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Direct mass measurements of ¹⁹⁴Hg and ¹⁹⁴Au: A new route to the neutrino mass determination?

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1. Introduction

ABSTRACT

The study of nuclear electron capture (EC) offers an exciting alternative for the determination of the neutrino mass. Whereas only tritium and ¹⁸⁷Re can be used in the case of β -decay experiments involving the anti-neutrino, a potentially large number of EC-nuclides can be used in experiments involving the monochromatic neutrino. This alternative to β -decay experiments requires an accurate measurement of Q_{EC} -values of appropriate candidates. In the present work we initiate a search for such a candidate and determined the Q_{EC} -value of the electron capture in ¹⁹⁴Hg by direct mass measurements of ¹⁹⁴Hg and ¹⁹⁴Au. The new Q_{EC} -value of 29(4) keV determined by the ISOLTRAP Penning-trap mass spectrometer at ISOLDE/CERN forbids the K-capture for ¹⁹⁴Hg. However, it allows a determination of the neutrino mass by a combination of a micro-calorimetric measurement of the de-excitation spectrum from *L*-capture in ¹⁹⁴Hg and a comparable Q_{EC} -value remeasurement by high-precision Penning trap mass spectrometery.

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The absolute mass of the neutrino is of fundamental importance for physics and cosmology. However, its determination is a very ambitious and challenging venture. Over the last decades many attempts have been undertaken to measure this elusively tiny quantity. For the antineutrino $\bar{\nu}_e$ an upper limit of $m(\bar{\nu}_e) < 2$ eV was obtained by tritium β -decay experiments [1]. In contrast, for the neutrino ν_e a less stringent limit of only $m(\nu_e) < 225$ eV was obtained from measurements of internal bremsstrahlung, which accompanies capture of orbital electrons (EC) by a ¹⁶³Ho-nucleus [2].

The idea of a determination of the neutrino mass by EC was put forward a long time ago. The first investigation was performed in the 1980s [3]: An upper limit of 1300 eV was found by observing the M-shell X-rays and Auger electrons for the EC in ¹⁶³Ho. At the same time A. de Rújula [4] came up with the idea to use calorimeters for a determination of the neutrino mass. Calorimeters can detect the total energy release of EC from electrons and photons (i.e. both non-radiative and radiative processes) but neutrino. Thus, such a calorimetric spectrum is a measure of the neutrino energy spectrum due to two body nature of EC. The endpoint of the calorimetric spectrum $E_{max} = Q_{EC} - m(v_e)c^2$ is independent of the capture states and their sum. As was pointed out in [4] the shape of this spectrum is not affected by natural widths of intermediate atomic states. Thus, a determination of the endpoint via the Curie-plot along with an independent measurement of the Q_{FC} value allows a determination of the neutrino mass. Until recently experimental means did not keep pace with requirements for precise measurements and to date the neutrino mass value from EC is stuck at the level of 225 eV. Thus, a dramatic improvement on the

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nuclide

Fig. 1. Total energies of the electron-neutrino Q_{ν} emitted in a capture of the K-shell electron (filled squares), of the L₁-shell electron (circles) and of the M₁-shell electron (empty square) as expected for nuclides with $Q_{EC} \leq 100$ keV. The uncertainties of most of the Q_{EC} -values [12] are smaller than the size of the symbols.

side of the neutrino is required if a test of CPT symmetry in the neutrino sector is considered.

The progress over the last years in both Penning trap mass spectrometry [6] and cryogenic micro-calorimetry [7,8] enables now a neutrino mass determination at a considerably increased level of accuracy [9]. Advanced Penning traps will provide an accuracy of 1 eV for the Q-value of EC in ¹⁶³Ho [10] and, similarly, cryogenic micro-calorimeters are able to determine electron binding energies B_i to a precision of about 1 eV [8]. These devices are able to detect the energy release from all atomic de-excitations (except via the neutrino) that occur after electron capture. Thus, a combined use of both methods allows a precise determination of the total neutrino energy and an analysis of the endpoint of the calorimetric spectra will provide an accurate value for the neutrino rest mass.

The best candidates for such measurements are nuclides with small Q_{FC}-values for which the EC results in an emission of a low-energy neutrino. This property is connected to a large relative contribution of the neutrino mass to the total neutrino energy and, thus, high sensitivity to the neutrino mass. To date, the best conditions are found in ¹⁶³Ho with a decay energy of $Q_{EC} = 2.56(2)$ keV [11] and with an expected total neutrino energy $Q_{\nu} \approx 500$ eV for the electron capture from the M₁-atomic shell. However, there is a variety of other potential candidates with Q_{EC}-values below 100 keV and with expected very small total energy of the emitted neutrino. Here, electron capture to the nuclear ground states as well as to excited nuclear states of the daughter nucleus can be considered. K- and L-shell electron-capture cases are presented in Fig. 1 together with EC of the M_1 -shell electron case in 163 Ho, where the EC from K- and L-shells is energetically forbidden. For some of the nuclides their masses have too large uncertainties. In order to find the most suitable candidate, accurate mass measurements of these nuclides have to be performed.

2. Experimental procedure

In this Letter we report on the Q_{EC} -value determination for the candidate ¹⁹⁴Hg. The choice of ¹⁹⁴Hg is based on several criteria: First, the half-life of 440(80) years is long enough to perform year-long micro-calorimetric experiments. Second, a source with sufficient strength can be produced, e.g., at ISOLDE/CERN. Third, the present Q_{EC} -value [12] based on [13–18] is not accurate enough to be useful for the purpose considered (see Fig. 1).

The Q_{EC} -value of ¹⁹⁴Hg was determined by direct measurements of atomic masses of ¹⁹⁴Hg and its daughter nuclide ¹⁹⁴Au.

Table 1

Properties of ¹⁹⁴Hg known prior to the present study and relevant to a subsequent determination of the neutrino mass: half-life $T_{1/2}$, Q_{EC} -value from [12] based on [13–18], binding energies of the K- and L₁-shell electrons B_i in ¹⁹⁴Au from [19], and the derived total energy of the emitted neutrino.

$T_{1/2}$	Q _{EC} /keV	i	B_i/keV	Q_{ν}/keV
440(80) yr	69(14)	K L ₁	80.72 14.35	-12(14) 55(14)

In Table 1 properties of ¹⁹⁴Hg relevant to this measurement are given. The total energy of the emitted neutrino Q_{ν} (see last column) is given by $Q_{\nu} = Q_{EC} - B_i$, where B_i is the binding energy of the captured electron. In this expression the recoil energy of the nucleus is neglected.

The present experiments were carried out at the ISOLTRAP Penning trap mass spectrometer at ISOLDE/CERN. The masses of ¹⁹⁴Hg and ¹⁹⁴Au were measured in separate runs within a month in between. The radioactive ion-beam production at the ISOLDE facility [20] and mass measurements with ISOLTRAP including data analysis have been described in detail elsewhere [21,22]. Here, only a brief summary of the experimental procedure is given. The ¹⁹⁴Hg and ¹⁹⁴Au were produced by 1.4-GeV protons impinging on thick uranium carbide targets coupled to the general purpose mass separator. The reaction products were ionized either in a hot plasma ion source (194Hg) or in the laser ion source RILIS (¹⁹⁴Au) as described in [23]. Mass-selected nuclides were delivered as a 30-keV continuous ion beam to the ISOLTRAP setup. There, the ions were stopped, cooled, and bunched in a linear radio-frequency quadrupole (RFQ). The ion bunch was then transferred to the first, cylindrical Penning trap for cooling and isobaric cleaning. The isobarically-cleaned bunch of singly-charged ¹⁹⁴Hg⁺ or ¹⁹⁴Au⁺ ions was then injected into a hyperbolic precision Penning trap. Here, a precise measurement of the cyclotron frequency $f_c = qB/(2\pi m)$ of ions with mass m and charge q, stored in a strong and homogeneous magnetic field *B*, is performed with a time-of-flight ion cyclotron-resonance detection technique [24]. To this end the magnetron motion is converted to the modified cvclotron motion by application of a quadrupolar radio-frequency field for a certain excitation time with a subsequent ejection of the ion towards a particle detector outside the magnetic field. The time-of-flight of the ion between the trap and detector is observed as a function of the excitation frequency (see the insets in Fig. 2). The minimum of the time-of-flight corresponds to the cyclotron frequency f_c of the ion. The magnetic field strength is calibrated by measuring the cyclotron frequency of a reference ion with wellknown mass. Thus, the mass of the ion of interest is deduced from the ratio of the two cyclotron frequencies.

For ¹⁹⁴Hg⁺ resonances were taken with excitation times of 6 s and 10 s, whereas for ¹⁹⁴Au⁺ all resonances were taken with excitation time of 10 s. Six cyclotron-frequency measurements for ¹⁹⁴Au⁺ were performed, the cyclotron frequency of ¹⁹⁴Hg⁺ was measured four times. For the calibration of the magnetic field strength the nuclides ¹⁹⁷Au⁺ and ¹³³Cs⁺ were used, respectively. The mass of both reference nuclides is known to an uncertainty better than 600 eV [12], i.e. sufficiently good for the present measurements. The presence of contaminant ions that may have been formed in the traps, e.g. by charge exchange, was accounted for by a count-rate class analysis of the cyclotron frequency as described in [22].

3. Results and discussion

In Fig. 2 the measured cyclotron frequency ratios f_c^{ref}/f_c are shown for ¹⁹⁴Au⁺ vs. ¹⁹⁷Au⁺, and for ¹⁹⁴Hg⁺ vs. ¹³³Cs⁺, $f_c(^{197}\text{Au}^+)/f_c(^{194}\text{Au}^+)$ and $f_c(^{133}\text{Cs}^+)/f_c(^{194}\text{Hg}^+)$. Table 2 lists

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