AN INVESTIGATION OF THE TECHNICAL FEASIBILITY OF USING PULSED OPTICAL PUMPING TO ELIMINATE THE LIGHT SHIFT AND TO IMPROVE THE LONG-TERM STABILITY OF PASSIVE RUBIDIUM FREQUENCY STANDARDS.

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Final Report for Period, 1 September 1977 to 30 April 1979
A pulsed optical pumping rubidium frequency standard has been constructed for the purpose of eliminating the light shift and investigating the long-term frequency stability in the absence of light shifts. The device used employs a very small physics package that occupies a total volume of only 260 cc.

Initial measurements of the light shift for this device showed that the...
light shift was reduced by about an order of magnitude: to approximately \(-1.5 \times 10^{-10}\) fractional frequency shift for a 30% reduction in light intensity. Under these conditions, the square-root of the Allan variance was determined to be of the order of parts in \(10^{12}\) for averaging times of 24 hrs and 48 hrs.

This residual "light shift" of \(-1.5 \times 10^{-10}\) appears to be due to magnetic field inhomogenity in the physics package which produces a frequency shift (pseudo light shift) that mimics a true light shift. When the C-field homogeneity was improved, this pseudo light shift was reduced from \(-1.5 \times 10^{-10}\) to \(< 1 \times 10^{-11}\) (limit of detectability). Unfortunately, due to time limitations, it was not possible to make long-term stability measurements under the conditions of improved C-field homogeneity and negligible pseudo light shift.

The short-term stability of the pulsed optical pumping frequency standard was measured and found to be

\[
\sigma_y(\tau) = 2 \times 10^{-11} \tau^{-\frac{1}{2}} \quad \text{for} \quad 1 \text{ sec} \leq \tau \leq 100 \text{ sec}
\]

This is comparable to commercial rubidium frequency standards, and exceeds the performance of most commercial cesium standards. This good short-term stability was made possible by a dual photocell compensation scheme that was developed as part of this work.
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INTRODUCTION

This is the final report for the work on pulsed optical pumping performed under ONR Contract No. N00014-77-C-0646. Work performed during the period 1 September 1977 to 31 August 1978 is described in a previous report: Annual Summary Report dated 31 August 1978 (ONR Report No. N00014-77-C-0646-001). Subsequent work is described in the present report and consists of two parts: (1) work performed during the period 31 August 1978 to 31 December 1978, and (2) work performed during 1979. Most of item (1) was the subject of a paper that was presented at the Tenth Annual Conference on Precise Time and Time Intervals (PTTI) that was held at the Naval Research Laboratory in Washington D.C. on 28-30 November 1978. A preprint of this paper is included as Appendix A of the present report. Item (2) has not previously been reported and is described in the next section, along with that portion of item (1) that is not contained in Appendix A.

WORK PERFORMED DURING 1979

Device Modifications

A number of device modifications were made during 1979. All of these modifications were made for the purpose of reducing device sensitivity to changes in ambient temperature, and to further improve long-term stability. Four device improvements were made, as described below.

(1) The previously used synthesizer board was replaced by a
new one having an improved temperature coefficient.

(2) A high-stability current regulator was constructed and used to supply the C-field current which should restrict fractional frequency changes due to changes in C-field current to <1x10⁻¹³.

(3) The previous resonance cell was replaced by a new, dual buffer gas cell designed to have a very small temperature coefficient.

(4) The PIN diodes used in the 10 MHz phase modulator were previously reported to be sensitive to changes in ambient temperature. For this reason these diodes were placed in a temperature-controlled oven.

Results

Upon completion of the device changes listed above, it was discovered (quite unexpectedly) during preliminary testing that the temperature coefficient of the new buffer gas resonance cell was very nearly the same as for the previous single buffer gas cell. The dual buffer gas cell is filled with 12 Torr of N₂ and 10.8 Torr of Kr. According to the data given by Missout and Vanier,¹ this combination of buffer gases should have a very small temperature coefficient in the absence of light shifts. Figure 1 shows the temperature dependence of output frequency for this cell as a function of cell temperature. The temperature coefficient (TC) of +5.5x10⁻¹⁰/°C (change in fractional frequency per degree centigrade) is not much different from the mean value of +6.5x10⁻¹⁰/°C for the previous
Figure 1. Dependence of output frequency ($\Delta f/f$) and resonance signal on cell temperature for a dual buffer gas rubidium resonance cell containing 10.8 Torr Kr and 12 Torr $N_2$. The slope of the curve marked $\Delta f/f$ corresponds to a TC of $+5.5 \times 10^{-10}/^\circ C$. 
**Figure 1**

Signal (Arbitrary Units)

- **Cell Temperature (°C)**
  - 74
  - 72
  - 70
  - 68
  - 66
  - 64
  - 62
  - 60
  - 58
  - 56

- **Fractional Frequency Offset**
  - $\Delta F/F$

- **Signal**

- 6
- 5
- 4
- 3
- 2
- 1
- 0
- -1
- -2
- -3
- -4
- -5
- -6
- -7
- -8
single buffer gas cell (Figure 2). The "theory" of Missout and Vanier, when applied to the single buffer gas cell, gives a predicted TC of $7 \times 10^{-10} /^\circ\text{C}$, in good agreement with the observed value. However, when applied to the dual buffer gas cell, it predicts a zero TC. In practice the TC is limited by one's ability to accurately measure the pressures of the two gases, but even so we expect a reduction in the TC (compared to a single buffer gas) by at least a factor of 5 to 10. This, of course, is contrary to what is observed. This seems to indicate that the TC's of different gases cannot be added algebraically to determine the TC of a mixture of gases. Another (related) possibility is that the results of Missout and Vanier were affected by light shifts, as they themselves have indicated. Thus, any results obtained in the absence of light shifts would be expected to differ from their results.

After these modifications were made, a trial run was conducted to determine if any improvement in stability had resulted. The trial run consisted of monitoring the frequency stability over a period of approximately 25 hours in a room where there were large variations in ambient temperature (up to nearly $10^\circ\text{C}$) during the test period. The results were unsatisfactory: the device still exhibits a significant temperature coefficient of output frequency, probably due mainly to the large TC of the resonance cell itself. Also, since the C-field power supply had been completely redesigned (modification (2), above) it was not convenient to use the C-field temperature compensation scheme that was used previously (pp.33, ONR Report
Figure 2. Dependence of output frequency ($\Delta f/f$) on cell temperature for a single buffer gas rubidium resonance cell. The experimental points are fit by two straight lines, A and B. The mean slope of these two lines corresponds to an average TC of $+6.5 \times 10^{-10}$°C.
No. N00014-77-C-0646-001). In view of these two considerations, the results obtained are not surprising.

At this point, the number of hours remaining under the contract had been nearly exhausted so that it was evident that it was not feasible to pursue these problems further. Nevertheless, it is felt that the problems encountered could have been solved if additional time had been available: In effect, it was necessary to discontinue the work during the debugging phase following the device modifications. Thus a fair test of the value of the modifications was not possible.

WORK PERFORMED DURING 1978 AND NOT REPORTED ELSEWHERE

Line Shape Distortion

During September 1978, test equipment was constructed (using Efratom internal funding) that allows both AM and FM line shapes to be automatically recorded on an XY recorder. This equipment was used to obtained line shapes for pulsed optical pumping apparatus. The results, which are shown in Figures 3-6, shed some light on the reason for the dependence of output frequency on microwave pulse duration that was reported previously (p. 34, ONR Report No. N00014-77-C-0646-001). Figure 3 shows the cw AM and FM line shapes. The AM line width (FWHM) is approximately 1.25 KHz and is due almost entirely to light relaxation. This is somewhat larger than the value of 1.1 KHz previously reported for the cw case (Curve B, Figure 14, p. 49, ONR Report No. N00014-77-C-0646-001), and may be
Figure 3. AM and FM line shapes for the pulsed optical pumping frequency standard when operated in the cw mode. The AM line width (FWHM) is 1.25 kHz. The location of the origin on the horizontal axis is arbitrary. The experimental conditions are given in Table 1.
TABLE 1. EXPERIMENTAL CONDITIONS FOR FIGURES 3 TO 8

<table>
<thead>
<tr>
<th>Figure number</th>
<th>Experimental Conditions $^{a,b}$</th>
<th>$T_L$ (msec)</th>
<th>$T_S$ (msec)</th>
<th>$T_\mu$ (msec)</th>
<th>$T_d$ (msec)</th>
<th>$T_L$ ($^\circ$C)</th>
<th>$T_C$ ($^\circ$C)</th>
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<tr>
<td>3</td>
<td>--------------------------cw operation------------------</td>
<td>130</td>
<td>66</td>
<td></td>
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<td></td>
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<tr>
<td>4</td>
<td>2.0</td>
<td>0.14</td>
<td>1.25</td>
<td>1.6</td>
<td>130</td>
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<td>0.14</td>
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<td>1.6</td>
<td>130</td>
<td>66</td>
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<tr>
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<td>2.0</td>
<td>0.14</td>
<td>0.30</td>
<td>1.6</td>
<td>130</td>
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<td>2.0</td>
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<td>o</td>
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<td>2.1</td>
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<tr>
<td>8</td>
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<td>4.5</td>
<td>4.8</td>
<td>130</td>
<td>66</td>
<td></td>
</tr>
</tbody>
</table>

$^a$ For all Figures, lamp no. 110 was used which contains both isotopes of Rb.

$^b$ The notation used is identical to that of ONR Report No. N00014-77-C-0646-001.
attributed to a higher lamp operating temperature.

Figure 4 shows the line shapes for pulsed optical pumping under the same experimental conditions. The AM line width (FWHM) is now reduced to 750 Hz. Subsidiary maxima and minima are now present in the wings of the line, which result from pulsing the microwave field and are similar to the line shapes obtained for single velocity molecular beams.\(^2,3\)

In Figure 4 the duration of the microwave pulse was 1.25 msec. Figure 5 shows what happens when the microwave pulse duration is shortened to 0.5 msec. The line shape is now very distorted and somewhat broadened. Figure 6, for which the microwave pulse duration is 0.3 msec, shows the distortion increases markedly as the microwave pulse duration is further decreased; at this point the line is grossly distorted and broadened. It is obvious that this distortion of the line shape will produce frequency shifts (large ones), when the device is operated as a frequency standard and that the sensitivity of output frequency to variations in microwave pulse duration found previously (Figure 12 of ONR Report No. 00014-77-C-0646-001, reproduced here as Figure 7), is related to this line distortion effect.

Although it is clear that line distortion will produce frequency shifts, it is not clear what is producing the line distortion. One possibility is that as the microwave pulse is made shorter, the rise and fall times, which are negligible for long duration pulses,
Figure 4. AM and FM line shapes for the pulsed optical pumping frequency standard when operated in the pulsed mode. The microwave pulse duration was 1.25 msec. The AM line width (FWHM) is 750 Hz. The location of the origin on the horizontal axis is arbitrary. The experimental conditions are given in Table 1.
Figure 5. AM and FM line shapes for the pulsed optical pumping frequency standard when operated in the pulsed mode with microwave pulse duration = 0.5 msec. The other experimental conditions are given in Table 1. The location of the origin on the horizontal axis is arbitrary.
Figure 6. AM and FM line shapes for the pulsed optical pumping frequency standard when operated in the pulsed mode with microwave pulse duration = 0.3 msec. The other experimental conditions are given in Table 1. The location of the origin on the horizontal axis is arbitrary.
Figure 7. Relative fractional frequency offset as a function of microwave pulse duration. Reproduced from ONR Report No. N00014-77-C-0646-001. Experimental conditions are given in Table 1.
Figure 7

\[ \frac{\Delta f}{f} \]

\[ \frac{1 \times 10^{-9}}{\text{MICROWAVE PULSE DURATION, } T_\mu \text{ (msec)}} \]
become relatively more important, resulting in distortion of the spectrum of the microwave exciting radiation. Another possibility is that for short microwave pulses (which produce less relaxation) there is some persisting of atomic coherence from one pulsed pumping cycle to the next. This could give a spurious "Ramsey pattern-like" effect.

Narrowest Attainable Line Width

Figure 8 shows an AM line shape for a microwave pulse duration of 4.5 msec. The line width (FWHM) is 190 Hz. This is the narrowest line width that we have observed with the pulsed pumping apparatus. This is significantly less than the previous narrowest line width of 360 Hz, and indicates that the minimum line width is most likely a function of the relaxation time for decay of the oscillating magnetization (T₂) rather than the relaxation time for decay of the population difference (T₁). (This is the case theoretically for cw operation in the limit of zero microwave power broadening.) For ⁸⁷Rb, Vanier ⁴ has shown (theoretically) that T₂ = (8/5) T₁. Using our experimental value of T₁ = 1.2 msec, we can calculate that T₂ = 1.9 msec. If the narrowest line width is due entirely to spin-exchange broadening, then FWHM = 1/(πT₂) = 170 Hz. Including a contribution to the line width of ~30-40 Hz due to inhomogeneity broadening gives ~200 Hz which is in approximate agreement with the experimental value of 190 Hz.

SUMMARY OF WORK CARRIED OUT UNDER THIS CONTRACT

This section gives the overall summary and conclusions for the
Figure 8. AM line shape for pulsed optical pumping frequency standard for a microwave pulse duration of 4.5 msec. This gives the narrowest line width observed to date for pulsed pumping, namely 190 Hz. Experimental conditions are given in Table 1. The location of the origin on the horizontal axis is arbitrary.
work carried out under this contract, and draws upon material presented in Appendix A, as well as in ONR Annual Summary Report No. 00014-77-C-0646-001.

1. Technical Feasibility

The design philosophy adopted in the present work is that it is not profitable to conduct laboratory experiments using principles and methods that cannot subsequently be applied to the development of a compact, lightweight, low power frequency standard. In keeping with this design philosophy, all experimental work described in this report has been carried out using a very small physics package (consisting of lamp, "integrated cell," microwave cavity, magnetic shield, photodetectors, and lamp electronics) that occupies a total volume of 260 cc (about equal to the volume of an orange). Using this physics package, it has been demonstrated that the method of pulsed optical pumping is technically feasible in the sense that signal-to-noise ratios can be obtained that are adequate for operating the device as a frequency standard. This is possible because of a dual photocell compensation method that was developed in the course of this investigation.

2. Short-Term Frequency Stability ($\tau = 1$ to 100 sec.)

The short-term frequency stability has been measured for the pulsed optical pumping frequency standard described above. The result is
\[ \sigma_y(\tau) = 2 \times 10^{-11} \tau^{-1/2}, \quad 1 \leq \tau \leq 100 \text{ sec.} \]

When the pulsed pumping apparatus is operated cw (no pulsing) the improvement in \( \sigma_y \) is less than a factor of two.

It should be pointed out that the \( \sigma_y \) obtained for pulsed pumping is comparable to that of present-day commercial rubidiums, most and exceeds that of commercial cesiums for the same averaging times. Also, it is possible that this \( \sigma_y \) could be improved, perhaps by a factor of two, with further work.

3. Long-Term Stability

Prior to homogenizing the C-field and eliminating the pseudo light shift (Appendix A, p.6), long-term stability data were taken over a period of ten consecutive days. From these data, the square root of the Allan Variance was computed for averaging times of 24 hours and 48 hours. The results obtained are

\[ \sigma_y(\tau) = (8.5 \pm 2.5) \times 10^{-12} \quad \text{for } \tau = 24 \text{ hours} \]
\[ \sigma_y(\tau) = (4.4 \pm 1.9) \times 10^{-12} \quad \text{for } \tau = 48 \text{ hours} \]

The uncertainties are those corresponding to one standard deviation, assuming white frequency noise.\(^5\)

These results indicate that the pulsed optical pumping frequency standard is not yet as good as our commercial units. However,
it should be emphasized that the pseudo light shift effect was present during these measurements; This would be expected to degrade the long-term frequency stability. (Due to this effect, a 1% change in light intensity will result in a fractional frequency shift of \(6 \times 10^{-12}\).) Because of the time factor (see p.6 of this report) it was, unfortunately, not possible to repeat these measurements using the uniform C-field coil (elimination of pseudo light shift effect), but if this were to be done, an improvement in long-term stability would most likely result.

4. **Light Shift**

Using the method of pulsed optical pumping, the light shift (fractional frequency shift due to a 30% reduction in light intensity) has been reduced by an order of magnitude compared to cw operation. The residual light shift (pseudo light shift effect) of \(1.5 \times 10^{-10}\) that remains has been further reduced to an undetectable amount (<\(1 \times 10^{-11}\)) by using a more homogeneous C-field. Thus, to the precision of the measurements, the light shift has been eliminated. (See Appendix A, p.6 and Table 5 for details.) It was not possible within the time frame of this contract to make measurements of the long-term frequency stability under conditions of no detectable light shift. This is unfortunate because the pseudo light shift effect is still sufficiently large that a 1% change in light intensity will produce a fractional frequency shift of \(6 \times 10^{-12}\).
5. Temperature Coefficient of Frequency

Over a 10°C range of ambient temperature, the temperature coefficient (TC) of frequency of the pulsed optical pumping frequency standard was previously found to be $+5 \times 10^{-12}/°C$ (with electronic temperature compensation, prior to most recent modifications). This is within a factor of two of the TC of the company's commercial rubidium. For the pulsed device, both the electronics and the Rb resonance cell are temperature sensitive, and it should be possible to make significant improvements in both areas. For example, by using a different combination of buffer gases in the resonance cell, it should be possible to reduce the resonance cell TC by nearly a factor of 10. This was attempted using a mixture of nitrogen and krypton as the buffer gas. The mixture used is predicted to have a low TC but this was not found to be the case experimentally (see p.2 of this report). Some modifications have been made to the electronics to reduce the TC there. At this writing, these modifications (p.6, this report) were still in the debugging stage and it is therefore too early to evaluate their effect.

6. Dependence of Frequency on Pulse Parameters

It is found that if the values of the pulse parameters of the pulsed pumping frequency standard are changed, the output frequency changes. For each pulse parameter investigated (light pulse duration, $T_L$; microwave pulse duration, $T_M$; dark period duration, $T_d$),
it is found that there is a range of values where the frequency changes quite rapidly, and another (separate) range of values where the frequency changes much more slowly. In general, it has been found that it is possible to choose parameter values so that the sensitivity of the output frequency to small changes in parameter values is minimized. In general, this can be done without having to sacrifice signal. This approach seems adequate for stabilities of a few parts in $10^{12}$ over days and weeks, but if stabilities of parts $10^{13}$ and $10^{14}$ are to be realized over the same periods of time (and longer), then it will be necessary to use a more fundamental approach.

The most sensitive parameter of the three mentioned is the microwave pulse duration, $T_\mu$. It has been found that the sensitivity of the output frequency to changes in $T_\mu$ is related to distortion of the line shapes as $T_\mu$ is reduced below 1 msec. At the present time, the cause of this line shape distortion is not known, but at least two possibilities exist (pp.6,14) that should be investigated in the future.

7. Position-Shift Effect

The "position-shift effect" occurs in frequency standards of the type used in this work. It is due to inhomogeneous broadening in the physics package, combined with spatially nonuniform optical
pumping and interrogation of the $^{87}$Rb atoms. In general, only two sources of inhomogeneous broadening are possible: that due to a nonuniform static magnetic field (C-field nonuniformity) and that due to a true light shift.

Based on the NBS findings$^6$ alone, it is virtually certain that the pseudo light-shift effect is due to the position-shift effect. Indeed, if there is no true light shift in the pulsed mode, as we believe to be the case, then the position-shift effect could be completely eliminated by homogenizing the static magnetic field (C-field); this would remove the only remaining source of inhomogeneous broadening. This has been done as part of this contract and it has been determined that this reduces the pseudo light-shift to an undetectable amount. Although there has, as yet, been no attempt to verify it experimentally, this modification should also reduce the sensitivity of the output frequency to changes in microwave power.

8. **Theoretical Model of Device Operation**

As part of this work, a simple theoretical model of device operation has been developed that provides a simple, conceptual picture of how the signal at resonance depends on device parameters, especially the pulse parameters. The theoretical predictions of this model are found to be in semiquantitative agreement with the experimental results. With further work it should be possible to extend this theory to allow predictions of line shapes as well. The importance of this model is that it provides a conceptual basis
for understanding the physics of device operation, which is important for a fundamental approach to device improvement.

9. Relaxation Times

A knowledge of relaxation times is also important for a fundamental understanding of device operation. One of the results of this work has been the development of a new method for the measurement of the relaxation time $T_1$ (time constant for decay of the population difference). This method is complementary to previous methods.\(^7\,4\)

Measurements of $T_1$ for our experimental conditions show that the dominant relaxation mechanism is spin-exchange. Our best set of measurements provides evidence for the presence of two distinct relaxation times. While this might be due to a spurious instrumental effect, it could also be due to two different values of $T_1$ for our apparatus: one for $^{87}\text{Rb}-^{87}\text{Rb}$ spin-exchange relaxation, and the other for $^{87}\text{Rb}-^{85}\text{Rb}$ spin-exchange relaxation. If this result is real, it would be of considerable interest because the $^{87}\text{Rb}-^{85}\text{Rb}$ spin-exchange relaxation rate has not been previously measured.

10. Line Shapes

The rubidium resonance line shapes for both pulsed and cw operation have been measured. From this, the following conclusions can be drawn: (1) For line widths greater than about 200 Hz, the line width is proportional to $1/T_\mu$ where $T_\mu$ is the duration of the micro-
wave pulse. (2) The "intrinsic" line width for pulsed operation in our apparatus is approximately 200 Hz and is due almost entirely to $T_2$-type spin-exchange relaxation. (3) For cw operation, the line width is approximately 1100 Hz and is due mostly to light relaxation. Therefore, for our experimental conditions, narrower line widths can be obtained when the apparatus is operated in the pulsed mode. (4) The line shapes for pulsed operation show small subsidiary maxima in the wings of the line. This type of line shape has also been observed in molecular beam experiments, and is due to the pulsing of the microwave radiation.

RECOMMENDATIONS FOR FUTURE WORK

1. Long-Term Stability Measurements

Additional phase comparison data should be taken for the pulsed optical pumping frequency standard over a period of several weeks, or more, under controlled environmental conditions. This will allow calculation of $\sigma_y(\tau)$ for $\tau > 24$ hours. This should be done using the more uniform C-field configuration to reduce the pseudo light shift effect to a negligible value, and after complete debugging of all recent improvements (modifications) to the apparatus. These measurements would most likely be limited by the verifiable (traceable to NBS) stability of our house standard to a precision of $< 4 \times 10^{-12}$ for $\tau = 24$ hours ($8.6 \times 10^4$ sec), and $< 2 \times 10^{-12}$ for $\tau = 1$ week ($6 \times 10^5$ sec). If these measurements were to yield a $\sigma_y(\tau)$ that were
limited by the stability of the reference, then it would be necessary to have access to a more stable (for \( \tau \geq 24 \) hours) reference to establish the true value of \( \sigma_y(\tau) \) for the pulsed optical pumping frequency standard.

2. Temperature Coefficient of Frequency

Work should be continued to find a buffer gas mixture having low temperature coefficient (TC). So far, only one gas mixture has been tried. Some effort should be made to understand how the absence of light shifts affects the TC of the resonance cell as a function of the buffer gas mixture.

The modifications to the electronics that were made to reduce the TC of this portion of the apparatus should be debugged and the residual temperature sensitivity of the electronics assessed experimentally.

3. Frequency Shifts due to Changes in Pulse Parameters

More work should be done in this area for the reasons discussed previously. Effort should be directed primarily to understanding the sensitivity of output frequency on microwave pulse duration, \( T_\mu \), since this is the most sensitive parameter. As least two possible causes of this effect have been mentioned; they should be investigated.

It is well known that the effect of pulsing a sinusoidal wave is to generate sidebands that are placed symmetrically about the carrier frequency. These sidebands are equally spaced, with the
spacing between adjacent frequency components being equal to 
\(1/T_c\) where \(T_c^{-1}\) = pulse repetition rate. The amplitude of these 
components is determined by the microwave pulse duration. The 
effect of these additional frequencies on device operation should 
be examined to see, for example, if it is possible to lock the 
atomic resonance to one of these components rather than to the 
carrier frequency. The possibility of frequency pulling by side-
band asymmetry should be studied in addition. Also, the large 
frequency shifts that occur as a function of \(T\mu\), when \(T\mu\) is small, 
(see Figure 7) may be related to the amplitude and location of 
these sidebands in some way. This is an area that also demands 
investigation. Efratom has just recently acquired a high resolu-
tion spectrum analyzer for the range 20 Hz to 40 GHz with which 
this spectrum could be studied.

4. **Theoretical Model of Device Operation**

The present model should be extended. Emphasis on model de-
velopment should be placed on the averaging processes that occur 
in the resonance cell, and on understanding the observed line shapes. 
For understanding device operation as a frequency standard, the 
theoretical approach should be extended to include FM operation.
REFERENCES


3. In the beam experiments, a molecule traveling through the apparatus passes through the rf (resonance) region. From its frame of reference, the molecule sees the rf as a pulse whose duration is equal to the time of flight through the rf region. The main difference between the beam case, and the gas cell case is the presence of relaxation for the gas cell atoms (for all practical purposes, molecules in the beam do not experience relaxation).


APPENDIX A

"Elimination of the Light Shift in Rubidium Gas Cell Frequency Standards Using Pulsed Optical Pumping"

by

Thomas C. English, Ernst Jechart and T. M. Kwon

from


Published as NASA Technical Memorandum 80250.
ELIMINATION OF THE LIGHT SHIFT IN RUBIDIUM GAS CELL FREQUENCY STANDARDS USING PULSED OPTICAL PUMPING

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ABSTRACT

It is well known that changes in the intensity of the light source in an optically pumped, rubidium, gas cell frequency standard can produce corresponding frequency shifts, with possible adverse effects on the long-term frequency stability. Since this so-called "light-shift" effect is due to the simultaneous presence of pumping light and interrogating microwave radiation, it can be eliminated, in principle, by alternately pulsing the pumping light and the microwave radiation so that there is no temporal overlap.

We have constructed a pulsed optical pumping apparatus with the intent of investigating the frequency stability in the absence of light shifts. Contrary to our original expectations, a small residual frequency shift due to changes in light intensity has been experimentally observed. Evidence is given which indicates that this is not a true light-shift effect. Preliminary measurements of the frequency stability of this apparatus, with this small residual "pseudo" light shift present, are presented. It is shown that this pseudo light shift can be eliminated by using a more homogeneous C-field. This is consistent with the idea that the pseudo light shift is due to inhomogeneity in the physics package ("position-shift" effect).

INTRODUCTION

At the present time, there is a real need for compact, lightweight, low power, highly stable atomic frequency standards. This is ex-
emphasized by programs such as that for the NAVSTAR Global Positioning System, or GPS for short. This program calls for state-of-the-art size, weight and power reductions for atomic frequency standards, and a long-term stability requirement of one part in $10^{13}$ over periods of approximately 1 to 12 days for phases II and III of GPS (1). Requirements such as these can be expected to be even more demanding in the future as the overall state-of-the-art of military technology moves forward.

Table 1 summarizes the state-of-the-art for small atomic frequency standards. The hydrogen and cesium devices have excellent stability over periods of weeks, months and even years. Rubidium is less satisfactory in this respect but has the advantage of much smaller size, weight and power consumption. The ideal frequency standard would combine the excellent long-term stability of hydrogen and cesium with the small size, weight and power consumption of rubidium. There are two possible methods of approach toward this goal. The first is to reduce even further the size, weight and power of hydrogen and cesium devices. While further small reductions might be possible, it is unlikely that these devices can be made to approach present day rubidiums in this respect. For example, even for cesium a factor of 9 in size and a factor of 7 in weight would be required. Moreover, relative to future rubidiums, factors of 18 and 12, respectively, would be required (2). We are therefore led to the second method of approach, which is to improve the long-term stability of rubidium devices without significantly increasing size, weight, or power consumption. Table 2 shows that there is reason to believe that this can be done.

The best reported stability for rubidium is several parts in $10^{14}$ for an averaging time of about 7 hours. Two such measurements have been reported (3,4), each on a different unit at different times and by different groups. In addition, a stability of parts in $10^{14}$ for $\tau = 6$ hr to 12 days for one rubidium device has been reported at this conference (this device uses an Efratom physics package) (1). This stability is on a par with cesium (5) and is only about a factor of 50 worse than the "best" reported stability for hydrogen (6). For times longer than 7 hours, the stability of most rubidium devices worsens, most likely due to uncontrolled changes in device parameters. If the device parameters that are changing could be determined and controlled, better long-term stability would result.

FUNDAMENTAL PHYSICAL EFFECTS

Figure 1 lists those fundamental physical effects that can adversely affect the stability of rubidium devices. Let us consider these effects one at a time.
Light Shift

The insert at the lower left of Figure 1 shows the two hyperfine energy levels of rubidium which determine the rubidium resonant frequency. In general, when a rubidium atom is illuminated by the light used for optical pumping, these two energy levels are Stark shifted by the electric field of the light, thereby producing a change in the atom's resonant frequency (7). This results in a frequency shift that is proportional to light intensity.

This effect can be eliminated by interrogating the atoms in the dark; that is, when the pumping light is absent.

Position-Shift Effect

The position-shift effect (8) is due to inhomogeneity in the physics package. The result of this inhomogeneity is to make the resonant frequencies of the rubidium atoms depend on their location within the cell, as indicated schematically at the lower right of Figure 1. This inhomogeneity is due mostly to C-field nonuniformity and also, to a lesser extent, to light shift nonuniformity. Since the optical pumping and the microwave interrogation are also nonuniform over the cell, the experimentally observed resonant frequency is a weighted average of the frequencies of the individual atoms in the cell. Now, if the intensity of pumping light were to change, then the weighted average would for example, shift so that those atoms in the center of the cell might be favored more than those at the ends. This would obviously result in an increase in the experimentally observed resonant frequency. We term this type of shift the "pseudo light shift" because it can mimic a true light shift.

Buffer Gas Shift

The buffer gas shift (9) occurs due to collisions between the rubidium atoms and the buffer gas atoms, and is dependent on buffer gas density and temperature, as well as on the buffer gas that is used.

Spectrum of Exciting Microwave Radiation

Frequency shifts due to the exciting microwave radiation are not usually a problem if care is taken to obtain a spectrally pure exciting frequency free of spurious and unwanted sidebands (10).

Magnetic Field

Since the magnetic field sensitivity (10) for rubidium is only about 2 x larger than for cesium, magnetic field sensitivity for rubidium
is not significantly more of a problem than it is for cesium.

It is very difficult to give estimates of the possible frequency changes due to these effects because they are a very strong function of the individual device configuration and parameters. In spite of this, we can still say that of the 5 effects listed here, the first 3 are the most important.

The objective of the experiments that we are carrying out is to improve the long-term stability of small rubidium devices. Today we will describe some preliminary results that lead toward this goal. These results deal with the reduction and elimination of the light-shift and the position-shift effects, and have been obtained with the expenditure of less than one man-year of scientific effort.

METHOD FOR ELIMINATION OF LIGHT SHIFT

Figure 2 shows the method that we have used to eliminate the light shift. This method was first suggested by Arditi [11]. First, the light is pulsed on and the atoms are optically pumped. Then the light is turned off and the interrogating microwave radiation is turned on. The microwave radiation is then turned off and the pumping light is turned on again. This basic cycle is subsequently repeated many times. Since the atoms are interrogated in the dark, the light shift should be eliminated. This, of course, assumes that the atomic coherence does not persist from one cycle to the next [12].

Before passing to the next slide, we note that in our experiments, the atomic transition is detected by optically monitoring the absorption of the pumping light. This is essentially the same detection method as that used in all conventional rubdiums.

Figure 3 shows a block diagram of the apparatus. The pulser alternately pulses the light and the microwave radiation at a 280 Hz rate, as shown in the previous slide, so that the atoms are interrogated in the dark. The remainder of the apparatus is a conventional frequency locked loop. The modulation frequency of 10 Hz is chosen to be about an order of magnitude smaller than the pulsing frequency so that the two signals can be separated by filtering before synchronous detection of the 10 Hz.

Figure 4 is a photograph of the physics package, which is a modified version of the physics package used in the Efratom, Model FRK rubidium frequency standard. The base of the rubidium lamp is at the right, and the magnetic shield that encloses the resonance cell and microwave cavity is at the left. The entire unit is less than 4 inches long.
The philosophy adopted in this work was that everything possible should be done to retain the small size.

RESULTS

Light shift measurements have been made on this apparatus and the results are shown in Table 3. Measurements were made of the fractional frequency shift resulting from a 30% change in light intensity. Two sets of measurements were made — one with the apparatus operated CW, and the other with it pulsed.

In the case of CW operation, we expect a large light shift. This was observed for each of two different rubidium lamps — lamp A and lamp B. These two lamps differ in the ratios of their rubidium isotopes. Lamp A produces a positive light shift, and lamp B a negative light shift. This is in agreement with the theory of the light shift in rubidium 87 as worked out by Mathur, Tang and Happer [7].

When the apparatus is operated in the pulsed mode we expect to see no light shift. Yet there is a change in frequency with light intensity. This change is about a factor of 10 smaller than the CW light shift, and we can tell that it is not a true light shift because it does not change sign in going from lamp A to lamp B. Other tests, which we will not describe here, also confirm this to be the case. For these reasons we dub this effect the "pseudo-light-shift effect." We will have more to say on this later.

Table 4 shows the results of some preliminary frequency stability measurements that have been made on our pulsed optical pumping apparatus. For pulsed pumping, the short-term stability is expected to be degraded somewhat by noise introduced in the pulsing process. The short-term stability for pulsed pumping has been measured for averaging times from 1 to 100 seconds and found to improve as $1/\sqrt{T}$ (footnote A in Table 4). This shows that we are dealing with white frequency modulation noise, as is usually the case for passive rubidium devices. The value of $\sigma_\tau$ for 100 sec is given in column 2 and can be compared with that for our small commercial rubidiums. The result for pulsed pumping lies between the spec for our two commercial models and is better than that of an HP 5062C cesium. The short-term stability of the pulsed pumping apparatus is therefore quite good, even in this preliminary stage, and can almost certainly be improved further.

The long-term stability was also measured for a 24-hour averaging time and a preliminary value of approximately 5 parts in $10^{12}$ was obtained. This is not yet as good as our commercial units.
After these stability measurements were made it was discovered that there were several device parameters that were not under tight control. These included significant second-harmonic contamination of the 10 Hz modulation, and frequency changes due to changes in barometric pressure. All of these parameter changes can be expected to produce frequency changes of parts in $10^{12}$, which is of the order of the observed instability over 24 hours. The pseudo-light-shift effect is not negligible at this level either, and it may be a contributor to the observed instability.

All of these parameters can be easily controlled except for the pseudo light shift. However, it was suspected that the pseudo-light-shift effect might actually be a manifestation of the position-shift effect, as mentioned earlier. To test this hypothesis, a new C-field was constructed for our physics package that greatly improved the homogeneity of the static magnetic field and which should therefore greatly diminish the position-shift effect.

Table 5 shows the result of using this new C-field. The first line of this table is a repeat of the data shown in Table 3 for the old C-field. The second line shows what happened when some small steel parts on the outside of the microwave cavity were removed. This improved the C-field homogeneity and also reduced the pseudo light shift by about 30%. Finally, the last line of the table shows the results for the new C-field. The pseudo light shift is now undetectable, of the order of parts in $10^{12}$ or less for a 30% change in light intensity.

To summarize, the residual light shift has now been reduced to an undetectable amount by using the method of pulsed optical pumping in conjunction with a homogeneous C-field. It is likely that the new homogeneous C-field will also have other beneficial effects, such as reduced sensitivity to changes in microwave power, but this has not yet been verified experimentally.

Our plans for the immediate future are to beat down the known sources of frequency instability to the level of parts in $10^{13}$ or below, and then to take additional long-term stability data. It is expected that this will lead to an improved long-term stability compared to the present value of about 5 parts in $10^{12}$ for $T = 24$ hours which was taken before the pseudo light shift was eliminated. Stability data over longer periods of time will also be taken to see if there is an improvement there.

Table 6 compares our preliminary results with those obtained by other investigators, namely, Arditi and Carver, who were the first to use the method of pulsed optical pumping for elimination of the light shift, and a Russian group that has done several man-years of work in
In their experiments, Arditi and Carver used a high sensitivity microwave receiver to detect the rubidium resonance. Because of the complexity of the electronics this method is not suitable for use in a practical device. The Russians used an optical detection method, that has the disadvantage of requiring 2 rubidium lamps. We also use an optical detection method but only one rubidium lamp is required. In addition, we have used a single rubidium cell that combines the filtering and resonance functions, thereby eliminating the need for a separate filter cell. For these reasons our physics package is extremely small, which is desirable in a practical device. In fact, this is the same physics package that is used in the Efratom small commercial rubidium frequency standards.

Arditi and Carver saw no light shift at the level of a part in $10^{-10}$. On the other hand, the Russians did observe true light shifts (due to persistence of the atomic coherence) but were able to minimize them by proper choice of operating conditions. In our experiments the light shift is undetectable so that if it exists at all, it is of order parts in $10^{-12}$ or less for a 30% change in light intensity.

As regards stability, Arditi and Carver made some short-term measurements at the level of about $1 \times 10^{-10}$. The Russians have not reported any stability measurements for reasons unknown to us. As already mentioned, we have measured the frequency stability for our apparatus and found it to be of the order of parts in $10^{-12}$. However, it should be emphasized that these measurements are preliminary and were made prior to elimination of the pseudo light shift.

**PROGNOSIS**

At the present time, the ultimate frequency stability attainable using the pulsed optical pumping method is not known. Possible limitations could be due to changes in buffer gas pressure, if they occur, and also to changes in the pulsing parameters, such as the durations of the light and microwave pulses. It is known that frequency shifts do occur due to changes in the pulsing parameters. We have investigated this phenomenon using the old C-field and have estimated that it certainly is important at the level of parts in $10^{-13}$.

Future efforts will concentrate on understanding and reducing frequency sensitivity to changes in pulsing parameters. Effort will also be devoted to devising a method for studying frequency shifts due to possible small changes in buffer gas pressure.
ACKNOWLEDGEMENTS

We would like to thank Werner Weidemann, Engineering Manager of Efratom Systems Corporation, for many helpful discussions, and for kindly making available both equipment and facilities, as needed. We also acknowledge the able technical assistance of Jeff Hayner, Henry Holtermann and John Hall.

REFERENCES


2. T. C. English, presentation on rubidium frequency standards as part of the Discussion Form on Atomic Frequency Standards, Session II of this conference (10th PTTI, 1978).


<table>
<thead>
<tr>
<th>DEVICE</th>
<th>LONG-TERM STABILITY</th>
<th>SIZE (LITERS)</th>
<th>WEIGHT (LBS)</th>
<th>POWER (W)</th>
<th>APPROX. COST (k$)</th>
</tr>
</thead>
<tbody>
<tr>
<td>SPACECRAFT</td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>H-MASER</td>
<td>$&lt; 1 \times 10^{-14}/10 \text{d}^A$</td>
<td>20-50$^A$</td>
<td>50-90$^A$</td>
<td>55$^A$</td>
<td></td>
</tr>
<tr>
<td>SMALL COMMERCIAL</td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>CESIUM</td>
<td>parts in $10^{12}/\text{yr}^B$</td>
<td>9</td>
<td>22</td>
<td>24</td>
<td>18</td>
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<tr>
<td>SMALL COMMERCIAL</td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>RUBIDIUM</td>
<td>$&lt; 1 \times 10^{-11}/\text{md}^B$</td>
<td>1</td>
<td>3</td>
<td>13</td>
<td>6</td>
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$^A$PROJECTED

$^B$TYPICAL MANUFACTURER'S SPECIFICATION
<table>
<thead>
<tr>
<th>DEVICE</th>
<th>STABILITY, $\sigma_y(\tau)$</th>
<th>AVERAGING TIME, $\tau$</th>
<th>REF</th>
</tr>
</thead>
<tbody>
<tr>
<td>SAO H-MAKER</td>
<td>$6 \times 10^{-16}$</td>
<td>11 HR</td>
<td>1</td>
</tr>
<tr>
<td>COMMERCIAL CESIUM</td>
<td>$2 \times 10^{-14}$</td>
<td>5 D</td>
<td>2</td>
</tr>
<tr>
<td>(HIGH PERFORM)</td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>SMALL COMMERCIAL</td>
<td>$3-4 \times 10^{-14}$</td>
<td>7 HR</td>
<td>3,4</td>
</tr>
<tr>
<td>RUBIDIUM</td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>GPS SPACE CRAFT</td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>RUBIDIUM (S/N 2)</td>
<td>$\leq 8 \times 10^{-14}$</td>
<td>6 HR TO 12 D</td>
<td>5</td>
</tr>
</tbody>
</table>

2. Percival, 5th PTI, 1973, p. 239.
TABLE 3
PSEUDO LIGHT SHIFT EFFECT FOR PULSED OPTICAL PUMPING

<table>
<thead>
<tr>
<th>RB LAMP</th>
<th>CW</th>
<th>PULSED</th>
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<tr>
<td></td>
<td>FRACTIONAL FREQUENCY SHIFT DUE TO LIGHT INTENSITY CHANGE ((\gamma)<em>{1o} - (\gamma)</em>{0.7o})</td>
<td></td>
</tr>
<tr>
<td>A</td>
<td>+1.4 x 10^{-9}</td>
<td>+1.5 x 10^{-10}</td>
</tr>
<tr>
<td>B</td>
<td>-1.6 x 10^{-9}</td>
<td>+2.4 x 10^{-10}</td>
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### TABLE 4
PULSED OPTICAL PUMPING
PRELIMINARY STABILITY RESULTS

<table>
<thead>
<tr>
<th>DEVICE</th>
<th>$\sigma_y$ (100 sec)</th>
<th>$\sigma_y$ (24 HR)</th>
</tr>
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<tbody>
<tr>
<td>PULSED PUMPING (PRESENT WORK)$^A$</td>
<td>$2 \times 10^{-12}$</td>
<td>$\sim 5 \times 10^{-12}$</td>
</tr>
<tr>
<td>EFRATOM FRK-H RUBIDIUM$^B$</td>
<td>$1 \times 10^{-12}$</td>
<td>(&lt; $1 \times 10^{-12}$)$^c$</td>
</tr>
<tr>
<td>FRK-L RUBIDIUM$^B$</td>
<td>$3 \times 10^{-12}$</td>
<td>------</td>
</tr>
<tr>
<td>NEWLETT-PACKARD 5062C CESIUM$^B$</td>
<td>$7 \times 10^{-12}$</td>
<td>(&lt; $1 \times 10^{-12}$)$^c$</td>
</tr>
</tbody>
</table>

$^A \sigma_y = 2 \times 10^{-11} \tau^{-1/2}, \; 1 \leq \tau \leq 100 \text{ sec}$

$^B$ MANUFACTURER'S SPECIFICATION

$^C$ UPPER LIMIT
TABLE 5
LIGHT-SHIFT MEASUREMENTS FOR DIFFERENT C-FIELD CONFIGURATIONS (LAMP A)

<table>
<thead>
<tr>
<th>C-FIELD</th>
<th>FERROMAGNETICS ON CAVITY</th>
<th>FRACTIONAL FREQUENCY SHIFT DUE TO LIGHT INTENSITY CHANGE ( \nu ) $\nu_{10}$ - $\nu_{0.710}$</th>
</tr>
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<tbody>
<tr>
<td>OLD</td>
<td>YES</td>
<td>$1.4 \times 10^{-9}$</td>
</tr>
<tr>
<td>OLD</td>
<td>NO</td>
<td>$1.3 \times 10^{-9}$</td>
</tr>
<tr>
<td>NEW</td>
<td>NO</td>
<td>$1.0 \times 10^{-9}$</td>
</tr>
<tr>
<td>STUDY</td>
<td>DETECTION SCHEME</td>
<td>LIGHT SHIFT</td>
</tr>
<tr>
<td>---------------------</td>
<td>--------------------</td>
<td>-------------------</td>
</tr>
<tr>
<td>ARDITI &amp; CARVER(^A) (1964)</td>
<td>MICROWAVE SUPERHET</td>
<td>NO ((&lt; 1 \times 10^{-10}))</td>
</tr>
<tr>
<td>ALEXSEYEV ET AL(^B) (1974)</td>
<td>OPTICAL (TWO RB LAMPS)</td>
<td>YES</td>
</tr>
<tr>
<td>PRESENT WORK</td>
<td>OPTICAL (SMALL PHYSICS PACKAGE)</td>
<td>NO ((&lt; 1 \times 10^{-11}))</td>
</tr>
</tbody>
</table>


FIGURE 1

FUNDAMENTAL PHYSICAL EFFECTS THAT CAN ADVERSELY AFFECT
THE LONG-TERM STABILITY OF PASSIVE RUBIDIUM DEVICES

- LIGHT SHIFT ($\sim 3 \times 10^{-11}$ for $\Delta I_{\text{light}}/I_{\text{light}} = 1\%$)
- POSITION-SHIFT EFFECT (UP TO PARTS IN $10^9$)
- BUFFER GAS SHIFTS ($\sim 1 \times 10^{-10}$/MILLITORR)
- SPECTRUM OF EXCITING MICROWAVE RADIATION
  (UP TO PARTS IN $10^9$)
- MAGNETIC FIELD (RB SENSITIVITY = $1.8 \times$ CS SENSITIVITY)
FIGURE 2
PULSING SCHEME

<table>
<thead>
<tr>
<th>PULSE PARAMETER</th>
<th>Typical Value (msec)</th>
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</thead>
<tbody>
<tr>
<td>LIGHT DURATION</td>
<td>2.0</td>
</tr>
<tr>
<td>DARK TIME</td>
<td>1.6</td>
</tr>
<tr>
<td>MICROWAVE DURATION</td>
<td>1.2</td>
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<tr>
<td>REPETITION RATE</td>
<td>280 Hz</td>
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</table>
FIGURE 3
BLOCK DIAGRAM OF APPARATUS

- RB LAMP
- PULSER (~ 280 Hz)
- RB RESONANCE CELL
- MICROWAVE SYNTHESIZER
- 10 Hz SIGNAL AMPLIFIER
- 6.834 GHz
- 10 Hz FILTER
- 10 MHz PHASE MODULATOR
- MODULATION
- MODULATION REFERENCE
- 10 Hz OSCILLATOR
- SYNCHRONOUS DETECTOR & INTEGRATOR
- 10 MHz VCXO
- SERVO CONTROL VOLTAGE
- OUTPUT (10.000... MHz)