Two-step pulse observation for Raman–Ramsey coherent population trapping atomic clocks

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We propose a two-step pulse observation method to enhance the frequency stability of coherent population trapping (CPT) atomic clocks. The proposed method is a Ramsey–CPT scheme with low light intensity at resonance observation and provides a Ramsey-CPT resonance with both reduced frequency sensitivity to the light intensity and a high signal-to-noise ratio by reducing the repumping into a steady dark state. The resonance characteristics were calculated using density matrix analysis of a Λ-type three-level system that was modeled on the ¹³³Cs D₁ line, and the characteristics were also measured using a vertical-cavity surface-emitting laser and a Cs vapor cell.

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Atomic clocks based on coherent population trapping (CPT) resonance have attracted attention as a means of fabricating very small frequency references, such as chip-scale atomic clocks.¹ CPT atomic clocks are in great demand for many applications, such as telecommunications, navigation systems, and synchronization of networks,² and such clocks are required for their high frequency stability.

Frequency stability in atomic clocks is generally represented by the Allan deviation and can be classified into two types, short term and long term, according to the averaging time of the Allan deviation. These two frequency stability types are degraded by different mechanisms. To enhance the short-term frequency stability, a high signal-to-noise ratio (SNR) and a narrow CPT resonance linewidth are required because the short-term stability is determined by the product of the SNR and the Q value. For long-term frequency stability, the light shift (or the ac Stark shift), which is a frequency shift caused by a light field, is a major limiting factor.³ Because the long-term stability degradation induced by the light shift is caused by laser light power fluctuations or aging of the optical elements, reduced frequency sensitivity to the light intensity is desired.

The Raman–Ramsey (RR) scheme for enhancement of the frequency stability of CPT atomic clocks has been investigated by a number of researchers.⁴⁻⁸ This scheme significantly reduces the transition in the light shift to one or two orders of magnitude lower than that under continuous wave (cw) illumination.⁷⁻⁹ In addition, because this scheme suppresses power broadening, a narrow linewidth can be obtained.⁵ Recently, a short-term stability of 3.2 × 10⁻¹³ r⁻¹/₂ was obtained, and stability as low as 3 × 10⁻¹⁴ was achieved at an averaging time of 200 s using a Cs vapor cell with the RR scheme.¹⁰ Moreover, when a cold Rb atom was used, a short-term stability of 4 × 10⁻¹¹ r⁻¹/₂ was obtained, and a long-term frequency stability of 3 × 10⁻¹³ was achieved by an averaging time of 5 h.¹¹

In our previous paper, we investigated the light shift in the RR scheme both theoretically and experimentally with the aim of enhancing the long-term frequency stability of CPT atomic clocks.⁹ The results showed that the light shift under the RR scheme is significantly reduced compared with that under cw illumination. We also found that the frequency variation of the light intensity is reduced by setting the observation time to be as short as possible. The main reason for this is that the atoms evolve toward a steady state during observation. In addition, we found that the light shift in the RR scheme is a nonlinear function of the light intensity, and reduced frequency sensitivity is obtained under a high light intensity beyond a set threshold value. Thus, the measurement conditions with both short observation times and high light intensity provide reduced frequency sensitivity to the light shift. However, the short observation time causes the short-term stability to degrade because of increases in both the Dick effect and the laser intensity noise.¹⁰ In addition, although the high light intensity provides a high transition population, it also leads to a reduction in the resonance signal because of increased repumping into the steady dark state.¹²⁻¹³ Therefore, it is difficult to maintain the SNR of the resonance while simultaneously reducing the variation in the light shift.

In this paper, we propose a two-step pulse observation method for enhanced frequency stability in CPT atomic clocks. The two-step pulse method is an RR scheme with low light intensity at observation. This method provides low variation in the light shift without reducing the SNR. We describe a light shift of the ⁶²S¹/₂ (F = 3 ↔ 4) state of ¹³³Cs as observed through CPT using the two-step pulse observation method. The light shift with the RR scheme is calculated using the density matrix. We also propose an estimation equation for the light shift under the RR scheme. This equation is useful for estimation of the light shift with the two-step pulse observation method.

The observation scheme for the two-step pulse observation method is shown in Fig. 1. The Ramsey-CPT resonance is observed by measuring the transmitted light intensity at observation time τₘ after the pulse rise. The two-step pulse observation maintains low light intensity for rIp until the observation point. Here r is defined as the observation intensity ratio, with a range of values from 0 to 1. When r = 1 is set, the two-step pulse observation method uses the same scheme as the conventional RR method. After the measure-
Light intensity approaches in under cw illumination, evolution time \( S \) light intensity and has two characteristics. The shift under the RR scheme in the absence of the in is shown in more detail in Ref. 8. The black solid line is selected as the excited state. The calculation method proportional to the light intensity \( \frac{I_p}{C_{11}} \), and \( \beta \) is required to reduce the systematic frequency shift. The free evolution time \( \tau_m \) and the atoms are prepared for the next measurement. A laser pulse with a duration of \( \tau - \tau_m \) irradiates the atoms for pumping of the steady dark state. After the free evolution time \( T \), a laser pulse irradiates the atoms again with light intensity \( r \). The light intensity \( I(t) \) for this scheme is written as

\[
I(t) = \begin{cases} 
  r I_p & (0 < t \leq \tau_m) \\
  I_p & (\tau_m < t \leq \tau) \\
  0 & (\tau < t \leq \tau + T)
\end{cases}
\] (1)

The calculated light shift in the RR scheme as a function of the light intensity is shown in Fig. 2. The \( D_1 \) line of Cs was used as the excitation transition, and the \( ^6\text{P}_{1/2}, F = 3 \) state was selected as the excited state. The calculation method is shown in more detail in Ref. 8. The black solid line is the calculated light shift under cw illumination, which is proportional to the light intensity \( \left( S_{cw} = \alpha_{cw} I_p \right) \). \( \alpha_{cw} \) is the slope of the light shift. The red line is the calculated light shift under the RR scheme in the absence of the influence of observation, \( S_{\tau_m=0} \). The free evolution time was set to 800 µs, and the \( \tau \rightarrow \infty \) steady-state solution is used as the initial condition to determine the evolution at later times. 13 The light shift in the RR scheme is a nonlinear function of the light intensity and has two characteristics. The first characteristic is that the variation under the RR scheme is equal to that under cw illumination with low light intensity. The reason for this is that the effective light field applied under the RR scheme is the same as that applied under cw illumination with low light intensity, because the low light intensity leads to a long coherence time. The second characteristic is that the light shift under the RR scheme approaches a saturation value \( S_{\text{lim}} \) as the light intensity \( I_p \) approaches infinity. This saturation value, \( S_{\text{lim}} \), is inversely proportional to the free evolution time \( T \left( S_{\text{lim}} = \frac{C_{\text{lim}}}{T} \right) \). When both of these characteristics are considered, the light shift estimation equation in the absence of an observation time, \( S_{\tau_m=0} \), is found to be

\[
S_{\tau_m=0} = \frac{\alpha_{cw} I_p}{\alpha_{cw} C_{\text{lim}} T P + 1} .
\] (2)

Under low light intensity conditions \( I_p \ll C_{\text{lim}} / (\alpha_{cw} T) \), because the first term of the denominator is negligible, the value of the light shift \( S \) approaches that produced under cw illumination. In addition, under high light intensity conditions \( I_p \gg C_{\text{lim}} / (\alpha_{cw} T) \), because the second term of the denominator is negligible, the light shift \( S \) converges on \( C_{\text{lim}} / T \)

\( (= S_{\text{lim}}) \). Therefore, Eq. (2) is satisfied using the above light shift characteristics under the RR scheme.

Under the influence of observation \( (\tau_m > 0) \), the light shift under the RR scheme then depends on \( C_{\tau_m} \). For large \( \tau_m \), the light shift under the RR scheme approaches that produced under cw illumination. Because the light intensity at observation is given by \( r \), the limit of \( S \) as the observation time \( \tau_m \) approaches infinity is

\[
S_{\tau_m \rightarrow \infty} = r S_{cw} = r \alpha_{cw} I_p.
\] (3)

From Eqs. (2) and (3), the light shift is shown to be a function that varies from the initial state to reach a steady value with increasing \( \tau_m \). We found that the time-varying function of the light shift follows a logistic function of \( \tau_m \). The light shift represented by a general logistic function is given by

\[
S = \frac{a_1}{a_2 e^{-a_3 \tau_m} + 1} \quad (a_1 > 0),
\] (4)

where the values of \( a_1, a_2, a_3 \) do not depend on \( \tau_m \). From the initial solution of Eq. (2) and the steady-state solution of Eq. (3), \( a_1 \) and \( a_2 \) equal \( r \alpha_{cw} I_p \) and \( r \alpha_{cw} C_{\text{lim}} T P + 1 \) respectively. When the time-dependent behavior of the CPT resonance is taken into account, \( a_3 \) is equal to the inverse of the pumping time \( \tau_p \), which is inversely proportional to the light intensity. 13 Thus, from Eqs. (2), (3), and (4), the light shift behavior in the two-step pulse observation \( S \) can be obtained as follows:

\[
S = \frac{r \alpha_{cw} I_p}{r \alpha_{cw} C_{\text{lim}} T P + 1} \left( 1 - e^{-a_3 \tau_m} \right) + 1.
\] (5)

We found that \( r \) has an optimal value for suppression of the frequency dependence of the observation time \( \tau_m \). The optimal \( r \) is derived as follows:

\[
r = \frac{S_{\tau_m=0}}{S_{cw}} = \frac{1}{\alpha_{cw} C_{\text{lim}} T P + 1}.
\] (6)

This means that \( r \) has a value such that the light shift in the steady state \( S_{\tau_m=\infty} \) is equal to the light shift in the absence of observation. Under this condition for \( r \), the light shift no longer depends on \( \tau_m \) and the light shift \( S \) equals \( S_{\tau_m=0} \).

The light shifts as a function of the light intensity under different values of \( r \) are shown in Fig. 3(a). The free evolution time \( T \) was 800 µs, and the observation time \( \tau_m \) was set to 10 µs. The results were then calculated numerically from the instantaneous values using density matrix analysis. The light shift was determined assuming that the dispersion spectrum given by the density matrix analysis is zero. The zero dispersion was found using Newton’s method. The light shift decreases with decreasing \( r \). Because the pumping rate increases with increasing light intensity, two-step pulse observation with a small \( r \) is effective for reducing the light shift under high light intensity. The light shift variation is also reduced by the use of a small \( r \). However, the offset shift remains at \( r \approx 0.00 \). Because the offset shift is attributed mainly to the free evolution time \( T \), a long free evolution time is required to reduce the systematic frequency shift.

Figure 3(b) shows the light shifts as a function of the observation time under different values of \( r \). The light intensity \( I_p \) was set to 2.0 mW/cm². The light shift is proportional to \( \tau_m \) because the observation time \( \tau_m \) is smaller.
than the pumping time $\tau_p$ in this range. The frequency variation of the observation time depends strongly on $r$ and decreases significantly with decreasing $r$ in the range from 0.25 to 1.00. In addition, the sign of the observation variation changes in the 0.00 < $r$ < 0.25 range. These results indicate that there is a value of $r$ between 0.00 to 0.25 that is optimal for suppression of the variation. The dashed line was plotted by setting $r = S_{\tau_m=0}/S_{cw}$; $r$ was 0.07 under this condition. Because the light shift is independent of the observation time setting, $r = S_{\tau_m=0}/S_{cw}$, it is not necessary to shorten the observation time to reduce the light shift. In addition, because a small $r$ leads to a low pumping rate, the resonance signal retains its high value, even if we use a longer observation time. Thus, the Ramsey-CPT resonance can be measured without reducing the resonance signal by two-step pulse observation.

A comparison of the results of the numerical calculations performed by density matrix analysis and the estimated values from Eq. (5) shows that the relative error of the results is no more than 0.05%. The residual of the relative frequency is on the order of $10^{-15}$, which is very small for CPT atomic clocks. Therefore, the proposed estimation equation is useful for practical purposes.

A contour map of the frequency variation of the light intensity is shown in Fig. 4(a). Because this variation decreases with increasing light intensity, the smallest variation using the conventional scheme ($r = 1.00$) is 0.562 Hz/(mW/cm²) at a light intensity of 2.4 mW/cm². The variation with two-step pulse observation decreases with decreasing $r$, and the smallest variation is obtained at $r = 0.00$. The variation at $r = 0.00$ was two to seven times smaller than that obtained using the conventional scheme. The variation reduction effect increases with increasing light intensity. The smallest variation with the two-step pulse observation method is 0.073 Hz/(mW/cm²), which is 7.6 times less than that obtained using the conventional scheme. The red dashed line indicates a locus satisfying the condition $r = S_{\tau_m=0}/S_{cw}$. The variation under this locus is higher than that under $r = 0.00$; however, the difference is no more than 10%. Therefore, a large reduction in the variance can be expected under this locus.

Figure 4(b) shows the frequency variation of the observation time. The variation when the conventional scheme is used increases significantly with increasing light intensity. This is because high light intensity leads not only to a high pumping rate but also to a large frequency difference between $S_{cw}$ and $S_{\tau_m=0}$. The variation in the observation time is dramatically suppressed under the condition $r = S_{\tau_m=0}/S_{cw}$. Because the contour interval is shorter for higher light intensities, the two-step pulse observation method provides a particular advantage under high light intensity excitation.

Figure 5 shows the experimental results for the light shift as a function of the light intensity for various $r$ values. These results were obtained using a Cs D1 line vertical-cavity surface-emitting laser (VCSEL) and a Cs vapor cell. The light intensity was modulated by an acousto-optical modulator (AOM), and the two-step pulse shape was formed using the control voltage of the AOM. Because the absolute resonance signal decreases with decreasing $r$, the density of the neutral density filter that was located in front of the photodetector was adjusted so that the resonance signal with the two-step pulse observation method was equal to that obtained using the conventional RR scheme. In this experiment, $r$ was set to 0.25 so that it did not saturate the photodetector voltage.
during pumping. The free evolution time was 800 µs, and the observation time was 10 µs. The experimental setup is described in detail in Ref. 8. The measured $\alpha_{cw}$ was 28.4 Hz/(mW/cm$^2$). The solid line is a fitting curve for the experimental data. The variation obtained using the conventional scheme was 1.62 Hz/(mW/cm$^2$). The variation obtained with two-step pulse observation ($r = 0.25$) was 0.42 Hz/(mW/cm$^2$), which is almost one-quarter of that obtained using the conventional scheme. This reduction is consistent with the calculated results.

In summary, we propose a two-step pulse observation method to enhance the frequency stability of CPT atomic clocks. The two-step pulse observation method provides a low variation in the light shift without reducing the resonance signal by reducing the steady state repumping. The calculation results show that there is an optimal value of $r$ for suppression of the frequency variation of the observation time. We also derive an equation for estimating the light shift in the RR scheme. The relative frequency differences between the results obtained using the estimation equation and those from the numerical calculations are on the order of $10^{-15}$, indicating that the estimation equation is useful for practical purposes. The measured light shift is reduced by the two-step pulse observation method, and the reduction in the light shift is reproduced well by the calculations.

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