Schottky Mass Measurements of Cooled Proton-Rich Nuclei at the GSI Experimental Storage Ring

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High-precision mass measurements of proton-rich isotopes in the range of $60 \leq Z \leq 84$ were performed using the novel technique of Schottky spectrometry. Projectile fragments produced by ^{209}Bi ions at 930A MeV were separated with the magnetic spectrometer FRS and stored and cooled in the experimental storage ring (ESR). A typical mass resolving power of 350 000 and a precision of 100 keV were achieved in the region $A \approx 200$. Masses of members of α chains linked by precise Q_α values but not yet connected to the known masses were determined. In this way it is concluded that ^{201}Fr and ^{197}At are proton unbound. [S0031-9007(97)03195-5]

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Precise knowledge of atomic masses is required for the understanding of nuclear gross properties and for many applications in other fields of physics, e.g., in astrophysics. In general, masses are well known for nuclei close to β stability, whereas for nuclei far off stability one has to rely mainly on extrapolations. Schottky spectrometry [1,2] of heavy ions at energies of several hundred A MeV has been used in the present experiment for direct mass measurements of proton-rich nuclei for elements from neodymium to polonium. In Schottky spectrometry the masses of stored and cooled ions are measured by recording their revolution frequencies via image currents in a highly sensitive capacity probe. For lighter nuclei ($A \leq 58$) the potential and power of this new method have been demonstrated before [3,4]. In the present experiment we have succeeded in extending the measurements to the region of $A \approx 200$. In the case of these heavy nuclei, experimental data on masses are especially scarce for refractory elements ($72 \leq Z \leq 78$) because the widely used ISOL (isotope separator on-line) technique could not access this region. The goals of our experiments to the backbone of known ones, and to measure the members at the end of α chains, thus providing new information on proton drip line nuclei linked via precise Q_α values.

The combination of an in-flight separator for fragments with a storage-cooler ring represents a unique tool for studying exotic nuclei [5]. By merging a cold electron beam with the circulating projectile fragments the momentum spread of the cooled ions can be reduced to $\Delta p/p \leq 10^{-6}$ when the number of stored ions with the same mass-over-charge ratio m/q is less than 10^4 [4,6,7]. The low momentum spread of stored beams obtained in this way, the high sensitivity [8], as well as the long storage time allow one to perform high-precision mass determination.

In the present experiment a 930A MeV ^{209}Bi beam accelerated in the synchrotron SIS is focused on an 8 g/cm^2 beryllium target placed at the entrance of the fragment separator (FRS) [9]. Projectile fragments ranging from the proton number of the primary beam down to $Z = 1$ are produced and emerge from the target. The FRS has been used as a pure magnetic-rigidity analyzer characterized by an acceptance of $\Delta(p/q)/(p/q) = \pm 1\%$, where p denotes the momentum of an ion. For the selected thickness of the target, mainly bar, H-like, and He-like ions emerge. The energy loss in this thick target caused only elements between $Z = 60$ and $Z = 84$ to be effectively transmitted through the FRS. Several field settings of the FRS were applied, each transmitting a different p/q band. The separated projectile fragments with mean kinetic energies of about 350 A MeV are injected into the ESR, where they are stored and electron cooled to an identical mean velocity determined by the velocity of the cooler electrons. In this case, the revolution frequency of each ion is defined by its mass-to-change ratio only, which is the basis for Schottky mass spectrometry (SMS). The stored ions circulate in the ESR (circumference ≈108 m) with frequencies of about 1.9 MHz. The Schottky signal induced in a pick-up probe is recorded for about 150 msec and subsequently frequency analyzed by fast-Fourier transformation (FFT). Mixing with 30 MHz transforms the spectrum of the 16$^{\text{th}}$ harmonic of the revolution frequency to the 100 kHz bandwidth of the frequency analyzer. A high signal-to-noise ratio is obtained by averaging up to 10^4 single FFT spectra which requires a measuring time of

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roughly 3 min. Figure 1 shows an example of a Schottky spectrum with 100 kHz bandwidth almost covering the ESR (m/q)-acceptance of 2.5%. In this case, the magnetic rigidity of the FRS was set to optimize the transmission of fully ionized ^{197}Bi fragments. The spectrum contains about 60 ion species in bare, H-like, He-like charge states. However, they cannot be resolved in 100 kHz overview spectra. An increased frequency resolution is obtained for the 10 kHz spectra mode of the frequency analyzer (Fig. 1, bottom).

Because of the large amount of peaks, the assignment of the different peaks to isotope and charge state represents a major step in the analysis. The observed frequency peaks are related to mass values by

$$\frac{\Delta f}{f} = -\alpha_p \frac{\Delta(m/q)}{m/q}, \qquad (1)$$

where f is the mean revolution frequency of the ion species and (m/q) is the corresponding mass-to-charge ratio. Δf is the difference of the frequencies corresponding to the considered $\Delta(m/q)$ value, either for calibration or mass measurement. The momentum compaction factor α_p depends on the ion optical operation mode of the ESR and denotes the ratio of the relative change in path length per turn to the relative change of the corresponding magnetic rigidity. For the purpose of peak identification, mass values taken from Ref. [10] and corrected for the missing electrons and their respective binding energies [11–13] allow one to compute the frequency differences for all tabulated nuclides in different charge states. The calculated frequency spectra exhibit a characteristic pattern and enable a computer assisted pattern recognition of mass spectra. This leads to an unambiguous assignment of mass number, atomic number, and charge state. After this identification procedure the spectra recorded with a smaller detection bandwidth of 10 kHz are used for mass determination. The mass value of the ion under investigation, e.g., ^{184}Pt, can be determined by comparing its revolution frequency to those of ions with known mass values. As an example, Fig. 2 shows a high-resolution spectrum taken with a (p/q)-setting of the FRS, different from that shown in Fig. 1. It contains six isotopes with unknown masses and another six with known masses. Each peak is fitted with a Gaussian and a linear background. The width (FWHM) of the peaks corresponds to approximately 500 keV and a high resolving power of $m/\Delta m$ (FWHM) $\approx$ 350 000. The momentum compaction factor ($\alpha_p \approx 0.14$) is calibrated according to (1) using neighboring peaks of isotopes with known masses. Over the full range of a 100 kHz spectrum, α_p variations of the order of 1% are observed. This can be caused by inhomogeneities and instabilities of the magnetic fields of the ESR. As we cannot precisely determine the global slope of the α_p as a function of m/q for the full acceptance of the ESR, the α_p calibration is restricted to a smaller portion of the frequency spectrum in the neighborhood of the masses of interest.

With α_p determined in this way new mass values can be assigned using several reference peaks.

An important feature of the SMS method is the possibility of using a large number of well-known masses for an accurate calibration. For instance, the signals of stable or long-lived isotopes such as H-like ^{151}Tb and bare ^{144}Pm are used for the calibration of nuclei far from stability, such as bare ^{184}Pt. Even more, since most of the ions are measured in three different ionic charge states thus appearing in several spectra surrounded by different reference nuclides, redundant and independent mass determinations are possible. As an example of this important feature, the mass measurement of ^{184}Pt is illustrated in Fig. 3. The error bars shown are due to a statistical error and a calibration error.

The statistical error accounts for uncertainties in the determination of the revolution frequencies. The frequencies of the 16$^{\text{th}}$ harmonic of about $f = 30$ MHz are determined

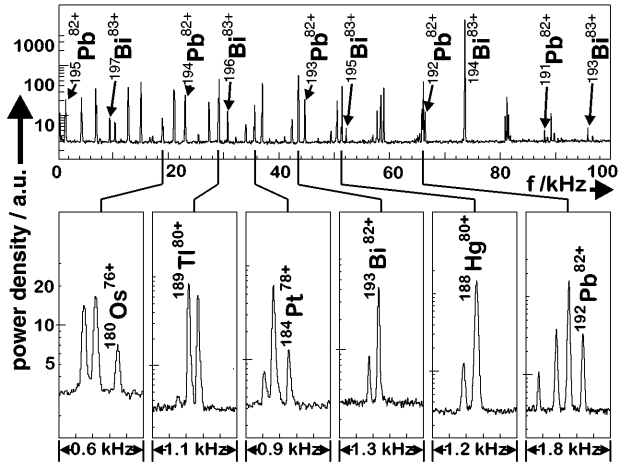

FIG. 1. Schottky frequency spectra for ^{209}Bi-projectile fragments separated with the FRS, and stored and cooled in the ESR. Top: low-resolution (100 kHz bandwidth) spectrum corresponding to the 16$^{\text{th}}$ harmonic of the revolution frequency, which covers nearly the full range of m/q values (2.5%) for ions accepted inside the ESR. Bottom: high-resolution spectra (10 kHz bandwidth) in which the nuclides under investigation are resolved.

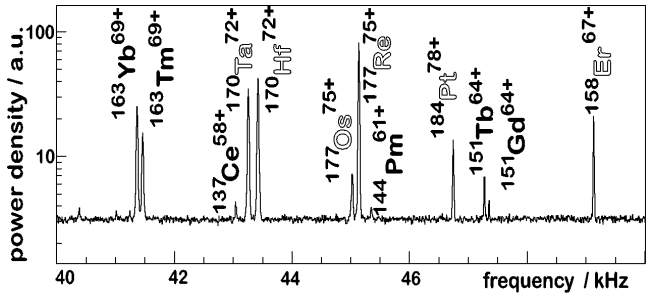

FIG. 2. High resolution Schottky spectrum (10 kHz bandwidth) for the mass determination of ^{184}Pt. The peaks of ions with known masses used for calibration are indicated by bold letters; nuclei with previously unknown masses are indicated by outlined letters.

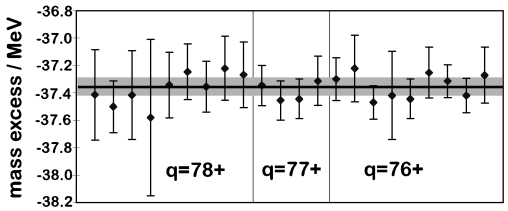

FIG. 3. Mass excess for ^{184}Pt as determined in several runs using different reference isotopes and in different ionic charge states q. The shaded area shows the statistical error band after averaging ($\sigma = \pm 40$ keV).

with an accuracy of about ± 1 Hz, i.e., with a relative uncertainty of $\delta f/f \simeq \pm 3 \times 10^{-8}$.

The calibration error accounts for uncertainties of the known mass values used for calibration. These errors range from a few 10 to 200 keV [10]. Our mass values are derived from relative measurements with known reference masses in the same Lorentz system, therefore, no additional errors have to be considered due to relativistic effects. The horizontal line and the shaded error band in Fig. 3 represents the weighted average of the individual data points and its error (e.g., $\sigma \approx \pm 40$ keV for the case of ^{184}Pt).

Finally, a systematic error has to be added which takes into account, e.g., the variation of α_p in time and as a function of the different ion orbits in the storage ring. This systematic error is determined by a comparison of already known masses: Mass values tabulated in Ref. [10] are compared to masses remeasured in this work by SMS. These multiple and independent direct measurements result in a normal distribution around zero. Therefore we are allowed to perform a χ^2 test in which we introduce our additional systematic error. This error of about 80 keV, which has to be added quadratically to the statistical and calibration errors mentioned above, now gives a perfect agreement or our SMS data and Ref. [10]. In the case of ^{184}Pt the final error results in $\approx$100 keV, corresponding to $\delta m/m \approx 5 \times 10^{-7}$ which is rather typical for the mass accuracy obtained by SMS.

Results of our measurements are shown in Table I. These nuclei are members of the two α-decay chains starting at ^{201}Fr and ^{200}Rn (Fig. 4). Their mass values are compared with masses calculated via Q_{alpha} values and with extrapolations given in Ref. [10]. Isomeric states of isotopes could only be measured in the present experiment if the half-lives are larger than 20 s.

Furthermore, the two states could only be resolved if the energy difference is equal to or larger than the experimental resolution [4]. Therefore the isomeric state of ^{193}Bi is too shortlived ($t_{1/2} = 3.2$ s) to be detected. In the case of ^{189}Tl, our result is compatible with the ground state mass within the experimental errors. In addition, the line shape analysis did not show any distortion. The inset of Fig. 4 shows a comparison of Q_α values deduced from the directly measured masses in this experiment

TABLE I. Mass values in units of u.

Nuclide	Mass (u)[a]	Mass (u)[b]	Mass (u)[c]
^{180}Os	179.95240(10)	179.95234(5)	179.95236(19)
^{184}Pt	183.95991(10)	183.95990(5)	183.95989(19)
^{188}Hg	187.96753(10)	187.96759(5)	187.96755(19)
^{192}Pb	191.97579(15)	191.97580(5)	191.97576(19)
^{189}Tl	188.97391(9)	188.97385(11)	188.97369(38)
^{193}Bi	192.98304(15)	192.98322(11)	192.98306(28)

[a]Determined in this experiment.
[b]Determined by SMS and Q_α values [14–18].
[c]Compared with extrapolations given in Ref. [10].

with precise Q_α values from the literature [14–18]. Both data agree within the error bars which are dominated by the SMS results due to the small uncertainties of the Q_α values (5–20 keV). Using the SMS data from Table I and the Q_α values from [14–18] as shown in Fig. 4, the masses for the short-lived nuclei ^{196}Po, ^{197}At, ^{200}Rn, and ^{201}Fr were determined. The precision for the masses which are derived by adding Q_α values from various starting nuclides can be clearly improved. For instance, the errors for the masses of ^{196}Po and ^{200}Rn are significantly reduced by using various directly measured masses (^{180}Os, ^{184}Pt, ^{188}Hg, and ^{192}Pb) of the same α chain, in addition to the Q_α values. The results are listed

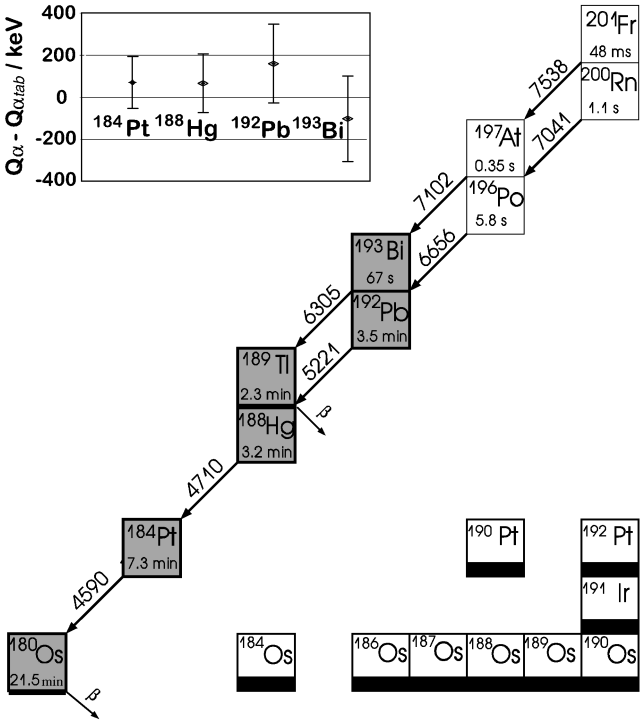

FIG. 4. Two α-decay chains for which six mass values are determined by SMS in the present work (shaded boxes). The half-lives and the Q_α values (keV) are indicated. Stable isotopes of platinum, iridium, and osmium are shown for orientation. The inset shows a comparison between directly determined $Q_{\alpha\text{-tab}}$ values and those from SMS (Table I).

TABLE II. Mass values for the short-lived nuclei at the top of the α chains (see Fig. 4) obtained by using our SMS mass values and Q_α values from Refs. [14–18] compared to extrapolations given in Ref. [10].

Nuclide	Mass (u)[a]	Mass (u)[b]
^{196}Po	195.98552(5)	195.98551(19)
^{200}Rn	199.99568(5)	199.99568(19)
^{197}At	196.99346(8)	196.99329(38)
^{201}Fr	201.00416(10)	201.00399(38)

[a]Refs. [14–18].
[b]Ref. [10].

in Table II. In the same way, the masses are determined for the odd-proton nuclides ^{197}At and ^{201}Fr.

Proton separation energies and pairing energies are of fundamental interest for the theoretical description of exotic nuclei [19]. For ^{201}Fr and ^{197}At, at the top of the α chain displayed in Fig. 4, we obtain

$$S_p(^{201}\mathrm{Fr}) = -(570 \pm 110)\ \mathrm{keV},$$

$$S_p(^{197}\mathrm{At}) = -(80 \pm 80)\ \mathrm{keV},$$

i.e., both nuclei are proton unbound. The proton separation energies are compared to different calculations [20–23]. These models yield for ^{201}Fr 280 keV [20], 0 keV [21], 80 keV [22], and 90 keV [23]; and for ^{197}At 380 keV [20], 190 keV [21], 260 keV [22], and 430 keV [23]. The property of being proton unbound does not necessarily mean that such nuclei are proton emitters, e.g., a rough estimate for the penetrability of protons through the Coulomb barrier of ^{201}Fr [24] yields a proton partial half-life of $T_{1/2}(p) \approx 10^8$ s. The partial α half-life is 69 ms [17], which means a proton emission from the ground state is very unlikely. The heaviest known proton emitter in this mass region is ^{185}Bi [25].

The results presented in this Letter were selected to demonstrate the power of Schottky spectrometry in combination with the FRS-ESR system for mass measurements. In an experiment of a few days, the masses of more than 280 isotopes have been determined. We succeeded in measuring about one hundred previously unknown or not accepted mass values [26,27]. These numerous results will be published elsewhere. We obtained in this experiment a resolving power for heavy nuclei as high as $m/\delta m$ (FWHM) $\approx$ 350 000, and the overall uncertainties of the mass determination are in the range from 80 to 200 keV. In the case of several links by measured Q_α values, the error can be reduced to $\approx$50 keV. The excellent sensitivity of SMS allows that even a single ion circulating in the ESR can be detected [8]. Unstable nuclides can be measured if their lifetime exceeds the time required for cooling and frequency analysis, which was of the order of one minute.

It should be noted that mother and daughter nuclei connected by a β-decay chain are often observed in the Schottky spectrum as close lying mass multipletts (see Fig. 2). SMS provides, therefore, not only direct mass spectrometry of nuclei far from instability but also a new approach for Q_β measurements of exotic nuclides.

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