Measurement of the $\beta^+$ and orbital electron-capture decay rates in fully-ionized, hydrogen-like, and helium-like $^{140}$Pr ions

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We report on the first measurement of the $\beta^+$- and orbital electron capture decay rates of $^{140}$Pr nuclei with the most simple electron configurations: bare nuclei, hydrogen-like and helium-like ions. The measured electron capture decay constant of hydrogen-like $^{140}$Pr$^{58+}$ ions is about 50% larger than that of helium-like $^{140}$Pr$^{57+}$ ions. Moreover, $^{140}$Pr ions with one bound electron decay faster than neutral $^{140}$Pr$^{58+}$ ions with 59 electrons. To explain this peculiar observation one has to take into account the conservation of the total angular momentum, since only particular spin orientations of the nucleus and of the captured electron can contribute to the allowed decay.

Various ways to influence nuclear decay rates have been tried by scientists since radioactivity was discovered. Their motivation reaches from the basic understanding of nuclear decay phenomena and astrophysical reactions to applications like the transmutation of nuclear waste. Small effects of up to a few percent have been observed in atoms by changing the environmental parameters such as pressure, temperature or electromagnetic fields $^{11,12}$. These changes are mainly attributed to modifications of the electron density at the nucleus. Significant modifications of the electron conversion rate have been measured when swift highly-ionized radioactive ions emerge from matter and their nuclear decay is determined in flight $^{13,14}$.

It has been predicted that the decay properties of highly-ionized nuclides can be altered dramatically: decay modes known in neutral atoms can become forbidden, new ones can be opened up. This can have substantial impact on the nucleosynthesis in hot stellar plasmas $^{13,14}$. Seminal results on decay studies with selected highly-ionized ions have been obtained with novel experimental tools using the combination of high-energy accelerators, in-flight separators and storage rings $^7$. For example, the electron capture and electron conversion decays become impossible in the absence of orbital electrons, i.e. in fully-ionized atoms. Thus, the pure $\beta^+$-decay branch has been measured in $^{52}$Fe$^{26+}$ ions $^8$ and the half-lives of isomeric states were found to be dramatically prolonged $^9$.

Bare $^{187}$Re$^{75+}$ ions decay, due to the new decay mode—the bound-state $\beta$-decay—by nine orders of magnitude faster than neutral $^{187}$Re atoms with a half-life of 42 Gyr $^{10}$. Note that the couple $^{187}$Re/$^{187}$Os is used as a cosmic clock. Bare $^{163}$Dy$^{66+}$ nuclei, being stable as neutral atoms, become radioactive, thus allowing the s-process, the astrophysical slow-neutron capture process of nucleosynthesis, to branch $^{11}$. We note, that a simultaneous measurement of $\beta$-decay to the continuum and bound states in $^{207}$Tl$^{81+}$ ions has been performed recently $^{12}$.

In the present experiment the $\beta^+$ and orbital electron-capture (EC) decays of bare nuclei and nuclei with one and two bound electrons have been investigated. The EC decay rates in hydrogen- and helium-like ions have been measured for the first time.

For this experiment we have selected the $^{140}$Pr (Z=59) nucleus. The neutral atom decays with 99.4% to the ground state of $^{140}$Ce via a pure Gamow-Teller $\beta$-decay with a change of the nuclear angular momentum by one unit ($\Delta I = 1$) and no parity change $^{13}$. The weak branches to the excited states in $^{140}$Ce can be neglected.
in our context. A proton in $^{140}$Pr can be converted into a neutron via a weak decay in two ways, namely via the EC decay whereby a monochromatic electron-neutrino is emitted ($p + e^- \rightarrow n + \nu_e$), or via a three-body decay in which the positron and the neutrino share the decay energy ($p \rightarrow n + e^+ + \nu_e$).

The experiment has been performed at the Gesellschaft für Schwerionenforschung (GSI), Darmstadt, Germany, where the combination of a heavy-ion synchrotron (SIS) \cite{14}, the in-flight fragment separator FRS \cite{15} and the ion storage-cooler ring ESR \cite{16} provides unique experimental conditions for decay studies of bare and few-electron exotic nuclei in an ultra-high vacuum ($\sim 10^{-11}$ mbar). It is possible to produce, separate and store exotic nuclei up to uranium with a well-defined number of bound electrons \cite{7 8 9 10 11 12}. Radioactive $^{140}$Pr ions have been produced via the projectile fragmentation of $^{152}$Sm ions/spill, accelerated by the SIS to 508 MeV/u. A 1 g/cm$^2$ thick beryllium target has been used. The fully-ionized, hydrogen- and helium-like $^{140}$Pr ions were separated in-flight by a two-fold magnetic rigidity analysis by means of the B$\rho$-\textDelta E-B$\rho$ separation method \cite{15} in the FRS and subsequently injected into the ESR. The flight time from the production target to the storage ring was a few hundred nanoseconds. The ion-optical settings of the FRS, the charge state distributions and the energy degraders used in this experiment are described in details in Ref. \cite{17}.

Stochastic \cite{18} and electron cooling \cite{19} were applied to the $^{140}$Pr$^{59+}$, $^{140}$Pr$^{58+}$ and $^{140}$Pr$^{57+}$ ions coasting in the ESR. The stochastic cooling provides fast pre-cooling at a fixed fragment velocity, corresponding to 400 MeV/u energy, thus reducing the overall cooling time to about 2 seconds. The cooling forces all stored ions to the same mean velocity and reduces the initial velocity spread, caused by the fragmentation reaction, to $\Delta v/v \approx 5 \cdot 10^{-7}$.

The unambiguous identification of cooled $^{140}$Pr$^{59+}$, $^{140}$Pr$^{58+}$ and $^{140}$Pr$^{57+}$ ions and their decay products has been achieved exploiting the time-resolved Schottky Mass Spectrometry \cite{20 21}. The latter is based on Schottky-noise spectroscopy \cite{22}, which is widely used for non-destructive beam diagnostics in circular accelerators and storage rings. The stored ions are circulating in the ESR with revolution frequencies of about 2 MHz. At each turn they induce mirror charges on two electrostatic pick-up electrodes. Fast Fourier Transform of the amplified signals yields the revolution frequency spectra, which provide information about the mass-over-charge ratios of the ions. The area of the frequency peaks is proportional to the number of stored ions, which is the basis for lifetime measurements \cite{8 9 10 11 12}. The details of the data acquisition system and of the data treatment can be found in Ref. \cite{21} and references cited therein.

In the EC decay the atomic mass changes but the atomic charge state is preserved. Therefore, this decay causes a sudden change in the revolution frequency of about 270 Hz (31$^{st}$ harmonics). An example for the $^{140}$Pr$^{58+} + e^- \rightarrow ^{140}$Ce$^{58+} + \nu_e$ decay is illustrated in Fig. \cite{1} where 195 subsequent Schottky frequency spectra are plotted as a water-flow diagram. Each spectrum is averaged over 10.5 s. It can be seen in

\begin{figure}[h]
\centering
\includegraphics[width=\textwidth]{schottky_frequency_spectra.png}
\caption{Schottky frequency spectra at the 31$^{st}$ harmonics of the revolution frequency taken subsequently as a function of time (195 spectra á 10.5 sec). In the EC decay of hydrogen-like $^{140}$Pr, the mass changes by 3.349 MeV/$c^2$ which leads to a small change in the frequency ($\sim$270 Hz). The intensity of the frequency lines is proportional to the number of stored ions. It can be seen that the intensity of the line corresponding to the parent ions $^{140}$Pr$^{58+}$ decreases in the course of time and that the intensity of the line corresponding to the daughter ions $^{140}$Ce$^{58+}$ increases.}
\end{figure}
Fig. 1 that the intensity of the peak at lower revolution frequency—corresponding to the parent ions $^{140}\text{Pr}^{58+}$—decreases steadily and that the intensity of the peak at the higher frequency—corresponding to the lighter daughter ions $^{140}\text{Ce}^{58+}$—increases. Examples of the decay and growth curves are shown in Fig. 2. Feeding of $^{140}\text{Pr}^{58+}$ or $^{140}\text{Ce}^{58+}$ ions via radioactive decays or reactions of other ions has been avoided by blocking the corresponding orbits in the ESR with mechanical slits.

Several measurements of the decay of $^{140}\text{Pr}^{59+}$, $^{140}\text{Pr}^{58+}$, and $^{140}\text{Pr}^{57+}$ ions have been performed. Decay curves of the parent ions have been fitted with an exponential function:

$$N_{Pr}(t) = N_{Pr}(0) \cdot e^{-\lambda t},$$

where $N_{Pr}(t)$ and $N_{Pr}(0)$ is the number of parent ions at the time $t$ after injection and at $t = 0$, the time of injection, respectively. For hydrogen-like and helium-like $^{140}\text{Pr}$ ions, the decay constant $\lambda$ is the sum of the EC decay constant $\lambda_{EC}$, the $\beta^+$ decay constant $\lambda_{\beta^+}$, and the loss constant $\lambda_{loss}$ due to collisions with residual gas atoms or pick-up of electrons in the electron cooler ($\lambda = \lambda_{EC} + \lambda_{\beta^+} + \lambda_{loss}$). The bare $^{140}\text{Pr}^{59+}$ nuclei can only decay via the $\beta^+$- decay-mode. Hence, the measured decay constant is the sum $\lambda_{\beta^+} + \lambda_{loss}$. The growth of the number of daughter ions from the EC decay of $^{140}\text{Pr}^{58+}$ into $^{140}\text{Ce}^{58+}$ nuclei and $^{140}\text{Pr}^{57+}$ into $^{140}\text{Ce}^{57+}$ ions is determined solely by the EC rate of $^{140}\text{Pr}$, whereas the loss of stable $^{140}\text{Ce}$ ions is determined only by $\lambda_{loss}$. Therefore, we can fit the number $N_{Ce}(t)$ of $^{140}\text{Ce}$ daughters as a function of time $t$ by using:

$$N_{Ce}(t) = N_{Pr}(0) \cdot \frac{\lambda_{EC}}{\lambda - \lambda_{loss}} \cdot [e^{-\lambda_{loss} t} - e^{-\lambda t}]$$

All measurements have presented consistent results. The averaged values for the $\lambda_{EC}$ and $\lambda_{\beta^+}$ decay constants converted to the rest frame of ions are presented in Table I. The mean loss constant has been determined to be $\lambda_{loss} = 0.0003(1)$ s$^{-1}$, which is within the error bars the same for the studied charge states of $^{140}\text{Ce}$ and $^{140}\text{Pr}$.

As can be seen from Table I, the measured $\beta^+$ decay rate is within the errors independent on the degree of ionization. This is expected, since the electron screening modifies the $\beta^+$ rate by less than 3% in fully-ionized ions compared to neutral atoms.

Previously, the EC from the K-orbit has been measured in implanted atoms by applying X-ray spectroscopy [24]. Such measurements have been performed for neutral $^{140}\text{Pr}$ in Refs. [25, 26, 27] and can be compared with our measurement on the helium-like ions. Using the values for the $^{140}\text{Pr}^{57+}$ ions we obtain $\lambda_{EC}/\lambda_{\beta^+} = 0.95(8)$, which agrees well with 0.90(8) from Ref. [26] and disagrees by about 2.5 standard deviations with 0.74(3) from Ref. [25] and with 0.73(3) from Ref. [27]. We note that it is the first time that this quantity could be measured directly in helium-like ions without the influence of other orbital electrons. These electrons modify the density of the K-electrons at the nucleus by about 1% [28], which has been neglected in the above comparison.

The striking result is—in spite of the fact that the number of orbital electrons is reduced from two in $^{140}\text{Pr}^{57+}$ ions to only one in $^{140}\text{Pr}^{58+}$ ions—that the EC-rate increases by a factor of 1.49(8). Moreover, the half-life of $^{140}\text{Pr}^{58+}$ with a single orbital electron, $T_{1/2} = \ln(2)/\lambda = 3.04(9)$ min, is even shorter than the half-life $T_{1/2} = 3.39(1)$ min [13] of the neutral $^{140}\text{Pr}^{0+}$ atoms with 59 orbital electrons.

Our result can be explained by taking into account the conservation of total angular momentum of the nucleus-lepton system. We note, that similar arguments have been used in Refs. [3, 29] to explain the de-excitation of nuclear excited states decaying via electron conversion in highly-ionized iron ions and in Ref. [30] to describe the muon-capture decay rates.

In the initial state $(i)$, the total angular momentum $F_i$ of a $^{140}\text{Pr}$ nucleus with spin $I_i = 1$ and a single bound K-electron with spin $s = 1/2$ can have two values of the

### Table I: Measured $\beta^+$ and EC decay constants obtained for fully-ionized, hydrogen-like, and helium-like $^{140}\text{Pr}$ ions.

| Ion      | $\lambda_{\beta^+}$ [s$^{-1}$] | $\lambda_{EC}$ [s$^{-1}$] |
|----------|-------------------------------|---------------------------|
| $^{140}\text{Pr}^{59+}$ | 0.00158(8) | — |
| $^{140}\text{Pr}^{58+}$ | 0.00161(10) | 0.00219(6) |
| $^{140}\text{Pr}^{57+}$ | 0.00154(11) | 0.00147(7) |
FIG. 3: Illustration of the EC decay of the hydrogen-like $^{140}$Pr$^{58+}$ ions to bare $^{140}$Ce$^{58+}$ ions.

The influence of the hyperfine-state of the electron on the EC-decay rate depends on the ratio of the statistical weights of the transition, i.e., $(2I_f + 1)/(2F_i + 1)=3/2$, which is in excellent agreement with our experimental result.

In summary, our experimental results have clearly revealed a fundamental property of the $\beta$-decay of highly-ionized atoms, which could not have been measured in any previous experiment. The description of the EC rate which is known from neutral atoms has to include the conservation of the total angular momentum, in particular when going to high atomic charge states, which prevail, e.g., in hot stellar plasmas during nucleosynthesis.

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