PRECISION SPECTROSCOPY OF HELIUM ATOM

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Quantum electrodynamic theory (QED) of the simplest bound-three-body system is now stringently tested by precise spectroscopic frequency measurements in Helium. Alternatively, comparison between He measurements and theory could be used for accurate determination of some fundamental quantities, as for example, the fine structure constant α, and the differences of nuclear charge radii between two $^3$He and $^4$He. In the following, we review our precise spectroscopic measurements on the 1083 nm Helium transition, connecting the triplet 2S and 2P states. Frequency differences among the 1083 nm measured frequencies give precise values of the fine structure (FS) and hyperfine structure (HFS) splittings of the $^2$P level in $^4$He and $^3$He respectively, and of the isotope shift (IS) between these two isotopes. Implications of these results in the α determination as well as for nuclear charge radii and structure are discussed.

1. Introduction

The QED-theory knowledge for bound-three-body-systems is of great importance because, when compared with spectroscopic measurements, fundamental physical quantities can be determined. Comparisons between theory and measurements in He-like atoms or molecular-ions, being more complicated than He, have been reported in this conference. In this way, antiproton-proton mass and charge ratios have been determined in antiprotonic He, but their final accuracy was limited, in part, by the present status of the QED-theory for this exotic atom. In this frame, the accuracy achieved for frequency measurements of transitions in stable He isotopes can be used as a stringent test of the QED theory for the simplest bound-three-body system, and therefore contributes by analogy to improve this theory for more complicated systems. Alternatively, this theory-experiment comparison applied to $^4$He has been used for more than three decades to give an accurate determination of the fine structure constant α by precise measurements and theory of the $^2P$ FS splittings. Also, the nuclear-charge-radius determination of He and He-like isotopes, by comparison of precise IS measurements and theory, is at present recognized as a powerful nuclear-model independent method, alternative to the usual one, i.e. e$^-$ scattering, almost for short-lived isotopes. The results have been used as a critical test of the effective low-energy nucleon-nucleon potential. The knowledge of this potential is fundamental for the theoretical calculation of the nuclear-charge-radius and other nuclear properties. Information about other nuclear properties can be achieved from precise measurements of hyperfine atomic transitions in He isotopes with non-zero nuclear spin.

Among all He transitions, the one at 1083 nm is perhaps the most studied and measured using different experimental approaches. It is particularly convenient because it starts from a metastable state (about 7900 s lifetime) that can be populated by an electrical discharge and because it gives access to the $^2S$ FS and HFS manifolds, by using microwaves as well as optical frequencies. Although the $^3$He nuclear-charge-radius has been determined by measurements of the $^2S \rightarrow ^2P$ transition at 389 nm, most determinations are given by measurements at 1083 nm. In addition, the Lamb-shift contribution to the $^2S$ and $^2P$ levels is larger than for other simple atoms and can thus be measured with higher accuracy even with less precise measurements of the atomic transitions. Here, we review the more than ten years long activity in Firenze on precise spectroscopy at 1083 nm. Preliminary results for absolute frequency measurements of the hyperfine $^3$He transitions at 1083 nm, and $^3$He-$^4$He IS at this wavelength are reported.

2. Experiment

In our experiment, fluorescence saturation spectroscopy in a metastable He beam, in absence of external magnetic fields, has been implemented for precise line center measurement of the 1.6 MHz HWHM $^2S_1 \rightarrow ^2P_j$ transitions at 1083 nm. This experimental approach has been described in detail elsewhere and it is schematically depicted in figure 1. The metastable beam of He$^+$ atoms is produced by a DC-discharge and both
\(^3\text{He}\) and \(^4\text{He}\) isotopes can be used. In the former case, indeed, a gas-recirculating system was implemented in order to minimize the quantity of gas used, due to the high cost of \(^3\text{He}\). Moreover, this system allows now to perform \(^3\text{He}-^4\text{He}\) measurements with a controlled gas mixture. After production, these atoms interact with the saturating laser standing-wave, in a volume screened from external magnetic fields with a \(\mu\)-metal cylinder and three pairs of Helmholtz coils, one for each spatial direction. The fluorescence from the excited \(^2^3P\) level as a function of the laser frequency scan is observed, as shown in Figure 2-a. A frequency modulation technique was used to enhance the saturation dip signal-to-noise (S/N) ratio and to eliminate the contribution of the residual Doppler fluorescence (see Figure 2-b/c).

In our experiment, the line center of He transitions is determined by a fitting procedure. For that, it is important to get a precise frequency scale on the x-axis. To this purpose, we phase-lock the scanning laser (slave) to a second (master) laser that, in turn, is frequency locked to a stable reference. The master-slave frequency difference is covered by a GPS-disciplined RF-

\[ f_1(s) = f(m) \pm f_1(\mu wa) = f_0 + N_m \times f_r \pm f_{cm} \pm f_1(\mu wa) \] (1)

Here, \(N_m\) is the order number of the OFS tooth nearest to the master and \(f_{cm}\) is the beat-note frequency between master and OFS. The line center in this scale is measured with a precision of about 5-15 kHz, limited only by the S/N and not by the master reference. In fact, the OFS-locked-master stability at 1 s is 160 Hz (Allan deviation of \(6 \times 10^{-13}\)), more than two orders
of magnitude better than the iodine-locked master-stability\(^b\). Moreover, a minimum accuracy of \(5 \times 10^{-12}\) and a better reproducibility of the He measurements are guaranteed by the new reference, used for the first time in the preliminary IS measurements presented here. It should be noticed that with this set-up we can perform "multiresonant precision spectroscopy"\(^a\), i.e., optical frequency measurements can be simultaneously measured together with their differences, by locking two different slave lasers resonant with different He transitions to two different teeth of the OFS\(^15\).

Statistical values for the He frequencies with an uncertainty of about 1 kHz are obtained by averaging several tens of single line centers measured as indicated above. Systematic corrections and uncertainties are then estimated to get the final measured value. We refer to our previous publications\(^2,3,5\) for a detailed description of systematic effects. Here, we simply note that the bigger effect in our measurements is due to mechanical effects of the light in the dynamic during the interaction time. These effects were well known to produce frequency shifts in accurate saturation spectroscopy measurements in atoms\(^16\), and they were observed and studied in He by us since the first FS measurements\(^17\).

3. Absolute frequency measurements of the \(2^3S \rightarrow 2^3P\) transitions

Lamb shift contributions to the He levels include QED and high order relativistic corrections to the non-relativistic ionization energy, which is calculated with high accuracy for this atom (about \(10^{-22}\) for the lower S states). Precise frequency measurements of transitions between these levels can be used to determine the Lamb shift contributions, and therefore to test the QED theory of ionization energies. We have performed this determination for what is, at present, the best measured Lamb shift in a simple atomic system, including Hydrogen and Deuterium: the \(^4\)He \(2^3S-2^3P\) Lamb shift\(^2\). The experiment measured the three \(2^3S \rightarrow 2^3P\) \((J=0,1,2)\) transitions with unprecedented accuracy for optical He transitions, by using the iodine-locked-master-slave spectrometer and the OFS. The resulting "spin-averaged" frequency of the \(2^3S \rightarrow 2^3P\) transition has an accuracy 30 times better than the previous interferometric measurement\(^18\), as shown in Table 1. Lamb shift of this energy interval with an accuracy of \(6.6 \times 10^{-7}\)

\(^a\)Due to the negligible contribution of the residual frequency noise of all other frequencies of Equation 1, the OFS-locked master stability is only limited by the \(f_0\) stability (i.e. by the quartz stability).

\(^b\)Previous measurements\(^14,18\) were given in parentheses.

<table>
<thead>
<tr>
<th>(f_0) (kHz)</th>
<th>(\Delta \text{QED (MHz)})</th>
<th>(QED (MHz))</th>
</tr>
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<tbody>
<tr>
<td>(2^3P) (F^3S-7)</td>
<td>276 736 405 624.6 (2) 531.2354 (35)</td>
<td>-1 253 949 (58)</td>
</tr>
<tr>
<td>Previous measurements(^14,18)</td>
<td>276 736 405 580 (70) 531.27 (7)</td>
<td>-1 253.9 (1)</td>
</tr>
<tr>
<td>Theory(^20)</td>
<td>276 736 406 100 (1100) 531 2.3 (1.1)</td>
<td>-1 253 75 (40)</td>
</tr>
</tbody>
</table>

Note: Uncertainties are given in parentheses.

was calculated by subtraction of the well known non-relativistic \(2^3S\) and \(2^3P\) ionization energies. Also, the Lamb shift of the \(2^3P\) level was calculated removing the best published value of the largest contribution of the \(2^3S\) Lamb shift, which limited the final accuracy. It is of particular interest to compare the accuracy of the experimental and theoretical determinations of these Lamb shifts. As shown in the Table 1, the theoretical values\(^20\) are between 7 and 300 times less accurate than the experimental ones, meaning a challenge for the He theory. Finally, we want to note that OFS-assisted measurements, as the ones described above, can revolutionize the He frequency metrology, because He one/two photon transitions fall in the visible/near IR region, where the OFS operates.

4. \(2^3P\) fine structure splittings in \(^4\)He: towards a determination of \(\alpha\)

Perhaps the best measured energy intervals in He are the FS splittings of the \(2^3P\) level in \(^4\)He due to the variety of experimental approaches used\(^3-8\). The interest of these measurements lies on the possibility to determine \(\alpha\) with an accuracy of 10 ppb by a measurement and a theoretical determination of the \(2^3P\rightarrow 2^3P\) (\(\nu_{01}\)) splitting with a 0.8 kHz accuracy. We have performed in the years two measurements of both \(\nu_{01}\) and \(\nu_{12}\) splittings by using first the He-locked-master-slave spectrometer\(^5\) and then the I\(_2\)-locked-master-slave spectrometer\(^3\). The results, shown in Figure 3, agree within one standard deviation. More important, an accuracy increment of a factor of two for the \(\nu_{01}\) interval in the case of the I\(_2\)-locked-master measurement was obtained due to the better master stability, that is the limiting factor for this interval. Also in this figure, the agreement with other measurements at the kHz accuracy level is shown. This agreement allows us to calculate a weighted-mean value for each interval considering only published measurements with an uncertainty better than 3 kHz:
$\nu_{12} = 2291.175.3(8)$ kHz (350 ppb)  
$\nu_{01} = 29.616951.8(6)$ kHz (20 ppb)

$\alpha^{-1} = 137.0359872(18)$ is calculated by using the above $\nu_{01}$ value and by considering the last updated theory\textsuperscript{21}. The result achieves the expected accuracy (13 ppb), but it is in disagreement with the present CODATA value\textsuperscript{22} by more than 90 standard deviations. Discrepancies between measured and calculated values (about 9 $\sigma$ for $\nu_{01}$ and 14 $\sigma$ for $\nu_{12}$) must be clarified before an $\alpha$ value from He can be considered in a new adjustment for this constant.

5. $^3$He-$^4$He isotope shift in the $2^3S$-$2^3P$ transition: nuclear-charge-radius determination

The energy difference for a given transition for two different isotopes (i.e., isotope shift) is determined by the difference of nuclear mass and by the different finite nuclear volume and charge distribution. 

$$\Delta \nu_{IS} = \Delta \nu_{MS} + \Delta \nu_{VS} \quad (2)$$

The volume-shift term $\Delta \nu_{VS}$ is dominated, in a first approximation, by an energy proportional to the difference between the square nuclear-charge-radius of both isotopes$^c$ $r_1^2 - r_2^2$, so that:

$$\Delta \nu_{IS} = \Delta \nu_{MS} + C_{nL1-nL2}(Z)(r_1^2 - r_2^2) \quad (3)$$

where the constant $C$ depends on the nuclear charge $Z$, and on quantum-numbers $nL$ of the levels involved in the transition. Therefore, an accurate $^c$Volume-shift corrections due to nuclear polarization or other relativistic recoil effects are negligible in He for IS measurements at the kHz level accuracy$^4$

$\Delta \nu_{IS}$ measurement and $\Delta \nu_{MS}$ calculation could determine an equally accurate value of $r_1^2 - r_2^2$. Moreover, if the radius of one of the two isotopes is well known, the value for the other can then be calculated. This is the case of He, where the nuclear-charge-radius of the $\alpha$ particle has been well measured ($6 \times 10^{-4}$)$^3$. The required accuracy must be, at least, higher than the volume-shift contribution, which for He is a small quantity (about 1.7 MHz for the 33 GHz $^3$He-$^4$He IS at 1083 nm). $\Delta \nu_{MS}$ for He is calculated with sub-kHz accuracy, due to the cancellation of most of the Lamb-shift contribution, which is nuclear-mass independent$^4$. With this $\Delta \nu_{MS}$ uncertainty, our preliminary 5 kHZ accurate measurement of $\Delta \nu_{IS}$ allowed us to determine $r_1^2- r_2^2$ with an uncertainty of $6 \times 10^{-4}$, that is the same uncertainty of the $^4$He nuclear-charge-radius presently known.

To perform the $\Delta \nu_{IS}$ measurement, we have measured the absolute frequencies of the $^3$He hyperfine transitions at 1083 nm by using the OFS-locked-master-slave spectrometer. In Figure 4 we show a record of the nine saturated-fluorescence transitions allowed between hyperfine levels of the triplet 2S and 2P states. Although we measured the frequency of five of these nine components, only in $2^3S_{1/2} \rightarrow 2^3P_{1/2}$ ($\nu_8$) and $2^3S_{1/2} \rightarrow 2^3P_{0,1/2}$ ($\nu_9$) transitions, consistent statistics was considered to extract some preliminary results. The final uncertainty was $1.5 \times 10^{-11}$ for
both transitions. More important, the $\nu_0 - \nu_8$ frequency differs by less than one standard deviation from the well known $2^3S$ HFS splitting, which is used as an accuracy test. The difference between one of these two $3^1H^4$ frequencies, and our previous measurement of $2^3S_1 \rightarrow 2^3P_0$ in $4^4He^2$ does not give the value of the $\Delta V_{1S}$, because the HF-shift must be removed from $\nu_8$ or $\nu_9$. Perturbative determination of HF-shifts is not possible due to the strong HF-interaction in $3^3He$. HF-shifts have been measured by using a phenomenological Hamiltonian to describe the HF-interaction, where HFS measurements or calculations enter as parameters. We have used the last published HF-shift values of the $2^3S_{1,1/2}$, $2^3S_{1,3/2}$ and $2^3P_{1,0,1/2}$ levels, to obtain a preliminary value of $\Delta V_{1S} = 33.668.074$ (5) kHz. More accurate and theory-independent results will be obtained when the measurements of all components (i.e. the $2^3P$ HFS measurements) will be completed. The very good agreement of our result with the last published measurement, highlights one of the characteristics of our method to measure microwave frequency intervals as a difference of absolute optical frequencies: the primary-standard traceability. In fact, our $3^3He$ and $4^4He$ measurements, performed in very different time periods with respect to the same frequency standard, give the same IS value of a previous $3^3He-4^4He$ heterodyne measurement. Taking into account the last calculated values for $\Delta V_{1S}$ and $C_{2S-2P}$, we have determined the $3^3He$ nuclear-charge-radius. The result, compared with other determinations in Figure 5, has an uncertainty of $2 \times 10^{-3}$ fm, which is the second even more accurate value. Finally, we want to note that the uncertainty of the $r_{3^3He} - r_{4^4He}$ difference can be even smaller than the $r_{3^3He}$ uncertainty. Therefore, we can conclude that accurate IS measurements in $3^3He$ and $4^4He$ and H-Tritium could, in principle, improve the knowledge of the proton radius.

References

1. In this book of proceedings see "Precise laser spectroscopy of antiprotonic Helium-weighing the antiproton", R.S. Hayaro and earlier references therein; and "Ultrasound molecular ions: towards precision spectroscopy of HD +", P. Blythe et al.
8. G. Gabrielse, Personal communication. To be published.