The influence of atmospheric helium on secondary clocks

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Abstract: We report a new error source for the frequency of glass-cell based secondary time-frequency standards. That is, a frequency shift by helium collision was perceived where the helium atoms were confirmed as from the atmosphere.

1. Introduction

Glass-cell based secondary time-frequency standards are the most popular laser-assisted frequency standards, playing a key role in space voyage, high-precision research laboratories, length-standard metrology, and the time base of high-end instruments in industry. However, the reported frequency accuracies were always much worse than what was expected, though high frequency stabilities have been achieved. We report here a new error source that can influence the frequency of those secondary time standards, that is, collision shift by helium atoms from the atmosphere. Our helium-cesium (He-Cs) collision experiment performed in high vacuum chamber amazingly agree with previous cells' data when expressed as a frequency-linewidth relation. Moreover, another independent helium diffusion experiment proved that our previous measurement error came from atmospheric helium, by which we solve the puzzle raised previously [1]. The obtained slope of the frequency-linewidth relation provides a yardstick for the degree of helium contamination

2. The crisis for glass-cell based secondary clocks and our solution

By measuring the frequency of cesium atom 6S→ 8S two-photon hyperfine transition, we were aware that the transition frequency of two of the ten cells used in [2] had blue-shifted with the same amount of 46-kHz during five years as is illustrated in table 1. Therefore, we conducted several experiments to verify the error source. That is, we directly measured The helium-cesium (He-Cs) collision shift with the helium pressure all the way down to be

Table 1. Absolute frequency and linewidth (FWHM), Cs 6S-8S hyperfine transitions

Cesium dispensor in 10^{-8} Vacuum (2019)				Cell#2 (2017)		Cell #3 (2017)	
Absolute frequency (kHz, F=4-4)*	Linewidth (kHz)	267 (3)	882(9)[#]	345(2)	1085 (6)[#]	385 (2)	1238 (13)[#]
Absolute frequency (kHz, F=3-3)**	Linewidth (kHz)	344 (3)	881(18)[#]	421 (2)	1108 (7)[#]	458 (2)	1283 (12)[#]
narrowest-linewidth cell and cell #2 and cell #3 in 2012							
Absolute frequency (kHz, F=4-4)*	Linewidth (kHz)	297[†] (10)	932 (11)[#,†]	300 (10)	1020 (11)[#]	338 (10)	1150 (13)[#]

* +364,503,080,000 kHz; **+364,507,238,000 [†]narrowest-linewidth glass cell in 2012 *(15)*

[#]Lorentzian part in "Lorentzian-transit" fitting [3].

smaller than what exists in the atmosphere; we put one of the aforementioned two cells inside a vacuum chamber filled with helium gas while use the other cell for frequency comparison to simulate the atmospheric helium contamination. From the aforementioned He-Cs collision experiment, we obtained the slope of 43.35 (25) kHz/mtorr and, surprisingly, most of the other frequency-linewidth relations obtained independently from different cells fall onto the same relation line as displayed in Fig. 1 (a); from the helium-diffusion experiment and with the use of the diffusion equation shown on the bottom-right of Fig. 1 (b) [4], we conclude that an addition of 1.23 (0.16)-mTorr helium from the atmosphere had simultaneously diffused into the two cells after 5 years, and it is in an excellent agreement with our He-Cs collision experiment where the 46-kHz pressure shift is exactly corresponding to 1.06 (23) mTorr! The aforementioned "43.35 (25) kHz/mtorr" provides an important information of knowing the amount of helium pressure existed in a suspicious cesium cell. Meanwhile, the linewidth-frequency relation in Fig. 2 offers the other significant information of identifying whether the alien collider in a cesium cell is helium or not. Fig. 1 (a) includes all the cells

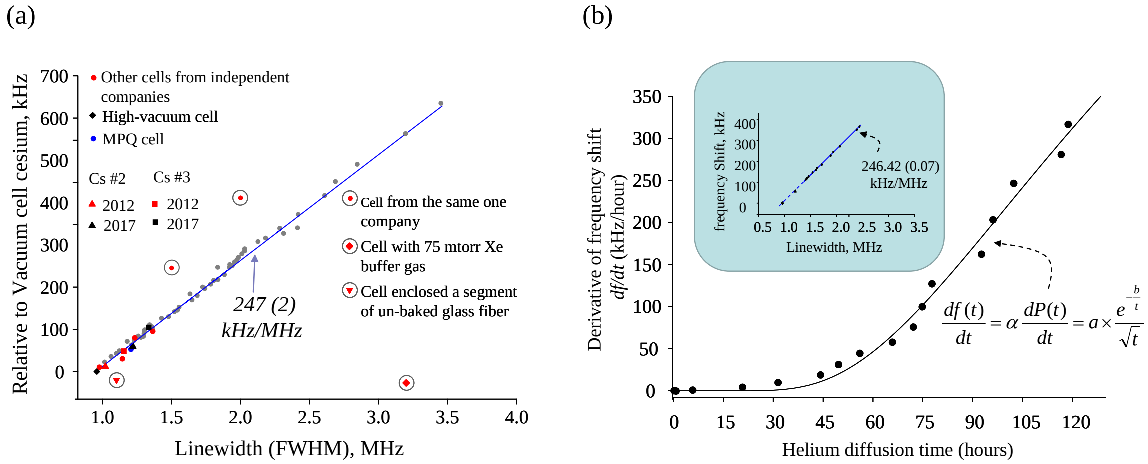

Fig. 1 (a) The linewidth- frequency relation from different experiments; gray color: data from the direct Cs-He collision experiment; red color: data taken from the ten cells mentioned in reference 15; blue color: date quoted from MPQ work in reference [5]; black color: data taken from Cs #2 and Cs #3 at 2017 and high vacuum cell at 2019; gray-circle marked data are extraordinary, see text. (b) Experimental data (black dots) taken from Cs #3 cell which was surrounded with 1.75-atm helium; the data were fitted (black line) by the "early time" diffusion formula [4] shown on bottom right, from which we are able to deduce, specifically, the helium diffusion coefficient *D* of Cs #3 cell, see text; α = 43.35 kHz/mTorr, value adopted from the He-Cs collision experiment; inset: the frequency-linewidth relation obtained in this experiment.

displayed in table 1 and we find that they all lie on the same blue relation line which is also the same linear fitting result of the gray dots. We can thus identify that the main colliders in table 1 are all helium since the data of gray dots are derived from the same He-Cs collision experiment. The amazing coincidence of the same linewidth-frequency relation in Fig. 1 (a) (except for four data marked with gray circle) provides us high confidence in explaining the puzzle raised in our previous experiment [2] and in solving the crisis of secondary standards addressed in reference [1]. Fig. 1 (b) illustrates the first derivative of the frequency shift with respect to time, by which we were able to determine the helium diffusion coefficient *D* of Cs #3. The data in Fig. 1 (b) is fitted by the so-called "early time" equation shown at its bottom-right [4]. We thus obtain the helium diffusion coefficient *D* of Cs #3 cell as *D*= 5.75 (24)$\times 10^{-9}$ cm^2/sec at 23^{0}C. This value is in a good agreement with that had been reported in reference [4] in which the value of *D* of Pyrex 7740 was determined as 7.74$\times 10^{-9}$ cm^2/sec at 27^{0}C. The value of *D* and *a* are then applied to the so-called "late time" [4] equation to extrapolate the amount of helium pressure of Cs #3 cell that had once diffused from atmosphere and we concluded that there was 1.23 (0.16) mTorr more helium pressure inside Cs #3 cell during the past five years. This value explains the mysterious same frequency shift of Cs #2 and Cs #3 in table 1, that is, 46 (5) kHz shift from the year of 2012 to the year of 2017. Note that, we already knew from the aforementioned He-Cs collision experiment that the shift of "46 kHz" results from 1.06 (23)-mTorr helium collision, which is amazingly consistent with the calculated 1.23(0.16) mTorr!

3. Conclusions

From all independent experiments reported here, we conclude that the atmospheric helium plays an important role in Cs #2 and Cs #3 contamination and most of the other cells in Figure 1 (b) bear the same suspicion. A high-vacuum chamber based spectrometer is suggested to be better candidate of secondary optical clocks, as a compact dispenser-based vacuum chamber has already invented having about 1.5-year operation time per dispenser [6]. Note that helium collisions would also influence the 6S-6P-6S two-photon transition cesium clock (CPT clock). Our results offer a frequency correction of a cesium frequency standard without bothering to use a frequency comb laser and atomic clock, by merely measuring its linewidth. In other words, the frequency-linewidth relation in the inset of Fig. 1 (b) could be used for judging the helium contamination, which is the most common alkali cell contamination as concluded in our study.

4. References

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