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Resonance ionization of holmium for ion implantation in microcalorimeters

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ABSTRACT

The determination of the electron neutrino mass by calorimetric measurement of the ^{163}Ho electron capture spectrum requires ultra-pure samples. Several collaborations, like ECHo or HOLMES, intend to employ microcalorimeters into which ^{163}Ho is implanted as an ion beam. This makes a selective and additionally very efficient ion source for holmium mandatory. For this purpose, laser resonance ionization of stable holmium ^{165}Ho was studied, using a three step excitation scheme driven by pulsed Ti:sapphire lasers. Five measurements with sample sizes of 10^{14} and 10^{15} atoms were performed for the efficiency investigation. In average, an excellent ionization efficiency of 32(5) % could be shown, demonstrating the suitability for ion beam implantation.

1. Introduction

Resonance ionization is an efficient and exceptionally selective ionization method. It is thus especially well suited for the production of radioactive ion beams, where both qualities are of major relevance. In several on-line facilities, for example at CERN-ISOLDE [1] or TRIUMF-ISAC [2], resonance ionization laser ion sources are used in conjunction with accelerator based production of exotic nuclides and are regularly requested by experimentalists. In addition, radioactive beams of long-lived isotopes can also be produced in off-line facilities from an external sample. An isotope of increasing interest is ^{163}Ho which decays with a half-life of 4570 a through electron capture. Its very low decay energy of $2833(30)_{stat}(15)_{sys}$ eV [3] makes it the prime candidate for calorimetric measurements of the electron neutrino rest mass [4]. Several collaborations investigating this decay have been started, including ECHo [5] and HOLMES [6]. To facilitate the desired high precision and low background measurements, highly pure ^{163}Ho is to be implanted into the calorimeters. Direct production in an on-line facility would introduce too many contaminants in the implanting ion beam. Therefore, a more intricate pathway has to be taken. In the case of ECHo, ^{163}Ho is produced in the high-flux nuclear reactor at ILL, Grenoble [7] by irradiating ^{162}Er. Following neutron capture to ^{163}Er, it decays with a half-life of 75 min by electron capture to ^{163}Ho. After irradiation, co-produced contaminants and the erbium target material are chemically separated. From this sample, a holmium ion beam is extracted by resonance ionization, mass separated to isolate ^{163}Ho and subsequently implanted into the microcalorimeters. High efficiencies in all process steps are needed to keep the required total sample amount reasonably low. Ion beam implantation offers the ability to precisely monitor the ion current during the process, allowing accurate quantification of isotope deposition. The combination with a resonance ionization laser ion source not only has the advantage of a high ionization efficiency but in addition further purifies the sample by exclusively ionizing the element of interest.

Resonance ionization of holmium has been successfully demonstrated at the Ion Source Test Facility 2 (ISTF2) at the Oak Ridge National Laboratory (ORNL) [8]: Liu and coworkers achieved an outstanding ionization efficiency of 40 %, surpassing specifications reported for all other elements in resonant laser ion sources. We report confirmation studies using stable ^{165}Ho at the RISIKO mass

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separator at the University of Mainz, where the ion beam implantation for the ECHo experiment is foreseen.

2. Experiment

The measurements were performed at the RISIKO mass separator (see Fig. 1) equipped with the pulsed titanium–sapphire laser system developed at the University of Mainz [9]. A known number of ^{165}Ho atoms was inserted into the sample reservoir. After evacuation of the system, the sample was evaporated, ionized by resonant laser radiation, mass separated and finally collected in a Faraday cup for ion beam current quantification. The ionization efficiency was calculated as the ratio of the number of detected ions to the initial sample size. Future implantations of ^{163}Ho will be performed in the same way, with the microcalorimeters positioned inside the Faraday cup, so that incoming ions will be implanted to a depth of a few nm in the absorber of the microcalorimeter.

2.1. Samples

The samples were prepared from commercially available Holmium(III) nitrate-pentahydrate ($Ho(NO_3)_3 \cdot 5H_2O$) with a purity of 99.9 %. This was transferred to aqueous solutions with mass concentrations of either $2.74(16) \cdot 10^{-2}$ g/l or $2.74(16) \cdot 10^{-3}$ g/l. 10 µl of these solutions, containing $1.00(6) \cdot 10^{15}$ (or $1.00(6) \cdot 10^{14}$, respectively) atoms of holmium were deposited on 99.8 %-purity titanium substrates of 6×6 mm^2 size and 12.5 µm thickness and evaporated to dryness. The chemical form of the residuum was likely $Ho(NO_3)_3 \cdot 3H_2O$. To exclude systematic errors or losses in the process, the samples were quantified by neutron activation in the research reactor TRIGA Mainz [10]. The nominal sample sizes could be confirmed within an uncertainty of 4 %. After placement in the sample reservoir, the remaining water is released by heating of the reservoir and the compound decomposes in several steps until at temperatures above 560 °C conversion into the form of the oxide Ho_2O_3 is completed [11]. It is partially reduced to elemental oxide in the equilibrium state of the redox reaction $2Ho_2O_3 + 3Ti \rightleftharpoons 4Ho + 3TiO_2$. At temperatures above the melting point of holmium under the applied high vacuum conditions, elemental Ho begins to evaporate leading to a shift of the equilibrium to the right side. Hence, the higher evaporation rate of Ho with increasing temperature is followed in response by higher reduction rate of Ho_2O_3 and thus providing a continuous supply of Ho for the ionization.

2.2. Ion source

The measurements were performed with a split, hot cavity-type laser ion source consisting of a main hot cavity and a separated sample reservoir. The reservoir is a tantalum capillary of 200 mm length and 1.1 mm inner diameter. It can be resistively heated to a temperature of 2000 °C at a maximum current of 100 A. For a measurement, the current is slowly ramped from 0 A to 100 A to sequentially drive the dehydration and reduction process and – once the melting point of Ho of 1460 °C is exceeded – to control the rate of released holmium atoms. The hot cavity serves as the atom-laser interaction volume. It is also made of tantalum with a length of 30 mm and an inner diameter of 2.2 mm. It is resistively heated by an electrical current of 270 A, to reach a maximum temperature of 2000 °C, which is kept constant during the measurement. The operating temperature is chosen to be high enough to prevent condensation of the holmium vapor, but at the same time as low as possible to minimize surface ionization of the atoms by contact with the hot surface. Ideally, the temperature should be equal over the complete volume, but in reality, the occurrence of temperature gradients is inevitable. Using computational fluid dynamics, a simulation model to reproduce the temperature distribution was created. It is based on the geometry and material properties of the individual parts. After calibration to pyrometrically measured temperatures, the coldest part of the ion source could be identified as the mounting point between the hot cavity and the sample reservoir. When operated at maximum temperatures of 2000 °C, the temperature at this point is around 1400 °C.

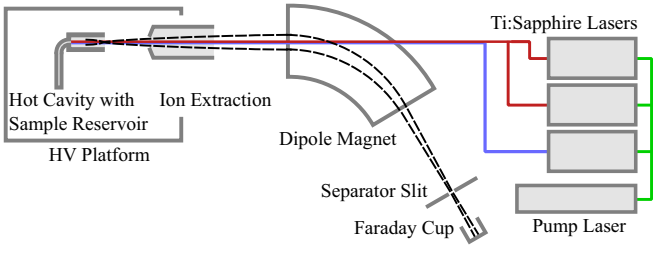

Fig. 1. Schematic overview of the experimental setup, showing the main components of the RISIKO mass separator and the connected Ti:sapphire laser system. For ion beam implantation, the microcalorimeters are mounted inside the Faraday cup.

2.3. Titanium–sapphire laser system

Laser ionization was achieved using the resonant three step excitation scheme with pulsed titanium–sapphire lasers shown in Fig. 2. The design of the Ti:sapphire lasers is developed by the LARISSA group at the institute of Physics of the University of Mainz. Its most recent iteration is presented by Rothe et al. in [12]. The lasers were pumped by a single commercial Nd:YAG laser operating at a repetition rate of 10 kHz. It delivered 15.5 W of frequency doubled light of 532 nm wavelength to each of the lasers. Tuned to the desired wavelengths, output powers of 2.8–3.2 W were achieved. The laser driving the ground state transition was frequency doubled with a BBO crystal placed in the resonator. In this configuration, the output power was 300 mW at the second harmonic frequency. Temporal synchronization of the 50 ns long pulses was achieved by adjusting the position of the pump beam focus in the Ti:sapphire crystal. The wavelengths were tuned by a birefringent filter and a solid etalon with 300 GHz free spectral range. The latter was stabilized with a piezo stepper motor adjusting the tilt angle. With the feedback from a HighFinesse WS6/600

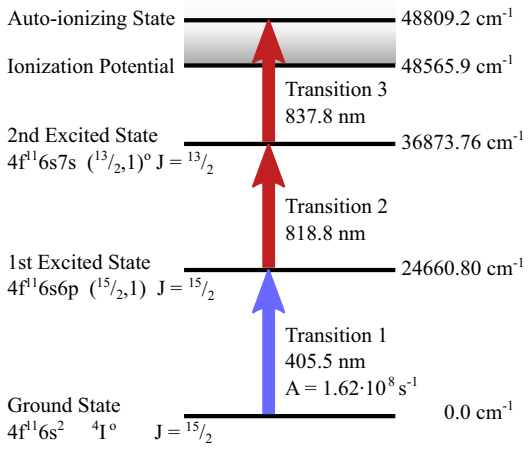

Fig. 2. Excerpt of the atomic level structure of holmium, showing the excitation scheme used in the measurements. Level energies and configurations are taken from [13], the ionization potential from [14].