First Penning Trap Mass Measurements beyond the Proton Drip Line

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The masses of six neutron-deficient rare holmium and thulium isotopes close to the proton drip line were determined with the SHIPTRAP Penning trap mass spectrometer. For the first time the masses of the proton-unbound isotopes ^{144,145}Ho and ^{147,148}Tm were directly measured. The proton separation energies were derived from the measured mass values and compared to predictions from mass formulas. The new values of the proton separation energies are used to determine the location of the proton drip line for holmium and thulium more accurately.

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The borderlines of nuclear stability are an important topic in modern nuclear physics. While the neutron drip line is experimentally still reached only for very light elements [1], the proton drip line was accessed up to heavy elements like protactinium [2,3]. The first experimental indication of direct (in contrast to β-delayed) proton radioactivity was found for an isomeric state, ^{53m}Co [4], while the first ground state proton emitter, ^{151}Lu, was discovered at GSI in 1981 [5]. Other proton-decaying nuclides like ^{147}Tm were observed shortly after [6]. Nowadays over 20 proton emitters beyond $Z = 50$ are known [7]. Their spectroscopic investigation provided important information on the underlying nuclear structure such as the ground state wave function beyond the drip line [8]. Whereas the detection of proton decay is sufficient proof of the unbound character of the emitting state, the inverse relation does not necessarily hold: The proton radioactivity of a proton-unbound nuclide may well be too weak to be detected, especially in close vicinity to the proton drip line. This is due to two effects. First, the Q value of direct proton decay Q_p, which is the negative of the proton separation energy S_p, strongly affects the decay rate. A small Q_p value results in a very low decay rate, i.e., a very long partial half-life, and thus a negligibly small branching ratio compared to the competing β decay. Second, it is experimentally very challenging to discriminate the low-energy proton against the background of β decay positrons. For these reasons proton emitters are generally found only at some distance from the proton drip line and cannot be used to delineate its location accurately. Accurate mass measurements of rare isotopes are an essential source of information for nuclear properties [9–12]. They can help to overcome these problems and to determine the Q_p value of the direct proton decay from the mass difference of the mother and the daughter nuclei. For nuclides close to the proton drip line this information is not accessible by decay spectroscopy for the reasons discussed above. In addition, so far unknown proton emitters can be identified. In this Letter we report the first direct mass measurements of proton-unbound rare isotopes and use the mass values to establish the location of the proton drip line for holmium and to unambiguously identify proton-unbound thulium isotopes.

The combination of fusion-evaporation reactions with in-flight separation provides a powerful technique for the production of neutron-deficient rare isotopes, in particular, of medium-heavy up to transuranium elements, and is presently the most competitive technique to reach the proton drip line beyond tin [1]. The SHIPTRAP facility [13] at GSI Darmstadt allows performing precision mass measurements of radioactive nuclides produced in fusion-evaporation reactions at the velocity filter SHIP [14]. A schematic overview of the setup is shown in Fig. 1.

The reaction products with energies of typically few 100 keV/u are kinematically separated from the primary beam and injected into the SHIPTRAP buffer-gas cell [15] through a thin metal foil serving as a degrader. The ions are thermalized in high-purity helium gas and extracted by a combination of electrical dc and radio frequency (rf) fields with gas flow through an extraction nozzle. The subsequent extraction rf quadrupole (RFQ) is operated as an ion guide and allows differential pumping. The precooled ions are

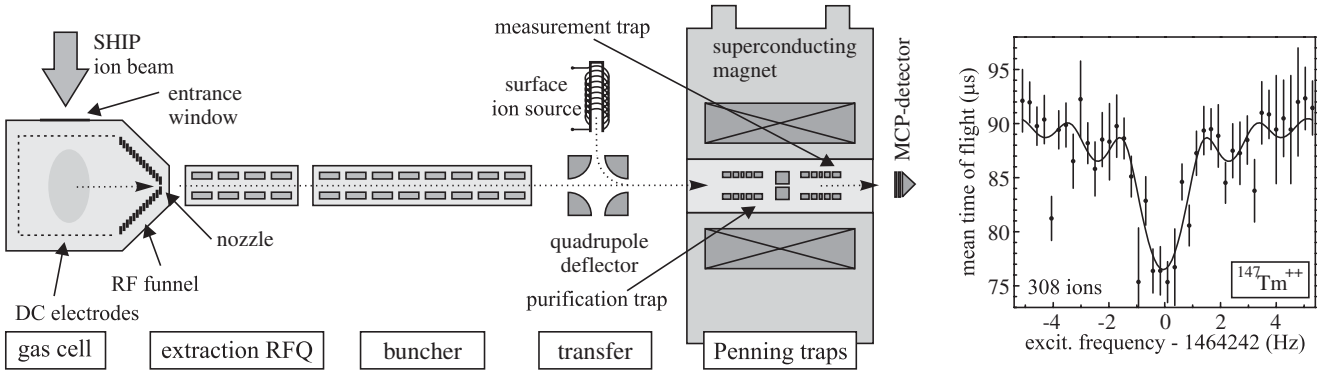

FIG. 1. Left: Schematic overview of the SHIPTRAP experiment. The radioactive ions delivered by SHIP are stopped (gas cell section), accumulated and cooled (buncher section), and transferred (transfer section) into the 7 T superconducting magnet (trap section). After isobaric separation (purification trap) the cyclotron frequency is determined (measurement trap). Right: A time-of-flight ion-cyclotron-resonance for ^{147}Tm.

accumulated in an RFQ cooler and buncher [16], where they are cooled in helium gas and extracted as a low-emittance bunched ion beam. The bunches are injected into a 7 T superconducting magnet housing two cylindrical Penning traps in two homogeneous-field regions. In the first Penning trap (purification trap) a mass-selective buffer-gas cooling technique [17] is applied to cool the ions and select them according to their mass-to-charge ratio. The mass resolving power achieved is sufficient to resolve nuclear isobars. The mass-selected ions are then transferred through a 3 mm orifice into the second Penning trap (measurement trap), where the mass is determined by measuring the cyclotron frequency

$$\nu_c = \frac{1}{2\pi}\frac{qe}{m_{\mathrm{ion}}}B, \tag{1}$$

for an ion of mass m_{ion} and charge state q in a magnetic field of flux density B. This frequency is measured by a time-of-flight ion-cyclotron-resonance (TOF-ICR) method [18]. A typical resonance obtained in this way is shown in Fig. 1. The magnetic flux density is calibrated by measuring the cyclotron frequency ν_{ref} of a reference ion with a well-known mass m_{ref}. Stable alkali ions from a surface ion source or alternatively carbon-cluster ions from a laser-desorption ion source [19] are used. Taking the frequency ratio $r = \nu_{\mathrm{ref}}/\nu_c$ the atomic mass of the nuclide can be calculated as

$$m = \frac{q}{q_{\mathrm{ref}}} r(m_{\mathrm{ref}} - q_{\mathrm{ref}} m_e) + q m_e, \tag{2}$$

where m_e is the electron mass and q_{ref} is the charge state of the reference ion. The binding energies of some eV for the outer electrons can be neglected for a mass uncertainty above 1 keV. A detailed description of the complete setup and the measurement procedure can be found in [20].

The data presented here were obtained in two experiments performed in October 2005 and December 2005. A primary ^{58}Ni^{14+} beam with an energy of 4.36 MeV/u and 4.60 MeV/u irradiated a 0.626 mg/cm^2 thick ^{92}Mo target to produce neutron-deficient isotopes in the terbium-to-thulium region. The gas cell was operated at a buffer-gas pressure of 50 mbar. The purification trap was operated with a mass resolving power of about 50 000 allowing only a particular isobar to be transferred to the measurement trap. Most of the mass measurements were performed with a cyclotron excitation time of 900 ms corresponding to a resolving power of 1.2×10^6. For each isotope several resonances were recorded. The presence of contaminant ions that may have been formed in the trap, for example, by charge exchange were accounted for by a count rate class analysis of the cyclotron frequency as described in [21]. Details about the uncertainty due to magnetic field changes are given in [22]. In total, the masses of 18 short-lived nuclides were measured in the two runs. A detailed description of the analysis, the mass evaluation, and the resulting mass values of all nuclides is given in [23]. Here we address the measured masses of the holmium ($Z = 67$) and thulium ($Z = 69$) isotopes and their impact on the proton drip line. These results are presented in Table I.

In comparison with the previous data the new results mainly agree with the measured ones but slightly deviate from the estimates. The previous mass value of ^{147}Ho was found to deviate by more than 3 standard deviations, its origin presently not being understood. The mass uncertainty was reduced by our data by up to a factor of about 30 with a relative mass uncertainty of 6.8×10^{-8} on average. Thus, an unambiguous interpretation of the proton separation energies is possible for all measured nuclei. Because of their unpaired proton the location of the proton drip line is much closer to the valley of stability than for elements with an even number of protons. The most exotic isotope measured was the proton emitter ^{147}Tm, with a half-life of 580 ms and a measured production cross section of only

TABLE I. Results of the mass measurements. The half-lives $T_{1/2}$ are listed according to [24]. Column three shows the weighted mean of the measured frequency ratios r. The values with $r > 1$ originate from the first run, where singly charged ions were measured relative to $^{133}Cs^{+}$, whereas in the second experiment ($r < 1$) doubly charged ions were measured relative to $^{85}Rb^{+}$. For ^{146}Ho and ^{147}Ho, which were measured in both experiments, an averaged value was used for the mass evaluation. The next two columns give the results from the atomic mass evaluation (M_{exp}) in comparison to the previous literature values (M_{AME}) [24]. Extrapolated masses are marked with #. The differences between the experimental and the previous data ($M_{exp} - M_{AME}$) are shown in column six. The last four columns list the proton separation energies obtained, respectively, from this work, from AME 2003 [24], from the finite-range droplet model (FRDM) [25], and from the Hartree-Fock-Bogoliubov mass formula (HFB-9) [26].

Nuclide	$T_{1/2}$	r	M_{exp} (keV)	M_{AME} (keV)	$M_{exp} - M_{AME}$ (keV)	S_p (keV)			
						This work	AME	FRDM	HFB-9
^{144}Ho	700 ms	0.847 655 803(54)	−44 609.5(90)	−45 200(300)#	590(300)	−271(16)	160(360)	260	−510
^{145}Ho	2.4 s	0.853 515 790(47)	−49 120.1(80)	−49 180(300)#	60(300)	−161(10)	−110(300)	260	30
^{146}Ho	3.6 s	1.098 111 737(78)	−51 238.2(70)	−51 570(200)#	330(200)	285(11)	570(200)	790	−130
		0.859 390 963(61)							
^{147}Ho	5.8 s	1.105 599 461(59)	−55 757.1(50)	−55 837(28)	80(28)	492(10)	570(40)	840	800
		0.865 250 860(44)							
^{147}Tm	580 ms	0.865 375 925(63)	−35 969.8(10)	−36 370(300)#	400(300)	−1066(13)	−1058(3)	−560	−780
^{148}Tm	700 ms	1.113 260 920(84)	−38 765(10)	−39 270(400)#	500(400)	−560(40)	−490(500)	10	−560

100–200 μb [6,27]. An average primary beam intensity of 200 particle-nA corresponds to a yield of 500–1000 ions per second in front of the SHIPTRAP gas cell. An example of a measured cyclotron resonance is shown on the right-hand side of Fig. 1. In total, within five hours 900 ions were collected in three resonances resulting in a mass value with a relative uncertainty of 7×10^{-8}.

The proton separation energy, S_p, defined as

$$S_p = B(Z, N) - B(Z-1, N) = -M(Z, N) + M(Z-1, N) + M_H, \quad (3)$$

where M_H is the atomic mass of hydrogen, allows distinguishing between the proton-bound ($S_p > 0$) and the proton-unbound ($S_p < 0$) nuclei or, in other words, to determine the proton drip line. From the present data the S_p values of the four nuclides ^{144,145}Ho and ^{147,148}Tm were determined to be negative. Table I shows the results in comparison with the results of the latest atomic mass evaluation (AME) [24] and the predictions of two global mass models, the microscopic HFB-9 [26] and the macroscopic-microscopic FRDM [25]. These two models were chosen, since they are able to reproduce the masses of the majority of nuclides within a few MeV. In particular, both mass models predict the masses of neutron-deficient nuclei fairly well. The S_p values predicted by the HFB-9 model are closer to the measured values while the FRDM predictions follow the experimental trend better; however, none of the models allows one to determine the drip line unambiguously. Our results agree well with the AME results that, due to their large uncertainties, did not allow an unambiguous assignment of the sign of S_p either. Only ^{147}Tm was known to be a proton emitter from decay spectroscopy [6,28], while for the other three nuclides the S_p values had prior to this work only been deduced from mass extrapolations [24] and from predictions of different mass models with large uncertainties.

The determination of the proton drip line on the basis of experimental data is visualized in Fig. 2. Here the S_p values for the holmium and thulium are plotted for their even-N isotopes only, thus avoiding the odd-even staggering due to the pairing energy. Three data points from the present work were included. The new S_p value obtained for ^{147}Tm agrees well with that determined previously [28]. The measurement of ^{147}Ho slightly shifts the previous result [24] to a lower separation energy. The value of ^{145}Ho was never measured before, and the measurements clearly show that ^{145}Ho is proton unbound. The data establish

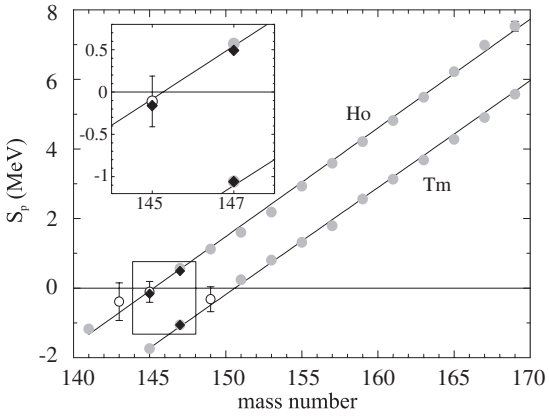

FIG. 2. Proton separation energies versus mass number for the odd-Z elements holmium ($Z = 67$) and thulium ($Z = 69$). Only the even-N isotopes are plotted. The circles show the previous measurements (full gray circles) and estimations (empty circles) taken from [24]. The data from this work are represented by black diamonds. Also shown are linear fits to the data. The inner box enlarges the interesting region around the drip line.

the location of the drip line between $A = 145$ and $A = 147$ for holmium. In the case of thulium we have shown that ^{148}Tm is already proton unbound. However, unless the value of $S_p(^{149}\mathrm{Tm})$ is also measured with a comparable accuracy the exact location of the drip line for thulium cannot be determined.

Whether the three new proton-unbound nuclides are accessible to investigations of direct proton decay can be answered by estimating the partial half-lives of this decay mode. In a simple, semiempirical approach applying the Wentzel-Kramers-Brillouin approximation the proton decay can be described similarly to the α decay [29]. The decay constant

$$\lambda_p = \frac{\ln 2}{T_p} = \nu_0 e^{-2C_p} \tag{4}$$

is in this picture the product of a frequency factor ν_0 and an exponential transmission term. The factor ν_0 is in first order the inverse of the characteristic nuclear time and mainly defined by the velocity of the proton and the radius of the nucleus. In the investigated region of the nuclear chart ν_0 has a typical value of 6×10^{22} Hz [29] and is more or less constant. The transmission term, usually called Gamow factor, describes the tunneling probability through the Coulomb and centrifugal potential barriers. The exponent C_p is the integral over the ''forbidden'' region of the potential. It depends mainly on the energy and angular momentum of the proton and can be calculated numerically. An angular momentum of 5 was estimated by comparing the calculated half-life of ^{147}Tm with the measured one. Only for ^{147}Tm the proton decay is an observable decay channel. While for ^{148}Tm with a partial half-life of $T_p = 10^{11}$ s it is experimentally very difficult to discriminate the proton from the β background, ^{144}Ho and ^{145}Ho are surely far out of reach for the study of proton decay as their partial half-lives for this decay mode are longer than the age of the universe.

In summary, we present the first Penning trap mass measurements of proton-unbound holmium and thulium isotopes with SHIPTRAP. The results show generally good agreement with the previously measured or estimated data but reduce the uncertainty by more than an order of magnitude. An analysis of the proton separation energies that were derived from the new mass values allowed pinning down the location of the proton drip line in this region unambiguously. Simple model calculations of the partial half-lives show that direct measurements of the proton decay by decay spectroscopy are not feasible or in the case of ^{148}Tm at least extremely ambitious.

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