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Demonstration of a Two-Photon Atomic Clock with Light Shift Suppression using Two-Colour Magic Wavelengths

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14. ABSTRACT
Both the University of Adelaide and AFRL have been constructing vapour-cell atomic optical frequency standards which aim to provide frequency stability performance matching or exceeding that of the highest-stability commercial frequency standards (e.g. a Microsemi MHM-2010 H-Maser), but do it with a ~10-fold reduction in SWaP. The fundamental approach is based around frequency locking a laser source to a two-photon 51/2 â†’ 55/2 transition in a heated Rb vapour cell. The AFRL clock takes the approach of a single colour excitation of this transition using two 778nm photons, while the Adelaide group uses two different wavelength photons at 780nm and 776nm for the excitation. In both cases, the excitation is performed using counter-propagating fields to provide a high level of suppression of Doppler broadening. In the AFRL case, this yields a transition bandwidth of around ~300kHz, while the Adelaide group obtains a degraded linewidth of 4MHz because of the use of the two colours. However, on the other hand, the more complex dual-colour scheme has an advantage in that it produces a 1000-fold increase in the strength of the two-photon interaction. The principal technical benefit of this increased interaction strength is to create a similar signal-to-noise ratio with a 1000-fold reduction in excitation power. Both frequency standards detect 420nm fluorescence to ensure that the two-photon excitation is frequency locked to the atomic transition. To that end, this project sought to reduce false light shift signals and improve the overall stability of the Adelaide clock by increasing the fluorescence signal which will significantly increasing the signal-to-noise ratio and minimize technical noise sources; experimentally eliminate light shifts and demonstrate a reduction in sensitivity to driving optical powers; and demonstrate that the experimental measurements are consistent with the theoretical expectations.

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Final Report: Demonstration of a Two-Photon Atomic Clock with Light Shift Suppression using Two-Colour Magic Wavelengths

Basic Research for AOARD Proposal 20IOA032 "Demonstration of a Two-Photon Atomic Clock with Light Shift Suppression using Two-Colour Magic Wavelengths", dated 31 MAR 2022

Principle Investigator: Chris Perrella[1] – University Research Fellow
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Background

Both the University of Adelaide and AFRL have been constructing vapour-cell atomic optical frequency standards which aim to provide frequency stability performance matching or exceeding that of the highest-stability commercial frequency standards (e.g. a *Microsemi* MHM-2010 H-Maser), but do it with a ~10-fold reduction in SWaP.

The fundamental approach is based around frequency locking a laser source to a two-photon $5S_{1/2} \rightarrow 5D_{5/2}$ transition in a heated Rb vapour cell. The AFRL clock takes the approach of a single colour excitation of this transition using two 778nm photons, while the Adelaide group uses two *different* wavelength photons at 780nm and 776nm for the excitation (see Figure 1). In both cases, the excitation is performed using counter-propagating fields to provide a high level of suppression of Doppler broadening. In the AFRL case, this yields a transition bandwidth of around ~300kHz, while the Adelaide group obtains a degraded linewidth of 4MHz because of the use of the two colours. However, on the other hand, the more complex dual-colour scheme has an advantage in that it produces a 1000-fold increase in the strength of the two-photon interaction. The principal technical benefit of this increased interaction strength is to create a similar signal-to-noise ratio with a 1000-fold reduction in excitation power. Both frequency standards detect 420nm fluorescence (see Figure 1) to ensure that the two-photon excitation is frequency locked to the atomic transition.

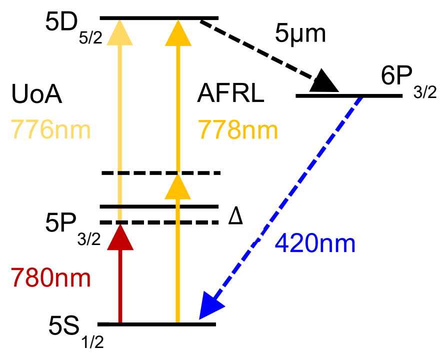

Figure 1: Two-colour two-photon Rubidium excitation scheme. UoA excitation scheme (left) and AFRL excitation scheme (right)

Motivation for the Research

At first sight, one might have believed that the lower powers used in Adelaide's two-colour scheme might have delivered an advantage over the AFRL single colour excitation by providing a smaller light shift [1-5]. However, a simple argument shows that this is not true. Ignoring complexities associated with polarisation and optical pumping, the unwanted optical light shift, Δf, depends on the laser frequency detuning from the intermediate $5P_{3/2}$ energy level, Δ, along with the laser intensities of the two counter-propagating fields, I_1 and I_2, in the form:

$$\Delta f \propto \frac{\mu_1^2\, I_1}{\Delta} - \frac{\mu_2^2\, I_2}{\Delta} \tag{1}$$

Where $\mu_{1,2}$ is the transition matrix element is of the 1st and 2nd transition of the two-photon transition. The excitation rate of the two-photon transition (which essentially sets the intrinsic performance of the clock) is proportional to the product of the driving laser intensities:

$$W \propto \frac{\mu_1^2\, \mu_2^2\, I_1\, I_2}{\Delta^2} \tag{2}$$

The transition rate, W, is directly related to the fluorescence from the atomic vapour. We detect 420nm fluorescence, see Figure 1, as a measure of successful excitation of the two-photon transition. We process this fluorescence signal to steer the lasers' frequencies to ensure they are tuned to the two-photon transition. The noise of the fluorescence signal, which limits the clock stability, is determined by the shot-noise, $\sqrt{W}$. Thus, the Signal-to-Noise Ratio is given by:

$$SNR \propto \frac{W}{\sqrt{W}} = \sqrt{W} \tag{3}$$

If we compare the two approaches with the same transition rate (such that the intrinsic frequency stability limits of the two clocks are about the same), and with the intensities of the two-colour beams approximately the same, $I_1 \sim I_2 \sim I$, then we see an optical light shift that is independent of the frequency detuning and depends only on the requested transition rate:

$$\Delta f \propto \frac{\mu_1^2 - \mu_2^2}{\sqrt{\mu_1^2\, \mu_2^2}}\, \sqrt{W} \tag{4}$$

Equation 4 tells us that both the two colour and single colour approach will yield approximately the same sensitivity to incident power fluctuations in circumstances where the dual colour excitation beams have about the same power.

Significance and Innovation

However, this analysis now points to a potential advantage of the dual colour scheme – if we adjust the power of each laser separately, we can alter the magnitude of the light shift while maintaining a given transition rate. In particular, by adjusting I_1/I_2 to be equal to μ_2^2/μ_1^2, we obtain a zero-light shift in Eqn. 1, while with $I_1\, I_2 = I^2$ we maintain the original transition rate. This approach can deliver a substantial advantage – that any power fluctuations of the two lasers do not translate into changes in the optical frequency. This offers the potential for a substantial reduction in the frequency instabilities in the output of the clock over the medium (hours) and long (days) observation periods.

A similar possibility exists for the single colour excitation AFRL clock. In this case, one can add an additional light field to co-propagate with the existing single colour excitation. This additional field would be detuned from the transition such that it will not add any additional atomic excitation but, through its own light shift, will have the effect of shifting the clock state transition by an amount with exactly the same magnitude as the clock probe light, but with the opposite sign. Thus, as with the two-colour clock, we will realize a substantially reduced frequency shift for any laser power fluctuations. Of course, this would come with the disadvantage of the increased complexity.

Project Outline

The project was broken into three tasks. The tasks aim to demonstrate the ability of the magic clock configuration to reduce the timing sensitivity of the clock to probe power fluctuations. The tasks aim to reduce false light shift signals and improve the overall stability of the Adelaide clock by *Increasing the fluorescence signal* which will significantly increasing the signal-to-noise ratio and minimize technical noise sources; Experimentally *eliminate light shifts* and demonstrate a reduction in sensitivity to driving optical powers; and demonstrate that the experimental measurements are consistent with the theoretical expectations.

Task 1: Improve Fluorescence Collections

Improving the fluorescence collected from the atoms will significantly increase the signal-to-noise ratio and minimize technical effects that masquerade as light shifts. This will allow higher precision tests of the residual light shifts once eliminated in Task 2, and will also greatly improve the stability of the clock.

Task 2: Eliminate Light Shifts

The careful characterisation of the light shifts, both experimentally and theoretically, that we have previously carried out will enable minimisation of the effect of light shifts in both the Adelaide clock and the AFRL clock. For the AFRL clock an additional laser will be installed that will counteract light shifts created by the laser probing the two-photon transition. For the Adelaide clock, the power and wavelength to the two lasers driving the transition will be tuned such that the light shift created by each laser is counteracted by the other.

Task 3: Performance Testing

Following from the minimisation of the light shifts, both labs will measure the performance of the clocks based on this light-shift free configuration. It is expected that by removing the sensitivity to light shift, the long-term stability of both clocks will be greatly increased. We will show this experimentally by performing long-term stability measurements that demonstrate the clock's insensitivity to power fluctuations.

Task 1: Improve Fluorescence Collections

A redesign of the 420nm fluorescence capture optics has been undertaken. Initially large area photodiodes were explored as options to increase the amount of fluorescence captured, however, tests showed that the photodiodes were particularly susceptible to interference from electronic pick-up due to low fluorescence power and large electronics amplification required. As a result, suitable photomultiplier tubes were chosen with the fluorescence collection optics redesigned to suit.

The optimised optical system for collecting significantly larger quantities of fluorescence is shown in Figure 2. Curved mirrors surround the Rb cell and focus the fluorescence onto two photomultiplier tubes either side of the cell. The ray tracing for light fluoresced in the transverse plane to the laser propagation (shown in Figure 2) predicts 53% capture efficiency for a point source at the centre of the cell. To verify this optical geometry a commercial package (Zemax) was used to verify the optical setup and extend the model into 3-dimensions to gain a better estimate of the collection efficiency. Expected collection efficiencies were still above 15%, a 200-fold increase in fluorescence capture leading to more than 10-fold increase in clock stability and light-shift measurement precision. The design was prototyped and a collection efficiency of 7% was measured, which a 50-fold improvement in collection efficiency and should result in a 7 improvement in the shot-noise floor and thus short-term performance of the clock with clock stability results reported below.

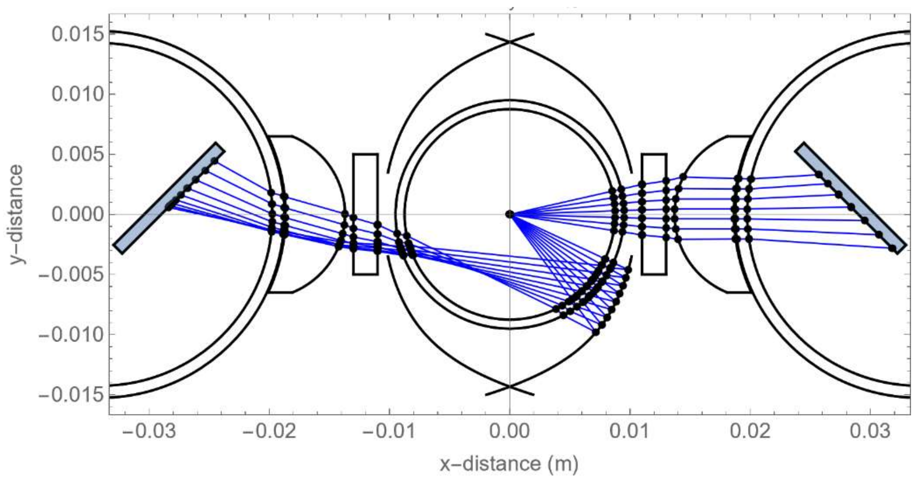

Figure 2: Cross section through the cell and collection optics. Ray tracing for a point source in the centre of the Rb cell (circles centred at (0,0) that represent the Rb cell cross section). The proposed fluorescence collection optics consist of mirrors above and below the cell that direct the light towards photomultiplier tubes centred at $\pm$ 35mm from the centre of the Rb cell.

Task 2: Eliminate Light Shifts

With the increased signal-to-noise ratio, light shift measurements were repeated with higher precision. Light shifts were induced by a tuneable laser that was combined with the probe 780 nm and 776 nm clock lasers, see Figure 1. The tuneable laser was able to be tuned from 770nm to 800nm and induce light shifts of the two-photon clock transition. The optical power of the tuneable laser was an order of magnitude greater than either of the two clock lasers, therefore, the light shifts are approximated as being induced entirely by the tuneable laser and not by the two clock lasers. The perturbing laser allowed measurement of the light shifts for a range of different wavelengths which were converted to differential polarizabilities and compared to an atomic theory model, see Figure 3. Excellent agreement can be seen between the theory (blue curve) and experimentally measured differential polarizability (red markers).

The atomic polarizability was found from relativistic atomic structure calculations. We use the high-precision correlation potential method to evaluate the relativistic atomic structure calculations, a method that works particularly well for the heavy alkali-metal atoms including Rb.

Table 1: Calculated reduced electric dipole matrix elements compared to experimental results. [a]Scaled SDpT, Ref.[1]. [b] Theory results are performed at SD level; "Expt." values from combination of theory and experiment, Ref.[2]. [c] Average of several experimental results, as referenced in Ref. [1]. [d] Reference [3]

$\|\langle k\|\|d\|\|5s_{1/2}\rangle\|$				$\|\langle k\|\|d\|\|5p_{1/2}\rangle\|$				$\|\langle k\|\|d\|\|5p_{3/2}\rangle\|$			
k	This	Other[a]	Expt.	k	This	Other[b]	Expt.[b]	k	This	Other	Expt.[b]
$5p_{1/2}$	4.2451	4.2498	4.233(2)[c]	$6s_{1/2}$	4.1343	4.146		$6s_{1/2}$	6.0305		
$6p_{1/2}$	0.3205		0.3235(9)[d]	$4d_{3/2}$	8.0246	8.017	8.051(63)	$4d_{3/2}$	3.6216		3.633(28)
$5p_{3/2}$	5.9919	5.9976	5.978(4)[c]	$5d_{3/2}$	1.3461	1.352		$5d_{3/2}$	0.6629		
$6p_{3/2}$	0.5215		0.5230(8)[d]					$4d_{5/2}$	10.8718		10.899(86)
								$5d_{5/2}$	1.9774	**2.334**[7]	**2.29(4)**[8]

The spectra of energy levels are discretised by placing the atom in a cavity. This improves the accuracy of the calculations by reducing the sum over (discrete and continuous) intermediate states in the polarizability to a sum over a pseudospectrum of discrete states, that match the physical states for the lowest-lying levels. To achieve this, we use a cavity of size 100 a.u.

The form of the total polarizability depends on the laser polarization and the direction of the magnetic field that determines the quantization axis. It is ultimately built up from scalar, vector (for circular/elliptical polarization), and tensor components. We have performed calculations of the scalar and tensor polarizabilities for the dynamic (Figure 3) and, as well as the relevant E1 matrix elements (Table 2) that are compared against available experimental and theoretical data. Along with checks of the stability of the results, and estimates for the size of missed correlations and other effects, these provide an excellent gauge of the accuracy of the calculations. Our results demonstrate an accuracy on the level of 0.1% for the E1 matrix elements and transition energies.

Fits to the experimentally measured differential polarizabilities allows us to extract the relevant E1 matrix elements and compare to theoretically calculated values. The measured E1 matrix elements are presented in Table 2 and show excellent agreement with theoretical calculations.

Table 2: Experimentally measured E1 matrix elements extracted from data in Figure 3.

Transition	$\langle J\vert d\vert J'\rangle$	discrepancy with theory
$5S_{1/2} \rightarrow 5P_{1/2}$	4.22(2)	+0.48%
$5S_{1/2} \rightarrow 5P_{3/2}$	5.79(2)	+3.4%
$5P_{3/2} \rightarrow 5D_{5/2}$	2.02(2)	-2.2 %

The above model and experimental verification enable tuning of the laser power and detunings from the intermediate state to be tailored to minimise the effect of light shifts. This has been implemented in the AFRL two-photon clock with the inclusion of an additional laser detuned from the clock transition [4]. For the UoA clock we will tune the power of both lasers, guided by the measurements and theory above to a power ratio of:

$$\left|\langle 5P_{3/2}||d||5S_{1/2}\rangle\right|^2 / \left|\langle 5D_{5/2}||d||5P_{3/2}\rangle\right|^2 = 9.2$$

In this configuration, common-mode power fluctuations of the 780nm and 776nm beams will produce opposing light shifts that cancel each other out.

Task 3: Performance Testing

The redesign of the fluorescence collection allowed a new method for monitoring the optical powers prior to the lasers entering the rubidium cell. The result of both of these changes is shown in Figure 4.

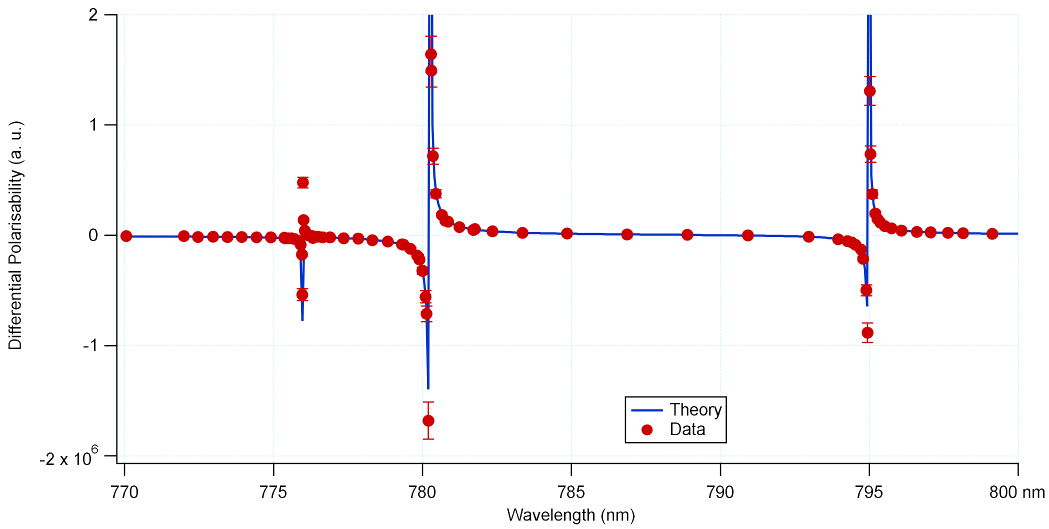

Figure 3: Measurements of the differential polarizabilities of the clock transition (red) and theoretical model (blue).

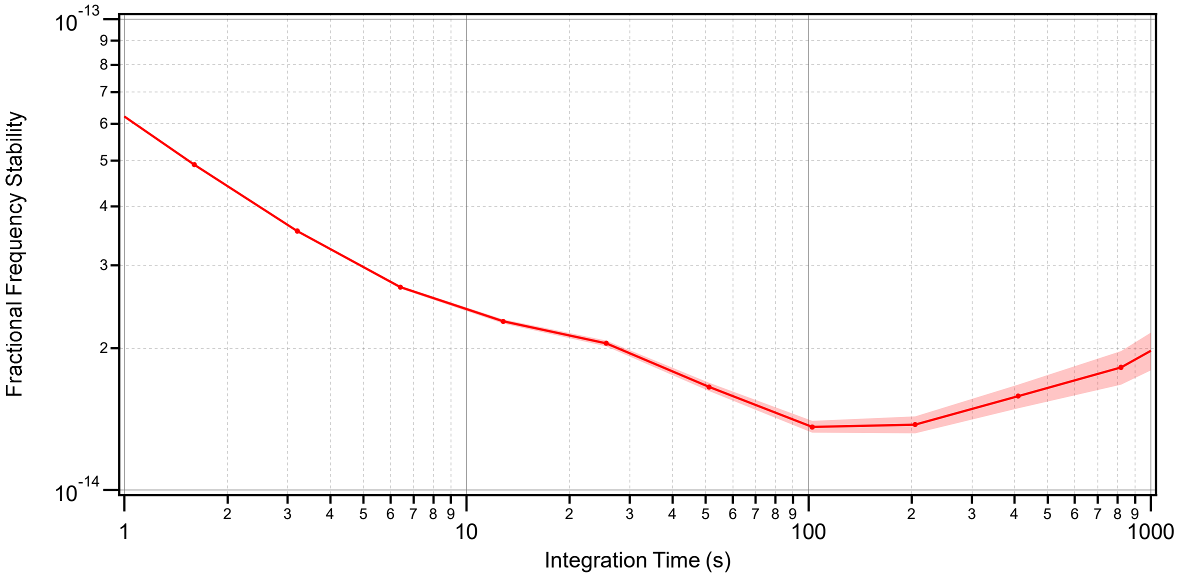

Figure 4: New two-photon clock stability measurements with improved fluorescence collection and power stability control loops implemented.

The new fluorescence collection has improved the short-term stability of the clock to 6×10^{-14} at 1s, a near 10-fold improvement from stability measurements prior to the new fluorescence capture optics. New power stabilisation control systems have improved the stability over integration times between 1 – 100s, with the 100s stability also improved by nearly 10-fold.

Future Work

A paper is being drafted to report the experimental measurements and calculations of the differential polarizability for the two-photon transition. To our knowledge, no one has presented such comprehensive theoretical calculations and experimental measurements for rubidium. It is expected that this paper will be submitted within the next month.

Long term stability measurements with tuned optical powers to cancel light shifts will be tested in the coming months. The optical clock is currently being fine-tuned for these measurements. A paper is also being drafted to present these results.

Project Outputs

We are expecting to be able to publish 3 papers from this project over the next year, with the first two listed here begin submitted in the coming months. These papers will report upon:

1. The stability of the clock with 1 second fractional frequency stability below 10^{-13} and stabilities at longer time scales reaching the 10^{-15} range.
2. Comparison of differential polarizability measurements and theoretical calculations for the two-photon transition.
3. Accurate measurement of atomic parameters using the precision of the clock's frequency stability and refinement of Dr Jacinda Ginges' atomic models.

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