

# EUROPEAN ORGANIZATION FOR NUCLEAR RESEARCH

Proposal to the ISOLDE and Neutron Time-of-Flight Committee  
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## SEARCH FOR NEW CANDIDATES FOR THE NEUTRINO-ORIENTED MASS DETERMINATION BY ELECTRON-CAPTURE

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### Abstract

This proposal is part of an extended program dedicated to the neutrino-mass determination in the electron-capture sector, which aims at ultra-precise mass measurements by Penning traps in combination with cryogenic micro-calorimetry for atomic de-excitation measurements. Here, precise mass measurements with ISOLTRAP are proposed for the orbital electron-capture nuclides  $^{194}\text{Hg}$  and  $^{202}\text{Pb}$ , as well as their daughters, with the goal to determine accurately their Q-values. These values are expected to be the smallest ones among a great variety of known electron-capture precursors. Therefore, these nuclides are strong candidates for an improved electron-neutrino mass determination. We ask for 8 shifts of on-line beam at ISOLDE for mass measurements of  $^{194}\text{Hg}$ ,  $^{194}\text{Au}$ ,  $^{202}\text{Pb}$ , and  $^{202}\text{Tl}$  at ISOLTRAP.



## Introduction and motivation:

Among the large variety of experiments exploring the neutrino properties, the absolute mass determination of the neutrino is a very ambitious and challenging venture that is of great importance for physics and cosmology. Over the last decades many attempts have been undertaken to find a neutrino mass different from zero. For the electron antineutrino, the lowest upper limit of 2 eV was obtained over this long period of time in the tritium decay experiments [1]. In contrast, the achievements in the neutrino mass measurements are stuck with results from the inner bremsstrahlung spectra of  $^{163}\text{Ho}$  and give a less stringent limit of 225 eV [2]. This very large difference in the neutrino-antineutrino mass limits shows the need to improve this “asymmetry” with a dramatic increase in precision for the neutrino mass in the electron-capture sector.

One of the first attempts to determine the neutrino mass by orbital electron capture in  $^{163}\text{Ho}$  was made at ISOLDE at the beginning of the 1980s [3]. The upper limit of 1300 eV was determined by observing the M-shell X-rays and Auger electrons. At that time the technical possibilities did not provide the required precision. However, the progress made over the last years in both Penning trap mass spectrometry [4] and atomic de-excitation micro-calorimetry [5], allows regenerating attempts for the neutrino mass determination at a new, considerably increased level of precision. In fact, the precision for masses (and thus decay energies) can reach 1 eV with the use of Penning traps [6]. Similarly, electron binding energies can be determined with a precision of 1 eV using micro-calorimeters. They are able to detect the energy release from all atomic de-excitations (except for that from the neutrino) that occur after electron capture, which is accompanied by the neutrino emission. Thus, the combined use of both methods allows the precise determination of the total neutrino energy and the analysis of the calorimetric spectra will provide the neutrino rest mass [7].

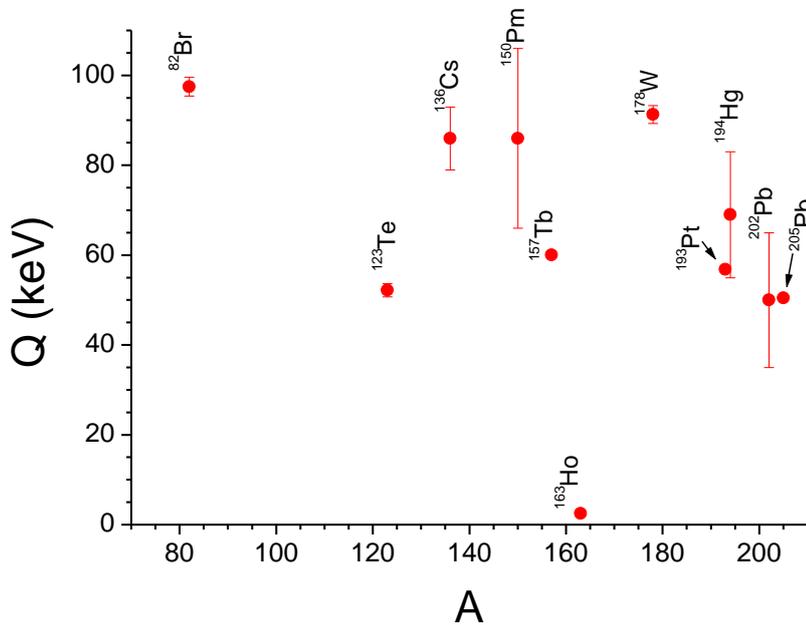


Fig. 1: Nuclides with the smallest electron capture Q-values.

The best candidate for such kind of measurements is a nuclide with the smallest decay energy in electron capture. This provides a prerequisite for a small total neutrino energy and subsequently leads to a high relative contribution of the neutrino rest mass in the total neutrino energy. The nuclides with Q-values less than 100 keV are shown in Fig. 1, where the Q-value is plotted as a function of the mass number. The electron capture with a Q-value of

about 2.5 keV in the case of  $^{163}\text{Ho}$  is an exceptional case. The expected total neutrino energy for M1-capture is at the level of about 500 eV. The possible level of the neutrino-mass value and the accuracy of measurements depend strongly on the magnitude of the Q-value and its precision. It is planned to perform ultra-precise measurements for  $^{163}\text{Ho}$  at the future Penning trap installations HITRAP at GSI [8] and MATS at FAIR [9]. Nevertheless, the Q-value for  $^{163}\text{Ho}$  might not be in a favorable range for an upper limit of the neutrino mass at the desired few-eV level of accuracy. Therefore, the question arises whether other nuclides exist, which are appropriate candidates for capture measurements.

Our proposal is dedicated to this problem: The ISOLTRAP mass spectrometer will be used to search for alternatives or additional candidates to  $^{163}\text{Ho}$  in the neutrino-oriented precision mass measurements.

### Physics case:

Besides  $^{163}\text{Ho}$ , there are ten nuclides (see Fig. 1) with a Q-value for the ground-ground state transition of less than 100 keV, which is the range of possible electron binding energy values in nuclides up to K-electrons in lead with  $B_K = 88$  keV. In order to be a candidate for the neutrino mass determination, a nuclide should have a value  $Q - B_i$  as small as possible, where  $B_i$  is the electron-binding energy for the  $i$ -th electron orbit of the atom. The analysis of decay information on radionuclides known so far (see [10] and [11]) shows that only  $^{194}\text{Hg}$  and  $^{202}\text{Pb}$  can be considered as possible nuclides for the neutrino-oriented precision mass measurements.

The Q-values taken from the mass-evaluation data of [11] are too imprecise for our goals (see error bars in Fig. 1). In addition, they are based mainly on the obsolete data for  $^{202}\text{Pb}$  [12,13] and data for  $^{194}\text{Hg}$  [14,15] and daughter nuclides [11]. The masses of these nuclides (mother and daughter) should be measured by Penning trap mass spectrometry. This will allow an improvement of the relevant Q-values with the required precision. Basic information about the nuclides  $^{194}\text{Hg}$ ,  $^{202}\text{Pb}$ , and their daughters is presented in Table 1. As an example, the decay scheme of  $^{194}\text{Hg}$  is shown in Fig. 2.

**Table 1:** Candidates and daughter nuclides for the neutrino-oriented precision mass spectrometry. Mass excess values and Q-values taken from [11].

Nucleus	$T_{1/2}$	Mass excess (keV)	$\delta m/m$	Q (keV)	i	$B_i$ (keV)	$Q_v$ (keV)
$^{194}\text{Hg}$	440(80) y	-32193(13)	$6.7 \times 10^{-8}$	69(14)	K	80.72	-12(14)
$^{194}\text{Au}$	38.02(10) h	-32262(10)	$5.7 \times 10^{-8}$				
$^{202}\text{Pb}$	$52.5(2.8) \times 10^3$ y	-25934(8)	$4.5 \times 10^{-8}$	50(15)	L	15.35	35(15)
$^{202}\text{Tl}$	12.23(2) d	-25983(15)	$5.4 \times 10^{-8}$				

In order to decide if  $^{194}\text{Hg}$  can be considered as a possible candidate for the neutrino-mass determination, we do not need a very precise mass measurement (see Table 1). Taking the neutrino total energy of  $-12 \pm 14$  keV, an improvement of the precision by a factor of 3 (up to 5 keV) will be enough to conclude whether subsequent ultra-precise mass measurements in future Penning trap experiments should follow. For the accurate determination of the Q-value for  $^{194}\text{Hg}$  the precise mass measurement on  $^{194}\text{Au}$  is also needed.

It is worthwhile to note that if the  $Q$ -value obtained does not match to the  $K$ -electron capture channel and energetically  $K$ -capture is forbidden, then in dependence of the  $Q$ -value the  $L$ -capture channel could be useful (see Fig. 2), especially taking into account the transition to the nuclear excited state at 35 keV in  $^{194}\text{Au}$ , though it is expected to be weak (less than 1%).

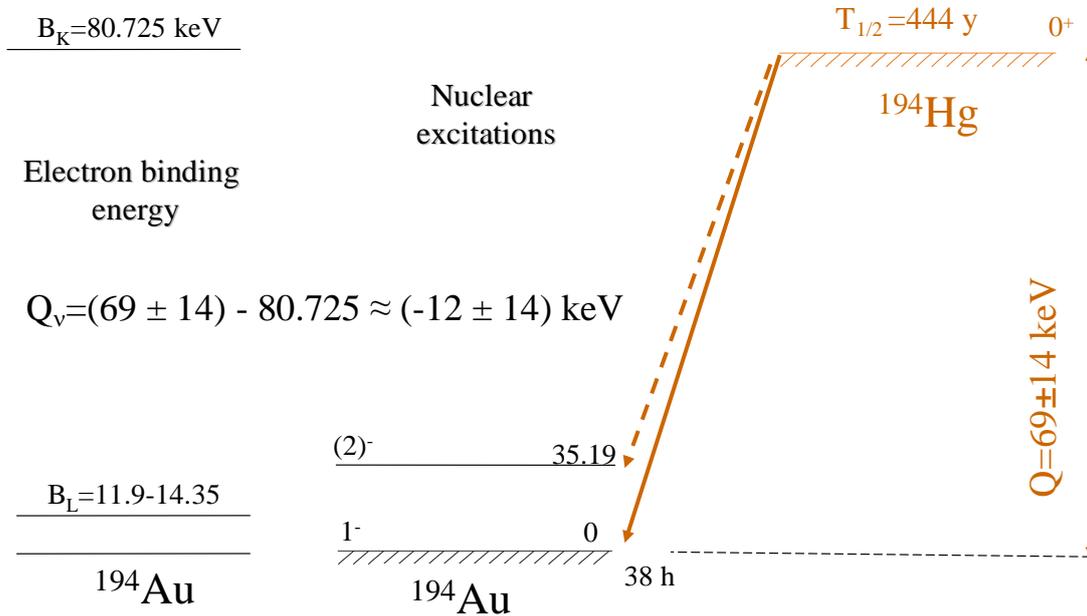


Fig. 2: Decay scheme of  $^{194}\text{Hg}$  and the nuclear and atomic energies of the daughter nuclide  $^{194}\text{Au}$ . All energies are given in keV.  $Q_\nu$  is the total neutrino energy for  $K$  capture (see also Table 1).

### Experimental setup:

The required mass measurements need to be performed with an uncertainty of a few keV, which corresponds to a relative uncertainty of the order of  $\delta m/m = 10^{-8}$  in the heavy mass region around  $A=200$  for an individual nuclide mass. It can and has been provided so far only by Penning trap mass spectrometers like ISOLTRAP at ISOLDE/CERN [16].

The ISOLTRAP experiment makes use of three ion traps for the preparation, purification, and mass determination of radioactive ions delivered by ISOLDE. The 60-keV ISOLDE continuous ion beam is first stopped, cooled, and bunched in a linear radio-frequency quadrupole (RFQ). The ion bunch is then transferred to the first, cylindrically shaped Penning trap for the removal of isobaric contaminants (mass resolution of up to  $m/\Delta m = 10^5$ ) that are still present after mass selection with the ISOLDE separator magnets. The isobarically cleaned ion bunch is finally injected into the hyperbolic precision Penning trap, where possible isomers can be removed by application of a resonant dipolar radiofrequency excitation.

The mass measurement principle is based on the very precise determination of the cyclotron frequency  $\nu_c = qB/(2\pi m)$  of ions with mass  $m$  and charge  $q$  that are stored in a strong and homogeneous magnetic field  $B$  [4]. With the time-of-flight cyclotron-resonance technique the gain of radial energy in the Penning trap after a quadrupolar rf excitation is probed by monitoring the change of the time of flight towards an external detector after axial ejection of the ions from the trap. The magnetic field strength is determined by measuring the cyclotron frequency of a reference ion with well-known mass. The mass of the ion of interest is finally deduced from the ratio of the two cyclotron frequencies.

The detection efficiency was recently increased by a factor of three to 90% by implementing a new ion detector that uses a Channeltron with a conversion dynode [17]. With this new detector setup almost true single ion counting is possible, which gives better control of ion contamination and beam times can be used more efficiently. The precision of the cyclotron frequency determination was also improved by a factor of about three, using time-separated oscillatory fields for the quadrupolar rf excitation in the precision trap (so-called Ramsey technique) [18]. The new method has been thoroughly tested for stable nuclides and was successfully applied in two on-line runs in 2006, when the masses of  $^{38,39}\text{Ca}$  and  $^{26,27}\text{Al}$  were measured with statistical uncertainties as low as  $2 \times 10^{-9}$  within a few hours.

For the four radionuclides that are considered in the present proposal further detailed information on the production yield is given below. All nuclides have lifetimes, that allow long excitation durations of the order of seconds for the cyclotron frequency determination. With the Ramsey technique it is planned to reach the present uncertainty limit of ISOLTRAP of  $\delta m/m = 8 \times 10^{-9}$  [19], which will give for all four cases an absolute mass uncertainty of 1.5 keV, i.e. enough to conclude if  $^{194}\text{Hg}$  and  $^{202}\text{Pb}$  are suitable candidates for the investigation of the neutrino mass. It is also planned to use the carbon cluster ion source to have an absolute mass reference.

#### **$^{194}\text{Hg}$ :**

According to the ISOLDE yield data base the yield as measured at the SC (0.6-GeV protons) is  $7.1 \times 10^9$  ions/ $\mu\text{C}$  for a molten Pb target and a hot plasma ion source. A contamination from Fr or Ra isotopes do not pose a problem, since these nuclides do not exist for the mass  $A=194$ . Possible contaminating nuclides might be  $^{194}\text{Bi}$ ,  $^{194}\text{Tl}$ , and/or  $^{194}\text{Pb}$ . However, the maximum yields of the order of  $5 \times 10^7$  ions/ $\mu\text{C}$  were obtained for all three nuclides for a UC<sub>x</sub> target with a RILIS ion source and protons from the PSB. A RILIS scheme for Hg is available and thus a more selective ionization process is possible.

#### **$^{194}\text{Au}$ :**

Radioactive Au beams have so far been obtained using Hg beams as precursor, producing Au nuclides after beta decay on a graphite substrate [20]. As in the case of Hg, a new RILIS ionization scheme has recently been developed for Au [21] with an efficiency of 3%. Another experiment proposal (P212), aiming at spectroscopy of  $^{201,203,205}\text{Au}$  [22], has estimated the production yields from a UC<sub>x</sub> target using the Silberberg-Tsao cross-section database. For  $^{201}\text{Au}$ ,  $^{203}\text{Au}$ , and  $^{205}\text{Au}$  a yield estimate of  $1 \times 10^6$ ,  $5 \times 10^5$ , and  $2 \times 10^5$  ions/ $\mu\text{C}$  has been calculated, respectively, with a prediction of a larger in-target production for the more neutron-deficient isotopes. The slow release is no problem for the long lived  $^{194}\text{Au}$  nuclide. Concerning possible contamination of the beam, the same arguments apply as in the case of  $^{194}\text{Hg}$ .

#### **$^{202}\text{Pb}$ :**

From a UC<sub>x</sub> target using a RILIS ion source, a yield of  $4.6 \times 10^6$  ions/ $\mu\text{C}$  is given in the ISOLDE yield database.  $^{202}\text{Ra}$  and  $^{202}\text{Fr}$  cannot contaminate the beam.  $^{202}\text{Bi}$  might be present in the beam, however, with the possibility to be cleaned in the preparation Penning trap of ISOLTRAP.

#### **$^{202}\text{Tl}$ :**

No yield is given in the ISOLDE data base. However, from the data shown for a UC<sub>x</sub> target using a RILIS ion source, a yield of  $1 \times 10^5$  ions/ $\mu\text{C}$  or more can be estimated. Possible contamination is the same as for  $^{202}\text{Pb}$ .

## Beam time request:

We ask for 8 shifts of on-line beam at ISOLDE. Although the Ramsey scheme will be applied for fast measurements, the longer excitation duration of the order of seconds and sufficient statistics to reach the goal of less than 2 keV for the uncertainty of the Q-value of  $^{194}\text{Hg}$  and  $^{202}\text{Pb}$  require more beam time for each mass. The measurements for  $^{202}\text{Pb}$  and  $^{202}\text{Tl}$  can be possibly combined in one run with a  $\text{UC}_x$  target and using RILIS for Pb and surface ionization for Tl. An overview of the proposed target/ion-source combinations is listed in Table 2.

Table 2: Overview of radionuclides, for which shifts are requested in this proposal.

Nuclide	No. of shifts	Target	ion source
$^{194}\text{Hg}$	2	Pb/ $\text{UC}_x$	RILIS/HP
$^{194}\text{Au}$	2	$\text{UC}_x$	RILIS
$^{202}\text{Pb}$	2	$\text{UC}_x$	RILIS
$^{202}\text{Tl}$	2	$\text{UC}_x$	RILIS/W-SI

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