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A new precise value of the absolute 2^3S_1, $F = 5/2 - 2^3P_2$, $F = 7/2$ transition frequency in $^7Li^+$

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Abstract. The absolute frequency ν_0 of the optical electric-dipole transition between the metastable $1s2s\ ^3S_1$, $F = 5/2$ and the short-lived $1s2p\ ^3P_2$, $F = 7/2$ hyperfine structure sublevels of the helium-like $^7Li^+$ ion in its rest frame was determined by combining Doppler-free saturation spectroscopy and laser heterodyning. One of two cw single-mode dye lasers was locked to the calibrated w-component of the [R(85) 26-0]-line in the (B-X)-system of the $^{127}I_2$-molecule at $\nu_R = 546\,462\,926.790(71)$ MHz. While crossing two counterpropagating beams of the other laser perpendicularly (lab angle $\alpha \approx 90°$) with a low-velocity ion beam and tuning its frequency over the Li^+ transition under study, the distance of the Lamb-dip center frequency ν_L from ν_R was obtained by mixing both laser frequencies with an avalanche diode and registering their beat frequency at about 4 GHz with a quartz-stabilized high-frequency counter. The Lamb-dip position ν_L was measured for different ion velocities β (in units of the velocity of light) in the range of $(3-4) \times 10^{-4}$. The parameters ν_0 and $\cos\alpha$ of the relativistic Doppler formula $\nu_0 = \nu_L\gamma(1 - \beta\cos\alpha)$ were extracted *via* a fit to the experimental data set (β, ν_L), providing $\nu_0 = 546\,466\,916.49(87)$ MHz and $\cos\alpha = 4.580(0.045) \times 10^{-4}$. This result of ν_0 differs noticeably from $\nu_0 = 546\,466\,918.79(40)$ MHz, measured by another group in a collinear-beam configuration at an ion beam propagating with $\beta = 6\times10^{-3}$.

PACS. 32.30.Jc Visible and ultraviolet spectra – 06.30.Ft Time and frequency – 07.57.Yb Other infrared, submillimeter wave, microwave, and radiowave instruments, equipment, and techniques

1 Introduction

Careful absolute-wavelength measurements of the spectral lines of atoms have a long and venerable tradition, reaching back to the nineteenth century. They contributed enormously to the foundations of the modern understanding of atomic structure. More recently, the electronic energy splittings of fundamental atomic systems, in particular hydrogen, helium, and their isoelectronic sequences became accessible to refined calculations taking into account also minute effects such as QED- and nuclear-structure corrections.

On this background, the Li^+ ion, the immediate isoelectronic neighbour of helium, has been subject to numerous experimental as well as theoretical studies (for early references see, *e.g.*, [1]). A significant fraction of these investigations dealt with the determination of absolute wavelengths or frequencies (both magnitudes being transformable into each other *via* the value of the velocity of light fixed by definition). It should be emphasized, however, that the value of an absolute frequency can only be fully exploited when making allowances for the influences originating from fine structure (fs), hyperfine structure (hfs), isotope shifts, and Lamb shifts. Therefore, the large amount of works performed on these effects in Li^+ deserves to be briefly addressed here in the following.

Most activities focused on the strong electric-dipole transition between the metastable $1s2s\ ^3S_1$ state ($\tau \approx 60$ s [2]) and the short-lived ($\tau \approx 43$ ns [3]) $1s2p\ ^3P_{0,2,1}$ multiplet at the wavelength $\lambda_0 \approx 548.5$ nm. The two stable isotopes ^6Li and ^7Li have nuclear spins of $I = 1$ and $3/2$, and therefore the fs-lines of the $2^3S_1 - 2^3P$ transition of the singly charged ions split into a large number of hfs-components. The energy diagram of the $1s2s\ ^3S_1$ and $1s2p\ ^3P$ terms of $^7Li^+$ is shown in Figure 1. Absolute-wavelength measurements of the fs- and hfs-components, based on the spectral analysis of the light emitted from a hollow-cathode discharge, provided also the fs- and hfs-splittings of the two terms [4–7]. A theoretical analysis of the $2\ ^3P$ hfs-splittings was given in [8]. The experimental knowledge of the splitting values was further improved by beam-foil spectroscopy [9,10], laser-scanning technique [1], combined laser-microwave spectroscopy [11], and in the recent past by laser heterodyning [12,13].

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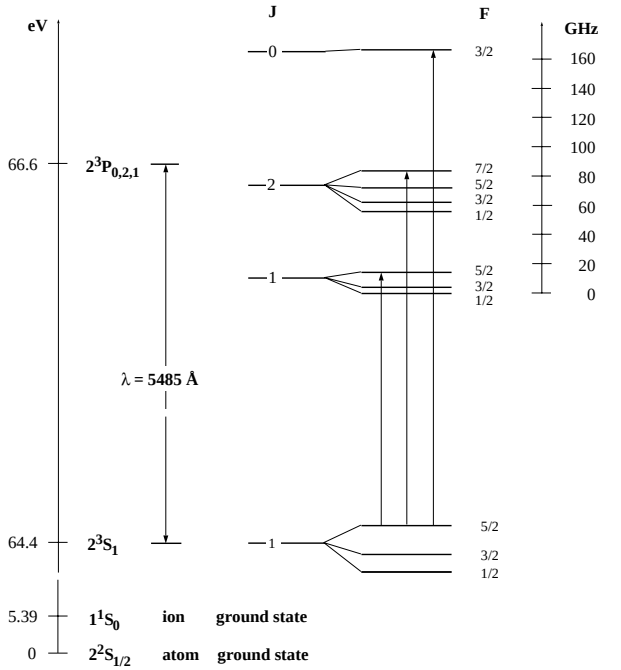

Fig. 1. Energy level diagram of the $1s2s\,^3S_1$ and $1s2p\,^3P$ terms of the $^7Li^+$ ion (taken from [12]).

Besides these results, absolute wavelengths were determined using laser fluorescence spectroscopy on a Doppler-tuned ion beam [14]. The absolute wavelength of one of the 2^3S_1–2^3P components was measured also with a traveling wavemeter by comparing the Li^+ line with the $\lambda = 633$ nm HeNe-laser line [15,16]. For this purpose, the HeNe laser was locked to a calibrated hfs component of the iodine molecule $^{127}I_2$. After reduction to the centers of gravity of the 2^3S_1 and 2^3P terms by means of the hfs-values from references [1,11], the 2^3S–2^3P transition energies including the 2^3P fine structure could be compared with theoretical values [17,18]. Since the latter calculation did not include QED corrections, the difference between the experimental and theoretical results was interpreted as the Lamb shift [14,19–21]. In addition, a comparison of this difference with a separate theoretical QED result [22] was also possible. Since then, further highly precise frequency measurements of the spectral line under study were performed for both $^6Li^+$ and $^7Li^+$ in a collinear-beam configuration [23,24]. Reference [24] contains also a detailed theoretical analysis of the hfs, isotope shift, and Lamb shift. Further, a very accurate calculation of the 2^3P fs splittings was performed in [25].

A renewed broader interest in Li^+ and its spectral properties was triggered by the advent of several ion storage rings, opening up the possibility of laser cooling and high-precision laser spectroscopy of ions propagating with relativistic velocities. In particular, an appropriate experimental configuration of an ion beam, overlapping parallel and antiparallel with two laser beams, which are tuned into resonance with two different Li^+ transitions, provides access to a sensitive test of the fundamental Doppler formula of the theory of special relativity. Experiments pursuing this aim were performed at the Test Storage Ring (TSR) in Heidelberg with $^7Li^+$ ions orbiting in the ring with the velocity $\beta = v/c = 0.064$ (c denoting the velocity of light) [26–29]. Similar experiments with $\beta \approx 0.3$ have been proposed for the Experimental Storage Ring (ESR) of GSI [30]. The $^7Li^+$ ion was chosen as a favourable candidate, since its spectrum allows suitable excitation schemes.

This paper reports on a new precise experimental value of the (2^3S_1, $F = 5/2$–2^3P_2, $F = 7/2$) transition frequency ν_0 in the $^7Li^+$ rest frame. The measurement was performed at a low-velocity ($\beta = (3\text{–}4) \times 10^{-4}$) ion beam. The absolute frequencies of other hfs-components of the 2^3S_1–2^3P_2 line multiplet, *e.g.*, those especially appropriate for relativity test experiments as mentioned above, can be immediately obtained from ν_0 when using the well-known hfs-splittings.

2 Experimental method

2.1 General configuration

The experimental method is based on a combination of Doppler-free saturation spectroscopy and laser heterodyning. It was applied successfully in a previous experiment to measure the 2^3P fine-structure splittings of $^7Li^+$ [12], and has been described in detail in reference [13]. Two counter-propagating light beams, obtained by dividing the beam of a cw single-mode dye laser, cross a slow (*e.g.*, $\beta = 3\times10^{-4}$, corresponding to a kinetic energy of ~ 300 eV) Li^+ beam at right angles. While scanning the laser frequency, the reemitted fluorescence is registered with a photomultiplier. Due to saturation, a Lamb dip occurs in the center of the excitation line profile. With a specific data acquisition technique, the Doppler-broadened background and thus its influence on the Lamb dip position is eliminated [13].

By means of heterodyning, the frequency distance of the Lamb-dip center from a selected hyperfine component with frequency ν_R of a spectral line in molecular iodine $^{127}I_2$ is measured. For this purpose, a second cw single-mode dye laser is locked to this hfs-component *via* lock-in technique with a stability of $\pm 1 \times 10^{-11}$ and a reproducibility of $\pm 1 \times 10^{-10}$, the latter value corresponding to ± 50 kHz. Two beams branched off from the main laser beams are overlapped parallel and sent to an avalanche photodiode. The diode produces the beat frequency ν_B of the two dye lasers in the range of several GHz, which is registered with a quartz-stabilized high-frequency counter. A Hewlett Packard Spectrum Analyzer 8593, Option 004, was used in the present experiment. Due to aging of the internal frequency reference of $\pm 1 \times 10^{-7}$/year, an uncertainty of $\pm 5 \times 10^{-7}$ was attributed to the beat frequencies registered with this spectrum analyzer. This is marginal in comparison with other sources of error. The absolute frequency ν_L of the laser when tuned to the Lamb dip center of the Li^+ transition under study is obtained immediately

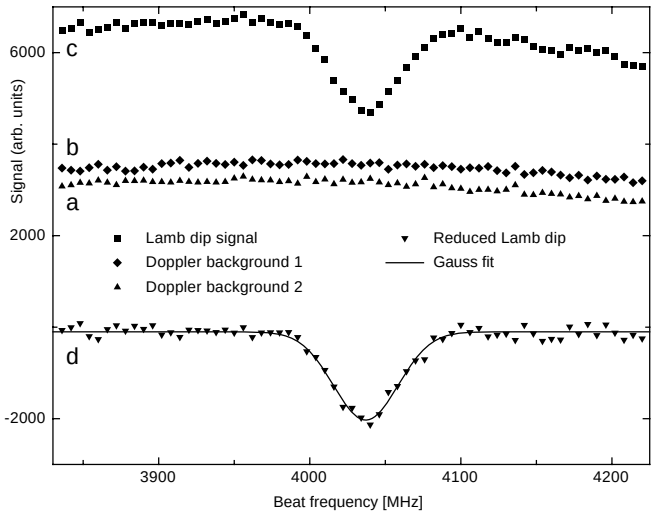

Fig. 2. Lamb-dip signal as a function of the beat frequency ν_B. Only every third data point is plotted in the diagram. The curves (a), (b), and (c) represent the intensity of the fluorescence re-emitted from the ion beam during the interaction with (a) the first laser beam, (b) the second laser beam, (c) both laser beams. The data acquisition of (a), (b), and (c) required 4-5 min, respectively. The signal (d) was obtained by the subtraction of (a) and (b) from (c). The data points of (d) were fitted with a Gaussian curve.

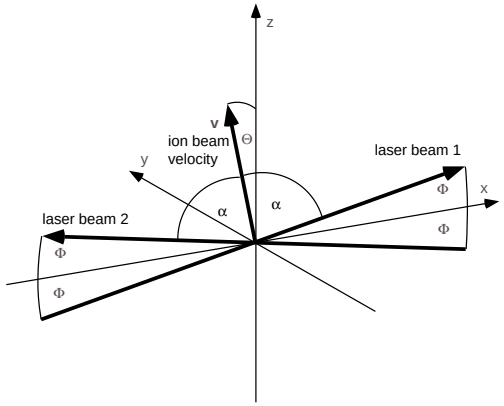

Fig. 3. Configuration of the ion beam and the laser beams, and definition of the angles α, θ and ϕ.

as the sum $\nu_L = \nu_R + \nu_B$, if the frequency ν_R of the iodine hfs-component is known. In fact, the experimental situation was characterized by $\nu_L > \nu_R$.

Figure 2 shows a Lamb dip prior to and after elimination of the Doppler-broadened background. It should be mentioned that compared with the Lamb dips reported in reference [13], the full width at half maximum was reduced in the present experiment by a factor of 3 by expanding the counterpropagating laser beams and lowering their intensity in the ion interaction region. The Lamb dip shape was caused by the natural linewidth as well as by transit-time and saturation broadening. When applying a Gaussian and alternatively a Lorentzian to the Lamb dip, the center frequencies agreed within the error bars, though the Lorentzian fitted the data less smoothly than the Gaussian. The Lamb dip center could be determined with an uncertainty of the order of ± 0.5 MHz, corresponding to roughly $\pm 1\%$ of the Lamb dip halfwidth.

The iodine cell used by us was compared with a cell which was produced and calibrated by the Physikalisch-Technische Bundesanstalt (PTB) in Braunschweig. The w-component of the [R(85) 26-0]-line in the (B-X)-system of the $^{127}I_2$-molecule served as reference transition. The frequency attributed to the w-component by the calibration procedure at the PTB was $\nu_R = 546\,462\,926.790(71)$ MHz [31]. The comparison of the cells was performed at the Physik-Institut of Mainz University by passing the same cw single-mode dye-laser beam through both cells and simultaneously recording at each cell the relevant section of the hfs-spectrum of $^{127}I_2$. The two signal curves, represented by the derivatives of the absorption line profiles of the reference transition coincided within the limit of resolution of about ± 50 kHz.

2.2 Procedure for the determination of ν_0

The configuration of an ion beam of velocity β (supposing for this consideration a highly collimated, monoenergetic beam) and a laser beam crossing each other with an angle α, as measured in the laboratory frame, is described by the relativistic Doppler formula

$$\nu_0 = \nu_L \gamma (1 - \beta \cos \alpha). \tag{1}$$

Here, ν_L is the transition frequency in the laboratory frame and $\gamma = (1 - \beta^2)^{-1/2}$ denotes the time dilation factor. The occurrence of a Lamb dip due to the simultaneous interaction of two counterpropagating laser beams of identical frequency with the same velocity class of ions ensures that each beam encloses the same angle α with the direction of propagation of these ions. If θ is the angle between the plane spanned by the laser beams and the ion beam direction, and if the laser beams enclose an angle 2ϕ, the geometry being illustrated in Figure 3 [32], then α can be expressed by θ and ϕ according to

$$\cos \alpha = \cos \theta \sin \phi. \tag{2}$$

In the following, it is assumed that the laser frequency ν_L represents the Lamb-dip center. If β and α are known, ν_0 can be extracted. The determination of ν_0 with a precision of 1 MHz or less requires the knowledge of α with an uncertainty of not more than a few μrad. It would be very difficult to measure α directly with such a precision. Therefore, we decided to exploit the following possibility: If ν_L is measured for two different values β_1 and β_2, while keeping α fixed, both ν_0 and α can be determined, provided that β_1 and β_2 have been measured in addition. This experimental procedure was justified, since we found a way to measure β with an uncertainty not significantly contributing to the final error bar of ν_0.

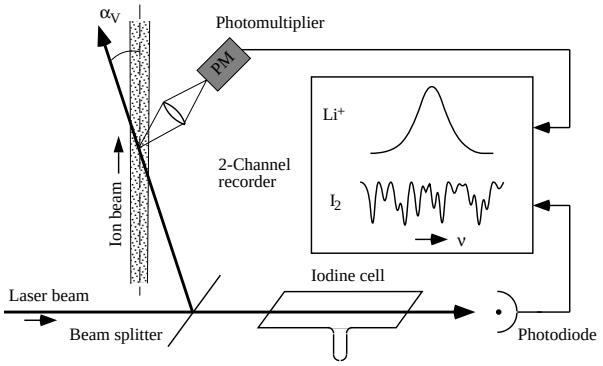

Fig. 4. Arrangement for the measurement of the Li^+ beam velocity.

At the same time it was possible to keep α constant within the required limits. This was accomplished by keeping the direction of the laser beams constant in space to within ± 1 μrad with the aid of a feedback system consisting of position-sensitive photodetectors and piezo-driven tilting mirrors [33,13]. Then, only changes of the direction of the ion beam, characterized by the angle θ, remain as a possible error source. According to (2), a change $d\theta$ of θ will cause a change

$$d\cos\alpha = -\sin\theta\sin\phi d\theta. \tag{3}$$

We estimate that changes of θ, which may especially occur when changing the acceleration voltage to produce the different β values, were below 1 mrad. With $d\theta = 1$ mrad, $\phi = 1$ mrad, $\nu_0 = 5 \times 10^{14}$ Hz, and $\beta = 3 \times 10^{-4}$, and further choosing $\theta = \pi/2$, where the effect reaches its maximum, the Lamb dip position experiences a shift $\delta\nu_L = \beta\nu_0\, d\cos\alpha \approx 2 \times 10^5$ Hz. Even this worst case is well covered by the final error bar of ν_0 (see Tab. 1).

2.3 Measurement of the ion-beam velocity

The method for the measurement of the ion-beam velocity [34] makes use of the Doppler effect and of ν_0, known from previous experiments with a precision by far sufficient for this purpose. The experimental arrangement is illustrated in Figure 4. A cw single-mode dye-laser beam is split into two beams, one crossing the ion beam at a laboratory angle α_V (chosen as near to 0° as our experimental conditions allowed), the other passing through an iodine cell. As the laser is scanned over the frequency range containing the Doppler-shifted line profile of the Li^+ transition under study, the excitation profile (represented by the intensity of the reemitted fluorescence) of the ions and the absorption lines of the iodine vapour occurring in this range are registered respectively with a photomultiplier and a photodiode.

The densely distributed I_2-lines have been calibrated with an uncertainty of ~ 60 MHz [35], and can therefore serve as frequency marks for determining the position of the Doppler-shifted Li^+ line with that precision. Due to the velocity distribution of the ion beam, the halfwidth was dominated by Doppler broadening and amounted to about 5 GHz. Thus, by comparing the Li^+ line profile with the I_2-spectrum the center frequency ν_L could be determined easily within 0.5 GHz. This provides a relative precision of ν_L of 1×10^{-6}. Since β was in the order of $(3{-}4.4) \times 10^{-4}$, it was sufficient to determine its actual value by means of the classical approximation of the Doppler formula

$$\nu_L = \nu_0(1 + \beta\cos\alpha_V). \tag{4}$$

The Doppler shift of the Li^+ center frequency is then given by

$$\nu_D = \nu_L - \nu_0 = \nu_0\beta\cos\alpha_V. \tag{5}$$

Setting α_V to 10° with an uncertainty of $\pm 1°$ and taking ν_0 from the literature [24] resulted in values of ν_D ranging from 160 to 240 GHz with an error of ± 0.5 GHz. The error of α_V of $\pm 1°$ is significantly larger than the divergence of the ion beam. This guarantees that the value of β and its error bar extracted from the measurement represent in any case also those ions that produce the Lamb dip signal. From equation (5) follow immediately the expressions

$$\beta = (1/\cos\alpha_V)(\nu_D/\nu_0) \tag{6}$$

and

$$\Delta\beta/\beta = [(\Delta\alpha_V \tan\alpha_V)^2 + (\Delta\nu_D/\nu_D)^2]^{1/2}. \tag{7}$$

By inserting the absolute values and uncertainties of α_V and ν_D given above, we obtained $\Delta\beta/\beta = 4.5 \times 10^{-3}$. This uncertainty satisfies fully the requirements of the measurement of ν_0, since it contributes less than 10% to the final error bar.

3 Measurement and result

The frequency distance $\nu_B = \nu_L - \nu_R$ between the ($2\,^3S_1$, $F = 5/2{-}2\,^3P_2$, $F = 7/2$) transition and the I_2-reference line was measured at three different ion beam velocities. Each velocity measurement required about 30 min and was performed prior to the acquisition of the Lamb-dip data. About 15 min were necessary to register a complete Lamb-dip spectrum, consisting of three laser scans (see Fig. 2). At the beginning, four measurements of the Lamb-dip position were performed at $\beta = 2.916(0.014) \times 10^{-4}$. The ion beam velocity was then increased by a factor of about 1.5, and a similar set of data was collected. As a third step, β was tuned back near to the first value, in order to check the reproducibility of the initial experimental conditions. The experimental results of β and ν_B are listed in Table 2.

The error attributed to each of the Lamb-dip center frequencies represents one standard deviation and results from the fit of a Gaussian curve to the Lamb-dip profile. Weighted means were calculated from the beat frequencies related to the same β, in order to facilitate the evaluation procedure by using only three (β, ν_L)-pairs. The error of the frequency counter amounts to ± 2 kHz and can

Table 1. Experimental values for the $2^3S_1,\ F = 5/2{-}2^3P_2,\ F = 7/2$ transition frequency ν_0.

Method	ν_0 (in cm^{-1})	ν_0 (in MHz)
Arc spectrum [4(b)][a]	18 227.42 (17)*	546 444 305 (5100)
Hollow cathode [5]	18 228.18 (7)*	546 467 089 (2100)
Cooled hollow cath. [7]	18 228.1687 (24)*	546 466 750 (72)
Traveling wavemeter [15]	18 228.17404(23)	546 466 910.1 (7.0)*
Laser spectroscopy [23]	18 228.17437 (17)	546 466 919.9 (5.1)*
Laser spectroscopy [24]	18 228.174336(13)	546 466 918.79 (40)*
This work	18 228.174256(29)	546 466 916.49 (87)*

[a] Schüler comments in a footnote that according to preliminary comparisons with lines of the neon spectrum, used as a standard, a correction of the Li reference line at 2741.29 Å by 0.1 to 0.2 Å would probably become necessary, and would respectively change also the value of the Li^+ transition wavelength at 5485 Å. He does, however, not give the sign of that correction. The error originates from the grating resolution as remarked in Table 1 of reference [4(b)].
* The star marks the value given in the respective publication. In order to facilitate an immediate comparison of the different results, this value was converted into the corresponding value expressed in cm^{-1} or MHz by means of the velocity of light, defined as $c = 2.99792458 \times 10^{10}$ cm/s.

Table 2. Li^+ beam velocities β and related beat frequencies $\nu_B = \nu_L - \nu_R$ (in MHz). Errors are one standard deviation.

$\beta \times 10^4$	ν_B	ν_B (weighted mean)
	4038.94 (0.36)	
2.916 (0.014)	4039.31 (0.44)	
	4040.08 (0.50)	4039.36 (0.24)
	4039.52 (0.46)	
	4046.90 (0.46)	
4.376 (0.018)	4046.44 (0.50)	
	4047.67 (0.49)	4046.89 (0.28)
	4046.55 (0.47)	
2.937 (0.015)	4040.09 (0.48)	4039.78 (0.34)
	4039.48 (0.47)	

thus be ignored compared with the statistical error of the Lamb dip position. The iodine frequency ν_R enters each ν_L with the root sum of squares of its 71 kHz calibration uncertainty and of a second contribution estimated to be ±100 kHz. This amount should cover the servo lock and reproducibility errors, respectively amounting to ±5 and ±50 kHz, as well as conceivable shifts with respect to the calibrated reference. Shifts may originate from conditions such as gas pressure and temperature of our iodine cell, differing from the cell calibrated at the PTB. We consider the 100 kHz-error as a conservative estimate.

The parameters ν_0 and $\cos\alpha$ of equation (1) were determined by means of the evaluation program "proFit" (Cherwell Scientific Publishing). For this purpose, ν_0 and $\cos\alpha$ were fitted to the data set of Table 2. Taking into account the individual experimental errors of all values (weighted fit), we obtained

$$\nu_0 = 546\,466\,916.49(87)\ \text{MHz}$$

and

$$\cos\alpha = 4.580(0.045) \times 10^{-4}.$$

For both results, the error in brackets is one standard deviation. The experimental values of ν_0 achieved by different authors are compiled in Table 1, in order to illustrate the enduring effort devoted to the precise measurement of this specific spectral line of Li^+. Our result should be compared in particular with the value $\nu_0 = 546\,466\,918.79(40)$ MHz, measured by another group in a collinear-beam configuration at an ion beam propagating with $\beta = 6 \times 10^{-3}$ [24].

4 Conclusions

We have remeasured the absolute frequency ν_0 of the $(2^3S_1,\ F = 5/2{-}2^3P_2,\ F = 7/2)$ hyperfine component of the helium-like $^7Li^+$ spectrum, *i.e.*, the frequency referring to the ion rest frame. The method combined Doppler-free saturation spectroscopy and laser heterodyning and was applied to a collimated ion beam propagating with a mean velocity β in the range of $(3{-}4) \times 10^{-4}$ (in units of the velocity of light).

The Doppler-shifted Lamb-dip position ν_L of the hyperfine component was measured for three different values of β with respect to a calibrated spectral line of molecular iodine. During the whole series of measurements, the direction of the laser beams was kept constant. Prior to each Lamb-dip signal acquisition, the respective β was determined from the size of the Doppler shift of the Li^+ transition under study. Here, due to the low velocity it was justified to use the classical rather than the relativistic Doppler formula. By applying the relativistic formula to the experimental data, both ν_0 and α were extracted.

Our result for ν_0 differs by 2.30 (0.96) MHz from the value measured in a collinear-beam experiment [24], the error in brackets being the root sum of squares of the respective 1σ-errors (see Tab. 1).

In the recent past, the $^7Li^+$ ion was chosen for storage ring experiments aiming at a test of the theory of special relativity [26–29]. A highly precise value of ν_0 is required for this purpose. From the present situation, we therefore draw the conclusion that a further measurement of ν_0 by means of a different method and with an increased precision will be indispensable. Apart from aspects of the theory of special relativity, refined storage ring experiments [30] could also contribute significantly to improve the techniques of precision laser spectroscopy on fast moving atoms or ions and to enhance the knowledge about subtle systematic errors.

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