Precise determination of the 1s Lamb shift in hydrogen-like lead and gold using microcalorimeters

S Kraft-Bermuth, V Andrianov, A Bleile, A Echler, P Egelhof, P Grabitz, S Ilieva, O Kiselev, C Kilbourne, D McCammon, J P Meier and P Scholz

1 Institute for Atomic and Molecular Physics, Justus-Liebig-University, Giessen, Germany
2 GSI Helmholtz Center for Heavy Ion Research, Darmstadt, Germany
3 Institute of Physics, Johannes Gutenberg University, Mainz, Germany
4 NASA/Goddard Space Flight Center, Greenbelt, USA
5 Department of Physics, University of Wisconsin, Madison, USA

E-mail: saskia.kraft-bermuth@iamp.physik.uni-giessen.de

Received 7 July 2015, revised 27 July 2016
Accepted for publication 4 August 2016
Published 16 February 2017

Abstract
Quantum electrodynamics in very strong Coulomb fields is one scope which has not yet been tested experimentally with sufficient accuracy to really determine whether the perturbative approach is valid. One sensitive test is the determination of the 1s Lamb shift in highly-charged very heavy ions. The 1s Lamb shift of hydrogen-like lead ($^{81+}$Pb) and gold ($^{78+}$Au) has been determined using the novel detector concept of silicon microcalorimeters for the detection of hard x-rays. The results of $(260 \pm 53)$ eV for lead and $(211 \pm 42)$ eV for gold are within the error bars in good agreement with theoretical predictions. To our knowledge, for hydrogen-like lead, this represents the most accurate determination of the 1s Lamb shift.

Keywords: high-precision x-ray spectroscopy, precision test of QED, microcalorimeters for x-rays

1. Introduction
The determination of the Lamb shift in hydrogen paved the way for the development of the theory of quantum electrodynamics (QED), and the accuracy of theory as well as experiment is remarkable [1, 2]. In theory, QED is generally calculated in a perturbation expansion in powers of the fine structure constant $\alpha$ as well as in powers of the term $(Z\alpha)$ which characterizes the nuclear binding field strength. In the very high electric fields surrounding atomic nuclei with high $Z$, the calculation of the QED corrections in powers of $(Z\alpha)$ may fail due to the high values of $Z \cdot \alpha \approx 1$. Therefore, the precise determination of the Lamb shift of the 1s as well as the $2s_{1/2}$ state in hydrogen-like very heavy ions has become an important tool for the investigation of QED in very strong Coulomb fields, both in theory [3–5] and experiment [6–9]. In theory, the accuracy of the calculations in all orders of $Z\alpha$ has been improved steadily over recent decades. Nowadays, a relative accuracy of $10^{-3}$ is obtained for the heaviest ions [5]. The experimental values of the Lamb shift are generally determined by measuring the transition energies of the Lyman transitions ($n = 2$ to $n = 1$). Whereas the Lyman transitions of the hydrogen atom are accessible by very precise spectroscopy methods [2], the Lyman-α transitions of high-Z...
ions lie in the x-ray regime. To obtain a precision of $10^{-5}$ or better, the energy resolution of the x-ray detector has to be in the order of $\Delta E/E \approx 10^{-3}$ in this photon energy range, which is not obtainable even with the best semiconductor detectors.

In the soft x-ray regime, considerable advantage in energy resolution has been obtained by using crystal spectrometers at an electron beam ion trap (EBIT) [8, 10]. For lithium-like uranium, the $2p_{3/2} - 2s_{1/2}$ transition has been determined by Beiersdorfer et al. with an accuracy of $5 \times 10^{-5}$. This accuracy allows the probing of QED two-loop calculations at a 30% level [8, 10]. By extrapolation, a value of the two-loop contribution to the $1s$ Lamb shift in hydrogen-like uranium was deduced with an accuracy of 50%. Independent measurements on lithium-like uranium, lead and gold performed by Brandau et al. at the GSI Helmholtz Center, using the method of dielectronic recombination, obtained an accuracy of $4 \times 10^{-4}$ [11] for the same transition, with the prospect of improving the accuracy further and reach $10^{-5}$. In all these measurements, the agreement between theory and experiment is excellent.

However, the situation is less favorable for the $1s$ Lamb shift in hydrogen-like heavy ions, which is usually derived from the transition energy of the Lyman-$\alpha 1$ transition, i.e. the transition $2p_{3/2} - 1s_{1/2}$. In contrast to lithium-like uranium or gold, which can be produced in a high-energy EBIT [10] in sufficient quantity for high-precision experiments, the production of hydrogen-like very heavy ions in experimentally relevant intensities is possible only at an accelerator facility [12–14]. Therefore, during the last two decades, a series of experiments to determine the $1s$ Lamb shift in hydrogen-like gold and uranium has been performed at the Experimental Storage Ring (ESR) at the GSI Helmholtz Center for Heavy Ion Research, and the experimental accuracy has been steadily improved [6]. The current best accuracy, obtained in experiments using a high-resolving segmented germanium detector positioned at the ESR electron cooler, is around 1% [15, 16]. One considerable limitation of the accuracy comes from the statistical error, which in turn is limited by the energy resolution of the used germanium detectors. Accordingly, alternative detector concepts with higher energy resolution for x-ray energies between 50–100 keV have been developed. The FOCAL crystal spectrometer [17, 18] uses the dispersion of x-rays in a silicon crystal in combination with position-sensitive germanium detectors. This detector concept has been demonstrated to obtain an excellent energy resolution of $\Delta E/E \approx 10^{-3}$ for x-ray energies around 70 keV. However, this excellent performance is limited to a very small dynamic energy range due to the Bragg condition. Also, the large size of the FOCAL spectrometer limits its active detector solid angle. This notwithstanding, the FOCAL spectrometer has been applied successfully at the ESR to investigate the $1s$ Lamb shift of hydrogen-like gold [18]. The results are expected in the near future [19].

An alternative approach, which combines good energy resolution with large dynamic range and a larger solid angle, is the concept of low-temperature microcalorimeters. Microcalorimeters detect the energy of an incoming x-ray photon as heat rather than by the creation of charge. This detection principle provides considerable advantage with respect to the energy resolution, because the excitation energy of a phonon is about 1000 times smaller than the excitation energy of an electron–hole pair in a conventional germanium x-ray detector. Accordingly, microcalorimeters for hard x-rays with x-ray energies in the range of 20–120 keV have been developed for application at the ESR [20–23] as well as for application at the EBIT and SuperEBIT at the Lawrence Livermore National Laboratory [24–26] and for $\gamma$-spectroscopy [27, 28]. In all cases, an excellent energy resolution of $\Delta E/E \approx 10^{-3}$ has been obtained. At the SuperEBIT at the Lawrence Livermore National Laboratory, the Lyman-$\alpha$ transitions of hydrogen-like xenon were determined using an array of silicon microcalorimeters with a relative accuracy of $6 \times 10^{-5}$ [26].

The application of microcalorimeters at the ESR at the GSI Helmholtz Center was first suggested by Egelhof et al. [29]. A first attempt using germanium microcalorimeters was made in 2003 [22], but the direct detection of the Lyman-$\alpha$ lines failed due to low detection efficiency at high x-ray energies as well as due to the small solid angle. Only an indirect determination of the transition energy with a very large error bar was obtained. In the following years, a setup of silicon microcalorimeters, which is specifically adapted to the experimental situation at the ESR, was developed. In this publication, we report the first experiments at the ESR with such silicon microcalorimeters in which the $1s$ Lamb shift in hydrogen-like lead ($Pb^{81+}$) and gold ($Au^{78+}$) was determined. Preliminary results were already reported in several conference proceedings [30–33]. Now, we present the final results on the transition energies as well as the values of the $1s$ Lamb shift. After presenting an overview on the experimental setup, the results of these two experiments are reported. The publication concludes with a discussion of the experimental uncertainties and of prospects for further improvements.

2. Experimental setup

Details of the microcalorimeters and the cryostat as well as the experimental setup at the ESR have been presented in [21, 30–32]. Therefore, we will only briefly summarize the most important aspects:

In general, in a microcalorimeter the incoming photon deposits its energy $E$ in an absorber of heat capacity $C$ and induces a temperature rise $\Delta T = E / C$. The temperature signal is then read out by a temperature-dependent resistor $R(T)$. A schematic layout of one detector pixel as well as photographs of the setup are shown in figure 1: The microcalorimeters consist of silicon thermometers [34, 35], which ensure a large dynamic range. On top, thin foils of tin or lead have been glued as absorbers in order to obtain high quantum efficiency for the relatively high x-ray energies [21]. As the amplitude of the temperature signals is inversely proportional to the heat capacity of the absorber, the active area of one pixel is limited
to about 0.3 mm$^2$. Therefore, the single microcalorimeters are combined into arrays of two rows with 16 single pixels in each row. For the two runs, two different microcalorimeter arrays with eight and 16 pixels, respectively, were deployed.

A scheme of the experimental setup at the ESR is shown in figure 2 on the left-hand side: The microcalorimeters were mounted inside a specially designed $^3$He/$^4$He dilution refrigerator [21], which is equipped with a copper cold finger inside a vacuum tube and several thermal radiation shields. This side arm fits inside a pocket in the ESR gas-jet target vacuum chamber. To separate the vacuum of the cryostat from the ultra-high vacuum of the ESR, an x-ray window of 50 μm stainless steel as well as four windows of aluminum-coated mylar were used. The combined x-ray transmission through these windows was 93% at an x-ray energy of 50 keV. On the right-hand side of figure 2, a schematic view of the ESR is displayed: Bare ions of lead or gold were injected from the heavy ion synchrotron. After deceleration, the ions were cooled with the electron cooler to a momentum spread of $\Delta p/p \approx 10^{-4}$ [16]. Then, the ions passed through a gas jet of argon or krypton, where some of them captured an electron and promptly emitted Lyman x-rays. As can be seen from the ultra-high vacuum of the ESR, an x-ray window of 50 μm stainless steel as well as four windows of aluminum-coated mylar were used. The combined x-ray transmission through these windows was 93% at an x-ray energy of 50 keV. On the right-hand side of figure 2, a schematic view of the ESR is displayed: Bare ions of lead or gold were injected from the heavy ion synchrotron. After deceleration, the ions were cooled with the electron cooler to a momentum spread of $\Delta p/p \approx 10^{-4}$ [16]. Then, the ions passed through a gas jet of argon or krypton, where some of them captured an electron and promptly emitted Lyman x-rays. As can be seen from the ultra-high vacuum of the ESR, an x-ray window of 50 μm stainless steel as well as four windows of aluminum-coated mylar were used. The combined x-ray transmission through these windows was 93% at an x-ray energy of 50 keV. On the right-hand side of figure 2, a schematic view of the ESR is displayed: Bare ions of lead or gold were injected from the heavy ion synchrotron. After deceleration, the ions were cooled with the electron cooler to a momentum spread of $\Delta p/p \approx 10^{-4}$ [16]. Then, the ions passed through a gas jet of argon or krypton, where some of them captured an electron and promptly emitted Lyman x-rays. As can be seen

Figure 1. The right-hand side photograph shows the side arm of the cryostat with the detector array mounted at its end (the array is protected by an aluminum cover with a thin aluminum foil). The left-hand side photograph shows the detector array used for the gold experiment, equipped with Sn absorbers. Next to it, a schematic of a single detector pixel is displayed [34].

Figure 2. Scheme of the experimental setup: x-rays were emitted after interaction of the ion beam with the internal gas-jet target of the ESR. The microcalorimeter array was mounted at an angle of $\theta \approx 145^\circ$ with respect to the ion beam; the distance between microcalorimeter and interaction region was approximately 350 mm. FOCAL1 and 2 denote the two parts of the double crystal spectrometer which was mounted in parallel [17].
from figure 2, the microcalorimeters were mounted at a backward angle of $\theta \approx 145^\circ$ with respect to the ion beam. In parallel, the double crystal spectrometer FOCAL (denoted as FOCAL1 and FOCAL2) [17] was mounted at two opposite ports under $\theta = \pm 90^\circ$ to have two independent detector systems in operation. The main experimental parameters for both experiments are summarized in table 1.

The high kinetic energy of the ions reduces the energy of the Lyman-α transitions by a factor of approximately two in the laboratory frame. To obtain the transition energy in the emitter frame, this Doppler shift has to be reversed according to the formula

$$E_{\text{emit},i} = E_{\text{lab},i} \cdot \frac{1 - \beta \cos(\theta_i)}{\sqrt{1 - \beta^2}}$$

(1)

where $E_{\text{emit},i}$ and $E_{\text{lab},i}$ are the energies of the Lyman-α lines detected with the $i$th pixel in the emitter frame, and in the laboratory frame, respectively. $\beta$ is the ion velocity, and $\theta_i$ is the average angle between the direction of the ion beam and the line perpendicular to the surface of the $i$th pixel. The individual angles $\theta_i$ were determined by first measuring the exact position of the cryostat with respect to the vacuum tube of the beam line by means of laser tracking and then measuring the position of each pixel relative to the center of the side arm by using a collimated movable $^{241}\text{Am}$ source [31, 32]. While it was possible with laser tracking to determine the position of the cryostat with respect to the vacuum chamber of the gas-jet target with high precision, the position of the gas-jet itself with respect to said vacuum chamber is less certain. Maximum overlap of ion beam and gas jet was obtained by maximizing the count rate of photons from the interaction point which were detected by a photo multiplier [36]. After this optimization, the position of the ion beam was determined using movable scrapers which are located in two positions upstream and downstream of the gas-jet target chamber. The position of the scrapers allowed the determination of the position of the interaction zone in the target chamber in the direction perpendicular to the ion beam vector. In contrast, the position of the interaction zone parallel to the ion beam vector could not be determined by direct measurements. Accordingly, Gassner et al performed an investigation with a movable slit system after the actual experiment [37].

### 3. Results

#### 3.1. Energy calibration

As the aim of the experiments was to absolutely determine the energy of the Lyman-α transitions as precisely as possible, great care had to be taken to calibrate the energy scale of the detectors. In the experiment on $^{208}\text{Pb}$, a $^{241}\text{Am}$ source was used, which emits $\gamma$-rays in a range of 15–60 keV. To have additional lines in the vicinity of the Doppler-shifted Lyman-α lines, a $^{133}\text{Ba}$ source with energies around 31 keV from its daughter $^{133}\text{Cs}$ was added. In table 2, all lines and their respective energies are summarized.

Due to the wide energy range of calibration energies, the non-linearity of the detector response function had to be taken into account by using a third-order polynomial. An example of a calibration function is given in figure 3. The order of the polynomial was determined in a separate calibration measurement with many calibration lines of high statistics. In the data analysis, an individual calibration was performed for each data sample [31].

As will be discussed in detail in section 3.5, the non-linear extrapolation introduced a considerable systematic error in the experimental transition energies. Therefore, in the second experiment on gold a different calibration source was chosen: $^{150}\text{Dy}$ decays with a half life of 144.4 days into $^{150}\text{ Tb}$, which emits $K\alpha$ radiation at 44.5 and 43.7 keV (see table 2).

### Table 1. The main parameters for the two experiments are summarized.

| parameter                      | Pb$^{208}$ | Au$^{198}$ |
|-------------------------------|------------|------------|
| Ion velocity $\beta$ [MeV u$^{-1}$] | 0.58647(1) | 0.47136(10) |
| Ion energy $E$ [MeV]          | 218.6      | 124.7      |
| Angle $\theta_0$ [°]          | 145.26(5)  | 145.16(4)  |
| Target gas                    | krypton    | argon      |
| Count rate per pixel [s$^{-1}$] | $3 \times 10^{-3}$ | $4 \times 10^{-4}$ |
| Expected transition energy of the Lyman-α1 x-rays [keV] | 42.5 | 45.5 |

1 Lyman-α1 denotes the $2p_{3/2} - 1s_{1/2}$ transition.

### Table 2. All calibration lines used are summarized with their respective line energies. The term $^{241}\text{Am}$ escape refers to escape photons in the tin pixels.

| Beam | Nuclide | Energy [eV] |
|------|---------|-------------|
| Pb   | $^{133}\text{Ba}$ | 80997(1) [38] |
| $^{241}\text{Am}$ | 95940.9(1) [39] |
| $^{133}\text{Cs}$ | $^{K\beta}$ | 34964(2) [40] |
| $^{241}\text{Am}$ escape | 34348.8(5) [39, 40] |
| $^{133}\text{Cs}$ | K$\alpha$1 | 30973.1(5) [40] |
| $^{133}\text{Cs}$ | K$\alpha$2 | 30625.4(5) [40] |
| $^{150}\text{Tb}$ | K$\alpha$1 | 44482.8(5) [40] |
| $^{150}\text{Tb}$ | K$\alpha$2 | 43744.6(5) [40] |

1 Weighted mean of $^{133}\text{Cs}$ K$\beta$ and $^{133}\text{Cs}$ K$\alpha$3, weighted with relative intensity.

2 Weighted mean of $E(^{241}\text{Am} - E(Sn K\alpha1))$ and $E(^{241}\text{Am} - E(Sn K\alpha2))$, weighted with relative intensity.

3 As the binding energy of the K shell of tin is smaller than $E(^{241}\text{Am}) = 59.5$ keV, the $^{241}\text{Am}$ x-rays may ionize the K-electrons of the absorber material. When the subsequently emitted K-α-x-rays of Sn escape the detector, the detected x-ray energy is reduced by the energy of the K-α x-rays. Accordingly, x-rays with smaller energy are observed as escape x-rays in the tin absorbers and can be used as additional calibration lines.
These $K\alpha$ lines lie in a range of $\pm 1$ keV around the Doppler-shifted Lyman-$\alpha$ lines of Au$^{78+}$, thus allowing the use of a linear calibration over this limited energy range [32].

### 3.2. Energy resolution

All results in the present analysis were derived using fits with Gaussians\(^9\). To determine the detector response function, in particular under consideration of the non-linear energy calibration, thorough characterization measurements were performed in the laboratory before conducting the experiments [21, 23]. Figure 4 shows an example spectrum of the prominent $^{241}$Am line at 59.5 keV. The line shape is described very well by a Gaussian. The non-linearity of the detector response function is negligible in the small energy interval of 150 eV.

\(^9\) As illustrated in figure 4, all widths listed in this article are full widths at half maximum (FWHM).

Investigation of the gas density yielded only a very small deviation of the gas-jet profile from a Gaussian shape [37] with a FWHM of around 6 mm. The density profile of the ion beam is also assumed to be Gaussian. Hence, it can safely be assumed that the x-ray emitting region is symmetric with respect to the central interaction point.

A first test experiment to investigate the performance of the detectors in the environment of a storage ring was performed on $^{91+}$U at a beam energy of 89 MeV u$^{-1}$ (corresponding to $\beta \approx 0.41$) [6, 30] and yielded an energy resolution of the Lyman-$\alpha$ lines of 115 eV which corresponds to $\Delta E/E = 2 \times 10^{-3}$ (see figure 5). This energy resolution was dominated by the so-called Doppler broadening. Due to the finite diameters of the gas jet as well as the ion beam, the x-ray emitting interaction zone forms a cylinder with a radius of $1 - 2$ mm [18, 36] and a length of $5 - 7$ mm [37]. As can be seen from

$$\Delta E_{\text{lab, Doppler}} = \Delta \cos(\theta_0) \cdot \frac{\beta}{1 - \beta \cos(\theta_0)} \cdot E_{\text{lab}} \quad (2)$$

the Doppler broadening is determined by the extension of the interaction zone which directly translates into $\Delta \cos(\theta_0)$. This causes a broadening of the x-ray lines, which is independent of the intrinsic energy resolution of the detectors. Due to their small lateral size of only 0.5 mm, the contribution from the extension of the detector pixel to the Doppler broadening is negligible.

Another important parameter is the velocity $\beta$ of the interacting ions. As can be seen from equation (2), low ion velocities as well as a small extension of the ion beam and gas jet are favorable. On the other hand, both Lamb shift measurements were performed in combination with the crystal spectrometer FOCAL [17], in order to have two independent detector systems to minimize systematic uncertainties. As crystal spectrometers generally pay for their excellent energy resolution with a rather limited dynamic range, the energy of the ions had to be chosen such that the Doppler-shifted Lyman-$\alpha$ lines were located in the dynamic range covered...
by the FOCAL spectrometer. For this reason, a relatively high ion energy was chosen. In combination with a relatively large gas-jet diameter of approximately 7 mm, for the lead measurement this resulted in an increased Doppler broadening at \( \theta = 145^\circ \) of \( \Delta E_{\text{Doppler}} = 220 \text{ eV} \). However, due to a worsened signal-to-noise ratio during the measurement as compared to laboratory conditions, the average intrinsic energy resolution of the detectors deteriorated to \( \Delta E_{\text{detector}} \approx 200 \text{ eV} \), so a total average energy resolution of \( \Delta E_{\text{Ly}-\alpha} \approx 300 \text{ eV} \) was obtained for the Lyman-\( \alpha1 \) line.

For the experiment on \( ^{208}\text{Pb} \), the readout electronics and shielding of the readout cables had been considerably improved, which resulted in an improved energy resolution in the laboratory of \( \Delta E = 50 \text{ eV} \) for all 16 pixels of the second detector array (see figure 4). However, during the actual experiment, the cryostat failed to reach the optimal working temperature. The detectors had to be operated at higher temperature, which lead to a considerably worse average energy resolution in the range of 150–280 eV. Therefore, the total average energy resolution was in the range of 190–300 eV for the Lyman-\( \alpha \) line, although the Doppler broadening was decreased to \( \Delta E_{\text{Doppler}} = 120 \text{ eV} \) due to the lower ion velocity \( \beta \) as well as a smaller gas-jet diameter of approximately 6 mm [37]. Due to these relatively large line widths, the natural line widths of the Lyman-\( \alpha \) transitions, which are in the order of 40 eV [41], are not taken into account in the Gaussian fits.

### 3.3. 1s Lamb shift of \( ^{208}\text{Pb} \)

In the following discussion, Lyman-\( \alpha1 \) denotes the \( 2p_{1/2} - 1s_{1/2} \) transition. Lyman-\( \alpha2 \), 3 denotes the blend of the \( 2s_{1/2} - 1s_{1/2} \) and \( 2p_{1/2} - 1s_{1/2} \) transitions, which are not resolved due to their small energy difference. Of the eight pixels of the test array, four were equipped with Sn and four with Pb absorbers. For both absorber elements, investigation in the laboratory yielded an excellent performance [21].

Figure 6 displays an example of a spectrum of one pixel with a Sn absorber. The Doppler-shifted Lyman-\( \alpha \) lines have a FWHM of \( \Delta E_{\text{Ly}-\alpha} = 257 \text{ eV} \). As can be seen from figure 6, x-ray background, which is a severe issue in experiments with germanium detectors [42] as well as crystal spectrometers [17], is negligible for microcalorimeters, because the single detector volumes are relatively small. Therefore, coincidence measurements with an additional particle detector, which are mandatory for experiments with germanium detectors and crystal spectrometers, can be omitted in the case of microcalorimeters, a fact that clearly simplifies the experimental demands and increases the overall statistics.

During the measurement, three of the eight pixels were disconnected from data acquisition, because they showed only high noise and no reasonable detector signals. Later investigation yielded that during cooling down the wires of these pixels had been damaged. Accordingly, data for only five pixels were recorded.

The Doppler-corrected results of these five pixels are displayed in figure 7 in dependence on the pixel position \( x_{\text{rel}} \), where \( x_{\text{rel}} = 0 \) denotes the position of \( \theta_0 \). The figure shows the results of the single pixels with their statistical uncertainties as well as the systematic uncertainties from the energy calibration. By weighted averaging, a weighted mean of \( E_{\text{Ly}-\alpha1} = (77919 \pm 10) \text{ eV} \) was obtained. Systematic uncertainties from the relative position of the cryostat and interaction zone as well as from the ion velocity \( \beta \), which are not reduced by averaging, are not included. To account for these uncertainties, an additional systematic error of 52 eV needs to be added quadratically. A detailed discussion of all uncertainties is given in section 3.5. Compared to our previous analysis presented in [31], the inclusion of two more pixels improved our statistical uncertainty by a factor of two.

To determine the 1s Lamb shift, the Lyman-\( \alpha1 \) transition energy was subtracted from the Dirac binding energy taken from [5] after adding the binding energy of the \( 2p_{1/2} \) state
Table 3. The experimental results for the 1s Lamb shift of hydrogen-like lead and gold (this work and [15]) are compared to the theoretical calculations from [5].

| Nuclide  | Experiment [eV] | Theory [eV] |
|----------|----------------|-------------|
| Pb$^{21+}$ | 260(53) | 244.6(2) |
| Au$^{78+}$ | 211(42) | 205.2(2) |
| Au$^{78+}$ | 202(8) [15] | |

from [4] and the energy difference $2p_{3/2} - 2p_{1/2}$, again from [5]. The result is stated in table 3 together with the theoretical values from [5]. Within the error bars, experiment and theory agree rather well. To our knowledge, our result represents the best experimental determination of the 1s Lamb shift for hydrogen-like lead up to now.

3.4. 1s Lamb shift of Au$^{78+}$

As Sn absorbers yielded a higher signal amplitude and lower baseline noise as compared to Pb absorbers, the array used for the experiment with gold was entirely equipped with Sn absorbers. Figure 8 displays a spectrum for one pixel for the Doppler-shifted Lyman-α lines together with the calibration lines. In contrast to the spectrum in section 3.3, a small background contribution is visible in this experiment. Its source is fluorescence from a very strong ytterbium calibration source of the FOCAL instrument. The level of this background was constant over the small energy range of the Lyman-α line, but it varied from pixel to pixel. In general, it was low enough to still reliably distinguish the Lyman-α line despite the low count rate and the relatively large line widths (see section 3.2). However, two pixels had to be discarded from the final analysis because, due to bad energy resolution and high background level, no Lyman-α line could be identified.

The results for the remaining 14 pixels are summarized in figure 9 after transformation into the emitter frame, in dependence on pixel position as explained for figure 7. In this case, the result for the energy of the Lyman-α transition is $E_{\text{Ly-α1}} = (71565 \pm 4_{\text{stat}} \pm 9_{\text{sys}}) \text{ eV}$. In addition, a systematic error of 41 eV has to be added to account for the systematic uncertainties from the angle $\theta$ as well as from the ion velocity (see discussion in section 3.3 and section 3.5).

Compared to the preliminary result published in [32], the final result is slightly shifted, because a determination of the exact position of the gas-jet target relative to the beam line revealed a small shift compared to the nominal position [37]. This shift is also responsible for the increase in systematic uncertainty.

The 1s Lamb shift was calculated similar to the one for lead and is listed in table 3 together with the theoretical calculations of [5]. The achieved overall uncertainty of ±42 eV is not comparable to the best result with germanium detectors obtained for the 1s Lamb shift on gold in [15] which has a precision of ±8 eV. However, as is detailed in section 3.5, the main contribution to the accuracy comes from the determination of the position of the interaction zone. The measurement of Beyer et al in [15] as well as the measurement on uranium in [16] was performed at the electron cooler of the ESR, where this systematic contribution was not present. Although the precision of this earlier experiment could not be improved, the result was confirmed with an independent detection technique. However, in both experiments the error bars are still too large to scrutinize theory.

3.5. Experimental uncertainties

Table 4 lists the different contributions to the experimental uncertainty for the final Lamb shift results. Comparing the two measurements on lead and gold, the overall experimental accuracy was improved, but the improvement is less pronounced than expected. The improvement in statistics is smaller than expected from the increase in the active detector area due to the fact that the number of stored gold ions in the ESR was about one order of magnitude smaller than
Table 4. All experimental uncertainties are listed. The total systematic error is the squared sum of the single contributions. For discussion, see text.

| Uncertainty [eV]                   | Pb$^{81+}$ | Au$^{78+}$ |
|-----------------------------------|------------|------------|
| Total                             | 52.6       | 41.9       |
| Statistical                       | 5.8        | 4.3        |
| Systematic total                  | 52.3       | 41.7       |
| Systematic from energy calibration| 8.4        | 6.1        |
| Systematic from δβ                | 0.4        | 1.4        |
| Systematic from cryostat position | 6.6        | 5.1        |
| Systematic from position of interaction zone$^1$ | 51.0 | 38.8 |
| Systematic from position of array center | 4.7 | 0.1 |
| Systematic from fit error$^2$     | —          | 4.0        |

1 Nominal position of interaction zone between the ion beam and gas-jet target.

2 For the gold measurement, the fit error in the position of Lyman-α1 was larger than expected by statistics (see text).

anticipated from the lead experiment (see table 1). However, it is evident from table 4 that in both experiments the uncertainty is dominated by systematic contributions.

- The contribution from the energy calibration has been reduced in the second experiment by using a linear energy calibration over a limited energy range (see section 3.1). It is mainly limited by the uncertainty of the position of the calibration lines, which can be improved by calibration sources with a higher count rate as well as by improving the energy resolution of the detectors. The uncertainty in the literature value of the energy of the calibration lines as listed in table 2 is still negligible at the present level of precision.

- The ion velocity β is generally derived from the voltage of the electron cooler, which had an absolute precision of ±5 V [16]. This translates into an uncertainty of $\Delta \beta/\beta = 1 \times 10^{-5}$ (see table 1) which in turn gives an uncertainty of 0.4 eV for the 1s Lamb shift for lead. This contribution is still small compared to the uncertainties from the calibration and angle $\delta$.

For the gold measurement, a problem with the determination of the cooler voltage occurred which lead to a decrease in accuracy of β by about a factor of 10. Accordingly, the systematic uncertainty is much larger in this second case. It has to be stated that this malfunction of a high-voltage measurement is not a general problem of the ESR instrumentation, but was an unfortunate coincidence which could not be resolved during the experiment.

- The dominant error contribution clearly remains the relative position of the detector array with respect to the interaction zone of the ion beam and gas-jet target as listed in the last entries of table 4. The position of the cryostat with respect to the position of the vacuum chamber of the gas-jet target was determined by means of a laser tracker with a relative precision of $10^{-5}$. This precision may be further improved by using additional markers on the cryostat itself as well as by exploring a transparent vacuum window at the vacuum chamber of the gas-jet target to directly measure the position of the center of the cryostat side arm. On the other hand, the determination of the position of the array center by means of a collimated x-ray source has been established with rather good accuracy and is negligible in the second experiment on gold.

By far the main issue is the position of the interaction zone relative to the external beam-line setup. As was detailed in section 2, the position of the interaction zone can only be determined in the direction perpendicular to the beam axis by direct means. But even though the relative position of the scrapers can be measured with an accuracy of 0.1 mm, their absolute position with respect to the gas-jet target is only known within ±1 mm. As no online method is available to investigate the position of the interaction zone parallel to the beam axis, Gassner et al. performed an according investigation after the experiment was completed [37]. However, due to time constraints, this measurement was performed almost six months after the actual experiment took place, and after dismounting the experimental setup. Therefore, the results of this investigation may not be entirely applicable to the situation during the actual measurement. A systematic error bar of the position of the interaction zone parallel to the ion beam vector of ±1 mm is assumed to incorporate this uncertainty. This introduces a considerable contribution to the systematic error, which has to be reduced in future experiments in order to obtain an accuracy which is comparable to the measurements performed at the electron cooler [16, 42]. Improved tools for beam diagnostics are currently under development [43, 44]. In addition, a new setup for the direct determination of the position of the gas jet is under construction [45].

- When fitting the data of the gold measurement with a Gaussian, it was found that the fit error for the peak energy was larger than expected by a purely statistical distribution. This larger fit error may be explained by the influence of the small background contribution to the Lyman-α1 peak from fluorescence x-rays from the very strong ytterbium calibration source of the FOCAL instrument. As discussed in section 3.4, this background varied from pixel to pixel. Its influence is stronger for pixels with poorer energy resolution and lower count rate in the Lyman-α1 line, because the limits of the Gaussian fit are not as well defined as in a clean spectrum. Accordingly, a χ²-analysis was performed to determine the optimal fitting limits. However, the position of the center of the Gaussian was not affected by small variations of these limits.

In future experiments, additional shielding will be applied in order to suppress this background, especially for low Lyman-α count rates.

In summary, the systematic uncertainties are dominated by the uncertainty in the determination of the Doppler shift. This was also the limiting factor on systematic accuracy in the
measurements of uranium and gold with germanium detectors [15, 16], albeit less pronounced due to the use of the electron cooler as target. Although improved tools to determine the ion velocity $\beta$ with higher accuracy are under development [46], ultimately the systematic uncertainty on $\beta$ will be the limit of accuracy for Lamb shift measurements at the storage rings. This is a fundamental difference to the experiments with microcalorimeters at the SuperEBIT, where Doppler shifts are not an issue and the precision is limited mainly by the statistical uncertainty [26].

However, the large dynamic range of microcalorimeters in combination with their excellent energy resolution offers an alternative approach to determine the Doppler shift. This approach makes use of the so-called Balmer lines. The transitions into the $n = 2$ levels of hydrogen-like ions are only subject to very small QED corrections [47]. Therefore, the precise determination of the Doppler-shifted transition energies of the Balmer transitions offers the possibility to determine the Doppler correction factor for the Lyman-$\alpha$ lines, which is independent on geometry or voltage measurements. For the experiments which are discussed here, the Balmer lines were Doppler-shifted to energies in the $5$–$10$ keV region. Because the microcalorimeters as well as the vacuum windows were optimized for high x-ray energies, the sensitivity below $20$ keV was too small to allow for a reliable determination of the Doppler shift. However, this disadvantage will be removed in the next-generation microcalorimeter arrays when a part of the array will be equipped with thinner absorbers optimized for x-ray energies below $20$ keV. In addition, with a new cryostat which is currently under commissioning, it will be possible to apply microcalorimeters at the electron cooler in analogy to the experiments in [15, 16].

4. Conclusion and perspectives

In this contribution, we report two successful experiments for the determination of the $1s$ Lamb shift in hydrogen-like heavy ions using silicon microcalorimeters at the GSI Helmholtz Center. The obtained results of $(260 \pm 53)$ eV for hydrogen-like lead and $(211 \pm 42)$ eV for hydrogen-like gold agree well with the theoretical predictions from [5] and—in the case of gold—with previous measurements using germanium detectors [15]. For lead, to our knowledge, this result even represents the most precise determination of the $1s$ Lamb shift.

However, to actually scrutinize the theory, an improvement in precision by at least one order of magnitude remains a mandatory task. With an accuracy of $0.5$ eV for the $1s$ Lamb shift of hydrogen-like uranium, the direct experimental investigation of two-loop contributions to the QED on a level of about $40\%$ would be feasible. Up to now, the only experimental value for this contribution was derived by Beiersdorfer et al [8] indirectly from measurements on lithium-like uranium with an accuracy of $50\%$.

The improvement of the statistical error will be obtained by increasing the number of detector pixels from 12, as of now, to one hundred in the next-generation microcalorimeter. To reduce the systematic uncertainty, the determination of the Doppler correction factor by using the Balmer lines has to reach an accuracy of $\sim 10^{-4}$. This is feasible with an energy resolution of $\Delta E/E \approx 10^{-3}$ which has frequently been demonstrated for x-ray energies below $10$ keV with such microcalorimeters [48]. As already discussed in section 3.5, the calibration error can also be improved by one order of magnitude by realizing the best energy resolution of $50$ eV in combination with calibration lines of higher intensity. Therefore, an improvement of the accuracy of the $1s$ Lamb shift to $\pm 1$ eV with current experimental techniques is within reach. To push the accuracy even further, it will be mandatory to reduce or even eliminate the Doppler correction from the measurements. To realize this purpose, two facilities with very low ion energies are currently under construction and commissioning at the GSI Helmholtz Center. The CRYRING [49] in combination with the ESR will provide very heavy highly-charged ions at energies down to $0.1$ MeV $u^{-1}$. In the HITRAP ion trap [50, 51], heavy highly-charged ions will be stored and cooled to rest, thus providing a source for Doppler-free spectroscopy on very heavy ions [52].

The future research facility FAIR [53] will provide high-intensity beams of rare nuclides, for which high-precision x-ray spectroscopy will be an interesting subject in itself. In addition, once the Lamb shift has been determined in stable isotopes with sufficient accuracy, the investigation of nuclear charge radii by means of the finite size contribution to the $1s$ Lamb shift will be a particularly interesting subject [23, 52], especially for rare or unstable nuclides.

Acknowledgments

We thank K Eberhard and J Runke from the Institute of Nuclear Chemistry of the Johannes Gutenberg Universität Mainz for producing the Dy-159 source. We also thank our co-experimentators from the FOCAL collaboration for good cooperation during the measurements. The helpful discussions during data analysis with T Stöhlker, H Beyer and T Gassner are gratefully acknowledged. The accelerator group of GSI, in particular the ESR operators, provided us with excellent, stable experimental conditions. This work was supported by the Emmy Noether Young Researchers Program of the Deutsche Forschungsgemeinschaft (DFG) under grant KR3721/1-1.

References

[1] Lamb W E and Retherford R C 1947 Phys. Rev. 72 241
[2] Niering M et al 2000 Phys. Rev. Lett. 84 5496
[3] Johnson W R and Soff G 1985 At. Data Nucl. Data Tables 33 405
[4] Beier T, Mohr P, Persson H, Plunien G, Greiner M and Soff G 1997 Phys. Lett. A. 236 329
[5] Yerokhin V A and Shabaev V M 2015 J. Phys. Chem. Ref. Data 44 033103
