Cs buffer gas collisional frequency shift: method and preliminary measurements

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***Abstract* — In this paper we propose separate measurements of the temperature coefficients of the Cs clock frequency shift caused by Cs-buffer gas (Ar, Ne, N_2) collisions, using the CPT interaction. The method of measurement of the temperature coefficients is described and the preliminary results for a Cs vapour cell containing 30 Torr of Ar are shown.**

I. 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].

The CPT requires to connect two ground states of an atomic specie to a common excited state through two phase-coherent light fields. When the frequency difference between both optical frequencies is equal to the hyperfine splitting, the atoms are trapped in a state superposition where the photons are not absorbed (dark state). The atomic medium transparency and the transmitted detected light are then maximal. The resonance linewidth is limited by the lifetime of the ground state hyperfine coherence created in the atomic medium.

Optimizing a CPT atomic clock then requires to increase the coherence lifetime and to enhance the signal-to-noise ratio (SNR) of the output-detected signal. To increase the coherence lifetime one adds buffer gas in the alkali vapour cell [3] or applies an anti-relaxation coating on the walls [4]. In vapour cells containing an additional buffer gas the frequency of the resonance is shifted (collisional shift) by several kHz per Torr. This shift is temperature dependent. In case of wall-coated cells, the frequency is also shifted (wall shift) by a value about of 100Hz. This shift is mostly ''aging'' dependent.

In SYRTE, it has been shown [5] that, the Cs pulsed CPT clock long term performance is limited by the Cs collisional shift fluctuations in the buffer gas cell. A metrological study of these shifts (in Cs) is then required.

The temperature dependence of the collisional shift can be written [6] as

$$\Delta\upsilon(T) = P_0\ (\ \beta + \ \delta(T-T_0) + \gamma\,(T-T_0)^2\ ), \qquad (1)$$

where p_0 is the buffer gas pressure in the cell at the temperature T_0 = 273K, β is the pressure coefficient (Hz/Torr), δ is the linear temperature coefficient (Hz/Torr K), γ is the quadratic temperature coefficient (Hz/Torr K^2) and T is the buffer gas temperature.

Normally, linear temperature shift is much greater than the quadratic shift, and for a given buffer gas cell we can consider that the temperature dependent collisional shift is proportional to the gas temperature. Using a mixture of gases having linear temperature coefficients of opposite signs, it is possible to cancel the linear temperature dependence at the chosen working temperature. Unfortunately, unlike for Rb, the temperature coefficients are poorly known for Cs (large uncertainty for the linear coefficients (tabl. 1), unknown quadratic coefficients). Recent chip-scale clock uses a buffer gas compensated (Ar and N_2) Cs vapour cell, but no details on the buffer gas mixture is given [7].

TABLE I. Cs TEMPERATURE COEFFICIENTS AVAILABLE IN THE LITERATURE

Buffer gas	Relative Pressure coefficient β/ν_0^*, $Torr^{-1}$	Relative Linear temperature coefficient δ/ν_0, $Torr^{-1}\,K^{-1}$	Reference
N_2	+89 10^{-9} +100.6 10^{-9}	**+80 10^{-12}** **+67.8 10^{-12}**	[8] [9]
Ne	+63.1 10^{-9} +70.9 10^{-9}	**+110 10^{-12}** **+15.2 10^{-12}**	[10] [9]
Ar	- 23.1 10^{-9} - 20.8 10^{-9}	**- 47.0 10^{-12}** **- 114.2 10^{-12}**	[8] [9]

* ν_0 – frequency of the hyper fine splitting of the ground state of Cs, 9.192631770 GHz

In this study we propose separate measurements of the temperature coefficients in Cs for of Ar, Ne, N_2 buffer gas using the CPT interaction. This method should provide better uncertainty that previous ones in Cs and also may reveal the value for quadratic temperature coefficients. Section II is

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devoted to the experimental setup description and to the important parameters we have to control. In Section III, the method of measurement of the temperature coefficients will be described and the preliminary results for a Cs vapour cell containing 30 Torr of Ar as a buffer gas will be shown.

II. EXPERIMENTAL SETUP

A. *General points.*

The Fig.1 shows a schematic view of the setup developed for the measurements of the temperature coefficients in Cs. It operates in continuous regime. The bichromatic field necessary to create the dark state is generated by a single diode laser phase modulated at 9.192 GHz (hyper-fine splitting in the ground state in Cs) using an external phase modulator.

The laser source is a laboratory-made interference filter stabilized external cavity diode laser (ECDL) at 852 nm [11]. It is frequency locked to a hyperfine component of the Cs D_2- line at 852 nm by saturated absorption in an auxiliary vapour cell (AbsSat). Slow fluctuations of the laser frequency are corrected by adjusting the cavity length with a PZT driver whereas fast fluctuations are corrected through the laser current I.

The acousto-optic modulator (AOM1) is used to shift the laser frequency in order to get a symmetrical CPT resonance at room temperature [12].

The electro-optic modulator (EOM) is driven by a 9.192 GHz signal and the dark state is created by the carrier and one of the first sidebands.

The synthesizer is driven by a Direct Digital Synthesis (DDS) which allows to sweep the frequency around the CPT resonance. In order to improve SNR we apply an additional frequency modulation (f_m is about several kHz) for the synchronous detection of the signal. It is a standard lock-in detection technique [13]. In our case the direct detection of the CPT signal typically shows a SNR about 8, whereas for the lock-in detection signal this ratio is about 100 (Fig. 2).

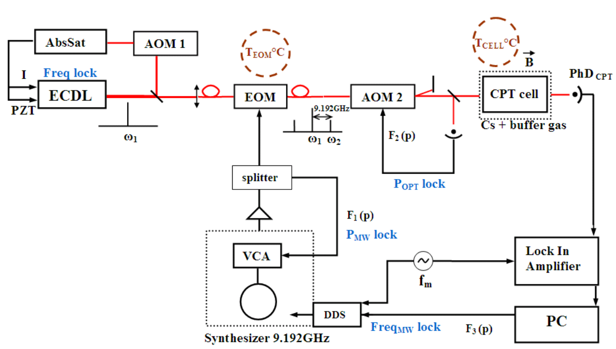

Fig. 1. Experimental setup. The light from an externally modulated laser source is sent through the CPT cell. Laser frequency, microwave power, laser intensity, clock frequency locking loops are shown.
ECDL – external cavity diode laser; AbsSat – saturated absorption; AOM – acousto-optic modulator; EOM – electro-optic modulator; VCA – voltage control attenuator; DDS – Direct Digital Synthesis; PhD – photo detector;

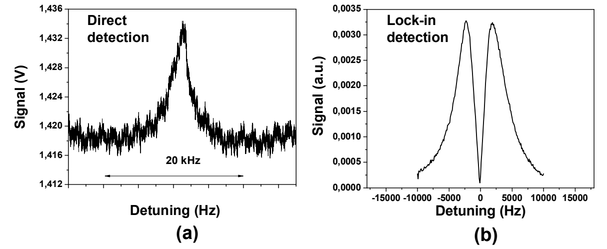

Fig. 2. (a) CPT resonance in case of direct detection; (b) Plot of the modulus R of the first harmonic at the lock-in output, where $R=(P^2+Q^2)^{1/2}$, P – in Phase signal, Q – in Quadrature signal.

We use 3 Cs cells measuring 20 mm long and 20 mm in diameter made of Pyrex containing 3 different pure buffer gas (not mixture): Ar (30 Torr), Ne (90 Torr), N_2 (30 Torr), respectively. The pressures were chosen to maximise the coherence lifetime at room temperature. In this paper only preliminary measurements performed with the cell containing 30 Torr of Ar are presented.

The optical power transmitted through the gas cell is detected by a photodiode. At the output of the lock-in amplifier, the signal is digitized and processed by a computer that drives the DDS frequency in order to lock the local oscillator on the hyperfine atomic resonance.

B. *Design and control.*

The frequency of the CPT resonance can be written

$$\nu_{00} = \nu_0 + f_1(\vec{B}) + [f_2(I_{laser}) + f_3(P_{MW}) + f_4(FM)] + f(T) + \ldots \quad (2)$$

where υ_0 is the unperturbed frequency linking the clock states $|F, m_F = 0>$ and $|F+1, m_F = 0>$; $f_1(B)$ is the Zeeman shift; The expression in the square brackets traduces light shift. The component $f_2(I_{laser})$ is the light shift due to the changes in the total laser intensity (carrier and sidebands). The component $f_3(P_{MW})$ is the light shift produced by changes in the power ratio between carrier and sidebands. The component $f_4(FM)$ is due to the frequency modulation applied to detect the CPT signal. The last term $f(T)$ is the temperature shift we intend to measure.

Special attention was paid to the design and control (thermal and magnetic) of the cell and to the radiation control in order to minimize all the possible shifts except the temperature shift.

- Cell design and control

1. The cell temperature T_{cell} is carefully measured with amagnetic thermistances. The cell is heated with a resistive wire. The heating wire was designed to minimize the stray magnetic field. The setpoint temperature is varied in the range from 25 °C to 60 °C. The fluctuations of the cell temperature near the set point are 3 – 5 mK. Such fluctuations correspond to a shift of the CPT resonance of about 0.1 Hz.

2. The cell is surrounded by a solenoid applying a static longitudinal magnetic field of 200 mG to raise the Zeeman degeneracy. The ensemble is surrounded by two

mu-metal magnetic shields. The fluctuations of the magnetic field are about ΔB = 10 μG and the corresponding shift is about 0.002 Hz.

- Radiation control

1. The total optical intensity is stabilized before the CPT cell (P_{OPT} lock). The residual intensity fluctuations are less than 0.1 %, which is 100 times better than without stabilization.
2. The fluctuations of the Carrier/Sidebands power ratio are mostly rejected by locking the microwave power applied to the EOM (P_{MW} lock).
3. A reproducible determination of the FM parameters is necessary in case of a non-symmetrical CPT resonance. The method to determine parameters (modulation frequency and modulation index) is described in Section III.
4. We have observed that the EOM temperature variations have a direct impact on the CPT resonance frequency. In non-stabilized case a 4 K fluctuations of the temperature correspond to a frequency shift of about 15 Hz. Since the EOM temperature is controlled at the level of 3 – 5 mK, the induced frequency shift can be neglected.

III. METHOD AND PRELIMINARY RESULTS

Frequency Modulation (FM) spectroscopy is a widely used, high resolution and sensitive spectroscopic technique developed by Bjorklund in the 1980's. The theoretical description of the FM signals was made for a Lorentzian profile [14, 15].

We have compared the theoretical results obtained for a Lorentzian line with experimental results obtained in the case of CPT signal. The line shape of the CPT resonance is not perfectly Lorentzian and this fact can explain that our results are only in qualitative agreement with the theory. Nevertheless we use this theoretical model to determine the FM parameters.

Our measurement method can be divided in 3 main steps:

- Determination of FM parameters
- Elimination of light shift
- Measurement of the temperature shift

A. *Determination of FM parameters*

The first step is a reproducible determination of FM parameters for each spectrum (versus laser intensity and microwave power).

The modulation index is fixed for all spectra, m=2. This value gives the maximum amplitude signal for a Lorentzian line shape [16].

The modulation frequency (f_m) is changed with intensity and microwave power in order to keep constant the ratio f_m/FWHM (full width at half maximum). This ratio is chosen to have the maximum amplitude of the demodulated signal.

The difficulty is that the FWHM must be measured in the direct signal and then can not be known very accurately because of the low SNR (Fig. 2 (a)).

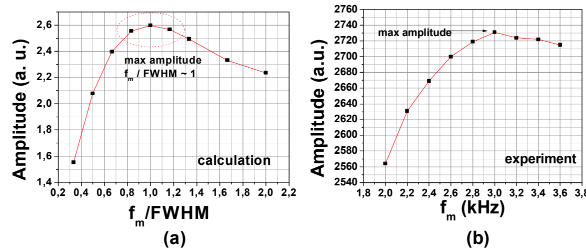

Fig. 3. (a) Result of calculation of the amplitude of the demodulated signal versus ratio f_m/FWHM; (b) Experimental results. Dependence of amplitude of the demodulated signal on f_m (FWHM is constant).

According to the model given in [14], the dependence of the amplitude of the demodulated signal on the ratio f_m/FWHM is shown on Fig. 3 (a). It can be interpreted in the following way: if we change f_m, the FWHM being constant, we pass through the maximum. In practice, for a given intensity and microwave power the linewidth of the CPT resonance is constant (direct signal). Then, as shown on Fig. 3 (b), we change f_m until getting a maximum of the demodulated signal amplitude. Fixing f_m at the maximum of the demodulated signal gives a reproducible method to keep constant the ratio f_m/FWHM, because the position of this maximum depends only on this ratio. f_m is adjusted for each value of the CPT resonance linewidth.

B. *Elimination of the light shift*

The resonance frequency is measured versus the total optical power for three different values of the microwave power (Fig. 4). The linear extrapolation as a function of the laser intensity converges for different microwave powers to the zero intensity shift. The uncertainties in the extrapolation to the zero intensity for different microwave powers will give the error bars in the graph of temperature dependence.

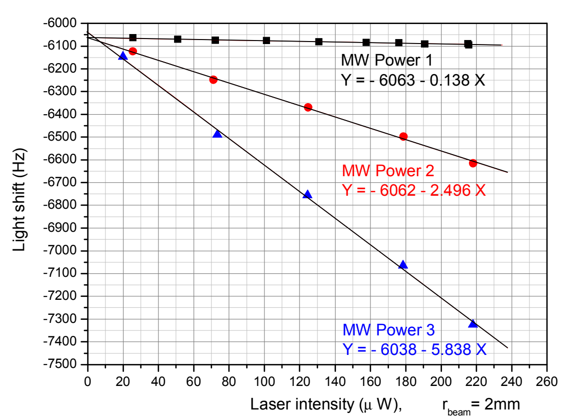

Fig. 4. The linear extrapolation as a function of laser intensity converges for different microwave powers to the zero intensity shift. (The shift is given with respect to υ_0 = 9 192 631 770 Hz. The quadratic Zeeman shift is not removed.)

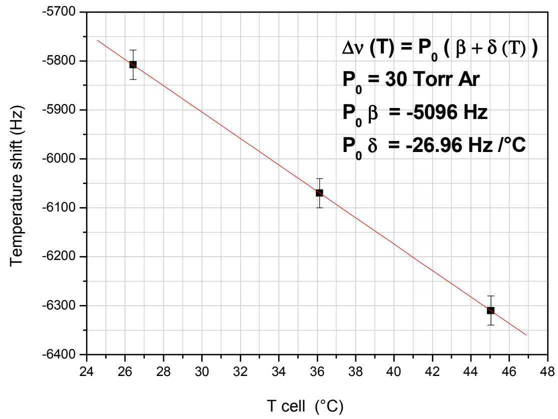

Fig. 5. Preliminary measurements for temperature shift in Cs vapour cell containing 30 Torr of Ar, where the frequency shift is given with respect to υ_0 = 9 192 631 770 Hz. The quadratic Zeeman shift is not removed.

C. Measurement of the temperature shift.

The results of the extrapolation at zero intensity for different temperatures of the cell give the temperature contribution to the frequency shift, assuming the Zeeman shift is constant (Fig. 5). As a preliminary result for the cell containing 30 Torr of Ar we obtain -27 Hz/°C, in reasonable agreement with the results obtained in [9], which would give a slope of -28.6Hz/°C.

IV. CONCLUSIONS AND PERSPECTIVES

In this paper a method for measurement of the temperature shift is proposed and the preliminary results for a cell containing 30 Torr of Ar is shown. The results obtained are in reasonable agreement with [9].

Next steps are to measure temperature shifts in cells with Ne and N_2 buffer gas.

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