

Precision RF Spectroscopy of Circular Rydberg Atoms

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INTRODUCTION

The Rydberg R is the best known fundamental constant in physics (precision of about 3×10^{-10}). It has been measured by various consistent optical spectroscopy experiments performed on atomic hydrogen [1, 2, 3]. The current precision is in fact limited by the reliability of wavelength measurements in the optical domain (1.6×10^{-10}). It would be very desirable to improve further the precision of this measurement.

A natural direction to improve the resolution of R measurements beyond the present level is to attempt the experiments at longer wavelengths, in a domain where the frequency of the radiation can be linked directly to the atomic time standard. A possible way is to measure R on microwave transitions involving what is called circular Rydberg atoms [4, 7].

THE CIRCULAR RYDBERG ATOM

Circular Rydberg atoms are highly excited atoms, i.e. Rydberg atoms (with a large principal quantum number, $n \geq 10$) with a maximum value of the angular momentum l and of its projection m along a quantization axis ($l = |m| = n-1$). These atoms are quasi-classical: the wave function of the valence electron has the shape of a thin torus lying around the classical circular orbit of the Bohr theory, whose radius is $a_0 n^2$ (where a_0 is the Bohr radius). These atoms realize exactly Bohr's atom model and are well adapted to the Rydberg constant metrology. The advantages of using these atoms for a Rydberg constant measurement are numerous:

- (i) These atoms have a very long natural life-time. Their radiative decay occurs entirely towards the $n-1$, $l=m=n-2$ lower circular level within a time proportional to n^5 , typically of the order of 10^{-3} s for $n=30$.

- (ii) Circular Rydberg atoms have non-permanent electric dipole moment and hence no linear Stark effect. Stray electric field shifts and broadenings, which are usually very important in the spectroscopy of the low angular momentum Rydberg levels, can thus be minimized and much better controlled.
- (iii) In circular atoms, the Rydberg electron remains always very far from the nucleus. Hence, all the contact terms, which become significant corrections at the 10^{-10} level in the optical experiments and which depend upon the not-so-well known proton form factor, are in circular states completely negligible. Lamb-shift corrections are also very small for these states. From the point of view of Q.E.D. corrections, circular atoms are, by far, the best candidate for R metrology.
- (iv) At the 10^{-11} precision level, it is not even required to perform the Rydberg measurement on a Hydrogen circular state : in alkali atoms, the circular electron sees the core made of the nucleus and the $Z-1$ remaining electron mainly as a point-like charge. The small correction due to the core polarizability can be expressed as a very small quantum defect in Lithium [5]. Actual knowledge of this polarizability corresponds to uncertainties in the 10^{-11} range.

We present the preliminary results of an experiment performed on quasi-hydrogenic circular Rydberg states of Lithium atoms whose microwave transitions are induced in a Fabry-Perot cavity [6]. We have observed transit time width-limited resonances between adjacent circular Rydberg levels at frequencies around 447 GHz and 270 GHz. Our results can presently be interpreted as a measurement of R with an accuracy of 6×10^{-9} . It is still one order of magnitude off the present accuracy of the optical experiments and two orders of magnitude away from the planned goal (10^{-11}). However this experiment is interesting since it has allowed us to study in detail the properties of circular Rydberg atoms and to analyze their behavior in electric and magnetic fields. This is an essential task to design carefully the final experiment and to control the various systematic effects susceptible to cause errors in the measurement. The improvement of the present experiment mainly involves the lengthening of the atom-microwave interaction time. This can be done either by slowing down the atoms or by using a long cavity geometry such as a Ramsey design split cavity.

THE EXPERIMENT

The circular atom microwave spectroscopy experimental set-up is sketched on Fig. 1-a. A thermal beam of Li atoms crosses three sections of the apparatus : the excitation, the microwave interaction region and the detection zone. The whole set-up is protected from room temperature thermal radiation by a liquid nitrogen cooled shield (which can be replaced in a later stage of the experiment by a liquid helium cooled one).

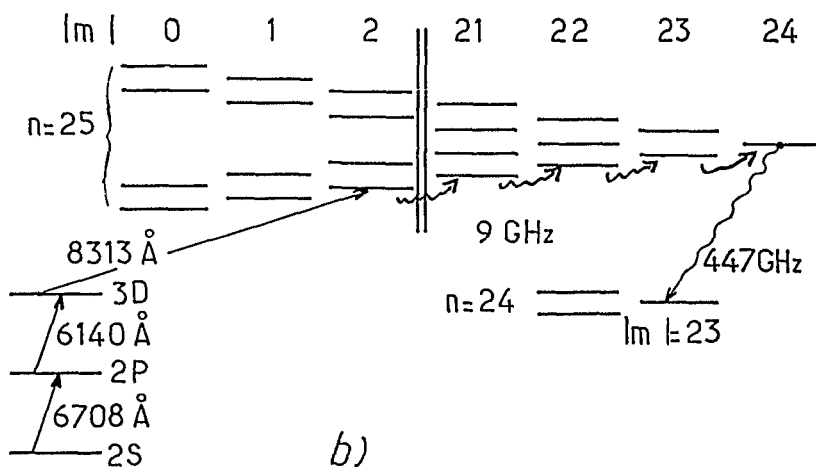
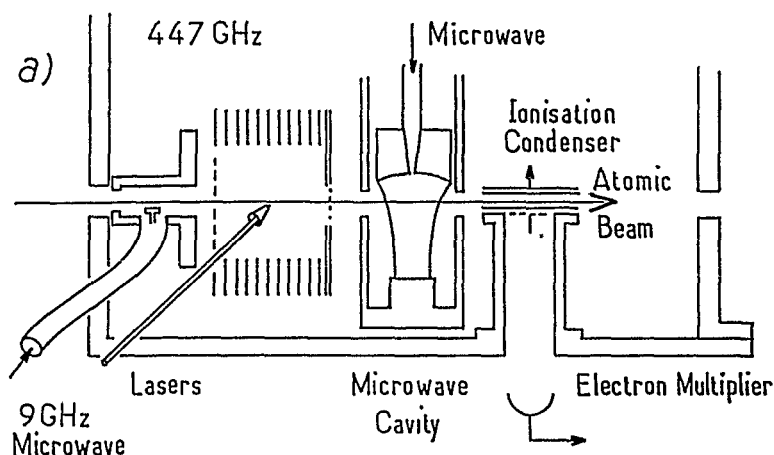


Fig. 1: a) Schematic of the experimental set-up
b) Diagram of the Li^7 levels involved in the experiment

The preparation of the Rydberg circular states is performed by pulsed laser excitation followed by an adiabatic microwave transfer method (A.M.T.M.) already described in ref. [8]. This method requires that the atom interacts with an homogeneous electric field F_1 , produced by the stack of equally spaced metallic plates shown on Fig. 1-a. This field removes the degeneracy of the various n -manifolds.

The laser excitation prepares the atoms in the lowest energy Stark state with $|m|=2$ in a chosen n -manifold ($n=24$ for example) (see Fig. 1-b). This excitation is itself a stepwise process involving three pulsed dye laser beams in resonance with the $2S-2P$, $2P-3D$ and $3D-n$, $|m|=2$, $n_1=0$ transitions respectively (wavelengths 6708, 6104 and about 8300 Å). From this Stark sublevel, the atoms undergo a succession of fast adiabatic passage transitions bringing them into the circular state of the manifold ($|m| = n-1$, $n_1 = 0$). These transitions, shown by the arrows on Fig. 1-b, are induced by a 9 GHz microwave field produced by an X-band klystron and the successive $m \rightarrow m+1$ steps are tuned in resonance one after the other by sweeping down the electric field F_1 and taking advantage of the slight frequency difference of the successive transitions (due to second order Stark effect). The microwave field is sent on the atom through a 25 mm diameter hole pierced in the stack of the Stark plates.

The circular atoms then cross a Fabry-Perot semi-cofocal cavity in which a resonant millimeter wave is fed through a coupling hole. The cavity, tuned by slight mirror translation, sustains a Gaussian mode, corresponding to a time-varying Gaussian envelope of the field seen by each atom as it crosses the cavity. The microwave around 447 GHz (for the one-photon 24-25 transition) is produced by a phase-locked backwave oscillator (Thomson-CSF) driven by a microcomputer, whose frequency can be stabilized, swept and measured continuously with reference to a time standard. After crossing the microwave cavity, the atoms enter into the Rydberg ionization zone, where they are selectively detected by the field ionization method [7]. A ramp of electric field $F_2(t)$ is applied after an appropriate delay between two metallic plates surrounding the atomic beam. This field reaches at different times the ionization threshold for the various n, m, n_1 states involved. It turns out in particular that the circular state of the n -manifold ionizes in a higher field than the lowest m levels. The time resolved ionization signals thus provide an unambiguous signature of the

circular Rydberg atoms. As the microwave frequency is scanned across a resonance line, the time-dependent Rydberg ionization signals change, since the circular n and $n+1$ or $n-1$ states have different ionization thresholds and appear at different times in the field ramp $F_2(t)$. The resonance signal can be conveniently plotted as a function of the microwave frequency. A computer is used to compute the circular state to circular state transfer rate (ratio of the peak intensities corresponding to the final and initial states populations) and this rate is plotted versus measured microwave frequency.

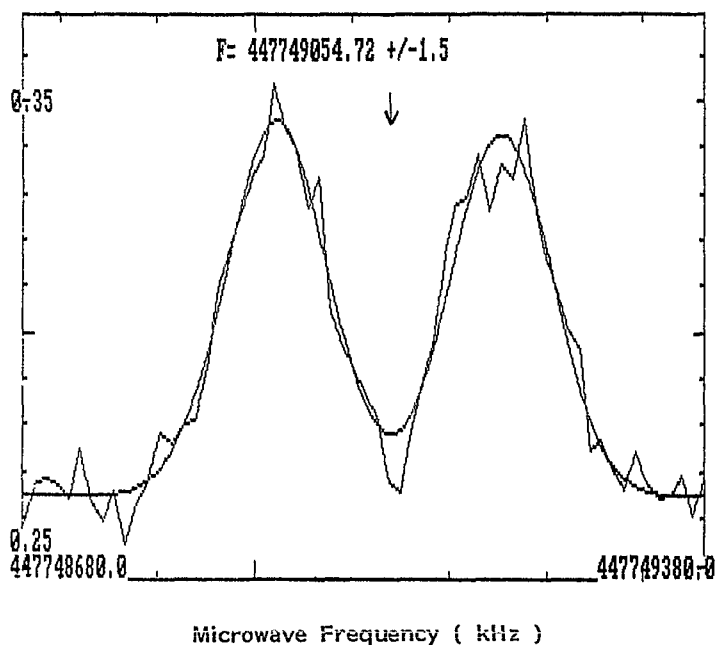


Fig. 2: $n=24$ to $n=25$ circular-circular transition microwave spectrum. The population transfer rate is plotted as a function of the microwave frequency. The smooth curve corresponds to the Gaussian fit. F is the fitted central frequency (arrow)

Fig. 2 shows such a typical resonance signal. It has been recorded with the atoms interacting with an electric E field (about 1V/cm) and a magnetic B field (about 0.1 Gauss) both oriented in the cavity axis vertical direction. The B field splits the line in two Zeeman components, corresponding to

both circular orbits' helicities. The E field shifts both Zeeman components by a small amount (about 7 kHz).

The Fig. 2 experimental points result from the averaging of 20 pulses recorded for each frequency. The frequency scan is performed randomly to avoid systematic drift effects. The width at half maximum of the resonance : 120 kHz, corresponds to the theoretical limit due to the finite transit time across the waist of the microwave Gaussian beam. The observed Zeeman components have not the same weight. This is just related to the resonant 447 GHz microwave source whose polarization is partially elliptical.

THE ANALYSIS OF THE RESULTS

We have performed a precise measurement of the $n=24$ to $n=25$ circular-circular transition in Lithium. This measurement is based on 63 atomic resonance spectra recorded in about the same conditions as the spectrum seen in Fig. 2.

Using a least squares iterative method, each spectrum is fitted with two Gaussian lines. The smooth curve seen on Fig. 2 corresponds to such a Gaussian fit. This procedure provides a rather accurate value of F_C , the frequency of the center of the two Zeeman lines. It provides also the F_C uncertainty interval. This interval is related to the difference between the best fit and the experimental spectrum, which is here mainly due to noise. Typically, for a sweep such as the one presented on Fig. 2, F_C is determined to within ± 2 kHz (variance).

The about 1V/cm E electric field is measured, and F_{exp} the unperturbed transition frequency is then deduced from F_C by subtracting to F_C the calculated Stark shift (the other corrective effects are negligible at our scale of accuracy). The E uncertainty, lower than 5%, corresponds to ± 50 Hz on the frequency, and can be neglected.

Fig. 3 summarizes the results. F_{exp} is given for each spectrum with its error bars. Data are seen in time order in three groups separated by a small vertical blank space, corresponding to three different days. The first group of data has been recorded with a first cavity, where the interaction time is shorter, making the linewidth typically twice as large. In this first group, the error bars are typically twice as large. The two following groups have been recorded with a second cavity.

The averaged value of F_{exp} (weighted by the inverse square of the uncertainties) is :

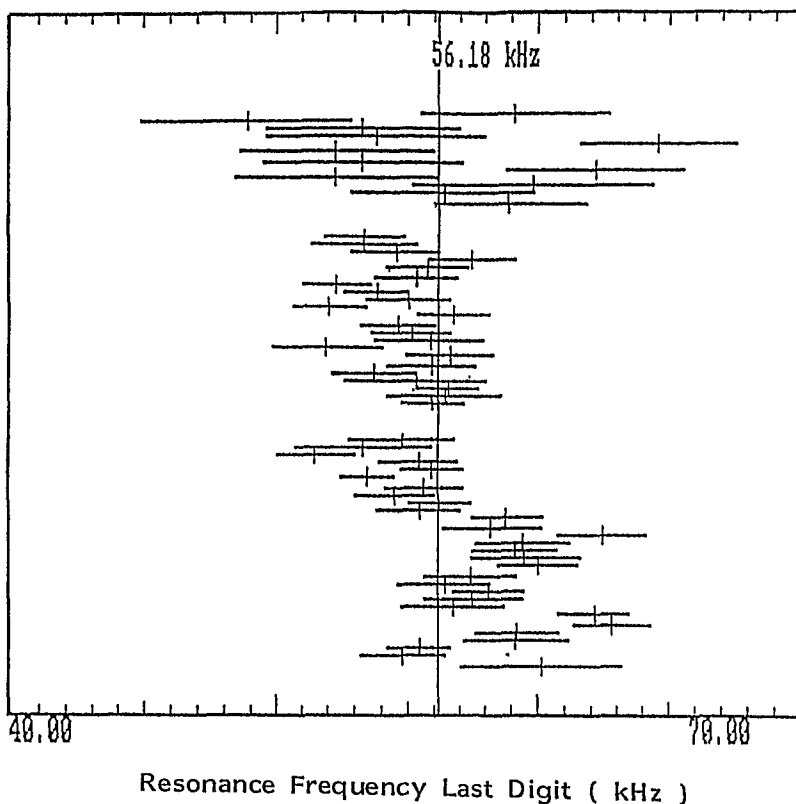


Fig. 3: Plot of the resonance frequency (447 749 056.18 kHz) of the $n=24$ to $n=25$ circular-circular transition of Li^7 . The vertical straight line corresponds to the data weighted average

$$F_{\text{exp}} = 447\,749\,056.18 \text{ kHz}$$

The theoretical predicted value is :

$$F_0 = 447\,749\,056.43 \text{ kHz}$$

In the F_0 calculation, we have not taken into account the following interactions :

- the hyperfine coupling (about 5 Hz on F_0),
- all the exchange or correlation effects between the core electrons and the Rydberg one (below 1 Hz),
- the Q.E.D. corrections, i.e. Lamb-shift (50 Hz),
- the B and E fields higher order terms (less than a few Hz).

The theoretical relative uncertainty on F_0 , except the 1.7×10^{-10} one due to the Rydberg itself [9], is about 4×10^{-11} . It is related to the actual one

percent precision on the static dipolar polarizability of the $LiI(1s)2$ ground state, needed to evaluate the electron-core interaction : the uncertainty on the atomic mass (about $1 \cdot 10^{-8}$) step is only at the $4 \cdot 10^{-12}$ level.

At first, we can note that the F_{exp} mean value is very close to F_0 (250 Hz), and therefore compatible with the very low uncertainty about 250 Hz (accuracy of $6 \cdot 10^{-10}$), that is precisely obtained if we assume that the noise on the spectra is the only cause of dispersion. However, as it appears clearly on Fig.3, this dispersion, characterized by the standard deviation of 2.7 kHz, is too large in comparison with the noise's amplitude. The effective distribution of F_{exp} suggests a dependance versus time, probably related to fluctuating systematic shifts.

In our case, the possible fluctuating systematics are the following :

- * The horizontal components of the magnetic field (In particular, the one parallel to the laser axis) cannot be exactly controlled. It results in a time-dependent drift within ± 300 Hz.
- * The cavity is tuned by maximizing the atomic transfer rate. By this way, the cavity center frequency equals F_c within about 25% of the cavity resonance half-width. The slow variation of the MW power through the linewidth shifts the line in direction of the cavity center frequency. Since the cavity has a rather low quality factor, this shift is at the worst ± 500 Hz. Due to thermal drift, the cavity tuning may vary at the time scale of an hour, and yield a time-dependent ± 500 Hz systematic.
- * For large atomic populations (about 1000 atoms detected), we have observed shifts of a few kHz, attributed to collective effects. Although all the reported measurements have been performed with less than 300 atoms, we are actually unable to exclude the possibility of a residual shift of about ± 500 Hz.

These three fluctuating effects, with their respective orders of magnitude, account for the observed standard deviation $\pm 6 \cdot 10^{-9}$ (± 2.5 kHz), which therefore represents the effective uncertainty on our measurement. Residual first order Doppler effect and relativistic corrections are negligible at this scale.

Finally, this preliminary experiment, where the microwave interaction time is two orders of magnitude shorter than the atomic lifetime, can be reinterpreted as a R Rydberg constant measurement with a $6 \cdot 10^{-9}$

accuracy. It demonstrates the feasibility of a competitive R measurement in frequency unit on microwave transition involving circular states.

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