IDS, using plastic scintillator and HPGe clover detectors to measure β- and γ-decays, respectively. Activity will be implanted into the movable tape of IDS, which will be used to remove unwanted, long-lived daughter and granddaughter activity from the measurement position(s). Following its recent upgrade, IDS will be equipped with 10 clover detectors at the implantation position to give a projected singles efficiency of 6.4% at 1 MeV (~50-100% improvement for singles, ~5 times improvement for γ-γ coincidences relative to [1]), and 2 clovers in close geometry to a newly commissioned secondary decay position, for recording the decays of long-lived activity. Arrays of plastic scintillators for β-particle tagging will be used at both positions, with efficiencies of ~30-40%.

The masses of ^{209,210}Hg will be measured using the MR-ToF MS of ISOLTRAP, from which a precision of the order ~50 keV is achievable. The mass measurement of ^{210}Hg is particularly critical and will be used to determine the strength of the proton-neutron interaction in δV_{pn}(^{212}Pb) with an estimated precision of ~15 keV.
The IS and hfs studies will be extended to ^{209,210}Hg, where the observed decay rates during LOI244 were sufficient for performing laser scans. Whilst only one peak will be present in the hfs for the 0^+ ground state of ^{210}Hg, the hfs of the $9/2^+$ state of ^{209}Hg will have a broad structure similar to the $9/2^+$ ground state of ^{207}Hg (≈60 GHz, see Figure 4 of [5]). Hence, operating RILIS in a broadband mode (~7 GHz linewidth) will provide sufficient resolution for extracting $\delta\langle r^2\rangle$ and μ values from the recorded data, whilst increasing the ionization efficiency by a factor of 2-3. Laser reference scans of ^{198,207,208}Hg will be regularly performed using the ISOLDE Faraday cup or ISOLTRAP's MR- ToF MS.

4 Beam time request

The following shift requests are based on estimates from the statistics recorded during LOI244, our previous experience performing hfs scans with IDS, and our expectations from nuclear structure systematics.
Decay and half-life measurements for $^{209\text{-}211}$Hg will be performed at IDS. To collect enough statistics for a thorough γ-γ coincidence analysis needed to construct decay schemes, we request 1 shift for measuring the decay of ^{209}Hg, 2 shifts for the decay of ^{210}Hg and 5 shifts for measurements of ^{211}Hg. We request 1 additional shift to test the feasibility of studying ^{212}Hg in the future. Therefore, we request **a total of 9 shifts for the decay and half-life measurements**.
We request **a total of 4.5 shifts for the IS and hfs measurements**. This will provide at least 20 counts in the maximum of the recorded hfs spectra, including accounting for the time needed to setup apparatus between each scan. Two scans of each isotope will be made to remove any systematic uncertainty due to the direction of the scan. Regular scans will be made of reference isotopes ^{198,207,208}Hg.
The precision mass measurements of ^{209,210}Hg will be performed at the ISOLTRAP mass-spectrometer. To collect enough statistics at low ion loads (to reduce the observed space-charge effects) we request 2 shifts for ^{209}Hg and 2 shifts for ^{210}Hg. Reference measurements of ^{206}Hg will be made regularly to check the yield constancy. Additionally, 0.5 shift is requested to study the ion beam composition for A=211 and 212. Therefore, we request **a total of 4.5 shifts for the precision nuclear mass spectrometry**.
In addition to the aforementioned dedicated measurements, 3 shifts are requested for: beam tuning from the ISOLDE separator to the experimental setups; optimisation of the laser systems; and for altering the target and ion source conditions for the mass measurements by maximising suppression of isobaric contamination. In summary, **we request a total of 21 shifts of beamtime** to complete the proposed decay, laser and mass measurements of $^{209\text{-}211}$Hg, and perform exploratory measurements for ^{212}Hg.

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1 Details for the Technical Advisory Committee

3.1 General information

Describe the setup which will be used for the measurement. If necessary, copy the list for each setup used.

☒ Permanent ISOLDE setup: *ISOLDE Decay Station, ISOLTRAP MR-ToF-MS, RILIS*

☒ To be used without any modification

3.2 Beam production

- Requested beams:

Isotope	Production yield in focal point of the separator (/μC)	Minimum required rate at experiment (pps)	$t_{1/2}$
$^{209-212}$Hg	0.5-5	0.2	~1 min

- Full reference of yield information: *LOI244 – production yields at the focal point of separator are estimated from the observed thallium to lead decays, as the branching ratios for the mercury isotope decays are unknown. However, we measured cumulative γ rates >0.5 Hz for transitions associated with the mercury to thallium β decay for each isotope, which is enough for the proposed work.*

- Target - ion source combination: *UC_x + quartz line*

- RILIS? *YES*

  ☒ Special requirements: *laser scanning*

- Additional features?

  ☐ Neutron converter:

  ☒ Other: *quartz transfer line*

- Expected contaminants: *Fr isotopes – will be removed by quartz line. Pb-Po isobars, yields will be reduced by optimising quartz line conditions – these isotopes are not an issue for the decay measurements as they have long half-lives.*

- Acceptable level of contaminants: *Not sensitive to stable/long-lived contaminants.*

- Can the experiment accept molecular beams? *No*

- Are there any potential synergies (same element/isotope) with other proposals and LOIs that you are aware of? *None that we are aware of that presently have shifts allocated.*

3.4 Shift breakdown

Summary of requested shifts:

With protons	Requested shifts
Target+quartz line optimisation Decay data taking, $^{209-212}$Hg Data taking, laser scans, $^{207-211}$Hg Data taking, mass measurements, $^{206,209-212}$Hg	1 9 4.5 4.5
Without protons	Requested shifts
Tuning stable beam from GPS to setups	2

3.5 Health, Safety and Environmental aspects

3.5.1 Radiation Protection

- If radioactive sources are required:
  - Purpose? *Energy and efficiency calibration of detectors.*
  - Isotopic composition? *^{137}Cs, ^{241}Am, ^{152}Eu, ^{133}Ba, ^{60}Co*
  - Activity? *<300 kBq, ISO standard calibration sources*
  - Sealed/unsealed? *Sealed*
- For collections:
  - Number of samples? *None*
  - Activity/atoms implanted per sample? *n/a*
  - Post-collection activities? *n/a*