

# MEASUREMENT OF Cs-BUFFER GAS COLLISIONAL FREQUENCY SHIFT USING PULSED CPT INTERROGATION

O. Kozlova<sup>(1)</sup>, R. Boudot<sup>(2)</sup>, S. Guérandel<sup>(1)</sup> and E. de Clercq<sup>(1)</sup>

<sup>(1)</sup>*LNE-SYRTE, Observatoire de Paris, CNRS-UMR 8630, UPMC  
61 Avenue de l'Observatoire, 75014 Paris, France  
[olga.kozlova@obspm.fr](mailto:olga.kozlova@obspm.fr)*

<sup>(2)</sup>*FEMTO-ST, Time & Frequency Dpt, CNRS UMR 6174, Besançon, France*

## INTRODUCTION

The coherent population trapping (CPT) phenomenon [1] has been investigated by several laboratories for a few years in view of applications to atomic clocks [2, 3, 4]. In such clocks a buffer gas is added in the alkali vapor cell in order to increase the interaction time by reducing the effect of wall collisions and in order to reduce the Doppler width by Dicke effect. However the collisions between alkali atoms and the buffer gas can shift the frequency of the resonance by several kHz per Torr, and this shift is temperature dependent.

The temperature dependence of the collisional shift can be written [5] as

$$\Delta\nu(T) = P_0 ( \beta + \delta(T-T_0) + \gamma(T-T_0)^2 ) \quad (1)$$

where  $P_0$  is the buffer gas pressure in the cell at the reference temperature  $T_0$ ,  $\beta$  is the pressure coefficient (Hz.Torr<sup>-1</sup>),  $\delta$  is the linear temperature coefficient (Hz.Torr<sup>-1</sup>.K<sup>-1</sup>),  $\gamma$  is the quadratic temperature coefficient (Hz.Torr<sup>-1</sup>.K<sup>-2</sup>) and  $T$  is the buffer gas temperature.

Using a mixture of gases having temperature coefficients of opposite signs, one can cancel the temperature dependence at the working temperature. Another possibility to cancel the temperature dependence using only one buffer gas is to work around the inversion temperature (for the buffer gases which shows strong quadratic temperature dependence). Unlike Rb, the temperature coefficients are poorly known for Cs.

In this study we propose separate measurements of the temperature coefficients for clock transition in Cs for N<sub>2</sub>, Ar, Ne buffer gases. To determine the value of the coefficient we have measured the shift of the clock frequency versus the cell temperature. The combination of two original techniques [6]: double-lambda scheme of CPT-resonance excitation and a temporal Ramsey interrogation technique allows us to have high contrast and narrow CPT resonances with reduced light shift dependence. This method provide a better frequency uncertainty than previous ones in Cs (Optical Pumping [7] or CPT [8] methods) and allows to reveal the quadratic shift dependence.

To reveal the coefficient an important parameter is the actual buffer gas pressure in the cell, which was measured from the shift of the optical transitions (D<sub>1</sub> line).

## MEASUREMENTS OF THE TEMPERATURE SHIFT OF THE MICROWAVE TRANSITION

### Experimental setup

The experimental setup used for temperature shift measurements is shown in the figure 1.

The optical radiations required to pump the atoms into the dark state are generated by two external cavity diode lasers (ECDL) tuned to the Cs D<sub>1</sub> line and phase locked with a frequency difference tunable around 9.192 GHz by comparison with a low noise microwave local oscillator referenced to a hydrogen maser. We use a so-called double-lambda scheme with lin per lin polarized laser beams propagating parallel to the static magnetic field applied to the cell. The atomic response is detected by transmission of the beam through the cell.

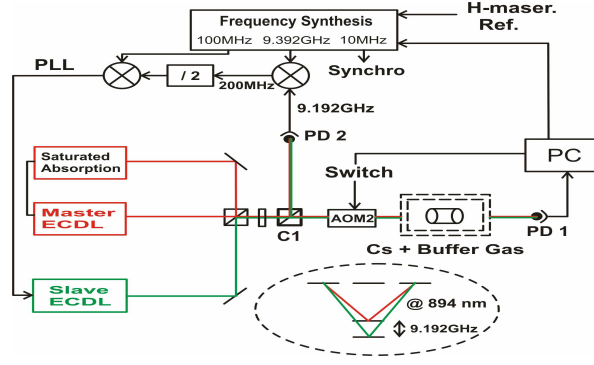


Fig.1. Experimental setup for temperature shift measurements. AOM: acoustooptical modulator, PD1: atomic signal photodetector, PD2: fast silicon photodiode, ECDL: external cavity diode laser. The insert shows the atomic energy levels involved in the double-lambda scheme.

A CPT pulse sequence where each pulse is used both for the coherence preparation and the atomic signal detection is applied. The light pulse durations are controlled by an acousto-optical modulator. A temporal Ramsey interrogation technique leads to narrow fringe widths scaling as  $1/(2T)$  not limited by the saturation effect. It has been shown [9] that in the pulsed regime the light shift dependence is greatly reduced.

We use sealed centimeter-scale Cs vapor cells filled with one buffer gas (30 Torr  $N_2$ , 30 Torr Ar or 90 Torr Ne). The temperature of the cell can be changed in the range from 25°C to 60°C and stabilized within the mK level. A static magnetic field is applied with a solenoid to raise the Zeeman degeneracy. The ensemble is surrounded by two mu-metal magnetic shields.

For each Cs-buffer-gas cell the resonance frequency is measured for different temperatures. Each measurement is performed by locking the synthesizer frequency on the CPT signal of the clock transition (Zeeman sublevel  $m = 0$ ). The measured frequencies are corrected for the Zeeman quadratic shift and for the light shift.

### Experimental results for temperature dependence of the collisional shift

The temperature dependence of the collisional shift was measured for three Cs cells filled with 30 Torr  $N_2$ , 30 Torr Ar or 90 Torr Ne (fig. 2). For the cells with buffer gases  $N_2$  and Ne the experimental points were fitted with 2<sup>nd</sup> order polynomial fit. The cell with Ar buffer gas perfectly fits with linear curve.

For 90 Torr Ne cell we have measure temperature dependence of the collisional shift using Ramsey technique in the range from 25°C to 65°C. Then from the fitting coefficients we find the inversion temperature around 77°C. To confirm the inversion temperature the measurement of the temperature dependence were made in the range from 70°C to 80°C, but in continuous mode (not pulsed Ramsey technique) (fig. 2c , grey color). The inversion temperature does not depend on the pressure, so no extra pressure measurement is needed. It is important to notice, that the presence of the inversion temperature at such temperature makes the buffer gas Ne very attractive for use in the Cs chip scale clocks.

For 30 Torr  $N_2$  cell from the fitting coefficients we estimate the inversion temperature around 157-158°C. For the temperature less than 157 °C  $N_2$  buffer gas leads a positive shift of the Cs clock transition.

The measured shift in Ar is linear in the range from 25° to 60°C. So inversion temperature cannot be predicted. The Ar buffer gas leads to a negative shift.

It is usual technique to cancel (or minimize) to the temperature dependence (at chosen working temperature) by making a mixture of two buffer gases with opposite signs. The precision measurement of the temperature dependence and coefficients we obtain (after pressure determination) will make more predictable the temperature range with canceled temperature dependence for a given Ar/ $N_2$  mixture.

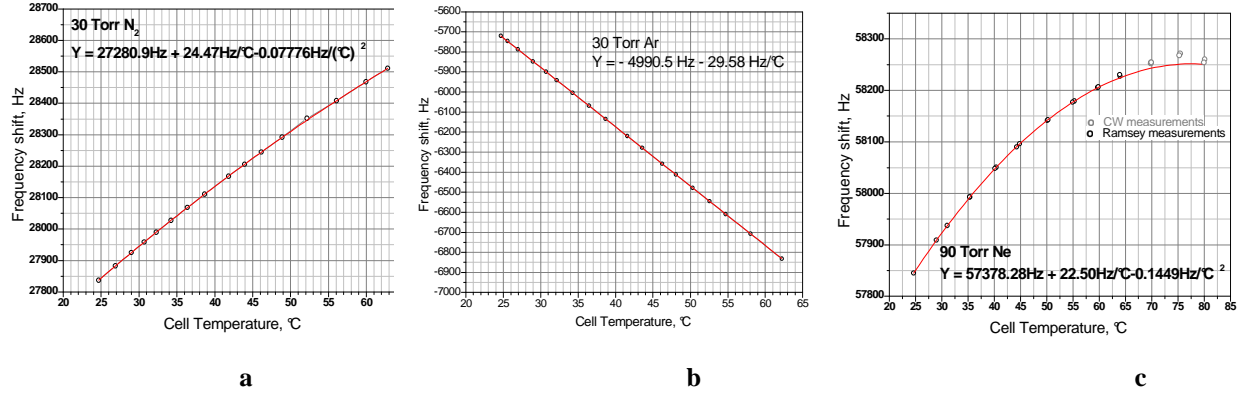


Fig. 2. Temperature dependence of the collisional shift for Cs cells with buffer gases:  
a) 30 Torr N<sub>2</sub> ; b) 30 Torr Ar ; c) 90 Torr Ne .

## BUFFER GAS PRESSURE MEASUREMENTS WITH THE OPTICAL TRANSITION SHIFT

### Experimental setup

The experimental setup for the measurement of the optical shift in Cs cells with buffer gas is shown in the figure 3.

We use Distributed Bragg Reflector laser diode (DBR) tuned to Cs D<sub>1</sub> line. Slow current modulation is applied to scan the frequency over 12GHz. Fast current modulation ( $f_m$ ) is applied in order to use phase sensitive detection technique. We are working with low intensities to avoid the effects of optical pumping. The power transmitted through the Cs cell and the Cs+buffer gas cell are detected by photodiodes PD1 and PD2 respectively. The PD0 is used to register the power changes during the frequency scanning. Then the power changes are eliminated from the PD1 and PD2 signals.

We use two techniques to measure the optical shift: absorption signal and 3rd derivative signal provided by lock-in-amplifiers (LA1 and LA2).

In the first case (absorption signal) the absorption signals from the Cs cell and the Cs+buffer gas cell are registered simultaneously. Then the absorption signal of Cs+buffer gas cell is fitted by a Voigt profile, the signal from the Cs cell by a Gauss profile. The positions of the peaks are extracted from the fit. To calibrate our measurements we correlate the difference in the peaks position of the Cs (without buffer gas) absorption curve with well known hyperfine frequency structure of D<sub>1</sub> line (fig. 4). The shift of the peak position in Cs+buffer gas cell compared to the Cs cell is the optical shift proportional to the buffer gas pressure.

The second method uses 3rd derivative signal provided by lock-in-amplifiers. Optical shift value is extracted from the zero crossing that corresponds to the peaks position (fig. 5). For the cell with 30 Torr N<sub>2</sub> the comparison of the optical shift values provided by the described methods gives the error bar within 2 MHz.

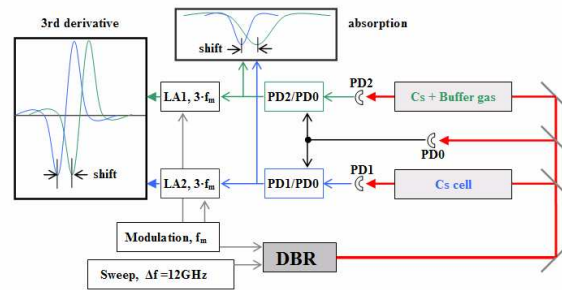


Fig. 3. Experimental setup for optical shift measurements.

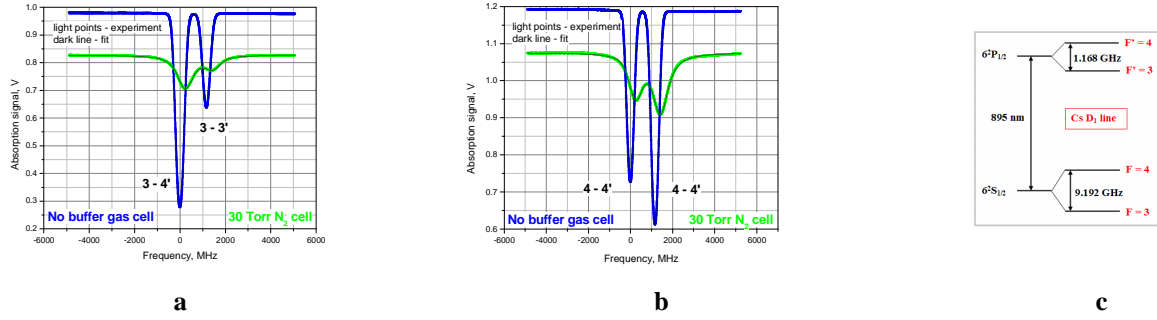


Fig. 4. Absorption method; a) Transitions  $F=3 - F'=4$  and  $F=3 - F'=3$ ; b) Transitions  $F=4 - F'=4$  and  $F=4 - F'=3$ ; c) Cs D<sub>1</sub> line level diagram.

### Buffer gas pressure determination

To determine the buffer gas pressure we use published coefficients [10,11] of pressure shift for optical transitions. The optical shift values for three buffer gas cells (30 Torr N<sub>2</sub>, 30 Torr Ar or 90 Torr Ne), pressure shift coefficients for the optical transitions and calculated values for the buffer gas pressure in the cells are presented in the table 1. The buffer gas pressure in N<sub>2</sub> buffer gas cell is expected  $30.3 \pm 3$  Torr at 0°C, in Ar cell  $26.3 \pm 4$  Torr, in Ne cell  $85.2 \pm 6$  Torr.

Table 1 Optical shift value, pressure shift coefficient and buffer gas pressure value.

Cell	Optical shift value at 22°C	Pressure shift value	Pressure value at 22°C	Pressure value at 0°C
30 Torr N <sub>2</sub>	$-259.8 \pm 4$ MHz	$-7.69 \pm 0.01$ MHz/Torr [10] $-8.23 \pm 0.02$ MHz/Torr [11]	$32.4 \pm 3$ Torr	$30.0 \pm 3$ Torr
30 Torr Ar	$-183.8 \pm 5$ MHz	$-6.47 \pm 0.01$ MHz/Torr [10]	$28.4 \pm 4$ Torr	$26.3 \pm 4$ Torr
90 Torr Ne	$-147.4 \pm 6$ MHz	$-1.60 \pm 0.01$ MHz/Torr [10]	$92.1 \pm 6$ Torr	$85.2 \pm 6$ Torr

### TEMPERATURE COEFFICIENTS RESULTS

In the table 2 previously published and new measured pressure and temperature coefficients for Cs clock transition are shown. For the first time the quadratic coefficients  $\gamma$  for N<sub>2</sub> and Ne have been determined. For Ar buffer gas, the quadratic coefficient  $\gamma$  is much smaller and we have estimated its upper limit.

Table 2. Pressure and temperature coefficients for Cs clock transition in N<sub>2</sub>, Ar and Ne buffer gases.

Buffer gas	pressure coefficient $\beta$ (Hz/Torr)	linear temperature coefficient $\delta$ (Hz/Torr K)	quadratic temperature coefficient $\gamma$ (Hz/Torr K <sup>2</sup> )	Reference
N <sub>2</sub>	<b><math>910.0 \pm 90</math></b>	<b><math>0.816 \pm 0.09</math></b>	<b><math>-(2.59 \pm 0.24) \cdot 10^{-3}</math></b>	Present
	$924.7 \pm 7$	$0.623 \pm 0.05$		[12]
Ar	<b><math>-189.8 \pm 33</math></b>	<b><math>-1.12 \pm 0.18</math></b>	$< 2 \cdot 10^{-5}$	Present
	$-191.4 \pm 3$	$-1.05 \pm 0.5$		[12]
	$-216 \pm 10$			[13]
	$-193 \pm 4$	$-1.00 \pm 0.08$		[14]
Ne	<b><math>673 \pm 48</math></b>	<b><math>0.264 \pm 0.019</math></b>	<b><math>-(1.7 \pm 0.1) \cdot 10^{-3}</math></b>	Present
	$647 \pm 4$			[14]
	$652 \pm 20$	$0.14 \pm 0.10$		[15]

## CONCLUSIONS

In this paper we presented separate measurements of the temperature coefficients for clock transition in Cs for N<sub>2</sub>, Ar and Ne buffer gases. The inversion temperature for Ne has been determined (77°C). For N<sub>2</sub> the inversion temperature is estimated at 157-158°C. We have reduced the uncertainties for linear temperature coefficients for Cs clock transition three buffer gases. The quadratic coefficients  $\gamma$  for N<sub>2</sub> and Ne have been determined for the first time. For Ar buffer gas, the quadratic coefficient  $\gamma$  is much smaller and we have estimated its upper limit.

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