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Wavelength-dispersive spectroscopy in the hard x-ray regime of a heavy highly-charged ion: The 1s Lamb shift in hydrogen-like gold

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Accurate spectroscopy of highly charged high- Z ions in a storage ring is demonstrated to be feasible by the use of specially adapted crystal optics. The method has been applied for the measurement of the 1s Lamb shift in hydrogen-like gold (Au^{78+}) in the ESR storage ring through spectroscopy of the Lyman x rays. This measurement represents the first result obtained for a high- Z element using high-resolution wavelength-dispersive spectroscopy in the hard x-ray regime, paving the way for sensitivity to higher-order QED effects.

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The theory of Quantum Electrodynamics (QED) has been tested for light atoms with extraordinarily high accuracy [1–6]. Yet, in the recent years, measurements on muonic hydrogen (combined with the state-of-the-art QED calculations), have produced inconsistency with the results obtained from hydrogen spectroscopy [7, 8]. This effect is commonly known as the “proton radius puzzle” and is still waiting for explanation. The experimental verification of the QED predictions is still significantly less precise in the domain of extreme field strength as experienced by an electron bound to a nucleus with high atomic number Z . In contrast to low- Z ions, bound state QED corrections are still a challenge for theory since they have to be treated in all orders of αZ . Here a very recent measurement of the hyperfine splitting in hydrogen- and lithium-like Bismuth has to be mentioned, showing a large disagreement with the QED predictions [9]. The QED corrections to the electronic binding energy, made up by the self energy and the vacuum polarization, are most important for the inner shells of high- Z systems

since they approximately scale as Z^4/n^3 [10], where n denotes the principal quantum number. Hydrogen and hydrogen-like ions are the most fundamental atomic systems where the QED effects can be calculated with high accuracy, thus offering a possibility of stringent experimental tests. Experimentally it requires the preparation of heavy hydrogen-like ions where notably the 1s Lamb shift can be accessed via x-ray spectroscopy of an $np \rightarrow 1s$ Lyman transition from which the calculated Dirac energy plus the small QED contribution of the np level are subtracted. Such measurements have initially been performed at lower Z where ion intensities were sufficient for the use of high-resolution techniques with low detection efficiency [11–16]. With the advent of heavy-ion accelerators and storage rings the investigations could be extended to the highest nuclear charges up to $Z = 92$ [17–20]. However the spectroscopy needed to be conducted with solid state Ge(i) detectors ensuring a high detection efficiency, although they soon faced their limits in spectral resolution.

To circumvent the low resolving power of semiconductor detectors, they were replaced by specially adapted crystal spectrometers, as will be reported in this letter, and by calorimetric low-temperature detectors yielding promising results in first storage-ring experiments [21, 22]. In the present experiment a pair of crystal spectrometers was used to measure the $1s$ Lamb shift of hydrogen-like gold accomplishing for the first time both, high- Z ions and high spectral resolution. Envisioned for a long time, the measurements have become feasible only recently because of the following developments: (i) adapted and optimized crystal-spectrometer optics, (ii) specially developed two-dimensionally position sensitive Ge(i) detectors for hard x rays with both energy and time resolution and (iii) a substantial increase of the ion-beam intensity in the Experimental Storage Ring (ESR) [23, 24].

One of the main challenges for the measurement of the $1s$ Lamb shift in a high- Z system is the preparation of a sufficient amount of ions in the desired charge state. In the case of gold, 79 electrons have to be removed from the atomic core, which can presently only be done efficiently with the help of a heavy ion accelerator. For this experiment the accelerator and storage-ring facility of the GSI Helmholtz Centre for Heavy Ion Research in Darmstadt, Germany has been utilized [25]. For this purpose, initially lowly charged ions are accelerated and successively ionized by passing through various stripping foils until a kinetic energy of about 300 MeV per nucleon (MeV/u) and very high charge states are reached. In this way more than 10^8 completely ionized gold ions per pulse can be provided. The ion beam is then transferred into the ESR experimental storage ring (see figure 1), where it can be cooled and decelerated to accomplish optimum experimental conditions [26]. The deceleration was done by means of two radio frequency (rf) cavities and down-ramping of the dipole magnets. The ion beam cooling (*i.e.* the reduction of the momentum spread of the stored ions) was achieved by the electron cooler which also defined the final velocity of $\beta = v_{ion}/c = 0.47136(10)$ of the ion beam [27, 28]. The relative momentum spread ($\Delta p/p$) of the cooled ion beam is typically in the range of 10^{-4} – 10^{-5} . On the opposite side of the electron cooler the internal gas target is located, where a supersonic gas jet can be overlapped with the circulating ion beam. Due to the low gas density of the order of $\sim 10^{12}$ atoms/cm², single collision conditions and reasonably long ion beam storage times of several tens of seconds can be guaranteed. In the present experiment, argon and krypton have been used as target gases. In each collision the ion has a chance to capture an electron from the target atom into an excited state which then decays directly or in a very rapid cascade to the $1s$ ground state of the newly formed hydrogen-like ion. About 1/3 of all down-charged ions decay (among other transitions in the cascade) via the Lyman- α_1 ($2p_{3/2} \rightarrow 1s$) transition, the accurate spec-

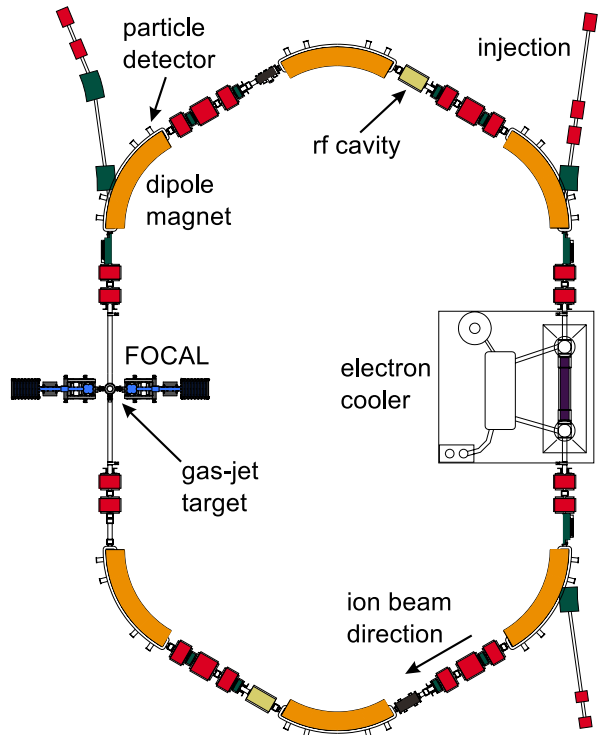


FIG. 1. Schematic view of the ESR storage ring. The experiment takes place at the gas-jet target, where the twin-crystal spectrometer FOCAL is located. In the upper left dipole magnet, a particle detector recording down-charged ions is located, which is used to apply a coincidence condition on the x-ray spectra.

troscopy of which is the main goal of the present experiment.

The Lyman- α_1 transition wavelength is measured by two twin spectrometers operated in the focussing compensated Laue (FOCAL) geometry. This type of spectrometer is well suited to find the right compromise between superior spectral resolving power and sufficient detection efficiency in the situation of very limited source strength and the presence of strong Doppler effects. Since the radiation source moves with relativistic velocity relative to the resting detector assembly (the laboratory frame) the velocity and observation-angle dependent Doppler effect has to be taken into account. The velocity of the ion beam is set by the electron cooler, however it seems unfeasible to aim for a determination of the actual observation angle with comparable accuracy. Due to this reason the dedicated FOCAL twin-crystal-spectrometer layout was developed [29, 30], where the two identical crystal spectrometer arms are aligned perpendicular with respect to the ion beam at both sides of the interaction chamber on one common line of sight. Both spectrometers (called FOCAL 1 and 2) are used to measure the Lyman- α_1 transition independently of each other leading to two distinct results for the wavelength $\lambda_{1,2}$. In this special geometry the observation-angle dependency

of the Doppler equation cancels out and the rest-frame transition wavelength λ_0 can be derived via

$$\lambda_1 + \lambda_2 = 2\gamma\lambda_0, \quad (1)$$

with the velocity dependent Lorentz factor γ .

The wavelengths $\lambda_{1,2}$ are measured with respect to a calibration line from an isotope enriched ^{169}Yb source. The strong and well known 63 120.44(4) eV γ transition [31] was selected as the main calibration line. The ion-beam velocity has been chosen such ($\beta = 0.471\,36(10)$), that the Doppler-shifted lab-frame energy of the Lyman- α_1 transition approximately coincides with this calibration energy thus avoiding systematic uncertainties due to large extrapolations. The wavelength comparison is made with respect to the dispersion plane defined by the crystals and detectors of the twin spectrometers.

The actual crystal-optics layout of one FOCAL spectrometer arm can be seen in figure 2. On the left side the x-ray source is located, emitting radiation in all directions. A part of it encounters the cylindrically bent silicon single crystal and may get Bragg diffracted on the (220) lattice planes. The crystal is cylindrically bent to a bending radius of $R = 2\text{ m}$ by means of a dedicated torque bending device. The diffracted x rays pass, independently from their wavelength, through the polychromatic focus and get registered in one of the position sensitive x-ray detectors. Due to the curvature of the crystal, the spatially wide beam of x-ray radiation is focused in a narrow line at the edge of the Rowland circle having a diameter exactly equal to the crystal bending radius. The position sensitive x-ray detectors are located close to the Rowland circle to make use of this focusing effect. Each FOCAL spectrometer arm is equipped with one germanium microstrip detector consisting of an 11-mm-thick germanium single crystal with both anode and cathode segmented into many strips [32]. The cathode is divided into 128 56-mm-wide and 250- μm -high strips, whereas the anode is segmented into 48 1.2-mm-wide and 32-mm-high strips. Each strip has its own amplifier and readout electronics and behaves, in this sense, as a single semiconductor detector. The strips on the front and on the back side are oriented perpendicularly with respect to each other allowing a two dimensional position reconstruction if front and back side strips are combined for events with the same measured energy. This way a $48 \times 128 = 6144$ pixel detector for hard x rays can be emulated. The narrow strips on the front are orientated perpendicularly to the dispersive direction of the spectrometer allowing for a more accurate position determination.

The intensity of x rays emitted from the Au^{78+} reaction products is too faint in order to allow the usage of a conventional crystal spectrometer geometry. For this purpose an *asymmetric* crystal cut has been applied with an angle deviation of $\chi = 2^\circ$ from the symmetric Laue case, where the reflecting lattice planes are orientated

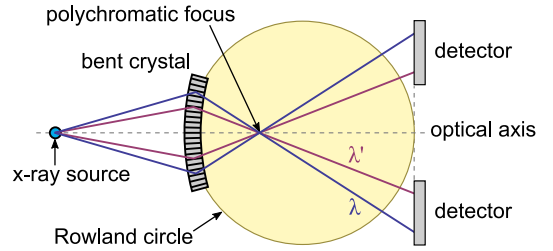


FIG. 2. Overview of the FOCAL crystal-optics layout. The source on the left emits x rays, which are diffracted by the bent crystal, if the Bragg condition is fulfilled. A position sensitive detector near the focusing Rowland circle records the impinge distance from the optical axis.

perpendicular with respect to the principal crystal faces. This asymmetric cut leads to a broadening of the Rocking curve and hence to an increased integrated reflectivity [29]. This way the efficiency could be enhanced by more than a factor of 20. The bent crystal is rotated by the angle χ to correct for the asymmetric cut, leading to symmetric but mirrored reflections above and below the optical axis. Both spectrometers are passively shielded by 15 mm thick lead plates and several thick tungsten diaphragms along the ray path to ensure that the majority of the detected photons stem from a diffractive process from the crystal. Additional background suppression was achieved by active shielding making use of the fact that the down-charged ion follows a different trajectory in the bending dipole magnets of the ESR storage ring where it was registered by a particle detector with a detection efficiency close to 100 %. X-ray events in the germanium detectors have been taken into account only if a singly down-charged ion has been coincidentally detected in the particle detector.

In this paper, the results from a first major production beam time with the fully equipped FOCAL spectrometer are presented. In three weeks of almost interruption free data taking about 1500 Lyman- α_1 photons per spectrometer arm could be collected. The Lyman lines are obtained from the measurement data by applying an appropriate energy and coincidence time condition. In this way almost background free lines are revealed as can be seen in figure 3 for one of the two spectrometers.

In this spectrometer not only the Lyman- α but also the Lyman- β transitions could be recorded spatially resolved. This shows the high resolving power of FOCAL. The slight tilt of the lines over several horizontal strips is caused by an effect called Doppler slating. Due to the spatial extent of the 2D detector a certain observation angle interval is covered, leading to higher Doppler shifted (laboratory frame) transition energies in forward angles relative to the backward direction. For illustrating reasons the 2D spectrum from figure 3 was projected according to the tilt angle onto the horizontal strip axis which is shown in figure 4.

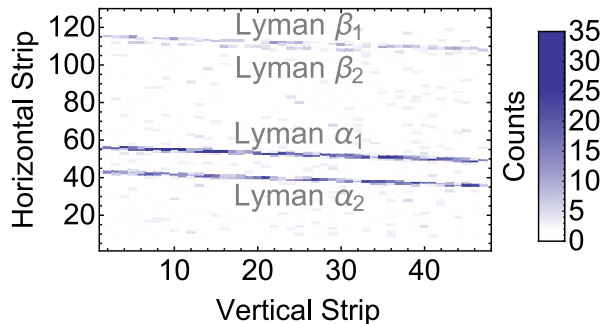


FIG. 3. Coincident x-ray spectrum as recorded by one of the FOCAL spectrometers.

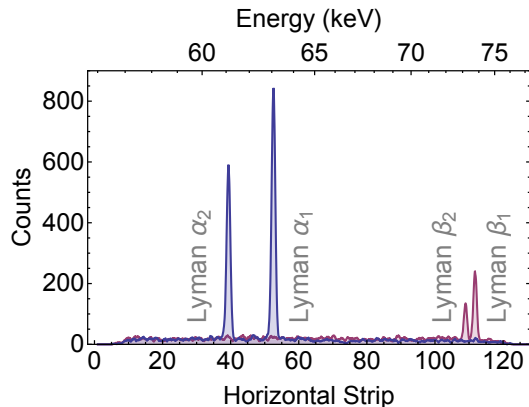


FIG. 4. The spectrum of the Lyman- α and - β doublets of Au^{78+} obtained by a projection of the respective two-dimensional intensity distribution shown in figure 3.

Since the spectrometer is operated as a wavelength comparator only the relative distance Δz_d between the main $^{169}\text{Yb-}\gamma$ calibration line and the Lyman- α_1 line matters.

This line spacing was determined by fitting a 2D model function to the original (not projected) measurement data for the Lyman and the ^{169}Yb calibration data. The fitting results can be found in table I. Here the minus sign indicates that the measured laboratory frame energy lies below the $^{169}\text{Yb-}\gamma$ line energy. Possible model dependencies and details of the line shape have also been addressed [30] by applying various fitting procedures resulting in only minor uncertainties in the line position which are subsumed under the statistical uncertainties.

TABLE I. Line Spacing between the main $^{169}\text{Yb-}\gamma$ calibration line and the Lyman- α_1 transition, and the measured spectrometer dispersion.

	Line Spacing (μm)	Spectrometer Dispersion
FOCAL 1	-35.2(5.1)	$1.905\ 29(53) \times 10^{-10}$
FOCAL 2	-51.8(3.6)	$1.909\ 74(52) \times 10^{-10}$

Besides the line spacing also the spectrometers dispersion D for both assemblies has to be measured. This is done by fitting in addition to the main $^{169}\text{Yb-}\gamma$ calibration line, the thulium $\text{K}\beta_{1,3}$ transitions which are present in the calibration source spectra. The results are again listed in table I.

With these findings and formula 1, a preliminary Lyman- α_1 transition energy can be derived $E_{\text{Ly-}\alpha_1}^{\text{prel.}} = 71\ 539.8(2.2)\text{ eV}$ which does not include any systematic effects so far.

In addition to the already mentioned statistical uncertainties a number of systematic effects has to be considered which do not only increase the total uncertainty but may also shift the final value of the Lyman- α_1 transition energy. In the following a survey of the performed investigations is given, and a compilation of the corresponding systematic effects and uncertainties are given in table II.

The first systematic effect was the temporal drift of the assembly during the three weeks of beam time. The drifts were monitored by the ^{169}Yb calibrations which were done every six hours. In total it was less than $100\ \mu\text{m}$ for both spectrometers. With the help of the numerous calibrations the effect could be minimized adding $\pm 2.8\text{ eV}$ to the total uncertainty.

If the moving x-ray source is shifted along the common line of sight between the two spectrometer arms, the FOCAL geometry corrects for that effect. However, if the source position (*i.e.* ion-beam-gas-target intersection region) is shifted out of that line (*i.e.* along the beam direction) this misalignment can not be corrected leading to a systematic deviation. For the actual position measurement of the gas-jet target a dedicated auxiliary experiment was performed in the aftermath of the FOCAL beam time [33] and the position of the gas-jet target was measured with an uncertainty of $\pm 0.30\text{ mm}$ revealing an offset of 0.25 mm in the ion beam direction. The corrected gas-jet position represents our present best guess. However, because of the long time passed between the main and the auxiliary experiment, we need to increase the position uncertainty to $\pm 1\text{ mm}$ in order to account for possible long-time changes (due to mechanical manipulations, venting and pumping, *etc.*). Fluctuations of this magnitude have previously been observed when checking the optical alignment of the gas-jet nozzles or when measuring the maximum overlap of the ion beam with the gas-jet. For the Lyman- α_1 transition energy it means a correction of 3.2 eV with an associated uncertainty of $\pm 13\text{ eV}$.

Also the uncertainty in the ion-beam velocity has to be considered, which is mainly caused by an insufficiently accurate calibration of the high-voltage terminal of the electron cooler. Another correction to be added is due to the space charge of the electron beam. Details concerning the evaluation of these corrections and the associated uncertainties can be found in [34–36]. The influence on the total uncertainty of this systematic effect is $\pm 4.3\text{ eV}$.

The last and strongest influence on the final value is given by the actual position of the germanium detector crystal inside the housing of the position sensitive x-ray detector. For its measurement, a dedicated beam time at the European Synchrotron Radiation Facility (ESRF) in Grenoble, France, has been conducted where an intense and narrow x-ray beam can be provided. The position sensitive detector was mounted on a movable table directly facing the x-ray beam. In small steps the detector was moved and the count rate of the strips as a function of the detector position was recorded. After the scan was completed the barycenter of this peaking curve was determined, defining the location of the germanium crystal relative to the outer fiducial marks which were also used during the original assembly of the FOCAL experiment at the ESR. The findings from the ESRF measurement lead to a systematic energy decrease of 11.6 eV with an uncertainty of ± 5.1 eV.

TABLE II. Individual contributions to the total Lyman- α_1 transition energy.

Contribution	Value (eV)
Preliminary Transition Energy	71 539.8(2.2)
Temporal Drift	-(2.8)
Gas-Target Position	+3.2(13.0)
Ion-Beam Velocity	-(4.3)
Detector-Crystal Position	-11.6(5.1)
Total	71 531.5(15.0)

Our final experimental value for the Lyman- α_1 transition energy including all statistical and systematic uncertainties (added quadratically) is given by $E_{Ly-\alpha_1}^{\text{exp.}} = 71\,531.5(15.0)$ eV.

TABLE III. The 1s Lamb shift of Au⁷⁸⁺ in eV.

Present Experiment	244.1(15.0)
Beyer <i>et al.</i> 1995 [37]	202.3(7.9)
Kraft-Bermuth <i>et al.</i> 2016 [21]	211(42)
Theory, Yerokhin and Shabaev 2015 [10]	205.2(2)

The experimental value for the 1s Lamb shift is obtained by subtracting the FOCAL value for the Lyman- α_1 transition energy from the theoretical value for the $2p_{3/2}$ binding energy, which is sufficiently well known [10]. The difference between this value and the Dirac value for the 1s binding energy yields the 1s Lamb shift. With the theoretical value for the $2p_{3/2}$ binding energy $E_{2p_{3/2}}^{\text{theo.}} = -21\,684.201(5)$ eV one obtains $\Delta E_{1s}^{\text{exp.}} = 244.1(15.0)$ eV. In Table III our result is compared to the experimental value obtained with a Ge(i) detector in an early experiment at the ESR electron cooler [37] and

to the experimental result reported for the calorimetric low-temperature detectors which was gained in the same beam time [21] as our present experiment. In the last entry of the Table the theoretical value of Yerokhin and Shabaev [10] is given. Our present value of the Lamb shift is higher than the theoretical value and the other experimental results by about 2.5 standard deviations of the estimated experimental uncertainty.

It is difficult at this stage to unambiguously pinpoint the reason behind this deviation. Without going into details of the other results which would be beyond the scope of this article, one can say that each of the measurements has been performed with different techniques, *i.e.* semiconductor detector at the electron cooler [37], microcalorimeter at the gas jet target [21] and thus are prone to different systematic effects. It is important to emphasize that even though we have performed very thorough and extensive studies of the various possible systematic effects, since this is the first measurement of its kind at the storage ring, potentially underestimated or unknown systematic effects can not be fully excluded. Therefore more measurements are required in order to clarify this issue.

In conclusion we performed a first measurement of the ground-state Lamb shift in a heavy H-like ion (Au⁷⁸⁺) using a high resolution crystal spectrometer in combination with a fast and dim source of hard x rays as present at a heavy-ion storage ring. The energy resolution corresponding to about 60 eV FWHM at 63 keV photon energy [30] surpasses the best semiconductor detectors by almost one order of magnitude. The achieved statistical uncertainty of 2.2 eV is groundbreaking for a crystal spectrometer operated in the region of hard x rays of H-like high- Z ions. Since storage rings are currently the only facilities routinely delivering high- Z hydrogen-like ions in large quantities, this measurement represents a very important milestone towards the challenging goal of achieving a sensitivity to higher-order QED effects as it is planned at the FAIR facility [38]. In a future run, particular effort has to be put into avoiding or reducing systematic uncertainties. The ion-beam velocity can already be determined with a much higher accuracy using a high-voltage divider from the Physikalisch-Technische Bundesanstalt (PTB) in the electron-cooler terminal, which will establish an absolutely calibrated velocity standard [39]. With a slightly modified assembly it will also be possible to measure the gas-target position relative to the detector-crystal position *in situ*, which will almost entirely eliminate these systematic uncertainties avoiding supplementary experiments altogether.

Furthermore, we would like to emphasize that this setup can also be applied for precision spectroscopy of heaviest helium-like ions (as well as other few-electron systems) which, taking into account the unprecedented resolution, would allow for resolving all the relevant fine structure levels for the first time. This is especially in-

teresting in the light of the recent controversy with the comparison between the experimental and theoretical results for helium-like ions [40–44].

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