Measurement of the β^+ and Orbital Electron-Capture Decay Rates in Fully Ionized, Hydrogenlike, and Heliumlike ^{140}Pr Ions

Yu. A. Litvinov,[1,2] F. Bosch,[1] H. Geissel,[1,2] J. Kurcewicz,[3] Z. Patyk,[4] N. Winckler,[1,2] L. Batist,[5] K. Beckert,[1] D. Boutin,[2] C. Brandau,[1] L. Chen,[2] C. Dimopoulou,[1] B. Fabian,[2] T. Faestermann,[6] A. Fragner,[7] L. Grigorenko,[8] E. Haettner,[2] S. Hess,[1] P. Kienle,[6,7] R. Knöbel,[1,2] C. Kozhuharov,[1] S. A. Litvinov,[1,2] L. Maier,[6] M. Mazzocco,[1] F. Montes,[1,9] G. Münzenberg,[1,10] A. Musumarra,[11,12] C. Nociforo,[1] F. Nolden,[1] M. Pfützner,[3] W. R. Plaß,[2] A. Prochazka,[1] R. Reda,[7] R. Reuschl,[1] C. Scheidenberger,[1,2] M. Steck,[1] T. Stöhlker,[1] S. Torilov,[13] M. Trassinelli,[1] B. Sun,[1,14] H. Weick,[1] and M. Winkler[1]

[1]*Gesellschaft für Schwerionenforschung GSI, Planckstraße 1, 64291 Darmstadt, Germany*
[2]*Justus Liebig Universität Giessen, Heinrich-Buff-Ring 16, 35392 Giessen, Germany*
[3]*Warsaw University, Hoza 69, 00-681 Warszawa, Poland*
[4]*Soltan Institute for Nuclear Studies, Hoza 69, 00681 Warsaw, Poland*
[5]*St. Petersburg Nuclear Physics Institute, 188300 Gatchina, Russia*
[6]*Technische Universität München, 85748 Garching, Germany*
[7]*Stefan Meyer Institut für subatomare Physik, 1090 Vienna, Austria*
[8]*Joint Institut for Nuclear Research, 141980 Dubna, Russia*
[9]*Michigan State University, East Lansing, Michigan 48824-1321, USA*
[10]*Manipal Academy of Higher Education, Manipal, KA 576104, India*
[11]*INFN-Laboratori Nazionali del Sud, I95123 Catania, Italy*
[12]*Universita di Catania, I95123 Catania, Italy*
[13]*St. Petersburg State University, 198504 St. Petersburg, Russia*
[14]*Peking University, Beijing 100871, China*

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We report on the first measurement of the β^+ and orbital electron-capture decay rates of ^{140}Pr nuclei with the simplest electron configurations: bare nuclei, hydrogenlike, and heliumlike ions. The measured electron-capture decay constant of hydrogenlike $^{140}\mathrm{Pr}^{58+}$ ions is about 50% larger than that of heliumlike $^{140}\mathrm{Pr}^{57+}$ ions. Moreover, ^{140}Pr ions with one bound electron decay faster than neutral $^{140}\mathrm{Pr}^{0+}$ atoms 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.

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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 [1,2]. 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 [3,4].

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 [5,6]. 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 (EC) and electron conversion decays become impossible in the absence of orbital electrons, i.e., in fully ionized atoms. Thus, the pure β^+-decay branch has been measured in $^{52}\mathrm{Fe}^{26+}$ ions [8] and the half-lives of isomeric states were found to be dramatically prolonged [9].

Bare $^{187}\mathrm{Re}^{75+}$ ions decay, due to the new decay mode—the bound-state β decay—by 9 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 $^{163}\mathrm{Dy}^{66+}$ 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 β decay to the continuum and bound states in $^{207}\mathrm{Tl}^{81+}$ ions has been performed recently [12].

In the present experiment the β^+ and orbital EC decays of bare nuclei and nuclei with one and two bound electrons have been investigated. The EC decay rates in hydrogenlike and heliumlike 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 β 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 ($p + e^- \longrightarrow n + \nu_e$), or via a three-body decay in which the positron and the neutrino share the decay energy ($p \longrightarrow n + e^+ + \nu_e$).

The experiment has been performed at the Gesellschaft für Schwerionenforschung (GSI), Darmstadt, Germany, where the combination of the heavy-ion synchrotron SIS [14], the in-flight fragment separator FRS [15], and the ion storage-cooler ring ESR [16] provides unique experimental conditions for decay studies of bare and few-electron exotic nuclei in an ultrahigh 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 [7–12]. Radioactive ^{140}Pr ions have been produced via the projectile fragmentation of $\sim 3 \times 10^9$ ^{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, hydrogenlike, and heliumlike ^{140}Pr ions were separated in flight by a twofold magnetic rigidity analysis by means of the $B\rho$-ΔE-$B\rho$ separation method [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 detail in Ref. [17].

Stochastic [18] and electron cooling [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 precooling at a fixed fragment velocity, corresponding to 400 MeV/u energy, thus reducing the overall cooling time to about 2 sec. The cooling forces all stored ions to the same mean velocity and reduces the initial velocity spread, caused by the fragmentation reaction, to $\delta v/v \approx 5 \times 10^{-7}$.

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 [20,21]. The latter is based on Schottky-noise spectroscopy [22], which is widely used for nondestructive 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 pickup 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 [8–12]. The details of the data acquisition system and of the data treatment can be found in Ref. [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 (31st harmonics). An example for the ^{140}Pr$^{58+} + e^- \longrightarrow$ ^{140}Ce$^{58+} + \nu_e$ decay is illustrated in Fig. 1, where 195 subsequent Schottky frequency spectra are plotted as a water-flow diagram. Each spectrum is averaged over 10.5 sec. It can be seen in Fig. 1 that the intensity of the peak at lower revolution frequency—corresponding to the

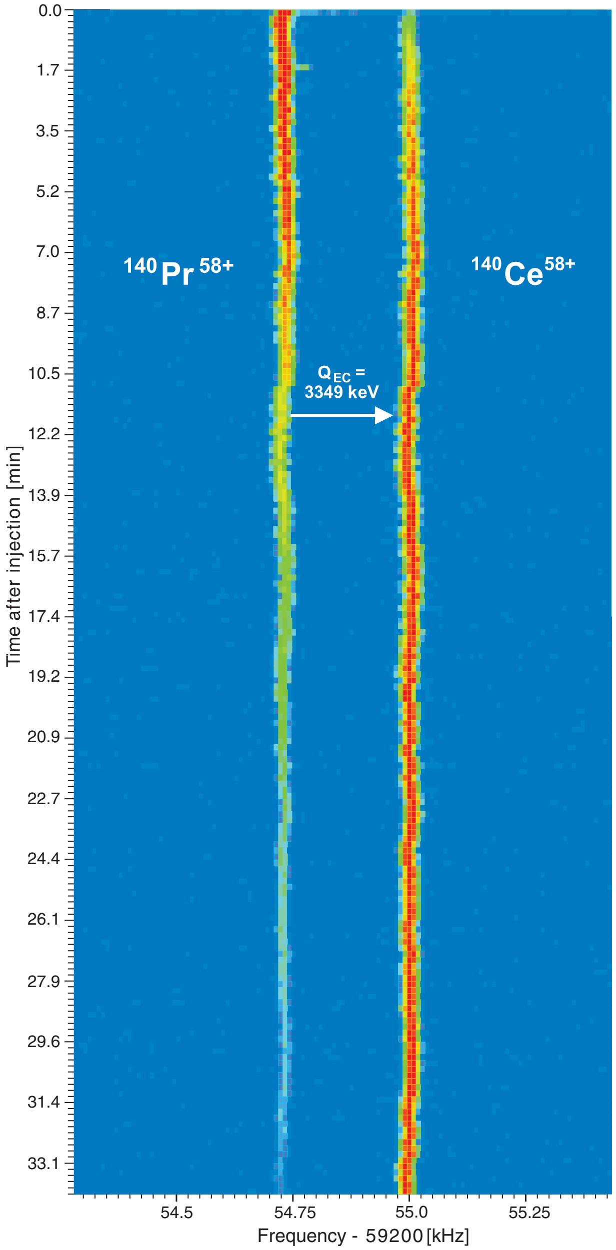

FIG. 1 (color). Schottky frequency spectra at the 31st harmonics of the revolution frequency taken subsequently as a function of time (195 spectra of 10.5 sec each). In the EC decay of hydrogenlike ^{140}Pr, the mass changes by 3.349 MeV/c^2 which leads to a small change in the frequency ($\sim$270 Hz). 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.

parent ions $^{140}\mathrm{Pr}^{58+}$—decreases steadily and that the intensity of the peak at the higher frequency—corresponding to the lighter daughter ions $^{140}\mathrm{Ce}^{58+}$—increases. Examples of the decay and growth curves are shown in Fig. 2. Feeding of $^{140}\mathrm{Pr}^{58+}$ or $^{140}\mathrm{Ce}^{58+}$ 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 $^{140}\mathrm{Pr}^{59+}$, $^{140}\mathrm{Pr}^{58+}$, and $^{140}\mathrm{Pr}^{57+}$ ions have been performed. Decay curves of the parent ions have been fitted with an exponential function:

$$N_{\mathrm{Pr}}(t) = N_{\mathrm{Pr}}(0)e^{-\lambda t}, \tag{1}$$

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

$$N_{\mathrm{Ce}}(t) = N_{\mathrm{Pr}}(0)\frac{\lambda_{\mathrm{EC}}}{\lambda - \lambda_{\mathrm{loss}}}[e^{-\lambda_{\mathrm{loss}}t} - e^{-\lambda t}] + N_{\mathrm{Ce}}(0)e^{-\lambda_{\mathrm{loss}}t}. \tag{2}$$

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

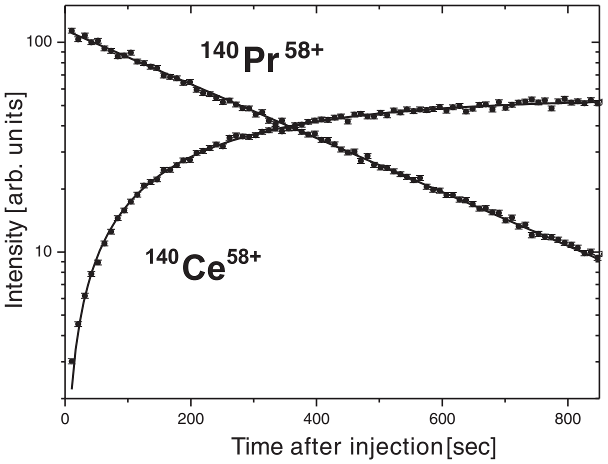

FIG. 2. Decay and growth curves of $^{140}\mathrm{Pr}^{58+}$ and $^{140}\mathrm{Ce}^{58+}$ ions as a function of time. The data points are shown in the laboratory frame and can be converted to the rest frame of the ions using the Lorentz factor $\gamma = 1.43$. The lines represent the fits according to Eqs. (1) and (2).

As can be seen from Table I, the measured β^+ decay rate is within the errors independent on the degree of ionization. This is expected, since the electron screening modifies the β^+ rate by less than 3% in fully ionized ions compared to neutral atoms [23].

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 $^{140}\mathrm{Pr}$ in Refs. [25–27] and can be compared with our measurement on the heliumlike ions. Using the values for the $^{140}\mathrm{Pr}^{57+}$ ions we obtain $\lambda_{\mathrm{EC}}/\lambda_{\beta^+} = 0.95(8)$, 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 heliumlike 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 $^{140}\mathrm{Pr}^{57+}$ ions to only one in $^{140}\mathrm{Pr}^{58+}$ ions—that the EC rate *increases* by a factor of 1.49(8). Moreover, the half-life of $^{140}\mathrm{Pr}^{58+}$ 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 $^{140}\mathrm{Pr}^{0+}$ atoms with 59 orbital electrons.

Our result can be explained by taking into account the conservation of the total angular momentum of the nucleus-lepton system. We note that similar arguments have been used in Refs. [3,29] to explain the deexcitation 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 the $^{140}\mathrm{Pr}$ nucleus with spin $I_i = 1$ and a single bound K electron with spin $s = 1/2$ can have two values of the hyperfine states, $F_i = I_i - s = 1/2$, if the spins of the nucleus and the electron are antiparallel, or $F_i = I_i + s = 3/2$ if the spins are parallel, as schematically illustrated in Fig. 3. In the final state (f), however, the total angular momentum can have only one value, $F_f = 1/2$, which is the sum of the zero angular momentum of the $^{140}\mathrm{Ce}$ nucleus $I_f = 0$ [13] and of the spin $s = 1/2$ of the emitted

TABLE I. Measured β^+ and EC decay constants obtained for fully ionized, hydrogenlike, and heliumlike $^{140}\mathrm{Pr}$ ions. The values are given in the rest frame of the ions.

Ion	λ_{β^+} (sec^{-1})	λ_{EC} (sec^{-1})
$^{140}\mathrm{Pr}^{59+}$	0.001 58(8)	⋯
$^{140}\mathrm{Pr}^{58+}$	0.001 61(10)	0.002 19(6)
$^{140}\mathrm{Pr}^{57+}$	0.001 54(11)	0.001 47(7)

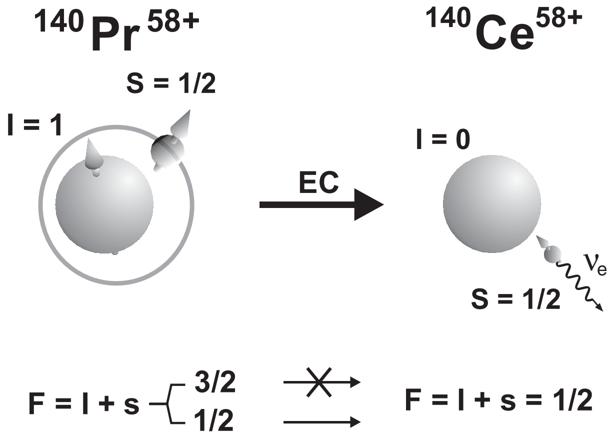

FIG. 3. Illustration of the EC decay of hydrogenlike $^{140}\mathrm{Pr}^{58+}$ ions to bare $^{140}\mathrm{Ce}^{58+}$ ions.

electron-neutrino. Hence, only transitions from the $F_i = 1/2$ hyperfine state can contribute to the decay to the final state. The decay from the $F_i = 3/2$ state would require that the emitted neutrino carries away two units of orbital angular momentum, which corresponds to a much slower (twice forbidden) β decay.

The $F_i = 1/2$ assignment to the lowest hyperfine state of $^{140}\mathrm{Pr}^{58+}$ follows from the positive magnetic moment μ of ^{140}Pr which has been deduced from the known magnetic moments of the neighboring odd-A nuclei of about $+2.5\mu_N$. For hydrogenlike ^{140}Pr, the relaxation time for the upper hyperfine state to the ground state ($\tau \approx 0.03$ s) is much shorter than the cooling time [31]. Electric and magnetic fields in the ring can, in principle, lead to a repopulation of the upper hyperfine level. Such repopulation, however, has not been observed in ESR experiments [32]. Thus, $^{140}\mathrm{Pr}^{58+}$ ions are dominantly stored in the pure $F_i = 1/2$ quantum state.

If a nucleus has $\mu < 0$ then the lower hyperfine state of the hydrogenlike ion is $F_i = I_i + s$. For instance, for hydrogenlike ^{64}Cu ions [$\mu = -0.217(2)\mu_N, I_i = 1$] [33], the ground state is $F_i = 3/2$ and it does not decay by an allowed EC decay to the ground state of ^{64}Ni ($I_f = 0$).

The influence of the hyperfine state of the electron on the EC decay rate at different temperatures has been investigated theoretically in Ref. [34], where significant changes in the decay rates have been predicted. The detailed theoretical description of our results is given in Ref. [35]. This work provides a systematic study of EC decay rates for hydrogenlike and heliumlike ions dependent on the nuclear spins. In the case of ^{140}Pr only one F_i state contributes to the decay. Then the EC decay rate depends on the ratio of the statistical weights of the transition, i.e., $(2I_i + 1)/(2F_i + 1) = 3/2$, which is in excellent agreement with our experimental result.

In summary, our experimental results have clearly revealed a fundamental property of β decay of highly ionized atoms, which could not 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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