Laser spectroscopy of the ground state hyperfine splittings of $^{209}$Bi$^{82+}$ and $^{209}$Bi$^{80+}$

Matthias Lochmann,1,2,3 Raphael Jöhren,4 Christopher Geppert,1,5 Zoran Andelkovic,2,3 Denis Anielski,4 Benjamin Botermann,3 Michael Bussmann,6 Andreas Dax,7 Nadja Frömmgen,2 Michael Hammel,2 Volker Hannek,4 Thomas Kühl,3,8 Yuri A. Litvinov,3 Rubén López-Coto,9,10 Thomas Stöhlker,3,9,8 Richard C. Thompson,10 Jonas Vollbrecht,4 Andrey Volotka,11 Christian Weinheimer,4 Weiqiang Wen,12 Elisa Will,2 Danyal Winters,3 Rodolfo Sánchez,3,5 and Wilfried Nörtershäuser1,2,3

1 Institut für Kernphysik, TU Darmstadt, Germany
2 Institut für Kernchemie, Universität Mainz, Germany
3 GSI Helmholtzzentrum für Schwerionenforschung, Darmstadt, Germany
4 Institut für Kernphysik, Universität Münster, Germany
5 Helmholtz Institut Mainz, Universität Mainz, Germany
6 Helmholtz-Zentrum Dresden-Rossendorf, Germany
7 Department of Physics, University of Tokyo, Japan
8 Helmholtz-Institut Jena, Germany
9 IOQ, Friedrich-Schiller-Universität Jena, 07743 Jena, Germany
10 Department of Physics, Imperial College, London, UK
11 Institut für Theoretische Physik, TU Dresden, Germany
12 Institute of Modern Physics, Lanzhou, China

(Dated: February 3, 2014)

We performed a laser spectroscopic determination of the 2s hyperfine structure (HFS) splitting of lithiumlike $^{209}$Bi$^{80+}$ for the first time and repeated the measurement of the 1s HFS splitting of hydrogenlike $^{209}$Bi$^{82+}$. Both ion species were subsequently stored in the Experimental Storage Ring at GSI and cooled with an electron cooler at a velocity of $\approx 0.71$ c. Pulsed laser excitation of the M1 HFS-transition was performed in anticollinear and collinear geometry for Bi$^{82+}$ and Bi$^{80+}$, respectively, and observed by fluorescence detection. We obtain $\Delta E^{(1s)} = 5086.3(11)$ meV for Bi$^{82+}$ and $\Delta E^{(2s)} = 797.50(18)$ meV for Bi$^{80+}$. A specific difference between the two splitting energies can be used to test QED calculations in the strongest static magnetic fields available in the laboratory independent of nuclear structure effects. Our results confirm the large relativistic contribution of $\approx 70\%$ to the interelectronic interaction at a level of $2\%$, but the value for $\Delta E^{(1s)}$ disagrees with the literature value. Our results pave the way to trap-assisted spectroscopy, which will finally enable a test of QED in the regime of strong electromagnetic fields.

Quantum electrodynamics (QED) is generally considered to be the best-tested theory in physics. In recent years a number of extremely precise experimental results have been achieved, such as the $g$-factor of the free electron [1] or of a single electron bound to a Si nucleus [2], which are in excellent agreement with theoretical QED predictions but apply only moderate electromagnetic fields. However, for systems exhibiting extreme electrical fields, like heavy, highly charged ions, only a few tests with much lower accuracy have been reported, e.g. in [3]. QED tests in extreme magnetic fields of $B > 10^5$ T as can be found in heavy, highly charged ions of isotopes with nuclear spin [4], are still pending.

The uncertainty in the theoretical calculations of the 1s ground-state hyperfine structure (HFS) splitting $\Delta E^{(1s)}$ of hydrogenlike $^{209}$Bi$^{82+}$ is determined by the uncertainty of the Bohr-Weisskopf (BW) effect, i.e. the spatial distribution of the nuclear magnetic moment. It completely masks the QED-effect of interest and thus hampers its experimental validation. However, the nuclear effects can be significantly reduced in a specific difference [7]

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

with $\Delta E^{(2s)}$ being the HFS splitting value of lithiumlike $^{209}$Bi$^{80+}$ and the parameter $\xi = 0.168 86$ chosen in a way to cancel the BW corrections. The parameter $\xi$ is mainly determined by the behavior of the 1s and 2s wavefunctions at the atomic scale and is therefore remarkably stable with respect to different nuclear-model distributions. Thus, the cancellation of the BW-effect is to a large extent model independent and the specific difference can be calculated to high accuracy. The total theoretical value $\Delta' E = -61320(6)$ meV [8, 9] is mainly determined by the one-electron Dirac term, the screened QED contribution, and the interelectronic-interaction corrections. Recent achievements are connected with the rigorous evaluations of the screened self-energy [10, 11] and vacuum-polarization [8] corrections, as well as with the two-photon exchange diagrams [9]. The total QED contribution is about $0.229(2)$ meV [8], and the interelectronic-interaction part gives $-29.740(3)$ meV [9]. It should be noted, that the interelectronic-interaction calculated in the nonrelativistic limit yields only $\approx -9.5$ meV [12]. Thus almost 70% are caused by relativistic effects, which can be completely (to all orders in $\alpha Z$) taken into account only within the rigorous QED approach. Thus, investigations of the specific difference allow the many-electron QED effects in extreme electric and magnetic fields to be
tested.

Laser-spectroscopic measurements of $\Delta E^{(1s)}$ yielded $5084.1(8)\text{ meV}$ already in 1993 \cite{13}. In the case of $^{209}\text{Bi}^{80+}$, only a much less precise, indirect X-ray emission spectroscopy measurement in an EBIT with $\Delta E^{(2s)} = 820(26)\text{ meV}$ \cite{14} was reported. Three attempts to measure the HFS-transition in $^{80+}$Bi with laser spectroscopy failed within the past 13 years. This initiated discussions about flaws in the experiment, the theoretical calculations or possible deviations from QED. Trap-assisted laser spectroscopic experiments are under preparation, promising relative accuracies orders of magnitude better for both charge states \cite{15}. However, knowing a more precise starting value for $^{80+}$Bi is a prerequisite to find the transition in these trap experiments. Here, we report the first direct measurement of the HFS-splitting in $^{80+}$Bi.

The laser spectroscopy experiment was performed at the GSI accelerator facility. First $^{82+}$Bi and then $^{80+}$Bi ions were produced at an energy of about 400 MeV/\text{u} and then injected into the Experimental Storage Ring (ESR) \cite{16} (see Fig. 1). About 10 s after injection, the electron beam velocity in the electron cooler \cite{17} determines the ion’s momentum spread of $\Delta p/p \approx 10^{-4}$. The electron-cooler cathode was operated at approximately $-214\text{ kV}$. The ESR orbit length is about 108.5 m and the ion’s revolution frequency $f_{rev} \approx 2\text{ MHz}$. A radio-frequency (RF) voltage with twice the free-revolution frequency – measured with the Schottky analysis – was applied to an RF cavity in the ESR, forcing the ions to circulate in two bunches of about 6 m length each \cite{18}. One of these bunches served as a reference for residual-gas fluorescence background subtraction, whereas the other one was irradiated with the pulsed spectroscopy laser and provided signal photons on resonance.

The relativistic Doppler effect shifts the resonance wavelengths from $\lambda_{0}^{(82+)} \approx 244\text{ nm}$ \cite{13} in the ion frame to $\lambda_{lab}^{(82+)} \approx 590\text{ nm}$ in the laboratory frame for anticollinear excitation of $^{82+}$Bi and the predicted value of $\lambda_{0}^{(80+)} \approx 1555\text{ nm}$ \cite{15} to $\lambda_{lab}^{(80+)} \approx 640\text{ nm}$ for collinear excitation of $^{80+}$Bi. Both wavelengths were produced with a SIRAH Cobra Stretch dye laser, pumped with about 600 mJ of the second harmonic of a SPECTRA PHYSICS Quanta Ray Pro 290-30 Nd:YAG laser. The typical pulse energy of the dye laser was $\approx 100\text{ mJ}$ at $< 10\text{ ns}$ pulse length, 30 Hz repetition rate and $\approx 18\text{ GHz}$ spectral line width. 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. Laser beam position and pointing after transport to the interaction zone ($\approx 80\text{ m})$ were stabilized using a commercial laser-beam stabilization system (Aligna 4D, TEM Messtechnik).

The optical detection region is located opposite to the electron cooler (see Fig. 1). At relativistic ion velocities, fluorescence emission is non-isotropic and not monoenergetic in the laboratory frame. At $0.71\text{c}$ about $30\%$ of the fluorescence photons are emitted under an forward angle of $\lesssim 30^\circ$ and have wavelengths for $^{80+}$Bi in the range of $640\text{ nm} \lesssim \lambda_{lab} \lesssim 850\text{ nm}$. This fits well to the spectral sensitivity of a Hamamatsu R1017 photomultiplier tube (PMT) with a maximum quantum efficiency of $16\%$ for photons emitted in the forward direction. A new optical detection setup was developed to detect these photons. Its main element is an off-axis parabolic mirror with a central slit through which the ions pass \cite{19}. Photons emitted at angles of $1^\circ \lesssim \alpha \lesssim 20^\circ$ to the flight direction are efficiently directed to the PMT R1017 especially selected for high sensitivity. For the detection of fluorescence photons of $^{82+}$Bi a different mirror system mounted at a radial distance of $\approx 10\text{ cm}$ from the ion beam, successfully employed in previous beamtimes (e.g. in \cite{20}) was used.

The photon events detected with the PMTs were processed by a GSI-developed field-programmable-gate-array-based device operating as a multi-hit TDC (Time to Digital Converter). The events were recorded relative to the bunching RF, with a time resolution of $1/(300\text{ MHz})$. Time windows for assigning detected photons to the signal bunch or the reference bunch are set
and optimized offline during data analysis.

The resonance signal of hydrogenlike ions was remeasured with the old mirror system to ensure ion-beam-laser overlap and to test the new data acquisition system. After changing to Bi$^{80+}$, the transition in lithiumlike ions was detected; a typical background corrected fluorescence signal, normalized to the ion current in the ESR, as a function of laser wavelength is shown in Fig.2 The error bars are based on Poisson statistics. This spectrum consists of data from three successive laser scans with 51 wavelength steps each, combined to a single graph. The laser wavelength was not actively stabilized, but continuously measured with a wavemeter for each wavelength step individually. Error-weighted fits with a Gaussian function without background yielded an FWHM of ≈100 GHz and statistically distributed fit residuals.

The data of in total 7 scans for hydrogenlike Bi$^{82+}$ were combined to two graphs similar to Fig.2 with an average of χ²_red = 1.00. For lithiumlike Bi$^{80+}$, 72 scans yielded 24 similar graphs with an average of χ²_red = 1.06. The error weighted average of the central laser wavelengths in the laboratory frame are

\[ \lambda_{\text{lab}}^{(82+)} = 591.183(09)(01)(18)(17) \text{ nm}, \quad (2) \]

\[ \lambda_{\text{lab}}^{(80+)} = 641.112(01)(05)(18)(17) \text{ nm}. \quad (3) \]

The four uncertainty contributions account for the statistical uncertainty, uncertainties in the angular alignment between ion beam and laser, possible effects of ESR operating parameters and the laser wavelength calibration, respectively. The effects of the alignment and the operating parameters in principle cause uncertainties in the ion velocity and direction. For simplicity, they are transformed here to laboratory frame wavelength uncertainties. The quadratic sum yields 26 pm uncertainty for Bi$^{80+}$.

The crucial step in the analysis is the transformation from the laboratory frame to the ion's rest frame, or rather the ion speed determination from the electron cooler voltage using

\[ \beta = \sqrt{1 - \gamma^{-2}} = \sqrt{1 - \left(1 + \frac{-eU_c}{m_e c^2}\right)^{-2}}, \quad (4) \]

where e is the elementary charge, $U_c$ the electron accelerating potential difference, and $m_e$ the electron mass.

Starting from the electron-cooler set-voltages during the spectroscopy of Bi$^{82+}$ (−213.900 kV) and Bi$^{80+}$ (−213.890 kV), we took into account several corrections $\delta U$ and uncertainties $\Delta U$: (a) The voltage calibration based on repeated comparative measurements between a high-voltage voltmeter (HVDVM 131 from Heinzinger) and the electron-cooler voltage supply. Shits of $\Delta U_{\text{shift}} \approx 50$ V on timescales of < 1 year were observed. The HVDVM was calibrated in 2001 at the PTB Braunschweig in the voltage range −100 kV to +130 kV and recalibrated in the range 0 to +100 kV in 2012. Assuming that the ratio of deviation between HVDVM and PTB-measured voltages between the positive and negative branch was constant between 2001 and 2012, yields $\delta U_{\text{cal}} = -131(108)$ V. Removing this constraint, the uncertainty becomes about twice as large. A possible $\Delta U_{\text{shift}}$ between the beamtime in 2011 and recalibration in 2012 was accounted for. Subsequent attempts to improve the voltage calibration were hampered by technical defects and resulting major repairs both in the voltage supply and in the voltmeter shortly after the measurements. (b) Space-charge induced corrections to the effective electron acceleration potential difference caused by electron beam and ion bunch amount to $\delta U_{\text{sp-ch}} = 27.2(42)$ V and 19.4(42) V for hydrogenlike and lithiumlike bismuth, respectively. (c) A short term reproducibility of the electron cooler voltage supply of $\Delta U_{\text{rep}} = 25$ V was measured. (d) Finally, a mismatch between the bunching frequency $f_{\text{RF}}$ and twice the measured free revolution frequency $2 \cdot f_{\text{rev}}$ was observed during the measurement of Bi$^{80+}$, resulting in a difference between the ion speed and the electron speed in the cooler, corresponding to $\delta U_{\text{RF}} = 67(5)$ V.

For Bi$^{82+}$ we obtained with (a)-(c) the effective acceleration voltage $U_c = -214.00(11)$ kV to be used in Eq. 1. Using (a)-(d) for Bi$^{80+}$, we obtained $U_c = -213.93(11)$ kV. The rest frame HFS-transition wavelengths are then calculated using the relativistic Doppler formula $\lambda_0 = \lambda_{\text{lab}} \cdot \gamma (1 + \beta)$ for Bi$^{82+}$ and Bi$^{80+}$, respectively. The values are summarized in Tab. I. Our value for Bi$^{80+}$ is two orders of magnitude more precise than the only experimental value reported so far, determined indirectly via X-ray emission spectroscopy to 1512(50) nm [14]. The value for hydrogen-
like Bi\textsuperscript{82+} deviates by twice our uncertainty from \cite{13}, although obtained with a similar experimental setup at GSI.

The right column of Tab. \ref{tab:1} contains the wavelengths converted to transition energies and the specific difference $\Delta^*E$, calculated using Eq. \ref{eq:4}. Our experimental results fit well to the theoretical value of $-61.320(6)\text{meV}$ \cite{8,9} and confirm the large contribution of the relativistic effect on the interelectronic-interaction at the level of about 2%.

We assume the electron cooler calibration to be unchanged during the consecutive spectroscopy of both charge states. Thus, we reinterpret the rest frame transition wavelengths and the specific difference in Tab. \ref{tab:1} as functions of a common variable: the voltage miscalibration $\delta U_{\text{mis}} := U_{\text{vel}} - U_c$, where $U_{\text{vel}}$ is the voltage corresponding to the real ion velocity according to Eq. \ref{eq:4}. The transition wavelength of Bi\textsuperscript{80+} (rest frame) is shown in Fig. \ref{fig:3} as a function of $\delta U_{\text{mis}}$ (black line). The voltage calibration uncertainty $\Delta U_{\text{cal}} = 108$ V from (a) is illustrated by the arrow and the two vertical lines. All other uncertainties (from Eq. \ref{eq:2} \ref{eq:3} and (b)–(d)) are represented by the ($1\sigma$-)shaded area along the diagonal line.

Two alternative values for lithiumlike Bi\textsuperscript{80+}, which are both independent of the voltage calibration are also plotted. The value of

$$\lambda_{\text{theo}}^{80+} = 1555.32(27)\text{nm}$$

was obtained in \cite{9} from Eq. \ref{eq:1} using the calculated value of $\Delta^*E = -61.320(6)$ meV from theory \cite{8,9} and Klaft’s experimental value for Bi\textsuperscript{82+} \cite{12}. It is independent of our measurement. On the other hand, we can use our results for $\lambda_{\text{lab}}^{82+,80+}$ from above and make use of the relation

$$\lambda_{\text{lab}}^{82+} \cdot \lambda_{\text{lab}}^{80+} = \lambda_0^{80+} \cdot \lambda_0^{82+}.$$ 

This holds because Bi\textsuperscript{82+} was measured anticollinearly and Bi\textsuperscript{80+} collinearly, but only if both measurements were performed at the same velocity. However, the small velocity difference between our measurements, corresponding to an electron cooler voltage difference of only 69 V, can be corrected for and leads to a wavelength shift of $-0.086\text{nm}$ for $\lambda_{\text{lab}}^{80+}$ in the laboratory frame. This value is almost independent on the absolute cooler voltage. Using again Klaft’s value for $\lambda_0^{82+}$ and solving Eq. \ref{eq:6} for $\lambda_0^{80+}$ we obtain

$$\lambda_0^{80+} = 1553.97(29)\text{nm}.$$ 

Obviously, these two wavelengths, both derived from Klaft’s value \cite{13}, do not agree within their uncertainties (shaded areas). Even worse, they are inconsistent since they require contradicting miscalibration voltages. We interpret this as falsification of either $\Delta^*E$ in \cite{8,9} or the HFS-splitting in \cite{13}. Since the uncertainty stated by Klaft \cite{13} is clearly dominated by the voltage uncertainty, this might have been underestimated at that time.

Alternatively, based on our values from Eqs. \ref{eq:2} \ref{eq:3} and assuming the theoretical value of $\Delta^*E$ \cite{8,9} to be correct, we can determine a ‘reconstructed’ velocity (and therefore $\delta U_{\text{mis}}$), that brings our experiment and theory into agreement. The corresponding correction $\delta U_{\text{rec}} = 17(24)\text{V}$ can be used to calculate improved transition wavelengths for both ion species. Applying numerical error propagation through all calculation steps, the more precise HFS transition wavelengths

$$\lambda_{\text{rec}}^{82+} = 243.768(11)\text{nm} \text{ for Bi}^{82+}$$

$$\lambda_{\text{rec}}^{80+} = 1554.61(7)\text{nm} \text{ for Bi}^{80+}$$

are reconstructed. We emphasize, that these values rely on the theoretical value of $\Delta^*E$, but are independent of the problematical voltage calibration and the HFS-splitting for Bi\textsuperscript{82+} obtained in \cite{13}.

In summary, we have remeasured the HFS-splitting in Bi\textsuperscript{82+} and – thanks to a newly developed optical detection
system – for the first time observed and directly measured the HFS-splitting in Bi$^{80+}$ using laser spectroscopy. The experimental uncertainty is dominated by the electron cooler voltage calibration uncertainty. Our results confirm the calculated relativistic effect on the interelectronic interaction at a level of about 2%.

The evaluation of a measurement of the lifetime of the upper HF-state of Bi$^{80+}$, performed after the wavelength measurements, is underway. It will provide an independent consistency check of the experimental data. Together with the results of this paper and with theoretical predictions similar to \[23\], this will test the QED-contribution on a level of a few percent.

This work has been supported by BMBF under contract number 06MS9152I and 05P12RDFA4, from the Helmholtz Association under contract VH-NG-148, and from HIC4FAIR (contract). T.S., Y.A.L. and W.W. acknowledge support from the Helmholtz-CAS Joint Research Group HCJRG-108, and W.W. and Y.A.L from the BMBF grant in the framework of the Internatonale Zusammenarbeit in Bildung und Forschung Project-No. 01DO12012. M. L. acknowledges support from HGS-Hire. Helpful discussions with V. Shabaev are acknowledged. Support from the accelerator and storage ring divisions at GSI is gratefully acknowledged. We thank S. Minami and N. Kurz at the GSI experiment electronics department, for their support in the data acquisition and the VUPROM development. We thank Spectra Physics and TEM Messtechnik for their support and Sirah Lasertechnik for outstanding service during our beamtime.

* current address: IFAE, Bellaterra, Spain

[1] D. Hanneke, S. Fogwell and G. Gabrielse, Phys. Rev. Lett. 100, 120801 (2008).
[2] S. Sturm, A. Wagner, B. Schabinger, J. Zatorski, Z. Harman, W. Quint, G. Werth, C. H. Keitel, K. Blaum, Phys. Rev. Lett. 107, 023002 (2011).
[3] T. Beier, Phys. Rep. 339, 79 (2000).
[4] A. Gumberidze et al., J. Phys.: Conf. Ser. 58, 87 (2007).
[5] P. Beiersdorfer et al., J. Phys. B 43, 74032 (2010).
[6] V. Shabaev et al., Int. J. Mass Spec. 251, 109 (2006).
[7] V. M. Shabaev, A. N. Artemyev, V. A. Yerokhin, O. M. Zherebtsov and G. Soff, Phys. Rev. Lett. 86, 3959 (2001).
[8] O. V. Andreev, D. A. Glazov, A. V. Volotka, V. M. Shabaev and G. Plunien, Phys. Rev. A 85, 022510 (2012).
[9] A. V. Volotka, D. A. Glazov, O. V. Andreev, V. M. Shabaev, I. I. Tupitsyn, and G. Plunien, Phys. Rev. Lett. 108, 073001 (2012).
[10] A. V. Volotka, D. A. Glazov, V. M. Shabaev, I. I. Tupitsyn, and G. Plunien, Phys. Rev. Lett. 103, 033005 (2009).
[11] D. A. Glazov, A. V. Volotka, V. M. Shabaev, I. I. Tupitsyn, and G. Plunien, Phys. Rev. A 81, 062112 (2010).
[12] V. M. Shabaev, M. B. Shabaeva, and I. I. Tupitsyn, Phys. Rev. A 52, 3686 (1995).
[13] I. Klaft et al., Phys. Rev. Lett. 73, 2425 (1994).
[14] P. Beiersdorfer, A. L. Osterheld, J. H. Scofield, J. R. Cre spo López-Urrutia, and K. Widmann, Phys. Rev. Lett. 80, 3022 (1998).
[15] Z. Andelkovic et al., Phys. Rev. A 87, 033423 (2013).
[16] B. Franzke, Nucl. Instr. Meth. Phys. Res. B 24/25, 18 (1987).
[17] M. Steck, P. Beller, K. Beckert, B. Franzke and F. Nolden, Nucl. Instr. Meth. Phys. Res. A 532, 357 (2004).
[18] W. Wen et al., Nucl. Instr. Meth. Phys. Res. A 711, 90 (2013).
[19] V. Hannen et al., J.Instr. 8 9018 (2013)
[20] P. Seeleg et al., Phys. Rev. Lett. 81, 4824 (1998).
[21] M. Lochmann, Dissertation, Johannes Gutenberg Universität Mainz, Germany (2013).
[22] R. Jöhen, Dissertation, Westfälische Wilhelms Universität Münster, Germany (2013).
[23] D. A. Glazov, A. V. Volotka, V. M. Shabaev, I. I. Tupitsyn, and G. Plunien, Phys. Lett. A 357, 330 (2006).