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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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We present the first identification and energy measurement 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 by means of a Bragg crystal spectrometer. Using the  $1s^2 2p\ ^2P_{3/2} \rightarrow 1s^2 2s\ ^2S_{1/2}$  transition in Li-like uranium as reference, we obtained an accuracy in the order of 0.01%, which provides, at present, the most accurate test of many-body and QED contributions in very heavy He-like ions.

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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 for the high- $Z$  regime. 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, 2, 3]. Experimentally, the progress achieved is documented by a technique recently introduced [4], which allows to isolate the two-electron contributions to the groundstate ionization potential [4, 5, 6]. Here an accuracy has been achieved which 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. Beside 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 ideal candidates to study parity violation in atomic effects [7, 8]. A level crossing of two states 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 atomic structure theory for the excited states in He-like ions is still pending. For this purpose the  $\Delta n = 0$   $1s2p\ ^3P_2 \rightarrow 1s2s\ ^3S_1$  intrashell transition, appears to be of particular relevance (a partial level scheme of a high- $Z$  He-like ion is shown in Fig. 1). An attempt of an energy measurement was re-

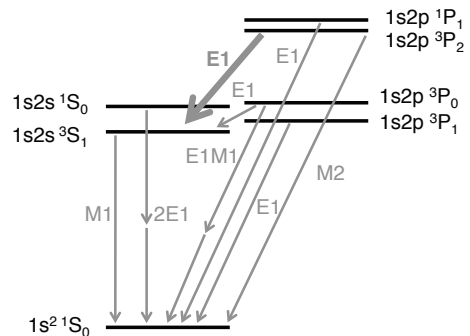


FIG. 1: Level scheme of helium-like uranium and major decay branches. The bold arrow indicates the transition studied.

ported for the  $1s2p \rightarrow 1s2s$  intra-shell transitions for He-like uranium using the electron-beam ion trap (EBIT) at Lawrence Livermore National Laboratory (LLNL) [14]. 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, 9, 10]. However, for higher values of  $Z$ , valuable information on excited states have been obtained only via lifetime measurements of the  $1s2p\ ^3P_0$  level [11, 12, 13].

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 large spectral range with a moderate spectral resolving power. The focussing crystal spectrometer serves as an accurate wave-

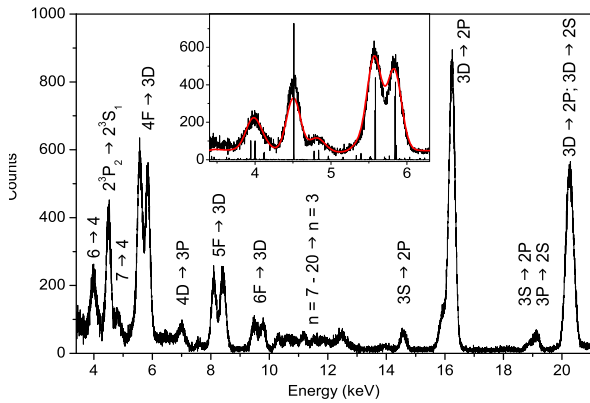


FIG. 2: The x-ray spectrum (color online), originating from 43.57 MeV/u He-like uranium ions, as recorded by the Ge(i) detector. The energies correspond to the emitter frame. The inset shows a zoom around the  ${}^3P_2 \rightarrow {}^3S_1$  transition with the vertical bars and the continuous (red) curve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

length 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 high accuracy of the experiment achieved,  $\Delta E/E \approx 10^{-4}$ , enables us to test, for the first time, the QED contributions of the electron–electron interaction for excited states in a very high- $Z$  He-like ion.

The experiment was performed at the ESR storage ring at the GSI in Darmstadt. Here a hydrogen-like uranium beam with up to  $10^8$  ions was stored, cooled and decelerated to an energy of 43.57 MeV/u with a momentum spread of close to  $\Delta p/p \approx 10^{-5}$ . He-like excited ions were formed by electron capture during the interaction of the ion beam with a supersonic nitrogen 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 a principal numbers  $n \leq 20$  [15, 16, 17] allowing for an efficient population of the  $1s2p\,{}^3P_2$  level via cascade feeding. This 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 to the  $1s2s\,{}^3S_1$  state by an electric dipole, E1, intra-shell transition, with a branching ratio of 30% [18]. In the emitter frame of reference the x-ray photons arising from the  $2\,{}^3P_2 \rightarrow 2\,{}^3S_1$  transition have an energy of 4510 eV.

The x rays were simultaneously detected by the Ge(i) solid state detector mounted under an observation angle of  $35^\circ$  relative to the ion-beam direction and by the Bragg crystal spectrometer mounted at  $90^\circ$ . Both instruments were separated from the ultra-high vacuum of the gas target chamber by 100  $\mu$ m-thick beryllium windows transparent for the low-energy x rays.

The crystal inside the Ge(i) detector 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. The count rate of the crystal spectrometer was found to be consistent with the count rate of the Ge(i) detector taking into account the estimated detection efficiencies and relativistic transformations of the solid angles at  $35^\circ$  and  $90^\circ$ .

A survey spectrum 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 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 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 an extension 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 [20]. 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 [20, 21, 22].

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  $\Theta_B = 46.0^\circ$ . A vacuum pressure of about  $10^{-5}$  mbar was maintained inside the spectrometer 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 at  $758.05 \pm 0.46$  mm from the crystal, i.e., about 182 mm outside the Rowland circle. The ion velocity was selected such as to Doppler shift the x-ray radiation under study to a photon energy of about 4308 eV in the laboratory frame, i.e., into 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 propose a zinc plate was mounted in a removable support between the target chamber and the crystal. In this configuration, the spectrometer had an efficiency of  $\sim 8 \times 10^{-7}$  and a resolution of  $\sim 2$  eV.

The uncertainty of the observation angle of  $\pm 0.04^\circ$  originates from 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 rather than from a stationary source. For this purpose the  $1s^2 2p^2 P_{3/2} \rightarrow 1s^2 2s^2 S_{1/2}$  transition in Li-like uranium has been used, accurately measured at the LLNL EBIT [23, 24]. Similar to the He-like system, Li-like ions were obtained by electron capture into He-like uranium ions. In order to Doppler shift the line of the Li-like ions close to where we observed the He-like ions, a kinetic energy of 32.63 MeV/u was required. Detecting both Li- and He-like uranium transitions in the same narrow spatial region of the CCD detector, i.e., at nearly the same Bragg angle, resulted in a drastic reduction of systematic effects such as the influence of the linear dispersion of the spectrometer.

The data were acquired during a period of about 4.5 days. To check 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 in case of the Li- and He-like U, respectively. Over the whole experimental run, the spectrometer was very stable, with a shift of  $\pm 3 \mu\text{m}$  of the zinc-line reflections on the CCD (corresponding to  $\pm 0.2$  pixels).

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  $\Delta \Theta_B$ . Using the Li-like intra-shell transition as calibration, with energy  $E_{\text{Li}} = (4359.37 \pm 0.21)$  eV [23, 24], 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 [21]. Such

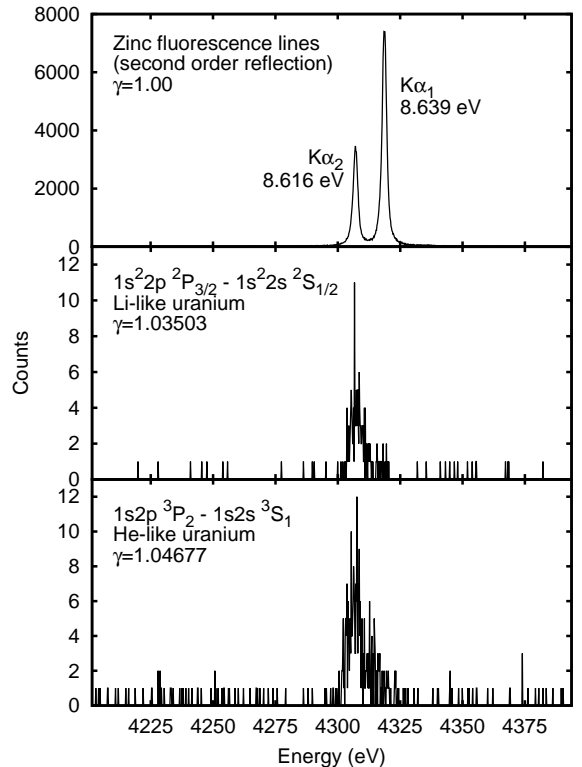


FIG. 3: X-ray spectra measured with the crystal spectrometer: Zn  $K\alpha$  doublet in second order (top) and the intra-shell transitions of Li and He-like U (middle and bottom, respectively).

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. The result of such projections is presented in Fig 3. These spectra are characterized by a very low background constrained only by the energy cuts and cluster analysis of the CCD raw data. We note that the shape of the line is slightly asymmetric. Such an asymmetry is, at present, not well understood. Due to the low statistics, no accurate investigation of this issue could be preformed. For this reason, a zeroth-order hypothesis was applied and the line position was calculated from the mean of the data distribution along the dispersion axis. The mean value was found in a self-consistent way regarding varying spectral intervals around the line center.

We obtained  $\Delta x = 44 \pm 57 \mu\text{m}$  corresponding to an uncertainty of 2.2 pixels on the CCD camera. Using this value, we found  $E_{\text{He}} = 4510.31 \pm 0.43 \pm 0.24$  eV, where the first uncertainty is statistical and the second one refers to the estimated systematic uncertainty. The dominant source of the systematic uncertainty is the limited accuracy of the calibration line, which contributes 0.21 eV. The uncertainty of the observation angle gives a contribution of  $\pm 0.11$  eV, drastically reduced compared to

TABLE I: Comparison of the experimental transition energy, in eV, for the  ${}^3P_2 \rightarrow {}^3S_1$  transition in He-like uranium with various theoretical predictions.

Experiment	4510.31 $\pm$ 0.50
Indelicato 2008 [25]	4510.30
Kozhedub 2008 [26]	4509.85 $\pm$ 0.07
Artemyev 2005 [3]	4510.03 $\pm$ 0.24
Plante 1994 [2]	4510.46
Chen 1993 [29]	4510.65
Drake 1988 [30]	4510.01

$\pm 0.9$  eV when a stationary calibration source is used. Other systematic uncertainties, including those originating from the beam velocity, are negligible.

As presented in Table I, 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 [25] represents a Multiconfiguration Dirac-Fock (MCDF) 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 contrast to [25], 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 [25] 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 [26] the value from [3] has been improved using a new value of the mean-square nuclear-charge radius for uranium [27] and the two-loop one-electron QED corrections for the excited states calculated in [28]. Also in [26] 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.

A more stringent test of the theoretical predictions can be obtained from the relative measurement of He- and Li-like uranium intra-shell transitions, where the experimental systematic uncertainty is reduced to 0.11 eV. In addition, theoretical uncertainties due to the finite nuclear size and the one-electron QED effects, are also drastically reduced. In this case, the experimental uncertainty is well below the two-photon QED contributions as presented in Table II. Our value agrees within less than 1.5 standard deviations with the *ab initio* calculations, while

TABLE II: 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.

	$E_{\text{Breit}}$	$E_{\text{rad}}^{\text{QED}}$	$E_{\text{non-rad}}^{\text{QED}}$	Total
Experiment				50.94 $\pm$ 0.45
Indelicato 2008 [25]	51.61	-1.66		49.96
Kozhedub 2008 [26]	51.48	-1.14	-0.04	50.30 $\pm$ 0.03

there are about two standard deviations from the MCDF predictions.

In summary, we report the first clear identification of the  $1s2p\ {}^3P_2 \rightarrow 1s2s\ {}^3S_1$  transition in He-like uranium. We measured the transition energy with a relative uncertainty of  $10^{-4}$ , which is currently the most accurate test of many-body and QED contributions in excited levels of very heavy He-like ions. Differential measurements pave the way for increased sensitivity. The two-electron QED contributions could be tested via the relative measurement of He- and Li-like uranium  $n=2$  intra-shell transitions.

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