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# Observation of the $2p_{3/2} \rightarrow 2s_{1/2}$ intra-shell transition in He-like uranium

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**Abstract.** - We present the first observation of the  $1s2p\ ^3P_2 \rightarrow 1s2s\ ^3S_1$  transition in He-like uranium. The experiment was performed at the internal gas-jet target of the ESR storage ring at GSI exploiting a Bragg crystal spectrometer and a germanium solid state detector. Using the  $1s^22p\ ^2P_{3/2} \rightarrow 1s^22s\ ^2S_{1/2}$  transition in Li-like uranium as reference and the deceleration capabilities of the ESR storage rings, we obtained the first evaluation of the He-like heavy ion intra-shell transition energy.

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He-like ions are the simplest multi-body atomic systems. Investigations of these ions along their isoelectronic sequence up to the heaviest species uniquely probe our understanding of correlation, relativistic, and Quantum Electrodynamics (QED) effects. In recent years, substantial progress in the investigations of these fundamental systems has been achieved in the high- $Z$  region, in which the nucleus Coulomb field strength and electron velocity, both proportional to  $Z\alpha$ , are very high. In theory, benchmark calculations have been reported where even second order QED effects were considered in a rigorous way for both the ground-state as well as for the first excited states [1–3]. Experimentally, progress has been mainly achieved by a technique recently introduced [4], which allows for the iso-

lation of the two-electron contributions to the ionization potential of the ground-state [4–6]. Here, the achieved uncertainty already approaches the expected size of higher-order QED contributions [5, 6]. However, for the excited levels at high- $Z$  ( $Z > 54$ ) virtually no experimental data on binding or transition energies are available. Besides the general importance of such data for atomic structure investigations, the great interest in more detailed information about the excited levels in high- $Z$  He-like ions is motivated by their relevance for the study of the influence of the electroweak interaction on the atomic structure. Since many years, high- $Z$  He-like ions are under the discussion as an ideal candidates for the study of atomic parity violation effects [7, 8]. The ability to observe parity-violating transitions critically depends on the relative binding energies of the excited states. A level crossing of two states

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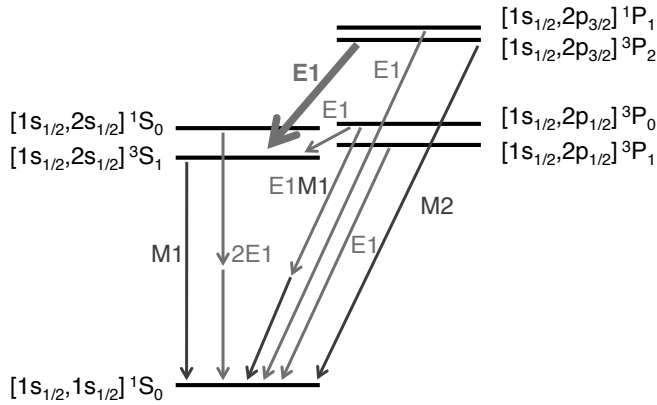


Fig. 1: Level scheme of helium-like uranium and major decay branches. The bold arrow indicates the transition studied.

with different parity, namely the  $1s2p\ ^3P_0$  and the  $1s2s\ ^1S_0$  levels, is predicted to occur close to  $Z = 66$  and  $Z = 92$ . However, a benchmark test of the atomic structure theory for the excited states in He-like ions is still pending. Therefore, a precise measurement of the  $\Delta n = 0$   $1s2p\ ^3P_2 \rightarrow 1s2s\ ^3S_1$  intra-shell transition energy is of particular importance. (a partial level scheme of a high- $Z$  He-like ion is shown in Fig. 1). An attempt of an energy measurement was reported for the  $1s2p \rightarrow 1s2s$  intra-shell transition for He-like uranium using the electron-beam ion trap (EBIT) at Lawrence Livermore National Laboratory (LLNL) [9]. There, the desired  $1s2p\ ^3P_2 \rightarrow 1s2s\ ^3S_1$  transition could not be identified unambiguously. This is in contrast to lighter ions up to  $Z = 54$ , where energy measurements of the  $1s2p \rightarrow 1s2s$  transitions have been performed by spectroscopy in the visible to the far UV region [3, 10, 11]. For higher values of  $Z$ , information on excited states has been obtained only via lifetime measurements of the  $1s2p\ ^3P_0$  level [12–14].

In this Letter we present the first clear identification of the  $1s2p\ ^3P_2 \rightarrow 1s2s\ ^3S_1$  intra-shell transition in He-like uranium and its energy measurement. For the purpose of the experiment, we used a standard Ge(i) solid-state detector and a new Bragg spectrometer specially designed for accurate spectroscopy of fast ions. The two instruments are complementary: The Ge(i) detector has a high detection efficiency and covers a wide spectral range with a moderate spectral resolving power. The focussing crystal spectrometer serves as an accurate wavelength comparator in a narrow wavelength interval. In concert, the present measurements allow for an unambiguous identification of the spectral lines observed plus an accurate determination of the transition wavelength or energy.

The experiment was performed at the ESR storage ring at the GSI in Darmstadt. Here, a beam of  $\sim 4 \times 10^7$  hydrogen-like uranium ions was stored, cooled and decelerated to an energy of 43.57 MeV/u. The ion beam's momentum spread was close to  $\Delta p/p \approx 10^{-5}$ , and its width

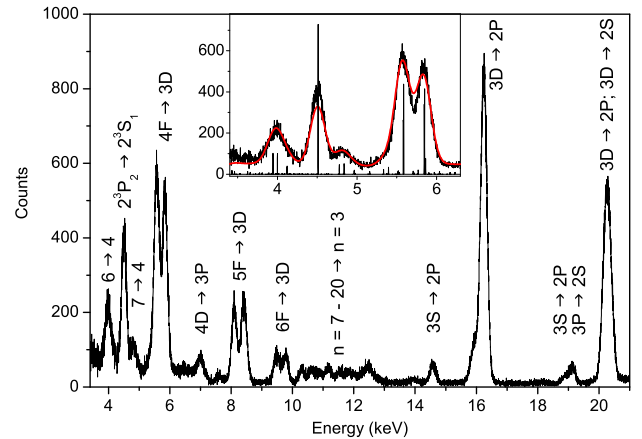


Fig. 2: The x-ray spectrum (color online), originating from 43.57 MeV/u He-like uranium ions, as recorded by the Ge(i) detector. The energies correspond to the emitter frame. The inset shows a zoom around the  $^3P_2 \rightarrow ^3S_1$  transition with the vertical bars and the continuous (red) curve representing the results of a full cascade calculation [19].

was about 2 mm. He-like excited ions were formed by electron capture during the interaction of the ion beam with a supersonic nitrogen gas-jet. The gas-jet had a width of about 5 mm and a typical areal density of  $10^{12}$  particles/cm<sup>2</sup>, which guaranteed single-collision conditions in the ion–target interaction. At the selected velocity, electrons are primarily captured into the shells with principal numbers  $n \leq 20$  [15–17]. This allows for an efficient population of the  $1s2p\ ^3P_2$  level via cascade feeding.

The  $1s2p\ ^3P_2$  excited state mainly decays in two ways (Fig. 1): to the ground state via a magnetic quadrupole (M2) transition, with a branching ratio of 70% and a decay time of  $4.9 \times 10^{-15}$  sec, and to the  $1s2s\ ^3S_1$  state by an electric dipole (E1) intra-shell transition, with a branching ratio of 30% and a decay time of  $1.2 \times 10^{-14}$  sec [18]. In the reference frame of the projectile, the x-ray photons arising from the  $2^3P_2 \rightarrow 2^3S_1$  transition have an energy of 4510 eV.

The Ge(i) solid state detector and the Bragg crystal spectrometer were mounted under observation angles of  $35^\circ$  and  $90^\circ$ , respectively. Both instruments were separated from the ultra-high vacuum of the gas target chamber by  $100\mu\text{m}$ -thick beryllium windows transparent for the low-energy x rays.

The Ge(i) detector crystal has a diameter of 16 mm and therefore provides a relatively large solid angle. An x-ray collimator was mounted in front of the Ge(i) in order to limit the horizontal acceptance angle thereby reducing the Doppler broadening to match the corresponding Doppler width with the intrinsic line width of the detector, amounting to 250 eV at 5.9 keV photon energy.

A survey spectrum recorded by the Ge(i) detector is displayed in Fig. 2 with prominent lines originating from  $n' \rightarrow n$  transitions, with  $n = 2 - 4$ . The existence of

lines originating from high  $n'$  to  $n = 2$  indicates that the  $^3P_2$  state is mainly populated by cascade feeding which is also supported by a theoretical study [15]. The inset in Fig. 2 shows a zoom of the low-energy region of the spectrum. The vertical lines in the inset mark the line energies and intensities obtained by a full cascade calculation [19] whereas the continuous (red) line represents a fit of the theoretical model taking into account the experimental energy resolution. The good over-all correspondence between experiment and model gives confidence for the correct identification of the strong line at the expected energy of the  $2^3P_2 \rightarrow 2^3S_1$  transition.

The measurement using the crystal spectrometer provided a much higher accuracy for the spectral line position. In this case, the observable energy range, principally limited by the ion beam diameter and its distance from the crystal, was in the order of  $4308 \pm 40$  eV – much narrower than the linewidth observed with the Ge(i). For an unambiguous identification of the He-like uranium intra-shell transition, the experimentally well known  $1s^2 2p^2 P_{3/2} \rightarrow 1s^2 2s^2 S_{1/2}$  transition [20,21] in Li-like uranium has been used as reference. Similar to the He-like system, Li-like ions were obtained by electron capture into He-like uranium ions. In order to park the Li-like transition close to the He-like line, we used the Doppler effect selecting a kinetic energy of 32.63 MeV/u for Li-like ions (to be compared with 43.57 MeV/u for He-like ones). This way both Li- and He-like uranium transitions appeared at nearly the same Bragg angle and consequently in the same narrow spatial region of the position-sensitive detector.

The crystal spectrometer was mounted in the Johann geometry for the detection of x rays with a corresponding Bragg angle  $\Theta_B$  around  $46.0^\circ$ . A cylindrically bent germanium (220) crystal with size of  $50 \times 25$  mm<sup>2</sup> and a radius of curvature of 800 mm was installed. The spectrometer did not need any collimation because the imaging properties of the curved crystal were used to resolve spectral lines from fast x-ray sources nearly as well as for stationary sources [22]. For this purpose it was necessary to place the Rowland-circle plane of the spectrometer perpendicular to the ion-beam direction. In such a configuration the spectral lines appear slanted in the image plane of the spectrometer with their slope proportional to the ion-beam velocity [22–24].

Photons diffracted off the crystal were detected with a windowless x-ray charge-coupled device (CCD), Andor DO420, placed at  $D = 575.75 \pm 0.65$  mm away from the crystal, corresponding to a Bragg-angle setting of  $\Theta_B = 46.0^\circ$ . The spectrometer was kept under vacuum conditions (residual gas pressure  $\sim 10^{-5}$  mbar) to assure proper working conditions for the CCD camera and to keep x-ray absorption low. The interaction volume between the ion beam and the nitrogen jet was located  $758.05 \pm 0.46$  mm away from the crystal, *i.e.* about 182 mm outside of the Rowland circle. The ion velocity was selected to observe the x-ray radiation under study with a photon energy of about 4308 eV in the laboratory frame, *i.e.* in the vicinity

of the 8.6 keV  $K\alpha_{1,2}$  lines of zinc observed in the second order of diffraction. At the same time, this matches the spectrometer configured to a Bragg angle of  $46^\circ$ . The zinc lines, produced by fluorescence with a commercial x-ray tube and a zinc target, were used as calibration and for stability controls. For this purpose a zinc plate was mounted on a removable support between the target chamber and the crystal. In this configuration, the spectrometer had an efficiency of  $\sim 7 \times 10^{-7}$  and a resolution of  $\sim 2$  eV for the first order Bragg reflection.

The data were acquired during a total period of about 4.5 days. To check for the stability of the spectrometer, daily calibrations with the zinc target were performed. During each accelerator cycle – consisting of injection into the ESR, cooling, deceleration, cooling, measurement – the CCD acquired data for 25 and 50 seconds for Li- and He-like U, respectively. Over the whole experimental run, the spectrometer was very stable, with a shift of  $\pm 3$   $\mu$ m of the zinc-line reflections on the CCD (corresponding to  $\pm 0.2$  pixels). For the transition in He-like uranium, a total number of about 300 counts in a net-time of  $\sim 24$  hours was accumulated. For the Li-like ions, about 160 counts in a net-time of  $\sim 5$  hours were recorded. As can be observed in Fig. 3, the two intra-shell transitions could be identified unambiguously. These spectra are characterized by a very low background constrained only by the energy cuts and cluster analysis of the CCD raw data. No particular shields, other than the spectrometer stainless steel and aluminum structure, were used. We note that the shape of the lines corresponding to the fast ion emission is slightly asymmetric. Such an asymmetry is, at present, not well understood. No relevant satellite transitions, due to a possible but improbable electron double capture [25], or parasitic fluorescence lines could be found in this energy region, not even when higher reflection orders are considered. Atomic cascade effects, like feeding and decaying time of the  $1s 2p^3 P_2$  level, which, due to the Doppler effect, could introduce an asymmetry in the x-ray energy distribution, are also unlikely to happen [15]. How far a non-uniform x-ray reflectivity over the crystal surface<sup>1</sup> could cause irregularities in the spectral response of the spectrometer is difficult to judge.

Using the Li-like intra-shell transition as calibration, with energy  $E_{Li} = 4359.37 \pm 0.21$  eV [20,21], the energy of the He-like  $2^3P_2 \rightarrow 2^3S_1$  transition could be evaluated. This reference line has been chosen instead of the  $K\alpha_{1,2}$  zinc lines to reduce the systematic uncertainty. The accuracy of the observation angle, equal to  $0.04^\circ$ , is due to the spatial uncertainty of the fast-beam x-ray source defined by the gas-jet and ion-beam positions. This caused a systematic uncertainty of about  $\pm 0.9$  eV on the fast-ion transition-energy measurement when the Zn  $K\alpha$  lines are used as reference. This problem can be circumvented by using a calibration line originating from the fast ion beam

<sup>1</sup>After the experiment, the crystal surface was surveyed by the x-ray optics group from the Institute for Optics and Quantum Electronics in Jena.

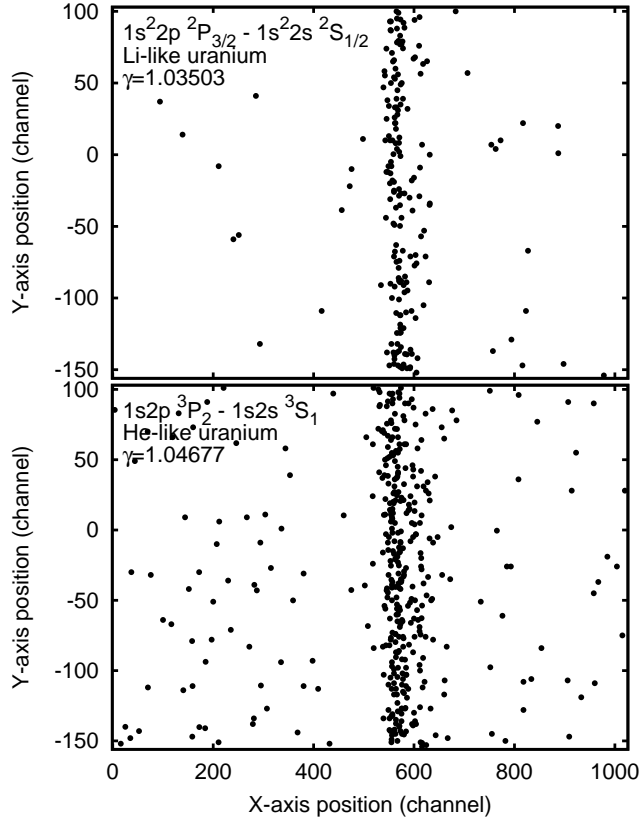


Fig. 3: Reflection of the Li-like (up) and He-like (down) uranium intra-shell transitions on the Bragg spectrometer CCD. The transition energy increases with the increasing of x-position. The slightly negative slope of the line is due to the relativistic velocity of the fast ions.

rather than from a stationary source.

Starting from Bragg's law in differential form,  $\Delta E \approx -E \cot \Theta_B \Delta \Theta_B$ , one obtains an approximate dispersion formula valid for small  $\Delta \Theta_B$ . The energy of the He-like  $2^3P_2 \rightarrow 2^3S_1$  transition  $E_{\text{He}}$  is given by the simple formula

$$\frac{E_{\text{He}}}{\gamma_{\text{He}}} \approx \frac{E_{\text{Li}}}{\gamma_{\text{Li}}} \left( 1 + \frac{\Delta x}{D \tan \Theta_B} \right), \quad (1)$$

where  $\Delta x = x_{\text{He}} - x_{\text{Li}}$  denotes the distance between the He- and Li-like U line images on the CCD and  $D$  the distance between crystal and CCD.  $\gamma_{\text{He}} = 1.04677$  and  $\gamma_{\text{Li}} = 1.03503$  are the Lorentz factors corresponding to the velocities of stored H- and He-like ions, respectively. Their values are determined by the accurately known voltages of the electron cooler.

More complicated is the evaluation of the distance  $\Delta x$  between the two spectral lines from the fast ions, characterized by a slope due to the Doppler effect [23]. Such a slope could not be determined experimentally due to the low statistics and it was calculated from simple geometrical considerations. The measured two-dimensional images have been rotated by the calculated slope before projecting them on the spectrometer dispersion plane and measure

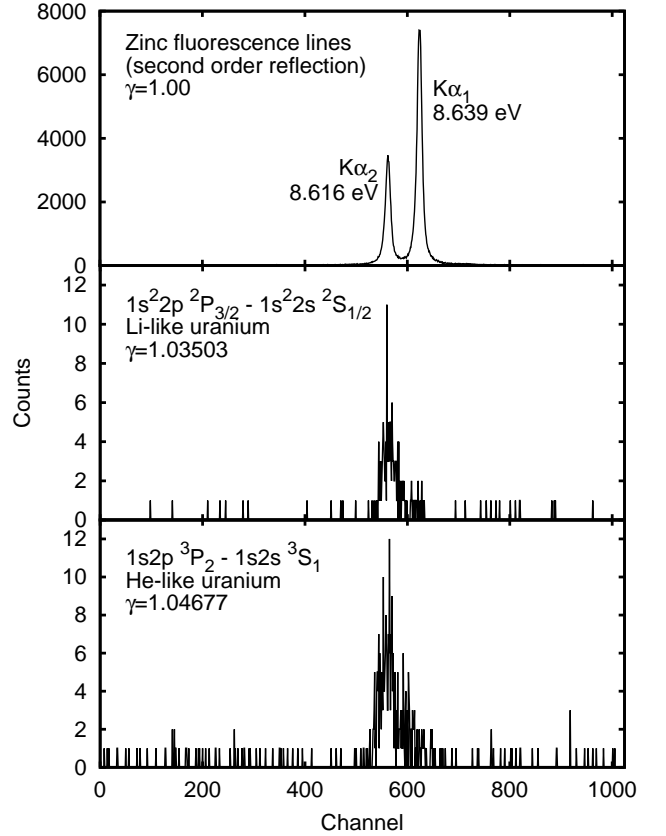


Fig. 4: X-ray spectra measured with the crystal spectrometer: Zn  $K\alpha$  doublet in second order (top) and the intra-shell transitions of Li and He-like U (middle and bottom, respectively).

the lines relative distance. The result of such projections is presented in Fig 4.

Due to the low statistics, no accurate investigation of the peaks asymmetry could be performed. The systematic effect induced by this asymmetry has been estimated by comparing the line position measurements obtained from different approaches. For this propose, a robust statistical analysis of the data set, like the median distribution, and a series of fit adjustment have been applied. The median value was found in a self-consistent way regarding varying spectral intervals around the line center. The fit adjustments were obtained via a likelihood function maximization for Poisson histograms [26], well adapted for low-statistics data sets, using different distributions such as Gauß-Cauchy curve, Gaussian convoluted with an exponential and a sum of two Gaussian. From this analysis, a scattering of  $\approx 110 \mu\text{m}$  of the  $\Delta x$  value was measured, with a corresponding statistical uncertainty of about  $65 \mu\text{m}$ . The value of  $\Delta x = +47 \mu\text{m}$ , obtained via the median position calculation, has been considered as reference for further calculations. The energy of the  $1s 2p^3 P_2 \rightarrow 1s 2s^3 S_1$  transition was measured to be

$$E_{\text{He}} = 4509.71 \pm 0.48_{\text{stat}} \pm 0.83_{\text{asym}} \pm 0.24_{\text{syst}} \text{ eV}, \quad (2)$$

The first uncertainty is statistical, and the second one is

Table 1: Comparison of the experimental transition energy, in eV, for the  ${}^3P_2 \rightarrow {}^3S_1$  transition in He-like uranium with various theoretical predictions. The experimental uncertainty is obtained from the quadratic sum of the different uncertainties listed in Eq. (2).

Experiment	$4509.71 \pm 0.99$
Theory	
Indelicato 2008 [28]	4510.30
Kozhedub 2008 [29]	$4509.86 \pm 0.07$
Artemyev 2005 [3]	$4510.03 \pm 0.24$
Plante 1994 [2]	4510.46
Chen 1993 [30]	4510.65
Drake 1988 [31]	4510.01

due to the peak asymmetry. The third term takes other sources of systematic errors into account, of which the limited uncertainty of the calibration line (0.21 eV) is the dominant one. The uncertainty of the observation angle gives a contribution of  $\pm 0.11$  eV, drastically reduced compared to  $\pm 0.9$  eV when a stationary calibration source is used. Other systematic uncertainties, including those originating from the beam velocity, are negligible (see [27]).

As presented in Table 1, the measured transition energy for He-like uranium agrees well with all theoretical predictions, which, however, reflect different approaches. The value provided by P. Indelicato [28] represents a Multiconfiguration Dirac-Fock (MCDHF) calculation, obtained with the code developed by P. Indelicato and J.-P. Desclaux [18]. Radiative QED corrections  $E_{\text{rad}}^{\text{QED}}$  to the inter-electronic interaction, *i.e.* QED screening effects, are taken into account via the self-consistent treatment of the one-loop vacuum polarization and the Welton approximation for the screened self-energy. The value obtained by Artemyev et al. in [3] is calculated *ab initio*. In [3] all QED corrections of first and second order in  $\alpha$  are taken into account within the rigorous framework of QED. In addition to [28] the non-radiative QED contribution to the inter-electronic interaction  $E_{\text{non-rad}}^{\text{QED}}$ , which is the difference between the contribution of the inter-electronic interaction diagrams calculated within the rigorous QED approach and within the Breit approximation, is calculated. Later in [29] the value from [3] has been improved using a new value of the mean-square nuclear-charge radius for uranium [32] and the two-loop one-electron QED corrections for the excited states calculated in [33, 34]. Also in [29] the non-QED contribution of three- and more photon exchange diagrams has been calculated using the relativistic configuration-interaction method. Two-electron QED effects contribute to the  ${}^3P_2 \rightarrow {}^3S_1$  transition energy with  $E_{2\text{el}}^{\text{QED}} = E_{\text{rad}}^{\text{QED}} + E_{\text{non-rad}}^{\text{QED}} = 0.76$  eV [3], which is close to the present experimental uncertainty.

In addition to the absolute energy measurement of the  $2{}^3P_2 \rightarrow 2{}^3S_1$  transition, the relative measurement of He- and Li-like uranium intra-shell transitions could be evalu-

Table 2: Experimental and theoretical energy difference between He- and Li-like intra-shell transitions in eV. Contributions from one- and two-photon exchange diagram,  $E_{\text{Breit}}$ , and from two-photon QED,  $E_{\text{rad}}^{\text{QED}}$  and  $E_{\text{non-rad}}^{\text{QED}}$ , are presented. The experimental uncertainty is obtained from the quadratic sum of the different uncertainties.

	$E_{\text{Breit}}$	$E_{\text{rad}}^{\text{QED}}$	$E_{\text{non-rad}}^{\text{QED}}$	Total
Experiment				
$50.34 \pm 0.96$				
Theory				
Indelicato [28]	51.61	-1.66		49.96
Kozhedub [29]	51.48	-1.14	-0.04	$50.30 \pm 0.03$

ated. In contrast with the previous case, the experimental systematic uncertainty, peak asymmetry excluded, reduced from 0.24 to 0.11 eV. Theoretical uncertainties due to the finite nuclear size and the one-electron QED effects, are also drastically reduced. A comparison between our value and different predictions is presented in Table 2. As we can observe, all theoretical values agree with the experimental value within one standard deviation.

In summary, we report the first clear identification of the  $1s2p{}^3P_2 \rightarrow 1s2s{}^3S_1$  transition in He-like uranium. In addition we could measure the transition energy of such transition with a relative uncertainty of  $2 \times 10^{-4}$ , which is currently the most accurate test of many-body and QED contributions in excited levels of very heavy He-like ions. Differential measurements between different charge states of the same fast ion pave the way for increased sensitivity via the reduction of the systematic uncertainty in both experimental and theoretical sides. In the present experiment, the accuracy was principally limited by the statistics and the observed peak asymmetry of the fast-beam spectra. Hence, the improvement of the experimental setup and an increasing of the acquisition time will allow in future experiments for more stringent tests of the two-electron QED contributions in heavy highly charged ions.

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