Absolute Frequency Measurement of the 1S-3S Transition in Hydrogen

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Abstract. This paper deals with high resolution spectroscopy of hydrogen and deuterium atoms. The 1S-3S and 2S-6S/D transitions have been used to determine the ground state Lamb shift with an accuracy of 46 kHz. The aim of the present experiment is to make an absolute frequency measurement of the 1S-3S transition. We present in this paper the improvement on the experiment and the developpment of a new method to compensate the second order Doppler effect by the application of a magnetic field.

High-resolution spectroscopy of atomic hydrogen is of great interest to test predictions of quantum electrodynamics on this simple atomic system. The best resolution has been observed in Doppler-free two-photon experiments where an atomic beam is excited by two counterpropagating laser beams. The frequency of the laser is determined with frequency chains. These optical absolute frequency measurements superstead all the other measurements (interferometric measurement, radiofrequency measurement). In fact up to now, only 3 transitions have been studied with this process (2S-8S/D, 2S-12D and 1S-2S transitions in hydrogen and deuterium) [1], [2] and [3]. We proposed to realize a second absolute frequency measurement starting from the ground state. Indeed the accuracy of the measurement of the transitions from the metastable state (2S) are now limited by the ac-Stark shift.

To induce the 1S-3S two-photon transition, one needs a radiation source at 205 nm (see Fig. 1). This is produced by quadrupling in frequency a Titanium-Sapphire [4] laser at 820 nm. The first doubling stage in realized by a LBO crystal placed in an enhancement ring cavity. This radiation is sent to an other ring cavity containing a BBO crystal. The main improvement occurs on the first cavity. Up to now, it produced 500 mW at 410 nm light for an incident power of 2 W [5]. We replaced the previous crystal by a better one, giving 700 mW at 410 nm in the same conditions. Now the main losses in the cavity are due to the doubling process. The second doubling stage is still the limiting point of the experiment. Indeed, we produce only few mW of UV light [6].To avoid rapid degradation of the faces of the BBO crystal, the cavity is placed inside a clean chamber filled with oxygen. Moreover, the length of this enhancement cavity is

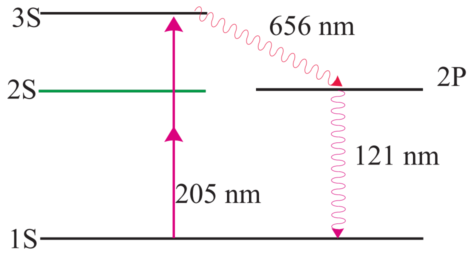

Fig. 1. Energy levels of hydrogen atom

modulated (modulation frequency of 15kHz) so as to be resonant only some of the time. We work in an intermediate regim in which the UV intensity consists of 3μs pulses at a frequency of 30kHz. This method prevents the generation, in the ring cavity, of the counterpropagative wave at 410nm, probably due to a photorefractive effect in the BBO crystal. To secure working of this cavity, the temperature of the crystal is now actively stabilized at 15°C. To observe the 1S-3S transition, we use an atomic beam. Atomic hydrogen is produced by a radiofrequency discharge, which is off-axis with respect to the atomic beam, and linked to a vacuum chamber by a 9 cm lenght of teflon tube. The atomic hydrogen flows through a teflon nozzle into the vacuum chamber which is evacuated by an oil diffusion pump. The atomic beam is carrefully delimited by two diaphragms to eliminate the stray light coming from the the hydrogen discharge. The atomic beam is also placed inside a linear buildup cavity formed by two spherical mirrors (radius of curvature 25 cm). The length of this cavity is 49 cm.(see Fig. 2). The uv beam emerging from the BBO crystal is corrected for astigmatism by

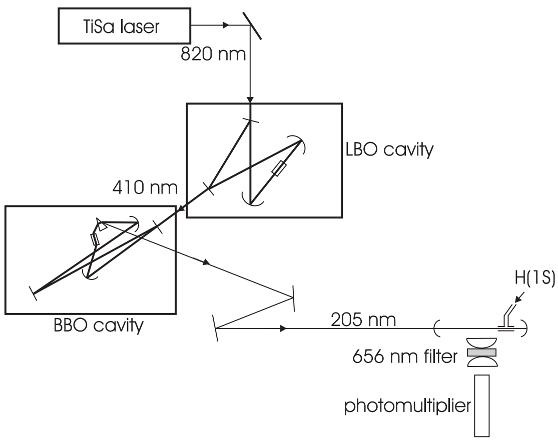

Fig. 2. 1S-3S apparatus

a spherical lens and a cylindrical lens and mode matched into the cavity with two more lenses. Inside the cavity the UV power is typically 10 mW and the beam is focused with a waist of 48 μm. The two cavity mirrors are mounted on PZT stacks and the length of the cavity is locked onto the UV frequency. The two-photon transition is detected by monitoring the Balmer-α fluorescence due to the radiative decay 3S-2P. The detection system was totaly removed. We use now a new photomultiplier freezed at -40°C, and the optical condensers are now anti-reflection coated. The electronic detection system has been improved. The UV radiation composed with pulses drives an electronic gate to enable or not the counting of the photomultiplier signal. In such a way, the noise of detection is decreased. All these improvements lead to an improvement by a factor 15 of the signal to noise ratio (see Fig 3, 4). The accuracy of the absolute measurement of

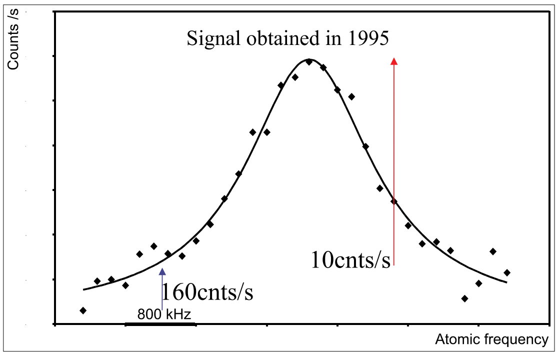

Fig. 3. 1995 signal

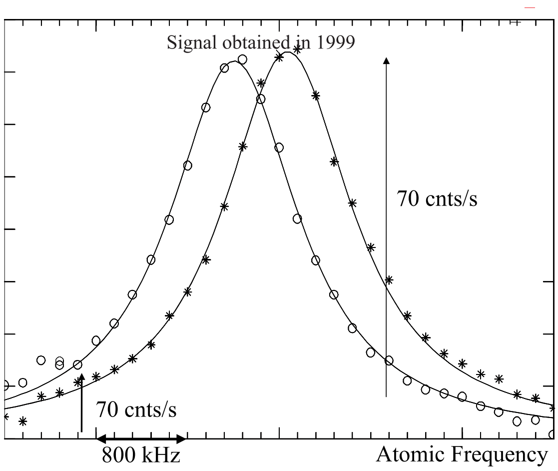

Fig. 4. 1999 signal

the 1S-3S transition is mainly limited by a systematic effect that is the second order Doppler shift. The second order Doppler effect is due to time dilatation in the atom frame. For a transition frequency ν_0 the corresponding frequency shift $\Delta\nu_D$ is given by

$$\frac{\Delta\nu_D}{\nu} = -\frac{\mathbf{v}^2}{2c^2} .$$

In the case of the 1S-3S transition in hydrogen and for an estimated velocity of v=3km/s, the shift is $\Delta\nu_D$=145 kHz. We can't measure the velocity distribution by observing the Doppler broadened 1S-2P transition at 121 nm with a colinear laser beam, because the production of Lyman-α radiation is very difficult. In 1991 a method to compensate or at least to measure this effect was proposed by F. Biraben [7]. The basic idea is to apply a transverse magnetic field B in the atom-laser interaction region. This field has two effects :

- Firstly the degeneracy between hyperfine sublevels is removed by the Zeeman effect.

- Secondly, in the atomic frame, it comes with a motional electric field $\overrightarrow{\mathbf{E}} = \overrightarrow{\mathbf{v}} \times \overrightarrow{\mathbf{B}}$ hydrogen atom are specially sensitive to this field. As this electric field is proportional to v, the corresponding shift of the S level due to the interaction with a near P level, is quadratic with v. Moreover, since the nearest level ($P_{1/2}$) is below the S one, this motional electric field can give a positive frequency shift of the transition $\Delta\nu_S$ able to compensate the negative shift due to the second order Doppler effect.

$$\Delta\nu_S = \frac{\mathbf{E}^2}{\Delta\nu_{S-P}} = \frac{\mathbf{v}^2\mathbf{B}^2}{\Delta\nu_{S-P}} .$$

With an appropriate choice of B the compensation is effective for all atomic velocities. As the velocity of the thermal beam of hydrogen atoms at 300 K is not well known, the magnetic field will be used to measure precisely the velocity distribution, and so the second order Doppler shift.

So the aim of our current experiment is to measure precisely the motional Stark effect and then deduce the velocity distribution. This measure has been made in two steps.

We have calculated exactly the Zeeman effect for the levels 1S, 3S and 3P. Indeed it is necessary to know the shift for all the hyperfine levels very well. These calculations are very classical and we just present the results in a Zeeman diagram (see Fig. 5). The most important part in the diagram is the crossing between the $3S_{1/2}$ (F=1, m_F=-1) and $3P_{1/2}$(F=1, m_F=0) levels, because the quadratic Stark effect is proportional to the square of the induced electric field and inversely proportional to the difference of energy between the two considered levels. Moreover the selection rules for the quadratic Stark effect in our case (E perpendicular to B) impose Δm_F=±1. So it is near this crossing that the motional Stark shift is large enough to be measured. In our calculations the Stark effect is introduced by the formalism of the density matrix [4] where the width of the levels are taken into account. The result of the calculation presented on

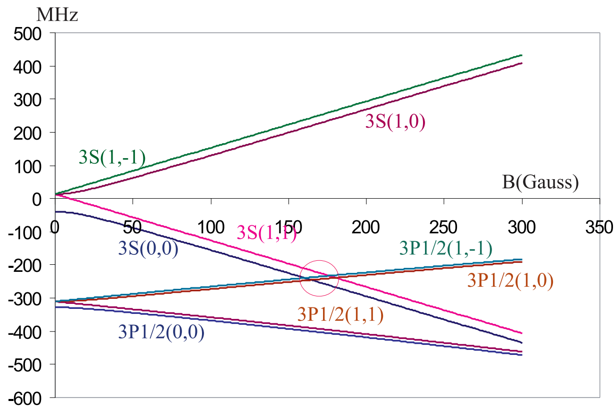

Fig. 5. Hyperfine Zeeman diagram of n=3 level

the figure 9 is a dispersive shape where we have plotted our experimental points. Note that the dispersion line is not adjust on the experimental points.

This crossing appears for a magnetic field about 180 Gauss (0.018 T). The magnetic field is created by two coils in Helmoltz configuration. The current circulating in the coils is given by a regulated magnet power supply delivering 7 kW under 0.25 Ω. The coils are realized with tubular copper frame (23 tr). The coils are refreshed by a water flow circulating inside the pipe. With this apparatus we can generate a magnetic field of 250 G (see Fig. 6). Afterwords it

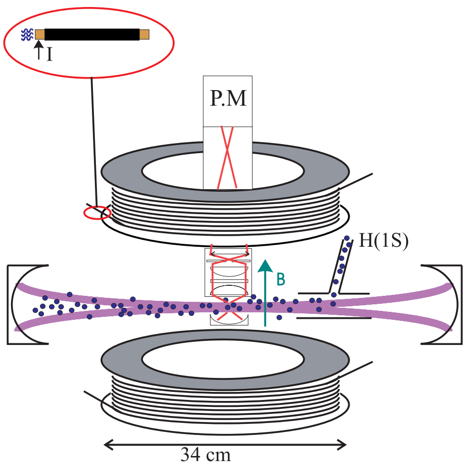

Fig. 6. Scheme of the coils

is necessary to calibrate the field seen by the atoms. For that we monitor the $1S_{1/2}$(F=1, m_F=0)- $3S_{1/2}$(F=1, m_F=0) two photon transition. At first order, this transition is frequency dependent with the magnetic field while the other transitions ($m_F=\pm$ 1) are not(see Fig. 7). For example, we present the recording

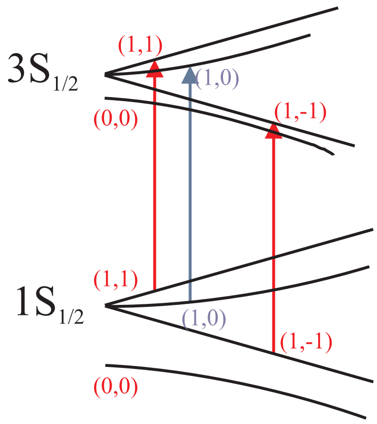

Fig. 7. Calibration of the magnetic field

of the 1S-3S transition in a magnetic field of 4G on the figure 8. For each magnetic

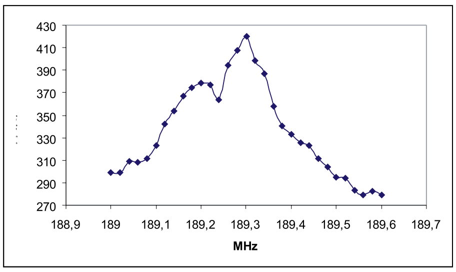

Fig. 8. 1S-3S signal with B=4G

field, we record :

- the $1S_{1/2}$(F=1, $m_F=\pm1$)- $3S_{1/2}$(F=1, $m_F=\pm1$) transitions with B
- the $1S_{1/2}$(F=1)- $3S_{1/2}$(F=1) transitions without B .

Each point on the figure 9 represents the frequency difference between these two lines for one magnetic field. These are preliminary results, the line shape used to extract the center of the two photon signal is very simple(see Fig. 9).

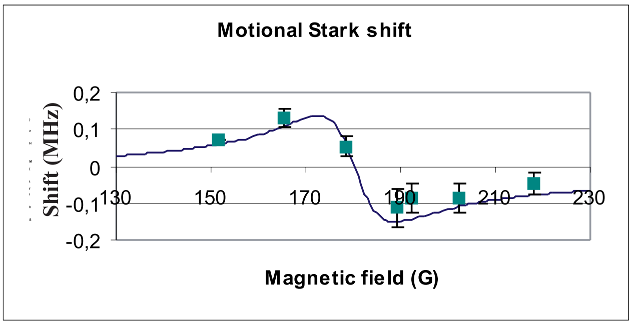

Fig. 9. The motional Stark shift

Conclusion

We have improved the signal to noise ratio of the experiment by a factor 15, this allows us to determine very precisely the 1S-3S transition frequency with a reasonnable integration time. The preliminary results for the second order Doppler effect are very stimulating. We plan to make more precise measurements of this effect. The frequency chain to make an absolute frequency measurement is in preparation in the Laboratoire Primaire du Temps et des Fréquences. The determination of the absolute frequency of the 1S-3S transition is planed within one year.

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