

Letter of Intent to the ISOLDE and N-ToF Experiments Committee (INTC)

**Development of the neutron-rich Hg beams from a UC/ThC target with a low-temperature quartz transfer line and RILIS at ISOLDE**

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**Abstract**

This Letter of Intent requests a test of the production of the neutron-rich Hg isotopes with masses heavier than  $A=208$ , by using a low-temperature quartz transfer line to suppress the strong isobaric Fr contamination, followed by the laser ionization with RILIS and mass separation with ISOLDE. 6 shifts are requested with either a  $UC_x$  or  $ThC_x$  target.

Provided a successful completion of these tests, several dedicated proposals for the nuclear and atomic spectroscopy, mass and beta-delayed neutron measurements will be pursued by the members of the present collaboration.



## 1. Introduction

In the recent years, an extensive successful laser spectroscopy campaign to study the neutron-deficient  $^{177-186}\text{Hg}$  and neutron-rich  $^{202,203,206,207,208}\text{Hg}$  isotopes has been performed by the RILIS-Windmill-ISOLTRAP collaboration [1,2]. It culminated in delineating the end of the region of the drastic shape staggering around  $^{181,183,185}\text{Hg}$  [1], discovered at ISOLDE 50 years ago [3], and identified the kink in charge radii of  $^{206,207,208}\text{Hg}$  ( $N=126,127,128$ ) when crossing the  $N=126$  neutron shell closure [2]. All these studies were performed with the laser-ionized Hg beams produced by the spallation of a molten lead target combined with a VADLIS (Versatile Arc Discharge and Laser Ion Source, an advanced version of RILIS).

At present, several research groups at ISOLDE and other facilities show a strong interest in the studies of the decay properties for the neutron-rich Hg beams. They are important for the astrophysical r-process network calculations [4], in particular in respect of the competition between allowed and first-forbidden  $\beta^-$  decays, as shown in a recent  $^{208}\text{Hg} \rightarrow ^{208}\text{Tl}$  study at ISOLDE [5]. Furthermore, the odd-even staggering and the  $N=126$  kink in the charge radii of the neutron-rich Hg isotopes have profound implications for the single-particle level structure in the region above  $N=126$ , see e.g. [2] and extensive references therein.

However, the neutron-rich Hg nuclei heavier than  $^{208}\text{Hg}$  cannot be accessed with the molten Pb target. While they can be produced with  $\text{UC}_x$  or  $\text{ThC}_x$  targets, the presence of extremely strong surface-ionized Fr isobaric contamination at masses  $A=209-213$  severely hampers the studies of respective isobaric Hg isotopes.

Therefore, with this LoI we propose to develop the beams of heavy Hg isotopes by using a quartz transfer line, which was proven to be able to cope with surface-ionized alkali species in the lighter mass regions, e.g. for the suppression of Rb isobars from neutron-rich Zn isotopes or Cs isobars from neutron-rich Cd beams, see e.g. Refs. [6].

## 2. Motivation for the studies of neutron-rich Hg isotopes and previously known data

The region of the neutron-rich nuclides with  $Z \leq 82$ ,  $N \geq 126$  south-east of the doubly-magic  $^{208}\text{Pb}$  is believed to play an important role for the astrophysical r-process. In particular, the  $A \sim 195$  mass peak in the elemental abundance plot is due to nuclei with  $Z \leq 82$  and  $N=126$ . The knowledge of several basic nuclear properties, such as half-lives, masses, deformation, competition between different decay modes and the probability for beta-delayed neutron emission is required for network calculations for such nuclei, see e.g. the recent review [4] and references therein. Furthermore, this region is of particular interest because first-forbidden  $\beta$  decays can successfully compete with allowed Gamow-Teller and Fermi

decays, thus impacting on the calculations of r-process nucleosynthesis abundances, see e.g. a recent  $\beta$ -decay study of  $^{208}\text{Hg} \rightarrow ^{208}\text{Tl}$  at ISOLDE [5].

However, due to experimental difficulties to reach this region of nuclei, only scarce information is available, as seen in Fig.1, which clearly shows how close the line of the known isotopes is to the doubly-magic  $^{208}\text{Pb}$ . For example, with the exception of  $^{208-211,213}\text{Tl}$  and  $^{207,208}\text{Hg}$ , masses for none of the 31 known nuclides from Os to Tl in this region have been measured.

Charge radii and magnetic moments are only known for  $^{208}\text{Tl}$  [7] and  $^{207,208}\text{Hg}$  [2]. In passing we note that the first measurement of the charge radius of  $^{209}\text{Tl}$  has been done in the LoI219 experiment in June 2022 by our collaboration at ISOLDE Decay Station (IDS).

Pb 208 52.4 $\alpha$ 0.00023 $\rho_{\text{ch}} < \text{BE-6}$	Pb 209 3.234 h $\beta^-$ 0.644 no $\gamma$	Pb 210 22.20 a $\beta^-$ 0.02, 0.06 $\gamma$ 47, 6 $\gamma$ , g $\alpha$ 3.72 $\alpha < 0.5$	Pb 211 36.1 m $\beta^-$ 1.4 $\gamma$ 405, 832 427	Pb 212 10.64 h $\beta^-$ 0.3, 0.6... $\gamma$ 239, 300... g	Pb 213 10.2 m $\beta^-$ 2.0... $\gamma$ 978, 258, 720 1171...	Pb 214 26.8 m $\beta^-$ 0.7, 1.0... $\gamma$ 352, 295 242...	Pb 215 147 s $\beta^-$ 184, 985, 839 1200... e $^-$	Pb 216 >160 ns $\beta^-$ ?	Pb 217 >160 ns $\beta^-$ ?	Pb 218 >160 ns $\beta^-$ ?	Pb 219 >160 ns $\beta^-$ ?	Pb 220 >160 ns $\beta^-$ ?
Tl 207 1.33 s 4.77 m $\beta^-$ 1.4... 1998... 1981	Tl 208 3.053 m $\beta^-$ 1.8, 2.4... $\gamma$ 2615, 583 511, 860, 277	Tl 209 2.162 m $\beta^-$ 1.8... $\gamma$ 1567, 465 117	Tl 210 1.30 m $\beta^-$ 4.2, 4.4... $\gamma$ 800, 296... 117	Tl 211 88 s $\beta^-$ 1190, 597 $\gamma$ 362, 316, 438... 117	Tl 212 96 s $\beta^-$ 804, 315, 157 $\gamma$ 675	Tl 213 46 s $\beta^-$ ?	Tl 214 >160 ns $\beta^-$ ?	Tl 215 >300 ns $\beta^-$ ?	Tl 216 >160 ns $\beta^-$ ?	Tl 217 >160 ns $\beta^-$ ?	138	
Hg 206 8.15 m $\beta^-$ 1.5... $\gamma$ 305, 650... g	Hg 207 2.9 m $\beta^-$ 1.8... $\gamma$ 351, 997 1637... m, g	Hg 208 -42 m $\beta^-$ ?	Hg 209 36 s $\beta^-$ 1424, 324 $\gamma$ 738, 208	Hg 210 >300 ns $\beta^-$ ?	Hg 211 >300 ns $\beta^-$ ?	Hg 212 >300 ns $\beta^-$ ?	Hg 213 >160 ns $\beta^-$ ?	Hg 214 >160 ns $\beta^-$ ?	Hg 215 >300 ns $\beta^-$ ?	Hg 216 >160 ns $\beta^-$ ?	138	
Au 205 31 s $\beta^-$ 1374 627 916 $\gamma$ 379, 487 446, 813	Au 206 >300 ns $\beta^-$ ?	Au 207 >160 ns $\beta^-$ ?	Au 208 >160 ns $\beta^-$ ?	Au 209 >300 ns $\beta^-$ ?	Au 210 >300 ns $\beta^-$ ?	132		134		136		
Pt 204 16 s $\beta^-$ 306, 602 895...	Pt 205 >160 ns $\beta^-$ ?	Pt 206 >300 ns $\beta^-$ ?	Pt 207 >300 ns $\beta^-$ ?	Pt 208 >300 ns $\beta^-$ ?	130							
Ir 203 >300 ns $\beta^-$ ?	128											
Os 202 >300 ns $\beta^-$ ?	Os 203 >300 ns $\beta^-$ ?											

Fig.1 A part of the Chart of Nuclides with  $Z \leq 82$ ,  $N \geq 126$ , with Hg isotopes highlighted in a red rectangle. The lower half-life limits of '>300/160 ns' were only based on the fact of survival of the ions during their time-of-flight from FRS [8-10], the actual half-lives are in the minutes-seconds range (Table 1).

Beta-decay half-lives for some of the nuclei in this region have been measured in the fragmentation studies at relativistic energies in an inverse kinematics  $^{238}\text{U}(1\text{A GeV}) + ^9\text{Be}(2.5\text{-}6 \text{ g/cm}^2)$  reaction with the FRS at GSI [8-10]. In the latter experiments [9,10] also the BEta-deLayEd Neutron (BELEN) detector was used, to deduce the probability for beta-delayed neutron emission. Statistics ranging from a few tens of implanted ions (e.g. for  $^{212,213}\text{Hg}$ ) up to a few hundreds of counts for more abundantly-produced  $^{208-211}\text{Hg}$  was collected, see Fig. 5 of [10]. The delayed neutron probabilities ( $P_n$ ) and half-life values deduced for some of Hg isotopes are shown in Table 1. However, due to absence of the gamma-ray detection at FRS in experiments [8-10], no beta-gamma decay data is known for  $^{210-216}\text{Hg}$ .

It is important to note that contradictory half-life values were reported for  $^{208,209}\text{Hg}$  in the literature. The much longer half-lives, by a factor of  $\sim 20$  and  $\sim 6$  for  $^{208,209}\text{Hg}$ , respectively,

came from the experiments at Lanzhou, in multi-nucleon transfer reactions of  $^{18}\text{O}$  on a thick lead target [11], which are incompatible, even within large uncertainties to shorter values from FRS [10], see comparison in Table 1 below. The half-life of  $^{208}\text{Hg}$  has been recently re-measured at ISOLDE [6], its value of 135(10) s is consistent with the FRS data of 132(50) s, but more precise. The large inconsistency of the half-life of  $^{209}\text{Hg}$  from Lanzhou [11] may also raise questions about the validity of their gamma-ray measurements attributed to beta-gamma decay for  $^{209}\text{Hg}$ .

Table 1. Beta-delayed neutron probabilities,  $P_n$ , and half-lives from the FRS measurements [9,10], partial tables are taken from [10].

TABLE II. $P_n$ results compared with theoretical predictions for the measured isotopes.								
Nuclei	$N$	$P_n(\%)$ (this work)	FRDM + QRPA (%) [67]	DF3 + cQRPA (%) [71]	RHB + RQRPA (%) [70]	KTUY (%) [69]	QRPA-HF (%) [72]	$Q_{\beta n}(\text{keV})$ [55,56] (extr. = extrapolated)
$^{208}\text{Hg}$	128		0.0	3.2	0.3	0.0		$-303.32 \pm 31.23$
$^{209}\text{Hg}$	129		0.0	2.8	0.5	0.0		$34 \pm 149$ (extr.)
$^{210}\text{Hg}$	130	$2.2 \pm 2.2$	0.0	9.3	0.6	0.0	71	$201 \pm 196$ (extr.)
$^{211}\text{Hg}$	131	$6.3 \pm 6.3$	0.81	7.5	0.8	0.0	11	$551 \pm 196$ (extr.)

TABLE III. Half-lives ( $T_{1/2}$ ) results, and some of the previous experimental data and theoretical predictions.						
Nuclei	$N$	Implanted ions	$T_{1/2}^{\text{expt}}$ (s) (this work)	Previous $T_{1/2}^{\text{expt}}$ (s)	FRDM + QRPA (s) [67]	DF3 + cQRPA (s) [68]
$^{208}\text{Hg}$	128	220	$132.2 \pm 50.0$	$2460_{-240}^{+300}$ [77]	168.9	12.1
$^{209}\text{Hg}$	129	583	$6.3 \pm 1.1$	$35.0_{-6}^{+9}$ [77]	33.6	3.7
$^{210}\text{Hg}$	130	512	$63.7 \pm 11.6$	$>300$ ns	41.2	5.2
$^{211}\text{Hg}$	131	253	$26.4 \pm 8.1$	$>300$ ns	14.9	1.9

### 3. Proposed program for the neutron-rich Hg isotopes at ISOLDE

ISOLDE is one of very few facilities world-wide, where the beams of the neutron-rich Au-Tl isotopes can be obtained. At ISOLDE, the neutron-deficient Hg beams are usually produced from a molten lead target, up to the heaviest  $^{208}\text{Hg}$ , see the discussion in [2], on the possible mechanisms, which involve the secondary particles for  $^{208}\text{Hg}$ . However, such a lead target cannot be used to produce even heavier Hg isotopes, the only available option being the use of a  $\text{UC}_x$  or  $\text{ThC}_x$  target.

It is important to mention here that the above-mentioned production of e.g.  $^{209-216}\text{Hg}$  isotopes at GSI showed that fragmentation/spallation reactions at these energies result in reasonable cross sections [8-10]. As ISOLDE uses a somewhat similar reaction, but in normal kinematics, and with 1.4 GeV protons, comparable production cross-sections can be expected also at ISOLDE.

However, experimental yields for the neutron-rich Hg isotopes with the  $\text{UC}_x/\text{ThC}_x$  targets are not yet known. The main reasons for this is that in the mass region of  $^{209-213}\text{Hg}$ , **very strong surface-ionized Fr isobaric contamination is present** (up to  $\sim 10^9$  ions/ $\mu\text{C}$ , [http://isoyields-classic.web.cern.ch/francium\\_isotopes.html](http://isoyields-classic.web.cern.ch/francium_isotopes.html)), being many orders of magnitude larger than the expected yields of the Hg isotopes of interest.

As the quartz transfer line is known to be able to suppress alkali elements [6], with this LoI we request a test of this method for the neutron-rich Hg isotopes in the presence of the strongly-produced isobaric Fr contamination.

So far, a number of physics groups showed strong interest in the development and the use of the neutron-rich Hg beams, thus their availability at ISOLDE will allow the following studies (in no particular order):

- a) Extending the HFS, IS and charge radii measurements to the most neutron-rich Hg isotopes which can be accessible beyond the heaviest measured by the laser spectroscopy -  $^{208}\text{Hg}$  [2]. Recently, the Windmill-ISOLTRAP collaboration performed a series of HFS and IS measurements for the long chain of  $^{177-186,202,203,206,207,208}\text{Hg}$  isotopes, see Fig. 2. The two main results were: a) the confirmation of the end of the shape staggering earlier observed in  $^{181,183,185}\text{Hg}$ , with the  $^{181}\text{Hg}$  approaching the spherical trend [1], and b) the observation of the kink in charge radii at  $N=126$  [2]. Both phenomena were successfully described by the state-of-the-art theoretical calculations, described in [1,2]. Provided the successful completion of this LoI, such measurements can be extended to even heavier Hg isotopes.

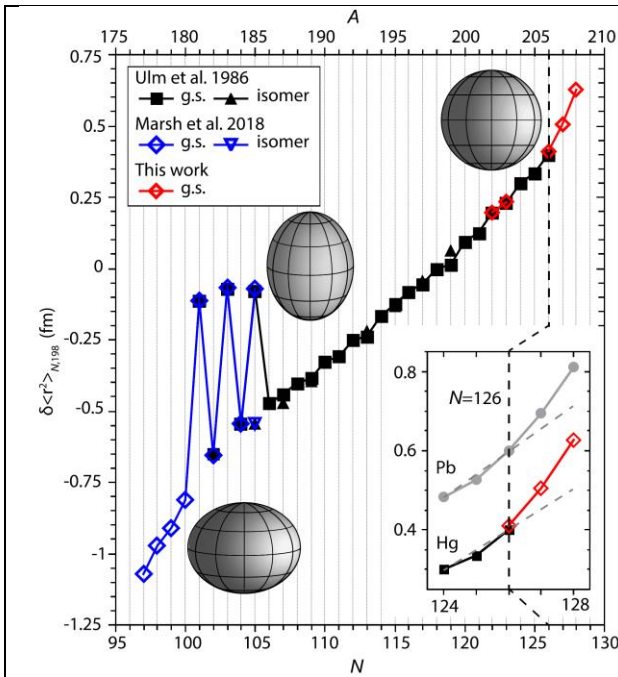


Figure 2 (taken from [2]). Systematics of the difference in mercury ground state (g.s.) and isomer mean-square charge radii. Blue and red symbols are the recent data from the Windmill-ISOLTRAP collaboration measurements: the blue points for  $^{177-186}\text{Hg}$  are from Nature Physics [1] and red points for  $^{202,203,206,207,208}\text{Hg}$  are from Phys. Rev. Lett. [2]. The inset highlights the kink at  $N=126$  and the neighboring odd-even staggering in both the mercury and lead isotopic chains. The lead isotopes are arbitrarily displaced from those of mercury for clarity. The sloped dashed lines through  $N=124$  and  $126$  in the inset are added to highlight the kinks. Statistical uncertainties are smaller than the data points.

- b) None of the mercury isotopes heavier than  $^{208}\text{Hg}$  have measured masses, they will be accessed in the follow-up experiments by the ISOLTRAP/MR-ToF MS colleagues. The measurements of the masses for the even-even isotopes in this region (including for Hg's) will allow the determination of the strength of the p-n interaction in nuclei close to  $^{208}\text{Pb}$ , an extensive discussion of the importance of this phenomenon was given by the 1<sup>st</sup> measurement of the mass of  $^{208}\text{Hg}$  at ESR in GSI, see e.g. Fig. 3 of [12].

- c) The half-lives of the heavy Hg isotopes are only known up to  $^{211}\text{Hg}$ , see Table 1. The GSI experiments [8] identified  $^{212-216}\text{Hg}$ , but no decay data were published. The detailed beta-decay measurements of the parent Hg isotopes at IDS will allow to study the structure and lifetimes (via dedicated fast-timing experiments, which are also planned within the follow-up program) of the excited states in respective Tl ( $Z=81$ ) daughter nuclides, for which only very limited information is known only up  $^{210}\text{Tl}$  (from the alpha-decay studies of  $^{214}\text{Bi}$ - $^{210}\text{Tl}$ ). These data will form an excellent basis for testing large scale shell model calculations in this region. Furthermore, first-forbidden beta decays are expected to dominate in these decays, they provide crucial information on the nuclear structure in this region, as shown in [5].
- d) The availability of the neutron detector at IDS will also allow the measurements of the beta-delayed neutrons probabilities for these nuclei ( $P_n$  values for only  $^{210,211}\text{Hg}$  are known so far, see Table 1.)

#### 4. Low-temperature quartz transfer line, RILIS and detection system

Previously, quartz transfer lines have been successfully employed at ISOLDE for suppression of Rb and Cs elements [6]. Similar to the Cd/Cs case, a low temperature quartz transfer line will be exploited in the present LoI. Judging from the known adsorption enthalpies on quartz (see Fig. 3 in [13]) the gain in Hg/Fr ratio should be comparable to or higher than for Cd/Cs.

However, the applicability of the method for strongly-produced alpha-decaying  $^{209-212}\text{Fr}$  contaminants has to be thoroughly investigated, before planning the full-fledged proposals/experiments. These isotopes have relatively long-half lives, which cannot be suppressed by the pulsed release method with the usage of delayed beam gate [14].

A standard 3-step RILIS laser ionization scheme used in [1,2] will be applied for ionization of Hg atoms in the hot metal cavity attached to the temperature-controlled quartz transfer line.

For the detection and identification of Hg isotopes the Isolde Decay Station (IDS) [15] will be used, equipped with a set of silicon and beta detectors, surrounded by an array of Clover Ge detectors. The nuclei will be implanted in a tape, which will allow to remove the longer-lived daughter products, and also to alleviate the issues due to the remaining Fr contamination, if it is not fully suppressed by the quartz line. For single-ion counting the use of Magne-ToF detector can also be considered.

#### 5. Beam time request and specific measurements within this LoI

We request **6 shifts** to perform the yield and beam purity measurements for  $^{209-216}\text{Hg}$ . Specific measurements will include the estimation of the Fr suppression as a function of the transfer line temperature, its dependence on the in-target production rate (proton beam

intensity), on the specific decay mode of the strongly-contaminating Fr isotopes (alpha/beta) and the lifetimes and chemical nature of possible precursors. We will also investigate other possible background contaminants, such as molecules with the same mass as the Hg isotopes of interest, which could jeopardize the mass measurements with the MR-ToF MS in the future dedicated experiments.

We will also determine the yields of several lighter Hg isotopes (e.g.  $^{180-185,208}\text{Hg}$ ) with this configuration, to compare them with those known from the measurements with the molten Pb target and with standard  $\text{UC}_x$  targets (see yield systematics in Fig.2 of S.Sels et al., [2]).

It is worth mentioning that the region of neutron-rich Hg isotopes is likewise interesting for the recently launched study on production yields using 1.7-GeV protons [16]. While it is not intended to include corresponding measurements in the LoI's program, we would like to point out the good synergies between the two campaigns, facilitating beamtime scheduling at ISOLDE.

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