# Effect of buffer gas ratios on the relationship between cell temperature and frequency shifts of the coherent population trapping resonance

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We studied the relationship between pressure ratio of the buffer gases (argon and neon) and the rate of coherent population trapping resonance frequency shift with cell temperature in $^{85}$Rb. We found that when the total pressure of the buffer gases varies within the range of 5–15 kPa, the frequency shift rate varies along a bell shaped curve. Every curve crossed the horizontal axis at two points that are roughly symmetrical with respect to the midpoint at 1:1. This allows us to minimize the rate of frequency shift by adjusting the pressure ratio of the buffer gases to these two points. © *2008 American Institute of Physics*. [DOI: 10.1063/1.2937407]

During the past few years, much interest has been demonstrated toward the development of the atomic clocks based on coherent population trapping (CPT).[1–5] One of the key advantages of this CPT technology is the possibility to substantially miniaturize the physical package of an atomic clock.[6] It can also reduce their consumed power. The atomic clock can be used in portable devices such as global-positioning system receivers. Energy levels of the atom are temperature sensitive. As a result, in real chip scale atomic clocks (CSAC), most of the energy is dissipated in temperature control. However, temperature drift still resents a limitation to the long-term stability of CSAC.

In a typical passive CPT atomic clock, a semiconductor laser is modulated by a local oscillator (LO). The LO is locked in the ground-state energy of the splitting frequency in alkali atoms. This modulation by the LO changes the single-mode light of the laser to a comb of optical sidebands. Two teeth of the comb (the two first-order sidebands or the carrier and one of the first-order sidebands) play the role of exciting the CPT resonance. When the frequency difference between the optical sidebands is equal to the frequency separation of the hyperfine split ground state of the atoms, the atoms are optically pumped into CPT state and no longer absorb incident light.[7] This leads to a reduction of fluorescence intensity from the vapor and an increase of the amount of light passing through the atoms. In a simple implementation of a CPT atomic clock, the frequency of the LO used to modulate the laser is stabilized onto the peak of the CPT resonance by a feedback loop. The frequency of the LO is used as a reference standard for output frequency.

Buffer gas is needed in the resonance cell because it reduces the line width of CPT resonance.[1] The buffer gas prevents wall relaxation by slowing down the diffusion of atoms. It increases the transit time of the atoms across the laser beam, and it prevents Doppler broadening in the typical implementation of CPT clock, in which a traveling wave is detected. In addition, at high alkali atom densities, the buffer gas has the effect of quenching the fluorescence. However, the atomic hyperfine frequency may be shifted as a result of collision between buffer gas atoms and alkali atoms. This is because the collisions perturbed the wave function of the rubidium atoms, which influences the hyperfine constant. Perturbations are sensitive to temperature, and although the cell is thermally isolated and its temperature controlled, residual temperature drifts still limit the long-term stability of CPT atomic clocks. Roughly linear shift rates, from several Hz/K to tens of Hz/K, were observed in rubidium.

Controlling temperature drift within an acceptable range presents a challenge and may put stringent requirements on the temperature stability of the physical package of a CPT atomic clock, especially after miniaturization. This may increase engineering-related constrains in the manufacture of CPT clocks and may require an increase of equipment size for thermally isolation.

In order to reduce the dependence of the atomic clock frequency stability on the temperature stability of the atoms, a mixture of different buffer gases is used to reduce the rate of frequency shift. The basic idea is to use two different buffer gases that produce opposite shift rates. In the case of rubidium, $Ar-N_2$ and $CH_4-N_2$ have been of particular interest because the nitrogen produces the required fluorescence quenching while the other partner is used to decrease the temperature coefficient.[8,9] Ar-Ne is another choice for rubidium clocks. In our experiment, to find the relationship between frequency shift rate and the ratio of Ar to Ne, three groups of small identical cylindrical cells with different buffer gas pressures were fabricated with mixtures of Ar and Ne. These cells were tested at varying temperatures.

Figure 1 shows a sketch of the experimental setup for measuring the frequency shift rate of CPT resonance with different buffer gases in the $^{85}$Rb cell. In this experiment, the vertical-cavity surface-emitting laser (VCSEL) beam was circularly polarized and attenuated, leaving 50 $\mu$W in a beam 3 mm in diameter at the entrance to the vapor cell. The cell is 0.8 cm long, 1 cm in diameter, and contains a mixture of natural rubidium (72% $^{85}$Rb and 28% $^{87}$Rb) with Ne and Ar. It is temperature controlled and magnetically shielded. When the VCSEL injection current is modulated, a substantial fraction of the laser power is transferred to the sidebands from the carrier band. Using the first lock-in amplifier, the VCSEL's wavelength was locked on the absorption spectrum from $5S_{1/2}$, $F=2$ to $5P_{1/2}$, $F=2$(795 nm), the carrier frequency is the same as $5S_{1/2}(F=2)\rightarrow 5P_{1/2}(F=2,3)$ transition

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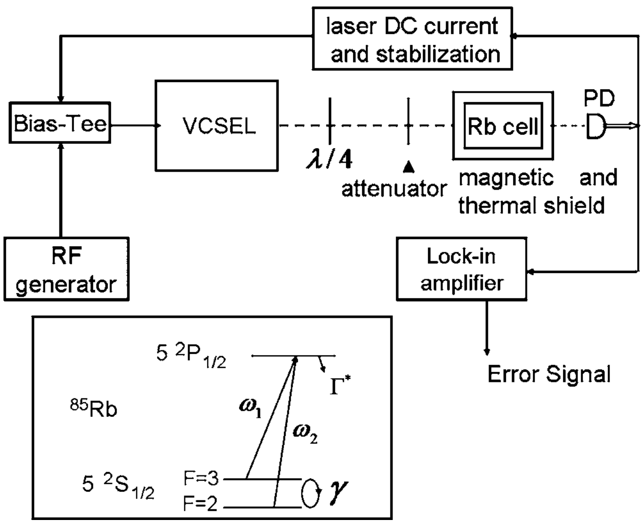


FIG. 1. Experimental setup for measuring the temperature shift rate of CPT resonance.

frequency. The first red sideband is roughly matched with the $5S_{1/2}(F=3)\rightarrow 5P_{1/2}(F=2,3)$ transition. The rf generator is frequency modulated at 190 Hz, and then the error signal (generated by the second lock-in amplifier) is monitored. The frequency of the rf generator is adjusted, and when the error signal is zero, the frequency of the rf generator is the CPT resonance frequency. The measurement was repeated for cells of different buffer gas ratios between 30 and 45 °C during which, frequency shift rates were obtained. This temperature range was chosen since the inside temperature of most running laboratory instruments is in this temperature range. To study the behavior of frequency change over a wider temperature range, we measured CPT frequency from 20 to 80 °C for two special cells (cell 1 and cell 2).

Figure 2 shows a plot of the clock frequency as a function of cell temperature for one cell (cell 1). This Rb cell contains a mixture buffer gas with Ar and Ne. The total pressure is 5 kPa and the ratio of Ar to Ne is 1:1.5. It can be seen from Fig. 2 that the relationship between frequency shift and temperature is approximately linear, with a rate of −3.8 Hz/K, between 30 and 45 °C. For this special cell, temperature instability of 1 mK leads to frequency instability in the $10^{-12}$ region. If the cell temperature varies within a 10° range, the frequency instability would be in the $10^{-8}$ region. For different cells, we can obtain different shift rates. The main purpose of our experiment is to find the possibility of reducing the shift rate by changing the buffer gas ratio.

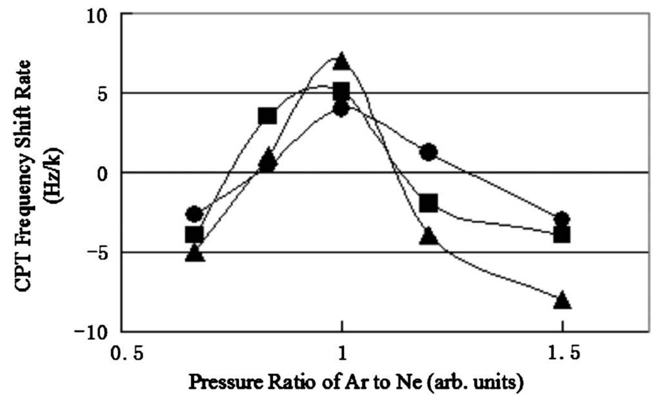


FIG. 3. Frequency shift rates for different buffer gas ratio and pressure. Circles correspond to group 1 cells with total buffer gas 50 kPa. Squares correspond to group 2 cells with total buffer gas 100 kPa. Triangles correspond to group 3 cells with total buffer gas 150 kPa.

We measured three groups of five cells each. All of them have the same size and shape. The three groups of cells have fixed total pressures of 5, 10, and 15 kPa respectively. In each group, the pressure ratio of Ar to Ne varies from 1:1.5 to 1.5:1. Figure 3 shows a comparison of shift rates among these 15 cells. It can be seen that the three curves have a similar bell shape and the frequency shift rates clearly have a strong relationship with the pressure ratio of Ar to Ne. Since the shift rate does not vary linearly with pressure ratio, the total effect of the two buffer gas is not the direct sum of the effects of each. When the pressure ratio is 1:1.2 or 1.2:1, the shift rate is close to zero. When the ratio is 1:1, the shift rate is positive, and when the ratio is 1:1.5 or 1.5:1 the shift rate is negative. From the figure, it can be inferred that there are two points on each curve where the frequency shift rate crosses the zero line. We can therefore optimize the buffer gas pressure to decrease the shift rate to approximately zero.

The lowest resulting shift rate among the 15 different cells is 0.4 Hz/K, which is shown in Fig. 4 (cell 2). In this cell, the total pressure is 5 kPa and the ratio of Ar to Ne is

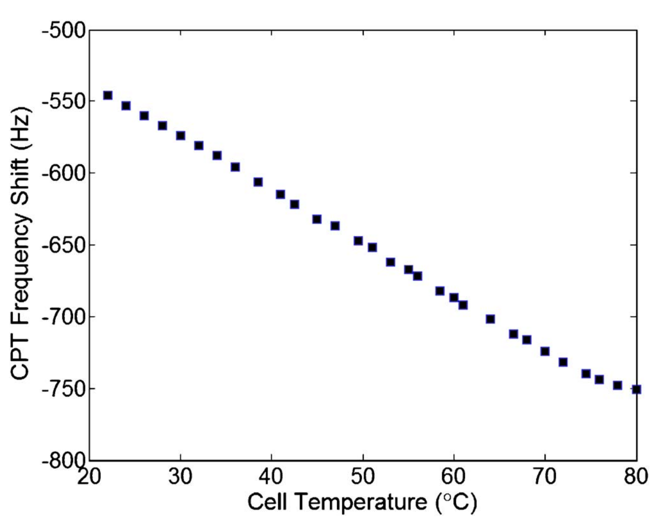


FIG. 2. (Color online) Frequency shift of CPT resonance as a function of cell temperature for cell 1.

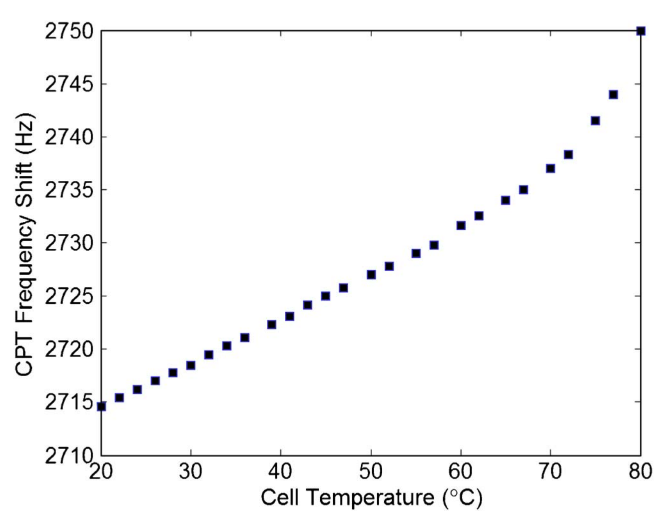


FIG. 4. (Color online) Frequency shift of CPT resonance as a function of cell temperature for cell 2.

1:1.2. On the one hand, the shift rate of this cell is opposite to that of cell 1. Since the two cells have the same characters but only different buffer gas pressure ratio, physically there is a point for the pressure ratio that the shift rate is equal to zero. On the other hand the shift rate magnitude of cell 2 decreases to about $\frac{1}{10}$ of the rate of cell 1. It means that if we use this cell, we will have the potential to improve the stability for ten times. In other words, if we want to achieve the same frequency stability through controlling the temperature, the requirement of temperature control for cell 2 will be $\frac{1}{10}$ that of cell 1. For cell 2, the temperature instability of 1 mK will cause frequency instability in the $10^{-13}$ region. Figure 4 shows that in the 20 to 60 °C range, when the cell temperature varies less than 10 °C, the frequency instability can reach $1.3 \times 10^{-9}$. Even if the cell temperature were to change from 20 to 60 °C, the frequency instability would be in the $10^{-9}$ region. When the cell temperature changes from 20 to 80 °C, the frequency instability will worsen to the $10^{-8}$ region. Compared to cell 1 (Fig. 2), we have obtained nearly a tenfold improvement in the frequency stability in the 20–60 °C range. This is done in a random buffer gas ratio test. Therefore, if we optimize the ratio carefully around the zero points, we may further reduce the temperature sensitivity of the CPT frequency. In rubidium cells, there may be diffusion and leakage of the buffer gases, which could potentially cause the pressure ratio and total pressure to change over time. This will lead to changes in stability. Therefore, it is crucial for us to maintain the buffer gases at constant pressure.

In conclusion, we have demonstrated that by optimizing buffer gas pressure and pressure ratio, it is possible to substantially reduce the temperature shift rate of CPT resonance. This may enhance the commercial feasibility of CPT clocks, especially in portable devices that may be subject to significant variations in environmental conditions. The method may also prove useful in improving the reliability of CPT atomic clocks by eliminating temperature-related frequency drifts. This idea allows its implementation in even miniature atomic devices.

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