Study of Rb 0-0 hyperfine double-resonance transition in a wall-coated cell

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INTRODUCTION

The idea to use alkali atomic vapour cells equipped with a wall coating for optical-microwave double resonance (DR) experiments began in the sixties. Experimentally it was demonstrated that coating the walls of the glass cell with chemically inert substances, such as paraffin increased the optical-pumping signals and reduced their line-width [1]. Although the idea to use wall-coated cells in an atomic clock was suggested by Robinson [2], it was not demonstrated due to the limitations in operating temperatures of such cells and other technological difficulties, e. g., in realizing and control of the coating quality. Part of this drawback has been overcome with laser pumped clocks, but a commercial product is yet to be realized. Recently, the interest in wall-coated cells for high precision spectroscopy and metrology is growing again because coated cells represent a good candidate to realize high-performance or micro-fabricated devices, such as miniaturized atomic clocks and/or atomic magnetometers [3].

The effect of collisions between alkali-atoms and widely used coatings, such as $(CH_2)_n$ and $(CD_2)_n$, have been studied in the sixties by Bouchiat [1], by using the method known as "relaxation in the dark" [4] that allows studying the relaxation due to the collision between the alkali and the coating averaged over the entire cell surface. The same technique has been used by Liberman for Cs atoms in paraffin-wall coated cells [5]. High-quality paraffin coatings have the advantageous property that polarized alkali atoms may bounce between the cell walls several thousand times before losing their ground-state polarisation. The long-lived atomic polarization in turn increases the Q-factor of the atomic resonance line and thereby improves on the short-term-stability of the clock. This can be explained by spin-flip effect [6]. Typical spin-relaxation times in wall-coated cells can be as large as ≈ 1 s. Therefore, narrow line-widths around 200 Hz or even as low as 10 Hz were observed [2]. Studies on the application of wall-coated cells for Rb frequency standards are reported in [7-10].

In this paper, we present our studies on double resonance using atomic ^{87}Rb in a wall-coated cell, in view of applications in a laser-pumped Rb atomic frequency standard. We study the systematic clock frequency shifts in a wall-coated cell, such as light shift, microwave power shift [11, 12] and shift due to cell temperature [13] (for the cell body, and for the Rb reservoir, also called the cell stem). We also report on our clock stability measurement using the wall-coated cell in DR scheme, and discuss the influence of the shift parameters first on the frequency stability of the clock.

DOUBLE RESONANCE IN WALL-COATED CELL

DR experiments were performed using a wall coated cell shown in fig.1. The inner walls of the cell are coated with paraffin and the stem of the cell acts as rubidium reservoir (^{87}Rb). The temperatures of cell volume (T_v) and cell stem (T_s) are maintained at a gradient to avoid formation of Rb droplets on the coated walls ($T_s < T_v$). Optical pumping is achieved using the light emitted around 780 nm (Rb D2 line) by a semiconductor laser diode (laser beam diameter of 1.9 mm at the cell), and microwave radiation is applied by placing the cell inside a TE_{011} magnetron cavity [11]. A schematic of the DR setup is shown in fig. 2. Active laser frequency stabilization to a sub-Doppler line (Rb D2 line) from a separate Rb cell is done with a lock-in feedback loop. A microwave synthesizer is used to drive the Rb ground-state "clock" transition ($F_1(m_F=0) \leftrightarrow F_2(m_F=0)$).

A typical DR clock signal is shown in fig. 3. We measure a very small DR linewidth of 642 Hz simultaneously with a good signal contrast of 11.3%, using a pump light power of 30 μW and a microwave power of -30 dBm injected into the cavity. The total wall-shift is approximately -368 Hz (see clock frequency systematic shifts section below).

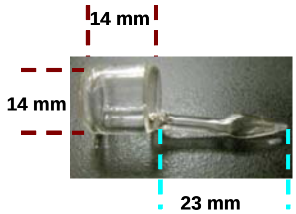

Fig. 1. Wall-coated cell filled with ^{87}Rb

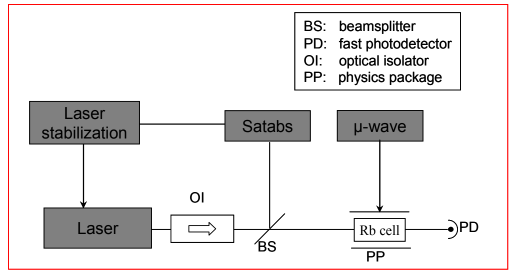

Fig. 2. Schematic of the DR experimental setup

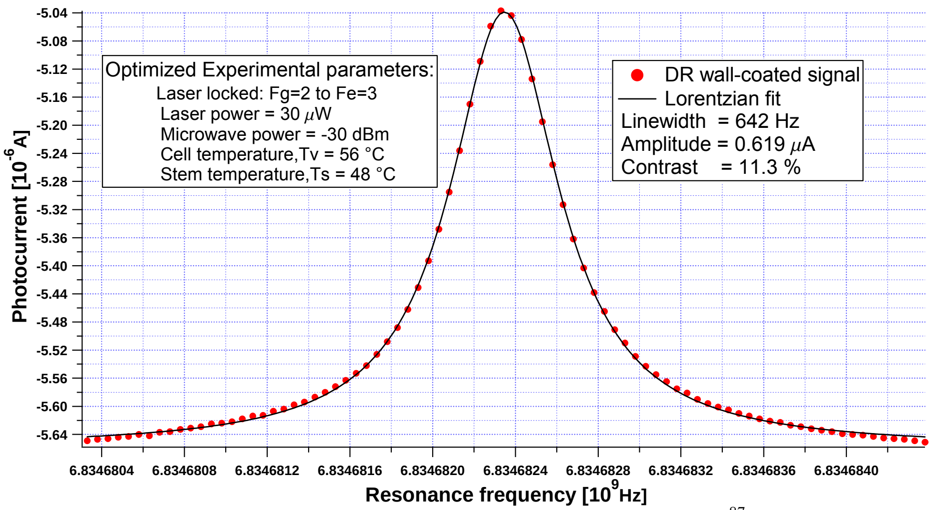

Fig. 3. Double resonance signal of the clock transition observed with a ^{87}Rb wall-coated cell.

Previous experiments suggest that in an uncoated glass cell the Rb atoms will depolarize by only one collision on the wall and hence the achievable DR linewidth is > 50 kHz, which would correspond to a clock short-term stability of ~ 10^{-9} at 1s, if not further degraded by time of flight (TOF). On contrary, relaxation in the dark experiments [4] to measure the *longitudinal relaxation time*, "T1" (c.f. fig. 4) show that in our wall-coated cell (14mm x 14mm) the polarization of the '*total hyperfine ground state population'* is maintained for up to 25 ms, which corresponds to ~ 500 wall collisions. This gives a much smaller width < 1 kHz, and thereby a better short-term stability for the clock. In comparison, the T1 relaxation time for a buffer gas cell (14mm x 23mm (Ar+N2, 21 mbar)) with similar experimental conditions is measured to be 5 ms, which is five times smaller than that for the wall-coated cell.

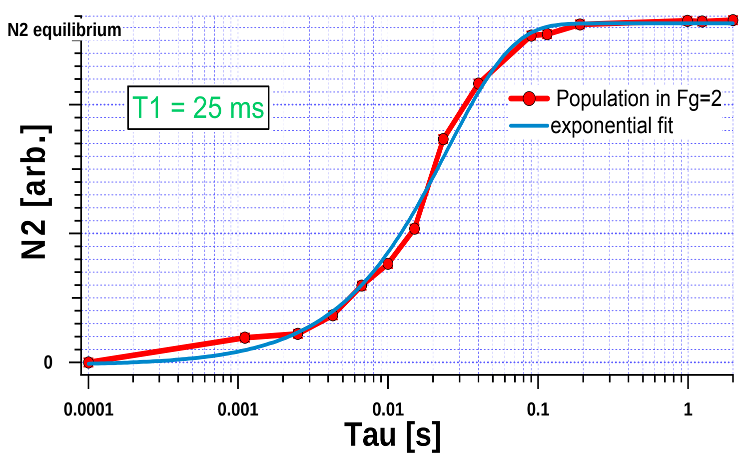

Fig.4. T1 relaxation time measured by relaxation in the dark technique.

In this experiment, an Acousto Optical Modulator (AOM) was used as a switch. Pump-laser switch-off time (tau in Fig. 4) was varied and the numbers of atoms remaining in the pump state were measured. It is important to notice that the T1 relaxation time is measured for the total hyperfine ground state, which includes the contribution from all the Zeeman levels. The coherence between two states, measured by the *transversal relaxation time*, "T2", is also an important parameter for the clock short term stability. Here, the T2 relaxation time is '*measured only for the clock transition*'. Extrapolation of the clock transition linewidth (FWHM) to zero pump-light intensity allows one to extract T2 (c.f. fig. 5) using FWHM = $1/2\pi$ T2 [14], when other sources of line broadening are negligible. For the cell used here (fig. 1), by subtracting the contribution of power broadening due to the microwave, we find T2 = 2.5 ms. This result of T2 is only preliminary and furthermore the comparison with that of the buffer gas cell for through understanding is underway.

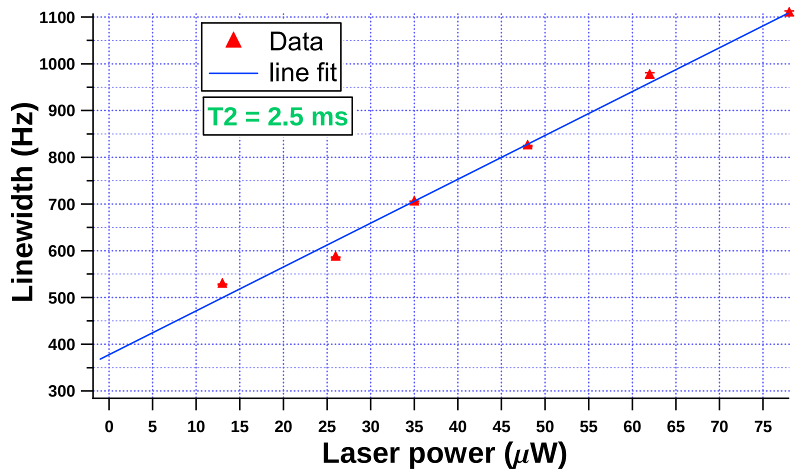

Fig.5. Double resonance width as a function of laser power.

CLOCK FREQUENCY SYSTEMATIC SHIFTS

The clock transition frequency is likely to be perturbed by various mechanisms. We focus our studies to the main and important ones that include AC Stark shift, here addressed as light shift (LS), microwave power shift and shifts due to temperature coefficients of the wall-coated cell. Our studies reported here focus on the case when the pump laser frequency is stabilized to the $F_g = 2$ to $F_e = 3$ transition in ^{87}Rb, as obtained from the separate, non-coated reference cell, except for the light-shift measurements. It is worth noting that this does *not* mean that the atoms probed by DR in the wall-coated cell are pumped on this transition, due to Doppler effect and velocity-changing collisions. The clock stability measurements and analysis reported further below were also performed with the laser frequency stabilized to this same reference transition.

Light Shift (Laser intensity)

At a fixed pump-light frequency detuning from the centre of the optical pump transition, the light-shift can be expressed as $\Delta\nu_{LS} = \alpha I_0$, where α is the light shift coefficient and I_0 is the intensity of the light. The line centre of each DR signal was measured as a function of laser power (c.f. fig. 4). The experiment was repeated by locking the laser frequency to different sub-Doppler transitions, which are shown in the insets on the right side of fig. 4.

The extrapolation of the data by line-fit to the zero laser power gives the indication of the frequency shift due to the wall coating only ("wall shift"), -368 Hz here. This wall shift depends on the cell temperature, and can vary for different coating material. From the slope of the linear fits, we extract the LS-coefficient α for the different pump-light detunings studied.

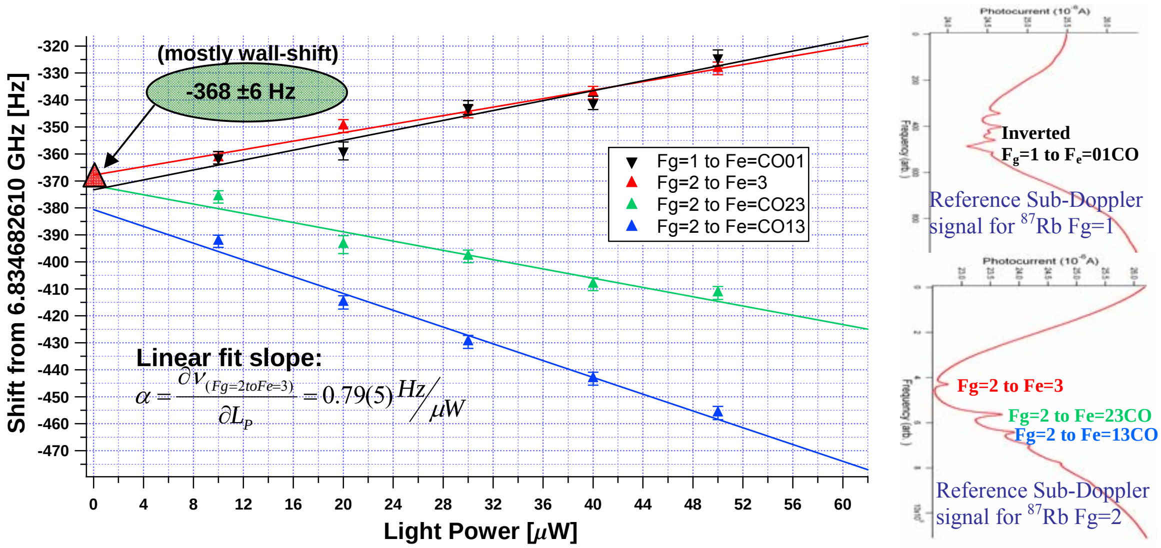

Fig. 4. Light shift in a wall-coated cell and the reference lines of laser locking are shown.

Estimated LS Contribution to clock instability at 10,000 s

In the continuous wave operation method of the clock studied here, the light shift will limit the achievable medium-term to long-term stability of the clock. For the laser frequency stabilised to the $F_g = 2$ to $F_e = 3$ transition, we find $\alpha = 0.8$ Hz/μW (from fig. 4); the stability of the laser intensity is $\sigma_{PL} = 5 \times 10^{-3}$ and used laser power, $P_{Laser} = 30$ μW. The LS contribution to the clock instability at 10,000 s can then be calculated as,

$$\sigma_\alpha = \frac{\alpha.\sigma_{PL}.P_{Laser}}{\upsilon_0} = 1.7 \times 10^{-11} \tag{1}$$

Expressed in terms of pump-light intensity, the light-shift coefficient in our case (D2 line, $F_g = 2$ to $F_e = 3$ transition) is $+1.16 \times 10^{-10}$ $mWcm^{-2}$. As a comparison, this is slightly lower than the value of -6×10^{-10} $mWcm^{-2}$ measured using CPT in a similar wall-coated cell (^{87}Rb D1 line, $F_g = 2$ to $F_e = 1$ transition) [15].

Light Shift Reduction

The effect of light shift can be reduced by several methods, for example by detuning the laser frequency or by using the pump laser in the pulsed mode, often called pulsed-optical-pumping (POP) [16]. The detuning of the laser can be done by using an Acousto Optical Modulator (AOM). We estimate the required laser detuning by plotting the light-shift coefficient as a function of laser frequency (c.f. fig. 5).

As seen in fig. 5, one can strongly reduce the light-shift coefficient towards zero value by detuning the laser frequency, for example by ~64 MHz when locking to the $F_g = 2$ to $F_e = 3$ transition. In this approach, slight adjustment of the AOM driving frequency can be used to fine-tune the light-shift coefficient α to zero-value within the required precision.

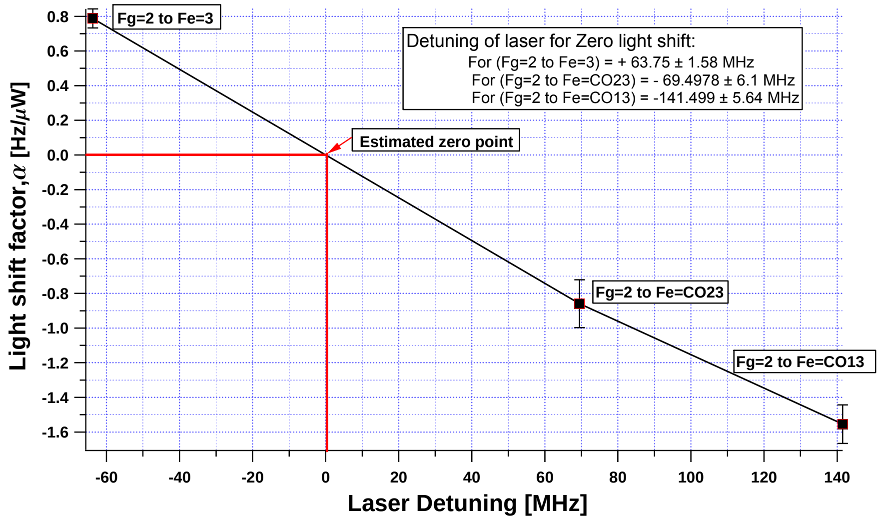

Fig. 5. Reduction of the Light-shift coefficient α in a wall-coated cell by control of the laser frequency detuning

Function of Microwave Power (Microwave Power Shift)

The clock-transition linewidth as a function of microwave power is shown in fig. 6. Up to 10 μW of microwave power, clearly a linear dependence (2.16 kHz/μW) is seen, after which the linewidth slowly goes towards saturation. The microwave power shift was measured for $F_g = 2$ to $F_e = 3$ transition.

Fig. 7 shows the shift of the clock transition as a function of microwave power. For a power of ≥ 1 μW we find a shift of 2.31 Hz/μW of microwave power. This shift is not negligible and one needs a good stability of the microwave synthesizer. The power stability of our synthesizer is estimated to be $\sigma_{\mu\text{-wave}} = 3 \times 10^{-4}$ at 10^4 s. Hence, the instability contribution of microwave power shift on the clock frequency at 10^4 s can be calculated as,

$$\sigma_{\mu-wave} = \Delta_{\mu-wave} . \sigma_{\mu-wave} . P_{\mu-wave} \Big/ \upsilon_0 = 1 \times 10^{-13} \qquad (2)$$

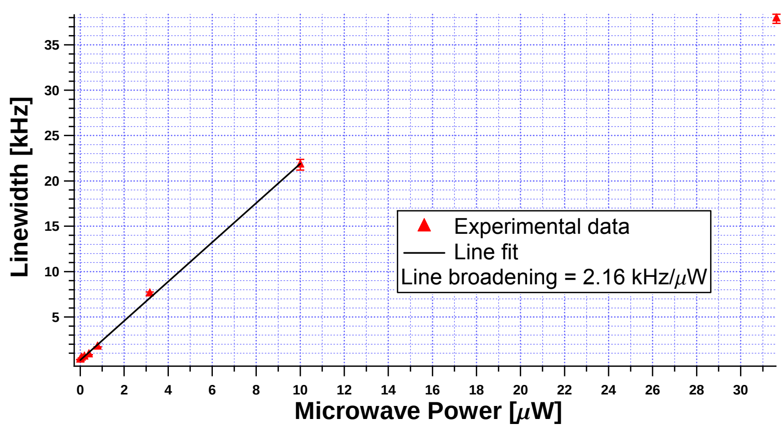

Fig.6. Double-resonance as a function of microwave power.

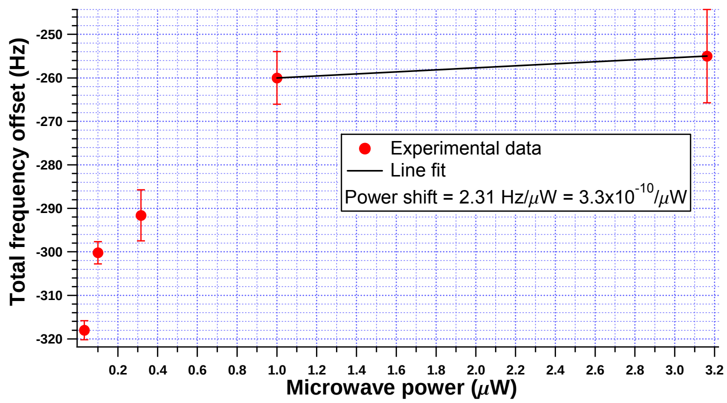

Fig. 7. Clock frequency shift as a function of microwave power.

We see the similar value as compared with buffer gas cells and as observed by Risely et al. [12]. Following to this, it is also shown in [12] that, any inhomogeneities in the cell are averaged in a wall-coated cell, and the effects are clearly observed in the case of buffer gas cell. Further to this, thorough studies are required to validate for the limitations between the wall-coated and buffer gas cells as a function of microwave power.

Function of Cell and Stem Temperatures

As shown in fig.1, the wall-coated cell used has two regions to be controlled; the cell body of which the controlled temperature is termed as T_v and the cell stem temperature as T_s. The temperature coefficient (TC) of a wall-coated cell is an integrated property of the coating material itself [13]. That is, the variations in the temperature will determine the physics of atoms interacting with the wall-coating and its impact on their polarisation state. Fig. 8 shows a negative overall shift [13] in the clock transition frequency from the unperturbed value (6.834682611 Hz), with increasing clock frequency for increasing cell volume temperature, T_v. The TC of the cell on the clock transition in first approximation is determined by the slope of the curve as +1.4(4) Hz/K. This corresponds to a temperature coefficient of $2x10^{-10}$/K, which is in good agreement with the value of $2.3x10^{-10}$/K measured using CPT and a nominally identical cell (^{87}Rb D1 line, [15]).

The influence of the TC on clock stability is a medium to long-term effect. By measuring the variations in the temperature control as $\sigma_T = 0.5$ mK, the estimation of the TC contribution at 10^4 s is given by,

$$\sigma_{TC} = {}^{TC.\sigma_T}\!/_{\upsilon_0} = 1\times 10^{-13} \tag{3}$$

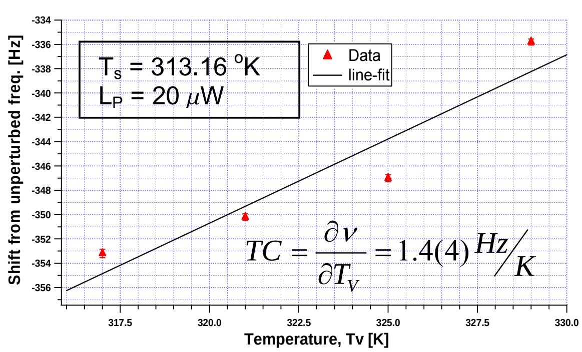

Fig. 8. Clock frequency shift as a function of cell volume temperature (T_v).

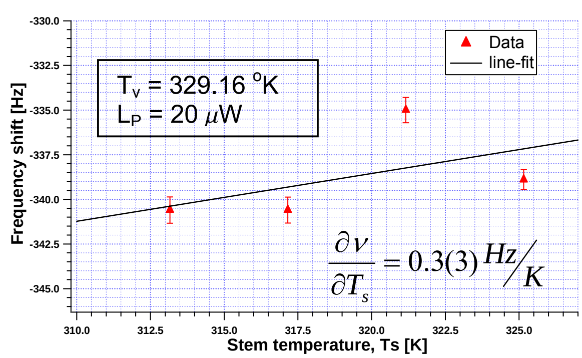

Fig. 9. Clock frequency shift as a function of cell stem temperature (T_s).

The variation in the clock transition frequency as a function of the stem temperature is shown in fig. 9 and amounts to 0.3 Hz/K. It is clear that the influence of this variation is smaller than that of the TC effect due to T_v, that will dominate the TC contribution to the clock instability. Still, the overall TC limitation to the clock stability is less than that from the light shift (c.f. fig. 4).

CLOCK SHORT-TERM STABILITY

The short-term stability of a passive rubidium frequency standard can be predicted as [17],

$$\sigma_y = \frac{N_{noise}}{\sqrt{2}.D.\nu_0}\tau^{-1/2} \tag{4}$$

Where N_{noise} is the noise measured in one Hertz bandwidth, ν_0 is the Rb ground-state hyperfine frequency and D is termed as the discriminator slope in the error signal close to the line centre, estimated as the ratio of the signal amplitude to the linewidth. Under typical experimental conditions, the shot-noise limit is $\sigma_y(\tau) = 3.6\text{x}10^{-13}\ \tau^{-1/2}$. Taking into account all experimental noise sources, we measure $N_{noise} = 5.1\text{x}10^{-12}$ A/Hz$^{1/2}$ (when microwave and laser are switched ON), and the measured discriminator slope of the DR signal is $D = 4.1\text{x}10^{-10}$ A/Hz. Therefore, the estimated short-term stability of our DR wall-coated clock is

$$\sigma_y = 1.3\times10^{-12}\tau^{-1/2} \tag{5}$$

Fig. 11 shows the experimentally measured short-term stability of the DR clock using the paraffin coated cell. The experimental parameters are as in Fig. 3, and the optical pumping was performed using a stabilized laser head [18] with a beam diameter of 0.83 mm. In these conditions, the light-shift coefficient was measured to be $9.3\text{x}10^{-12}$/μW. We measure a short-term clock stability of $\sigma_y(\tau) = 2\text{x}10^{-13}\ \tau^{-1/2}$, in good agreement with the estimated signal-to-noise limit (5). Also, the stability drift of < $7\text{x}10^{-12}$ at 10^4 s is in good agreement with the instability contributions of $6\text{x}10^{-12}$ from the light-shift, which is the dominating effect limiting the clock stability at these timescales.

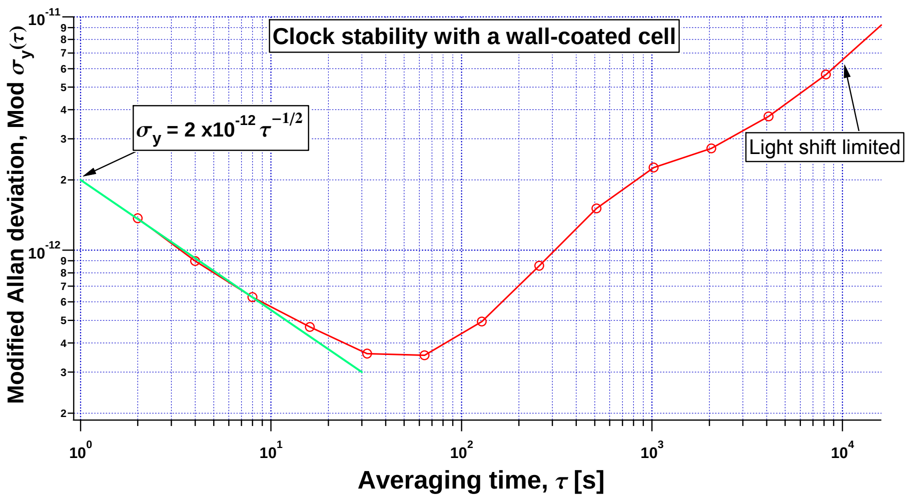

Fig. 10. Modified Allan deviation plot of the clock stability signal in a wall-coated cell

CONCLUSIONS AND FUTURE PROSPECTS

In this paper, we have studied the systematics of the double-resonance signal using a wall-coated cell using a TE_{011} type magnetron cavity, giving narrow linewidth and good signal contrast for the clock transition resonance line. The reported short-term clock stability of $2\text{x}10^{-12}\ \tau^{-1/2}$, proves the good suitability of wall-coated cells for obtaining state-of-the art short-term stability with Rb vapour-cell atomic clocks. To our knowledge, this result constitutes the first measured Rb clock stability obtained with a wall-coated cell using DR technique. Presently, the contribution of light shift is the main limiting factor for the clock stability at 10^4 s.

Though the long term stability in our measurement is limited by strong light shift effect, two methods to reduce the effect are possible: One by detuning the laser frequency using an AOM and the other by pulsed optical pumping. Ultimately, the medium-term clock stability is expected to be limited by the cell's temperature coefficient, and microwave shifts.

Our future studies foreseen include the light-shift reduction by detuning the laser frequency, as well as by laser intensity and frequency stabilization. It is also considered to study the medium and long-term behaviour (including frequency retrace and drift).

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