Laser Spectroscopy of Trapped Ra+ Ion
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Document Version
Publisher's PDF, also known as Version of record

Publication date:
2011

Link to publication in University of Groningen/UMCG research database

Citation for published version (APA):

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Hyperfine Structure of the $6d^2D_{3/2}$ Level in Trapped Short-Lived $^{209,211}$Ra$^+$ Ions

The hyperfine structure of short-lived trapped $^{211,209}$Ra$^+$ ions was investigated by means of laser spectroscopy\(^1\). The hyperfine structure constants $A$ and $B$ of the $6d^2D_{3/2}$ level were determined. There is a 2.2 standard deviation difference between the theoretical and the more accurate experimental value for the $B$ coefficient of $^{211}$Ra$^+$. These measurements provide a test for the atomic theory required for upcoming experiments on atomic parity violation and atomic clocks.

5.1 Motivation

We present here the results of on-line hyperfine structure laser spectroscopy of trapped, short-lived $^{211,209}$Ra$^+$ ions obtained at the TRIµP facility [76, 77, 82] of KVI. Hyperfine structure (HFS) is a sensitive probe of the atomic wave functions [87], and is used to estimate the accuracy of theoretical predictions of APV matrix elements [32, 70]. Up to now, no experimental value for the electric quadrupole hyperfine structure constant $B$ of the $6d^2D_{3/2}$ level in Ra$^+$ was available for such tests. The $B$ coefficient is also of particular interest for calculations related to a Ra$^+$ atomic clock [22].

\(^1\)Results in this Chapter are the basis of [89]. Here some additional information is provided.
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Figure 5.1: Level schemes of $^{211}$Ra$^+$ and $^{209}$Ra$^+$ with wavelengths derived from Ref. [28].

5.2 Experimental Setup

The experimental setup was discussed in detail in Chapter 3. For completeness, the key points are repeated below.

The isotopes $^{211}$Ra and $^{209}$Ra were produced in inverse kinematics by bombarding 2 mg/cm$^2$ pyrolytic carbon foils with, respectively, 8.3 and 10.3 MeV/nucleon $^{204}$Pb beams with typically $10^{11}$ particles/s from the AGOR cyclotron. The foils were mounted on a rotating wheel to distribute the dissipated energy due to beam stopping. The particles emerged from fusion-evaporation reactions $^{204}$Pb + $^{12}$C $\rightarrow$ $^{216}$−$^x$Ra + $n$, in which $x$ neutrons were liberated. The Ra isotopes were separated from the primary beam and fission products in the TRIP magnetic separator [75]. They were stopped and re-ionized to Ra$^+$ in a Thermal Ionizer (TI) [76]. Up to 9% of the stopped Ra$^+$ isotopes were extracted as a singly charged ion beam with 2.8 keV energy [77]. Rates of approximately 100 $^{211}$Ra$^+/s$ and 10-100 $^{209}$Ra$^+/s$ were extracted as verified employing a surface-barrier Si detector to detect alpha particles from radioactive decay (the nuclear lifetimes are 18.8 s for $^{211}$Ra and 6.6 s for $^{209}$Ra). The Ra$^+$ isotopes were passed through a Wien Filter which eliminated contaminant ions. The ion beam was subsequently electrostatically decelerated upon injection in a gas-filled Radio Frequency Quadrupole (RFQ) cooler [78]. Typically the RFQ was operated at a frequency of 500 kHz with a peak-to-peak RF voltage of $V_{RF} = 380$ V applied between neighboring rods. The ions were trapped at the end of the RFQ by a
suitable axial potential $V_{DC}$ (see Fig. 5.2). The effective potential depth was 13 V while the axial potential depth was 10 V. A Ne buffer gas was used to aid effective catching and trapping of the radioactive particles from the beam in the RFQ. Importantly, the buffer gas influenced the radiative lifetimes of the ions by means of optical quenching and (hyper)fine-structure mixing of the metastable levels [58]. This is an essential property for the measurements described in this Chapter: The gas enabled sufficient state-mixing to avoid dark state trapping (see below). Partial pressures of other contaminant gases were kept below $10^{-8}$ mbar.

Figure 5.2: Schematic overview of the end part of the RFQ.

Home-built Extended Cavity Diode Lasers (ECDLs) drive the optical transitions (see Fig. 5.1). Light to drive the $7s^2S_{1/2}-7p^2P_{1/2}$ transition at wavelength $\lambda_1 = 468$ nm came from NDHA210APA01 laser diodes from Nichia (see Fig. 3.11); the $6d^2D_{3/2}-7p^2P_{1/2}$ transition at wavelength $\lambda_2 = 1079$ nm was driven with light from LD-1080-0075-1 diodes from Toptica (see Fig. 3.12). The laser light was delivered to the ion trap with single-mode optical fibers. The beams were overlapped with polarizing beam splitters and a dichroic mirror and sent axially through the trap to minimize scattered light. They were focused to 1 mm diameter at the trap location. Typical laser beam intensities $I$ at the trap center were $I(\lambda_1) = 200 \, \mu W/mm^2$ and $I(\lambda_2) = 600 \, \mu W/mm^2$. Both laser light intensities were close to saturation values so the fluorescence rate at $\lambda_1$ was limited by the (hyper)fine-structure mixing rate. The wavelengths were monitored with two HighFinesse Angstrom WS6 VIS and IR wavelength meters. The frequency of the laser operating at wavelength $\lambda_2$ was referenced to an optical frequency comb (Menlo Systems) via an auxiliary laser which was detuned by a few GHz. The auxiliary laser served as an absolute fixed reference frequency. The frequency detuning of the spectroscopy laser light was determined by means of a beat note detected on a fast GaAs photo-diode (see Fig. 3.12). This enabled the measure-
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ment of a frequency offset of up to 4 GHz. The beat note frequency was mixed down employing an ADF4007 mixer from Analog Devices.

The transitions in Ra\textsuperscript{+} were detected via fluorescence light from the 7\textsuperscript{s}2\textsuperscript{S}_{1/2} - 7\textsuperscript{p}2\textsuperscript{P}_{1/2} transition at wavelength \( \lambda_1 \). Because of the 10% branching into the metastable 6\textsuperscript{d}2\textsuperscript{D}_{3/2} level, this fluorescence was only observed when both the 7\textsuperscript{s}2\textsuperscript{S}_{1/2} - 7\textsuperscript{p}2\textsuperscript{P}_{1/2} and 6\textsuperscript{d}2\textsuperscript{D}_{3/2} - 7\textsuperscript{p}2\textsuperscript{P}_{1/2} transitions were resonantly excited. The fluorescence light was imaged with a single lens of focal length \( f = 30 \text{ mm} \) inside the vacuum through a low-pass filter with 80% transmission for wavelengths shorter than 500 nm (Thorlabs FES0500) onto the photocathode of a photomultiplier (Hamamatsu R7449). The collection solid angle was 0.4 sr.

5.3 Results

5.3.1 Detection of the 7\textsuperscript{s}2\textsuperscript{S}_{1/2} - 7\textsuperscript{p}2\textsuperscript{P}_{1/2} Transition

The absolute wavelength of the 7\textsuperscript{s}2\textsuperscript{S}_{1/2} - 7\textsuperscript{p}2\textsuperscript{P}_{1/2} transition in 211\textsuperscript{Ra}+ was derived by scaling our previous measurements performed on 212−214\textsuperscript{Ra}+ [82] by the very accurate isotope shifts established by the ISOLDE collaboration at CERN [64]. However, no such information was available for 209\textsuperscript{Ra}+. Therefore, the absolute wavelength of the 7\textsuperscript{s}2\textsuperscript{S}_{1/2} - 7\textsuperscript{p}2\textsuperscript{P}_{1/2} transition in 209\textsuperscript{Ra}+ needed to be established. To this end, the frequency of the laser light at \( \lambda_1 \) was scanned over the 7\textsuperscript{s}2\textsuperscript{S}_{1/2} - 7\textsuperscript{p}2\textsuperscript{P}_{1/2} resonance. Nitrogen buffer gas at relatively high pressures of typically \( 6 \times 10^{-3} \text{ mbar} \) was used to sufficiently reduce the lifetime of the metastable 6\textsuperscript{d}2\textsuperscript{D}_{3/2} level by means of optical quenching. Therefore, no repump laser light at wavelength \( \lambda_2 \) was necessary. The frequency of the laser light at \( \lambda_1 \) was monitored with a HighFinesse Ångstrom WS6 VIS wavelength meter. The absorption line in Te\textsubscript{2} at frequency 640,146,536(70) MHz (no. 178 in Ref. [81]) provided absolute frequency calibration. The measured line shape is shown in Fig. 5.3. The frequency of the 7\textsuperscript{s}2\textsuperscript{S}_{1/2} - 7\textsuperscript{p}2\textsuperscript{P}_{1/2} transition in 209\textsuperscript{Ra}+ was established at 640,146,875(187) MHz. The IS of the center-of-mass of this transition with respect to the transition in 214\textsuperscript{Ra}+ (see Chapter 4) is 6,647(239) MHz where a conservative 1% uncertainty was assumed for the hyperfine structure constants \( A \) of the 7\textsuperscript{s}2\textsuperscript{S}_{1/2} and 7\textsuperscript{p}2\textsuperscript{P}_{1/2} levels [25, 64]. With the data from Ref. [64] the IS of this transition in 209\textsuperscript{Ra}+ with respect to 214\textsuperscript{Ra}+ is determined to be 11,230(239) MHz. This result can be compared to the value 11,702(15) MHz that can be obtained from linear interpolation of the data presented in Ref. [64].
5.3 RESULTS

Figure 5.3: (a) Absorption spectrum of Te$_2$. The solid black line represents a Gaussian fit to the data. The width of the resonance is 408(10) MHz; it is centered at 1166(122) MHz. The reduced $\chi^2 = 0.8$ at 24 degrees of freedom (d.o.f.). (b) Line shape of the $7s^2S_{1/2}$-$7p^2P_{1/2}$ transition in $^{209}$Ra$^+$. The solid black line represents a Gaussian fit to the data. The width of the resonance is 230(22) MHz; it is centered at 1505(122) MHz. The reduced $\chi^2 = 0.8$ at 40 d.o.f. An additional uncertainty due to the calibration of the frequency axis was taken into account in the determination of the position of the resonances. The offset count rate of the PMT signal includes scattered photons from the pump laser light at $\lambda_1$ of order $10^4$ counts/s as well as the dark count rate of the PMT (below $10^2$ counts/s).

The uncertainties of the PMT signal are based on the standard deviations of the bin contents as calculated using the TProfile class of CERN’s ROOT code.

5.3.2 Hyperfine Structure

The wavelengths of the light from two diode lasers operating at $\lambda_1$ were kept close to the $7s^2S_{1/2} F=3 - 7p^2P_{1/2} F'=3$ and $7s^2S_{1/2} F=2 - 7p^2P_{1/2} F'=3$ resonances to pump ions from both hyperfine ground levels. This pumping scheme ensured that no decays to the $6d^2D_{3/2} F'=1$ occurred. It enabled the study of the hyperfine levels $F=4,3,2$ of the $6d^2D_{3/2}$ level in $^{211}$Ra$^+$. The experiment was
performed at $1 \times 10^{-2}$ mbar Ne buffer gas pressure. Collisions of the ions with the Ne buffer gas mixed the hyperfine levels of the $6d^2D_{3/2}$ level, ensuring that no significant shelving to the metastable $6d^2D_{3/2}$ hyperfine levels occurred when one of the $6d^2D_{3/2}$ hyperfine levels was depopulated by the resonant laser light at $\lambda_2$. The frequency of the laser light at $\lambda_2$ was scanned over the resonances $6d^2D_{3/2} F=4.3.2 - 7p^2P_{1/2} F'=-3$. Many such scans were averaged in order to achieve a good signal to noise ratio (SNR). A typical resulting line shape is shown in Fig. 5.4. The solid line represents a fit of 3 Voigt profiles to the data. From the positions of the peaks the HFS constants $A$ and $B$ were extracted [90] (neglecting contributions from higher moments). These constants and corresponding HFS intervals are given in Tables 5.1 and 5.2, respectively. The theoretical predictions [32, 42], presented in Table 5.2, are in good agreement with the experimental values. The Gaussian line widths (see Fig. 5.4-5.6) of approximately 200 MHz imply [48] a temperature of the trapped ion cloud of about 1200 K.

The hyperfine levels $F=3,2,1$ of the $6d^2D_{3/2}$ level in $^{211}$Ra could be studied in a similar manner. In this case, the wavelengths of the light from two diode lasers operating at $\lambda_1$ were kept close to the resonances $7s^2S_{1/2} F=3 - 7p^2P_{1/2} F'=2$ and $7s^2S_{1/2} F=2 - 7p^2P_{1/2} F'=2$. This pumping scheme ensured that no decays to the $6d^2D_{3/2} F=4$ occurred. The frequency of the laser light at $\lambda_2$ was scanned over the resonances $6d^2D_{3/2} F=3,2,1 - 7p^2P_{1/2} F'=2$. Many such scans were averaged in order to achieve a good SNR. A typical resulting line shape is shown in Fig. 5.5. This data was taken at Ne buffer gas pressures varying between 1-5 $\times 10^{-2}$ mbar. The $6d^2D_{3/2} F=2.1 - 7p^2P_{1/2} F'=2$ resonances could not be resolved due to the close proximity of the resonances and large Doppler width. The combined HFS data sets $F=4,3,2$ and $F=3,2,1$ contain the HFS of the $7p^2P_{1/2}$ level. It is in agreement with the value given in Ref. [64], albeit with a larger uncertainty.

In case of the HFS of $^{209}$Ra a lower yield and shorter lifetime of this isotope resulted in a smaller signal. Nonetheless, the hyperfine levels $F=4,3,2$ of the $6d^2D_{3/2}$ level in $^{209}$Ra could be determined by averaging scan data over many hours. The frequency of the laser light at $\lambda_2$ was scanned over the resonances $6d^2D_{3/2} F=4,3.2 - 7p^2P_{1/2} F'=3$. Similar to the case of $^{211}$Ra, the wavelengths of the light from two diode lasers operating at $\lambda_1$ were kept close to the resonances $7s^2S_{1/2} F=3 - 7p^2P_{1/2} F'=3$ and $7s^2S_{1/2} F=2 - 7p^2P_{1/2} F'=3$. A Ne buffer gas was present at pressure $2.5 \times 10^{-2}$ mbar. The resulting line shape is shown in Fig. 5.6.

The extracted hyperfine structure constants are presented in Table 5.2 along with theoretical predictions. The theoretical values for the $A$ coefficients are in excellent agreement with our experimental values. A 2.2 standard deviation difference (taking a 10% theoretical uncertainty into account) is found for the $B$ coefficient of $^{211}$Ra. However, in the theoretical calculations several potentially significant contributions were not yet taken into account and as such the theory
Figure 5.4: Spectrum of the $6d^2D_{3/2} F'=4,3,2 \rightarrow 7p^2P_{1/2} F'=3$ transitions in $^{211}\text{Ra}^+$. The solid line represents a fit of 3 Voigt profiles to the data. The Gaussian widths ($1\sigma$) of the resonances are typically 200 MHz. The Lorentzian line widths vary between 20-100 MHz depending on laser light power and level multiplicity. The reduced $\chi^2 = 1.0$ at 27 d.o.f. The offset count rate of the PMT signal includes scattered photons from the pump laser light at $\lambda_1$ of order $10^4$ counts/s as well as the dark count rate of the PMT (below $10^2$ counts/s). The uncertainties of the PMT signal are based on the standard deviations of the bin contents as calculated using the TProfile class of CERN’s ROOT code.

<table>
<thead>
<tr>
<th>HFS interval</th>
<th>$^{211}\text{Ra}^+$</th>
<th>$^{209}\text{Ra}^+$</th>
</tr>
</thead>
<tbody>
<tr>
<td>$F = 4 - F = 3$</td>
<td>687(9)</td>
<td>673(28)</td>
</tr>
<tr>
<td>$F = 3 - F = 2$</td>
<td>407(7)</td>
<td>396(49)</td>
</tr>
</tbody>
</table>

Table 5.1: Hyperfine structure intervals in MHz as extracted from the model fit to the data of the $6d^2D_{3/2} F' \rightarrow 7p^2P_{1/2} F' = 3$ transition in $^{211,209}\text{Ra}^+$ presented in Figs. 5.4 and 5.6.

The extracted $A$ coefficients from $^{211,209}\text{Ra}^+$ data can also be compared with the value $A = 528(5)$ MHz obtained from $^{213}\text{Ra}^+$ [82] as a check of consistency. This number is scaled by $I/I' \times \mu_r^2/\mu_I$ [90], where $\mu_I$ is the nuclear magnetic moment taken from [25]. This scaling from the $^{213}\text{Ra}^+$ value yields 151(2) MHz for $^{211}\text{Ra}^+$ and 148(2) MHz for

value for the $B$ constant can be improved [91].
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Figure 5.5: Spectrum of the $6d^2D_{3/2} F=3,2,1 \rightarrow 7p^2P_{1/2} F'=2$ transitions in $^{211}\text{Ra}^+$. The solid line represents a fit of 3 Gaussian profiles to the data. The $6d^2D_{3/2} F=2,1 \rightarrow 7p^2P_{1/2} F'=2$ transitions could not be resolved.

$^{209}\text{Ra}^+$. These numbers are in excellent agreement with this work as presented in Table 5.2.

<table>
<thead>
<tr>
<th></th>
<th>This work</th>
<th>Theory</th>
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<tbody>
<tr>
<td></td>
<td>$^{211}\text{Ra}^+$</td>
<td>A 151(2)</td>
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<tr>
<td></td>
<td></td>
<td>B 103(6)</td>
</tr>
<tr>
<td></td>
<td>$^{209}\text{Ra}^+$</td>
<td>A 148(10)</td>
</tr>
<tr>
<td></td>
<td></td>
<td>B 104(38)</td>
</tr>
</tbody>
</table>

Table 5.2: HFS structure constants $A$ and $B$ of the $6d^2D_{3/2}$ level in $^{211}\text{Ra}^+$ and $^{209}\text{Ra}^+$ in MHz. The theoretical values were converted to $^{211}\text{Ra}^+$ by scaling with $I/I' \times \mu_I'/\mu_I$ [90] in case of $A$, where $\mu_I$ is the nuclear magnetic moment taken from [64]; the hyperfine structure constant $B$ was obtained by scaling with $Q_I'/Q_I$, where $Q_I$ is the electric quadrupole moment taken from [26]. *The theoretical uncertainty is at the percent level. **This uncertainty is due to the experimental uncertainty in $Q_I$ [26] used to scale from one isotope to the other. The theoretical uncertainty is at the 10% level.
5.4 DISCUSSION

In summary, on-line excited-state laser spectroscopy was performed on short-lived trapped Ra\(^+\) ions. Experimental values for the hyperfine structure constants \(A\) and \(B\) of the 6\(d^2\)D\(_{3/2}\) level in \(^{211,209}\)Ra\(^+\) were obtained. The \(A\) coefficients are in good agreement with theory. However, only marginal agreement is found between the theoretical and the more accurate experimental value for the \(B\) coefficient of \(^{211}\)Ra\(^+\) at 2.2 standard deviation difference. The accurate determination of HFS constants reduces the frequency uncertainty of a Ra\(^+\) clock (see Chapter 6). These measurements test the atomic theory at the percent level. Future experiments are aimed at improving the precision of the HFS measurements employing the microwave optical double-resonance technique. This is expected to decrease the experimental uncertainty by one or two orders of magnitude, providing a testing ground for future improved high-precision calculations of atomic theory, for which the necessary improved theoretical methods are currently being developed [91].

Figure 5.6: Spectrum of the 6\(d^2\)D\(_{3/2}\) \(F=4,3,2 \rightarrow 7p^2\)P\(_{1/2}\) \(F'=3\) transitions in \(^{209}\)Ra\(^+\). The solid line represents a fit of 3 Gaussian profiles to the data. The Gaussian widths (1 \(\sigma\)) of the resonances are typically 200 MHz. The reduced \(\chi^2 = 1.0\) at 39 d.o.f.
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