Laser spectroscopy of the ground-state hyperfine structure in H-like and Li-like bismuth

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Abstract. The LIBELLE experiment performed at the experimental storage ring (ESR) at the GSI Helmholtz Center in Darmstadt aims for the determination of the ground state hyperfine (HFS) transitions and lifetimes in hydrogen-like ($^{209}\text{Bi}^{82+}$) and lithium-like ($^{209}\text{Bi}^{80+}$) bismuth. The study of HFS transitions in highly charged ions enables precision tests of QED in extreme electric and magnetic fields otherwise not attainable in laboratory experiments. While the HFS transition in H-like bismuth was already observed in earlier experiments at the ESR, the LIBELLE experiment succeeded for the first time to measure the HFS transition in Li-like bismuth in a laser spectroscopy experiment.

1. Introduction

Highly charged ions provide a testing ground for QED calculations in extreme electric (up to 10^{16} V/cm) and magnetic (up to 10^4 T) fields that cannot be created in the laboratory with conventional methods (like lasers and superconducting magnets). This approach has been used since the 1990s with various isotopes in laser spectroscopy as well as in x-ray emission spectroscopy experiments (see references in [1]).

To put the results in a theoretical context precise QED calculations have to be performed, where a major issue is the large uncertainty of nuclear structure corrections. Particularly the uncertainty of the Bohr-Weisskopf effect, which arises due to the spatially smeared out magnetic moment distribution in the nucleus, is comparable in size to the total contribution of QED corrections and hinders a direct test of QED. To tackle this problem, Shabaev et al. [2] proposed to use a new approach by introducing the so-called specific difference $\Delta'E$ between the HFS splittings in H-like ($\Delta E^{(1s)}$) and Li-like ($\Delta E^{(2s)}$) configurations of the same isotope

$$\Delta'E = \Delta E^{(2s)} - \xi \Delta E^{(1s)} \ . \tag{1}$$

The parameter ξ that acts to cancel the contribution of the Bohr-Weisskopf effect in the specific difference is largely model independent and can be calculated to high accuracy [3, 4]. A suitable candidate for an experimental determination of the specific difference is the bismuth isotope ^{209}Bi. Here the ground state HFS transitions in both, the H-like and in the Li-like system, are accessible to high precision laser spectroscopy measurements.

While the $\Delta E^{(1s)}$ splitting in ^{209}Bi^{82+} has already been observed experimentally in earlier laser spectroscopy measurements at the Experimental Storage Ring (ESR) at the GSI Helmholtz Center in Darmstadt [5], searches for the HFS transition in ^{209}Bi^{80+} have been unsuccessful for a long time. The main obstacles for this case are the long transition wavelength predicted to be at $\lambda_{\rm theor} = 1555.3(3)$ nm [4] in the ions rest frame, which is outside the sensitivity of typical photo-multipliers used for fluorescence detection, and the long lifetime of the HFS state of $\tau_{\rm theor} = 82.0(1.4)$ ms [6] leading to low signal rates in the experiment.

The LIBELLE collaboration overcame these difficulties using a newly designed detection system [7] and managed to detect the HFS transition in ^{209}Bi^{80+} in 2011 for the first time in a laser spectroscopy experiment [8]. During the same experiment we also re-measured the hyperfine transition in ^{209}Bi^{82+} and made a first measurement of the lifetime of the HFS state in ^{209}Bi^{80+}. The accuracy of the experimental wavelength result was limited by uncertainties in the voltage calibration of the electron cooler present in the storage ring in order to cool the ion beam. The voltage of the device that ultimately determines the velocity of the cooled ions had an uncertainty of approximately $5 \cdot 10^{-4}$ limiting the overall precision of the measurement. For this reason a second beam-time with a in situ measurement of the cooler voltage, using a precision HV divider provided by the Physikalisch-Technische Bundesanstalt Braunschweig (PTB), and with an improved data acquisition system was performed in 2014. During this second beam-time both HFS transitions were re-measured with an expected accuracy $< 10^{-4}$ and lifetime data for the HFS states in ^{209}Bi^{82+} and ^{209}Bi^{80+} were taken.

These data will provide a first assessment of the QED contributions to the specific difference of the HFS transitions and pave the way for trap-assisted experiments like SPECTRAP [9] that will allow to study HFS transitions in highly charged ions at rest reaching relative accuracies in the 10^{-7} regime.

2. Experimental setup

For the LIBELLE experiment, either hydrogen-like or lithium-like bismuth ions were provided by the GSI accelerator system, injected into the ESR and stored at 400 MeV/u. This represents an ion speed of $\beta \approx 0.71$. This has the advantage, that the transition wavelengths are Doppler shifted into a region convenient for (anti)collinear laser excitation and fluorescence detection according to

$$\lambda_{\rm lab}^{\uparrow\uparrow} = \lambda_0 \frac{1}{\gamma(1+\beta)} \quad ; \quad \lambda_{\rm lab}^{\uparrow\downarrow} = \lambda_0 \frac{1}{\gamma(1-\beta)} \ . \tag{2}$$

Subsequently the ions are cooled in the electron cooler to a momentum spread of $\Delta p/p \approx 10^{-5}$. The acceleration voltage of the electron cooler finally also determines the velocity of the ions. The ions are compressed into two bunches of about 10 m length each by applying an RF voltage to one of the radio-frequency cavities installed in the ring. One bunch, referred to as signal bunch, is brought into overlap with the pulsed excitation laser inside the cooler straight section, while the other bunch is not illuminated by the laser and is used as reference for background correction (see figure 1).

Fluorescence photons emitted by the HFS resonance in H-like bismuth are reflected by a mirror system installed in the vacuum pipe towards two view-ports and detected by two solar blind photomultiplier. This mirror system, which has also been used in earlier studies of HFS states in highly charged ions at the ESR [10], collects mainly photons emitted at angles between $15°$ and $60°$ relative to the beam direction.
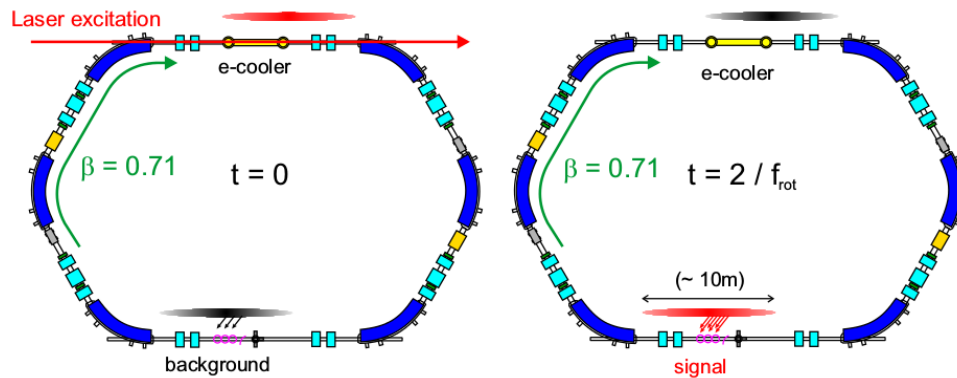
Figure 1. Schematic view of the experiment at the Experimental Storage Ring at GSI.

For the detection of the resonance in Li-like bismuth it is mandatory to collect the most forward emitted photons, as only these are Doppler shifted to a wavelength regime where the applied photomultiplier has the largest quantum efficiency. For this purpose a dedicated detection system was developed by the University of Münster [7]. It contains a movable parabolic mirror made of Oxygen-free high thermal conductivity (OFHC) copper with a central 3 cm slit for the ions to pass through. Fluorescence photons emitted at angles $\lesssim 20°$ relative to the ion beam are reflected by the mirror towards a highly selected low-noise photomultiplier tube model R1017 from Hamamatsu with a maximum quantum efficiency of 16% for the most forward emitted photons (see figure 2, left). The PMT is located in a Peltier cooler housing outside the vacuum beam-line.

Light at the Doppler-shifted transition wavelengths of about 590 nm and 640 nm for H-like and Li-like ions, respectively, was produced by a pulsed dye laser delivering a typical pulse energy of ≈ 100 mJ at ≤ 10 ns pulse length and 30 Hz repetition rate. Temporal overlap between laser pulse and ion bunch in the interaction zone inside the electron cooler was achieved by synchronizing the pump laser Q-switch signal with the bunch-generating RF voltage. By scanning the laser wavelength across a region around the predicted value for the HFS transition, this setup enabled the detection of the $2s$ hyperfine splitting in ^{209}Bi^{80+} (see figure 2, right).
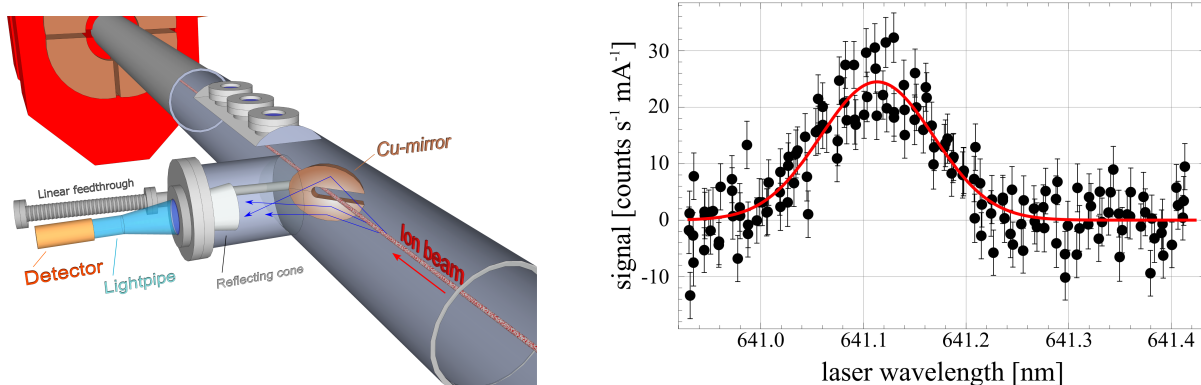

Figure 2. Left: Schematic view of the detection system for forward emitted fluorescence photons. Right: Wavelength scan over the resonance region of ^{209}Bi^{80+}, i.e. detected photon rate normalized to the beam intensity versus wavelength in the laboratory system [11].

3. Results of the 2011 beam-time

After data analysis of the data taken in 2011, the transition wavelength for H-like and Li-like bismuth in the laboratory frame were determined to be

$$\lambda_{\text{lab}}^{(82+)} = 591.183\,(26)\text{ nm} \quad ; \quad \lambda_{\text{lab}}^{(80+)} = 641.112\,(24)\text{ nm} \tag{3}$$

respectively [8]. In order to obtain the wavelength in the reference frame of the ions we used the voltage applied to the electron cooler (see [8]). However this voltage could not be measured directly and an extrapolation had to be performed. This introduces an additional systematic uncertainty. A thorough analysis with corrections extracted from previous work performed by Lochmann [11] and Jöhren [12] yielded the HFS transition wavelength in the rest frame of

$$\lambda_{\text{rest}}^{(82+)} = 243.76\,(5)(2)\text{ nm} \quad ; \quad \lambda_{\text{rest}}^{(80+)} = 1554.66\,(33)(10)\text{ nm} . \tag{4}$$

Here the first uncertainty arises from the voltage calibration while other uncorrelated effects are summed up in the second uncertainty contribution. For further details please see Ref. [8].

In this beam-time we were also able to measure the lifetime of the HFS state of Li-like bismuth. For this purpose a shutter was installed in front of the entrance window to the ESR blocking the pulsed laser beam in regular intervals. During the measurement the laser wavelength was fixed to the resonance wavelength. A cycle was started where, after the ions interacted with the laser, the shutter was closed to let the excited ions decay over a time window $[0, t_c]$ with $t_c \approx 1000$ ms. Subsequently the shutter was opened again for an interval $[t_c, 1500$ ms$]$ to repopulate the upper HFS state.

To extract the lifetime, a fit model was constructed taking into account the processes of excitation, stimulated emission and spontaneous emission of fluorescence photons in order to be able to use the data both from the excitation phases as well as from the pure decay phases of the measurement. The fit model is given by

$$f(t) = \begin{cases} s \cdot N_2(0)\exp\left(-\frac{t}{\tau}\right) & \text{for } t \le t_c \text{ (laser off)} \\ s \cdot \left[\frac{a}{b} + \left(N_2(t_c) - \frac{a}{b}\right)\exp\left(-b\,(t - t_c)\right)\right] & \text{for } t > t_c \text{ (laser on)} \end{cases} \tag{5}$$

where $a = \frac{g_2}{g_1}N\frac{\phi}{\tau}$, $b = \frac{1}{\tau}\left[\left(\frac{g_2}{g_1} + 1\right)\phi + 1\right]$ and $\phi = \frac{\lambda^3 u}{8\pi h}$. Here u corresponds to the spectral radiance and N to the total ion number, while $N_2(t)$ denotes the number of excited ions at a given time. g_1 and g_2 count the number of magnetic sub-states of the ground state ($F = 4$) and the excited state ($F = 5$) of the HFS transition, respectively, ϕ describes the time averaged laser power and s is a scaling factor taking into account experimental parameters like measurement time and observed solid angle.

The fit gives a preliminary result for the lifetime of the HFS state in $^{209}\text{Bi}^{80+}$ in the lab frame of

$$\tau_{\text{lab}}^{(80+)} = 106.9(8.0)\text{ ms} . \tag{6}$$

The given uncertainty is purely statistical. Separate fits of the decay and excitation phases resulted in lifetimes consistent to the fit over the combined data. As the lifetime measurement could use only 2-3 hours of the beam-time, there was no time to investigate systematic effects on the lifetime during these first measurements.

4. Preliminary results of the 2014 beam-time

To overcome the dominant uncertainty introduced by the electron cooler voltage a second beam-time took place in March 2014 with special emphasis on the voltage calibration. For this purpose a high-precision divider for voltages up to a maximum of 250 kV provided by PTB

Braunschweig allowed for the first time an in-situ measurement of the high voltage and improved the uncertainty to the $< 10^{-4}$ level. Data were recorded for the ground state HFS transitions in $^{209}\mathrm{Bi}^{82+}$ and $^{209}\mathrm{Bi}^{80+}$. Data analysis regarding the transition wavelengths is currently ongoing. To improve the statistics of the lifetime measurements and collect data for both $^{209}\mathrm{Bi}^{82+}$ and $^{209}\mathrm{Bi}^{80+}$, a data acquisition system with improved timing capability was set up. With this system it was possible to collect data for the lifetime of H-like bismuth whenever the laser was tuned on resonance (here the lifetime in the rest frame is $< 400\ \mu$s and therefore much shorter than the interval between laser shots of 33 ms). Due to the long lifetime of the HFS state in Li-like bismuth, a shutter system was required again in that case, but much more measurement time has been spent on this part of the measurements. Figure 3 shows results of the lifetime fits
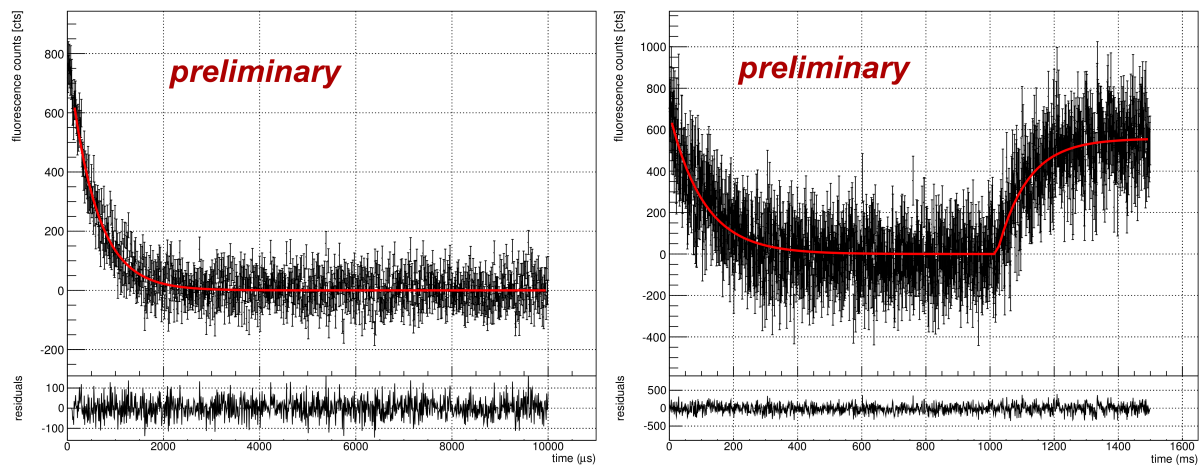

Figure 3. Detected fluorescence photons versus time in ms with preliminary fits of the lifetime data recorded in 2014 for $^{209}\mathrm{Bi}^{82+}$ and $^{209}\mathrm{Bi}^{80+}$.

for H-like and Li-like bismuth yielding preliminary values of

$$\tau_{\mathrm{lab}}^{(82+)} = 556(14)\ \mu\mathrm{s} \quad ; \quad \tau_{\mathrm{lab}}^{(80+)} = 106.8(4.2)\ \mathrm{ms} \tag{7}$$

in the laboratory frame. The fits currently contain only a subset of the data and systematic effects are not taken into account yet. Analysis of the full dataset and of possible systematic corrections is ongoing.

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