Stable CW laser based on low thermal expansion ceramic cavity with 4.9 mHz/s frequency drift

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Abstract: We describe a CW laser stabilized to a low thermal expansion ceramic cavity which has a lower frequency drift rate than cavities based on ultralow-expansion glass (ULE), which are widely used as optical references. Two identical optical cavities with spacers of different material, ceramic and ULE, were assembled and the optical frequencies locked to each of these cavities were compared. The optical frequency drifts of both CW lasers were measured to within a precision of $10^{-11}$ in one second over the course of one year. The ceramic cavity had a long-term frequency drift rate of 4.9 mHz/s and the ULE cavity had one of 23 mHz/s.

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1. Introduction

Ultra-stable and narrow-linewidth CW lasers are widely used in many applications, such as precision spectroscopy [1], atomic clocks [2], and tests of fundamental physics [3]. The short-term stability of the optical frequency of a CW laser is a crucial aspect for atomic clock applications, and when using ultralow-expansion glass (ULE) cavities as frequency references, it can reach $\Delta f / f \sim 10^{-15}$ level in one second [4,5]. Short-term stability is limited by the Brownian thermomechanical noise of cavity mirrors and spacers [6], and recently developed cryogenic single-crystal silicon cavities have shown to improve the short-term stability further down to $\Delta f / f \sim 10^{-16}$ level in one second [7–9].

Long-term stability is also an important aspect of CW lasers in some applications, such as the calibration of spectrographs in observatories [10]. For example, a stability of $10^{-10}$ level in one year is required to investigate Earth-like exoplanets [11]. Long-term instability is typically caused by material aging. The structure of conventional glass, like SiO₂, spontaneously changes from an unstable amorphous structure to a stable crystal one [12,13]. This material aging effect is called creep, or crystallization, and it limits the stability of ULE glass cavity stabilized CW lasers to $\Delta f / f \sim 10^{-9}$ in one year, with typical frequency drift values ranging from 20 mHz/sec [14] and 100 mHz/sec [13]. On the other hand, with cryogenic single-crystal silicon cavities, since there are no crystallization effects, the frequency drifts are lower, reaching under 0.2 mHz/sec [7], which corresponds to $\Delta f / f \sim 10^{-11}$ in one year. To date, cryogenic single-crystal silicon cavities are the most stable reference cavities, however, they are complex to develop and to operate, because their zero crossing temperature is at an ultra low temperature (124 K, 4 K or 0 K). When it is operated at 124 K, the zero crossing sensitivity of the coefficient of thermal expansion (CTE) is twenty times larger, compared to ULE cavities.

The low thermal expansion ceramic “NEXCERA” (Kurosaki Harima, CO, Japan) is another option as the spacer material of a high-finesse cavity. Hereinafter, a reference cavity based on this thermal expansion ceramic will be referred to as a “ceramic cavity”. NEXCERA is a cordierite (2MgO-2Al₂O₃-5SiO₂), which has a polycrystalline structure with a 1μm grain size and an amorphous layer between grains [15]. NEXCERA has a Young’s modulus of approximately 130 GPa and a Poisson’s ratio of approximately 0.30, which means high specific rigidity, compared to ULE that has the Young’s modulus of approximately 68 GPa.
and a Poisson’s ratio of approximately 0.17 [16]. NEXCERA does not have a porous microstructure like other ceramics, so a smooth surface with an averaged-roughness of sub-nm level and a flatness of less than \( \lambda/10 \) can be realized, which is the same as with glass materials [17]. At room temperature, NEXCERA has a zero thermal expansion state due to the negative and positive contributions of the thermal expansion coefficients of cordierite grains and the amorphous layers [18]. It has been reported that ceramic cavities have zero expansion at room temperature and that the zero crossing sensitivity of the CTE is approximately five times larger than that of ULE cavities [19]. Although, the long-term stability of a ceramic cavity has yet to be reported, it is expected to be higher than that offered by ULE cavities due to its polycrystalline structure. In terms of spacer material for optical reference cavities, low thermal expansion ceramics seem like good candidates as they enable ultra-low long-term stability at room temperature and Hz level linewidths. A precise comparison of the long-term drift of a cavity based on NEXCERA ceramic and ULE glass is thus necessary. To this end, we have developed two almost identical high-finesse cavities with different spacer materials, NEXCERA and ULE. ULE substrate mirrors are attached to the spacers by optical contact bonding. The absolute optical frequency locked to each cavity is measured with an Yb-fiber based optical frequency comb [20] over the course of 12 months. And from these experiments, we find that the long-term drift of the ceramic cavity is smaller than that of the ULE one.

This paper proceeds as follows. Section 2 is a description of the experimental setup and the properties of the CW laser stabilized to each of the two cavities under comparison. Section 3 details the long-term frequency drift measurements of the absolute frequency of the CW lasers stabilized to either of the two cavities using an optical frequency comb. Section 4 is a discussion of the experimental results. And finally, Section 5 is the summary and conclusion.

2. The CW laser stabilized to the ceramic cavity

Figure 1 is a schematic diagram of the CW laser stabilized to the ceramic cavity. The cavity spacer which has a length of 77.5-mm is made of the low thermal expansion ceramic “NEXCERA N117B”. It was mounted vertically [21] and processed into a rugby-ball shape [22]. The ceramic spacer rested on Teflon posts and a low thermal expansion ceramic ring in order to reduce radial forces caused by the thermal expansion of the vacuum chamber. Ideally, the mirror substrate and the spacer are both made out of the same material, however, NEXCERA is not transparent at 1 \( \mu \)m wavelength, so ULE glass was used instead as the mirror substrate. Both ULE substrate mirrors are concave with a 500 mm radius of curvature and they have SiO\(_2\) and Ta\(_2\)O\(_5\) dielectric multi-layer coatings. Using the cavity ring down method, the finesse of the ceramic cavity was measured to be 297,000 for the TEM\(_{00}\) mode and at a wavelength of 1064 nm.

The ceramic cavity was placed in an aluminum alloy heat shield and a stainless steel vacuum chamber. The vacuum chamber was evacuated at a pressure of 1 \( \times \) 10\(^{-5}\) Pa using a 20 l/s ion pump. To couple light, AR coated windows were fitted to the boreholes along the optical axis of both the chamber and the heat shield using Torr seal epoxy. These windows had a 4 degree tilt to avoid etalon effects. The vacuum chamber which included the ceramic cavity was mounted on a passive vibration isolation platform (Minus K technology, BM-4) and enclosed in a soundproof box built in-house, which consisted of 40-mm thick polyurethane foam and 3-mm thick aluminum plates. The ceramic cavity temperature was stabilized to the zero-crossing temperature (26.5 °C) to within a precision of 1 mK by heat radiation from the vacuum chamber through PID regulated heaters and water-cooled air.

The CW seed source was an external cavity diode laser (ECDL) built in-house which consisted of an AR-coated diode laser with an external cavity in a Littrow configuration [23]. The ECDL frequency was approximately 281.7054 THz, which corresponds to a wavelength of 1064 nm. The ECDL radiation was guided to the ceramic cavity by a polarization
maintaining single mode fiber (PMF). Locking the ECDL to the ceramic cavity resonance was realized with the Pound-Drever-Hall (PDH) method [24] using an electro-optical modulator (EOM) driven at 15 MHz. Feedback was carried out through the diode laser current with a fast bandwidth (10 MHz) PID controller and through a grating mounted on a piezoelectric transducer (PZT) in the external cavity with a slow bandwidth (10 Hz) PI circuit.

To compare to the ceramic cavity, a ULE cavity (Advanced Thin Films, CO, USA) was developed in tandem. The ceramic and ULE cavities were identical apart from the spacer material, which had a length of 77.5-mm. The rugby-ball-shaped ULE spacer was vertically mounted and the ULE mirrors used were concave-concave pairs that had radii of curvature of 500 mm. The finesse of the ULE cavity was 274,000, as measured by the cavity ring down method for the TEM\(_{00}\) mode at a wavelength of 1064 nm. The zero crossing temperature of the ULE cavity was approximately 26.5 °C and it was maintained in the same way as with the ceramic cavity. The ECDL optical frequency locked to the ULE cavity was approximately 281.7059 THz, corresponding to a wavelength of 1064 nm.

The optical beat note between the two CW lasers, each one stabilized to either the ceramic or the ULE cavity, was measured in order to study the thermal expansion in addition to the short-term stability. The beat note signal was detected with a photo diode and down-converted to 10 MHz. All measurement equipment was locked to a GPS-linked Rb clock (Stanford Research Systems, FS725), and the uncertainty of the measurement was limited by the stability of the Rb clock which was $\Delta f / f \sim 10^{-11}$ in one second.

The graph of Fig. 2(a) shows the measured temperature dependence of the beat note frequencies and parabolic curves fitted to them. The red (blue) data points represent the beat note frequency shift when the temperature of the ceramic (ULE) cavity was changed. The optical frequency was measured a day after changing the temperature of the chamber, when it was believed the cavity had reached a state of equilibrium. A frequency counter (Agilent 53132A) was used to measure the beat note frequency at a gate time of 1 sec, and means and standard deviations were averaged over a continuous period of 1000 sec. The resonant frequency $f$ dependence of the cavity temperature $T$ follows the relation $\Delta ff = 1/2 \frac{d\alpha}{dT} (T - T_0)^2$ ($\Delta f$: Resonant frequency shift resulting from the change in temperature, $\alpha$: Coefficient of thermal expansion, $T_0$: Zero crossing temperature). In this measurement, the resonant frequency shift $\Delta f$ corresponded to the beat frequency shift, and the resonant frequency corresponded to the absolute frequency of the CW laser locked to the reference cavity. In Fig. 2(a), parabolic curves following the relation $\Delta f = f \times 1/2 \frac{d\alpha}{dT} (T - T_0)^2$ were fitted to the measured data points. These fits show that the zero crossing temperature of both cavities is coincidentally the same, approximately 26.5 °C. Figure 2(b) is a plot of the coefficients of thermal expansion (CTE) of both cavities. These are obtained by taking the derivative of the beat note frequency shift of Fig. 2(a) and normalized against the absolute frequency (281.7054 THz for the ceramic cavity and 281.7059 THz for the ULE cavity), corresponding to $d(\Delta ff)/dT = d\alpha/dT (T - T_0)$. The coefficients of thermal expansion have a linear dependence with temperature. The slope $d\alpha/dT$ corresponds to the zero crossing sensitivity of the CTE.

The zero crossing sensitivity for the ceramic cavity was $4.9 \times 10^{-9}$ K$^{-2}$, whereas the one for the ULE cavity was $1.1 \times 10^{-9}$ K$^{-2}$. Therefore, the ceramic cavity is approximately five times more thermally sensitive than the ULE cavity. This result is consistent with previous reports [19]. Although the sensitivity to changes in temperature is greater for ceramic cavities, the zero crossing temperature can be maintained the same way as with ULE cavities.

Figure 3(a) is the power spectrum density and (b) is the Allan deviation curve of the beat note between the ceramic and ULE cavities. The power spectrum density was acquired with a RF spectrum analyzer (HP, 89440A) and the linewidth of the beat note was determined to be approximately 1 Hz. The Allan deviation plot was compiled from frequency counter
measurements (Agilent 53230A) acquired at a gate time of 50 ms over 1100 s and to which a linear frequency drift was subtracted from. A stability of $10^{-15}$ level at an averaging time of 10 s was found.

3. Absolute frequency measurement

The long-term stability of the ceramic- and ULE-cavity length was observed through the absolute optical frequency of each cavity-stabilized CW laser using an Yb-fiber optical frequency comb built in-house, as shown in Fig. 1, and which is described in detail in [20]. Here we only report the changes that were made to it for the purposes of the work presented in this article. The offset frequency was locked to a GPS-linked Rb frequency standard (Stanford Research Systems, FS725) and the beat note between one comb mode against the CW laser was locked to a reference frequency. The optical frequency of the stable CW laser was determined using the equation

$$f_{CW} = N f_{\text{rep}} \pm f_0 \pm f_b$$

(CW: CW laser frequency, $N$: mode number, $f_{\text{rep}}$: repetition frequency, $f_0$: offset frequency, $f_b$: beat note frequency between the $N$th comb mode and the CW laser). With the equation $f_{CW} = N f_{\text{rep}} \pm f_0 \pm f_b$ there is the uncertainty of the exact mode number ($\sim 3$) and the direction of the sign (4 possible combination of plus and minus). To unambiguously determine the absolute optical frequency, it was measured for multiple repetition frequencies. The uncertainty of these measurements was limited by the stability of the Rb clock, which was $\Delta f/f \sim 10^{-11}$ in one second and corresponded to, with respect to the optical frequency of the CW lasers, a few kHz in one second.

Figure 4 represents the absolute frequency of the ceramic- and ULE-cavity-stabilized CW lasers measured over the course of one year. The absolute frequency was measured with a frequency counter (Agilent, 53230A) set to a 1 s gate time and each data point was averaged over a time $\tau = 180$ seconds. The measurement error bars were on the order of kHz/$\tau^{1/2}$, which was smaller than the data points themselves on the plot. The absolute frequency drift of the ceramic-cavity-stabilized CW laser was 200 kHz/year; whereas the one for the ULE-cavity-stabilized CW laser was 900 kHz/year. The trend of the absolute frequency drift for the two cavity-stabilized lasers seemed to follow an exponential function, $f(t) = a + b \left( 1 - \exp(-t/c) \right)$. The R square correlation between the exponential fit and the measured absolute frequency drift of Fig. 4 was over 0.99 for both the ceramic and ULE cavities. The time constants of the exponential fits for the ceramic cavity data ($c = 779$ days) was smaller than that for the ULE cavity data ($c = 1105$ days).

Figure 5 shows the absolute frequency drift rates determined from the time differential of the exponential fits of Fig. 4. The solid lines are the time differentials and the dotted lines are extrapolations. The absolute frequency drift was approximately 8 mHz/s at the start of the measurement and it went down to 4.9 mHz/s after one year. This value was smaller than that
of the value for the ULE cavity, which was 23 mHz/sec. In 3 years from now, the absolute frequency drift rate of the ceramic-cavity-stabilized CW laser is projected to reach 1 mHz/sec ($\sim 10^{-10}$ in one year), which is lower than the value projected for the ULE-cavity-stabilized CW laser, as well as the value presently achievable by these systems.

Fig. 4. The absolute frequency drift of the CW lasers stabilized the ceramic cavity (Red) and ULE cavity (Blue) over one year. The absolute frequency was measured with a frequency counter set to a 1 s gate time and each data point was averaged over a time $\tau = 180$ s. The measurement error bars are of the order of kHz/$\tau^{1/2}$, which is smaller than the data points themselves.

Fig. 5. The absolute frequency drift rates of the ceramic cavity (red) and the ULE cavity (blue). They were deduced from the time differential of the exponential fits of Fig. 4. The solid lines are the time differentials and the dotted lines are extrapolations.

The beat note between the ceramic- and ULE-cavity-stabilized CW lasers was measured simultaneously. Figure 6 shows the difference between the absolute frequencies (orange triangle, as measured in section 3) and the direct beat note frequency (green square, as measured in section 2) between the two CW lasers. They virtually overlap and the exponential function fitted to the direct beat note frequency (green line) represents a frequency drift rate of 18.8 mHz/sec, which is equal to 23 mHz/sec (ULE cavity frequency drift rate) – 4.9 mHz/sec (ceramic cavity frequency drift rate). Therefore the subtraction of the two measured absolute frequencies reproduced the beat note frequency data well.
4. Discussion

The results of our experiments show that the drift rate of the ceramic cavity length is lower than that of the ULE cavity length. This slower drift might be owed to the polycrystalline structure of low thermal expansion ceramics, which have an amorphous layer at the cordierite grain boundaries. Such a performance would be useful for multiple applications with stringent demands on the long-term stability. One such example is in the field of astro-combs, which are used as frequency references in the precise measurement of the Doppler shifts of stars in order to obtain information about exoplanets. Ideally suited for such applications are multi-GHz frequency combs, which would need to demonstrate a stability of $10^{-10}$ level in one year to investigate Earth-like exoplanets, although their pulse energy is usually too small to be able to detect the offset frequency via the typical self-referencing $f$-$2f$ interferometer. Presently, low repetition-rate frequency combs combined to some filtering cavities are used for such purposes [10]. For example, a multi-GHz self-referencing frequency comb based on a CW laser and an EOM was recently shown to provide an extremely accurate frequency calibration against a frequency standard [25]. Nonetheless, the technique was challenging and required a complicated setup which included multiple amplification broadening and filtering stages. To overcome these limitations, we have been developing a direct multi-GHz frequency comb stabilized to a CW laser [26]. The advantage of doing so is ease of operation and scalability to higher repetition rates. While the stability is lower than that of self-referenced frequency combs, it will depend on that of the cavity the CW laser is stabilized against. At present, the long-term stability of ULE glass cavities is $10^{-9}$ in one year, which is insufficient for this purpose. The long-term stability of Si cavities is $10^{-11}$ in one year and while this is sufficient for the exploration of exoplanets, these cavities are complex to operate for non-experts. The long-term stability of the ceramic cavity presented in this work could be $10^{-10}$ in one year, which is sufficient for the illustrated purpose and the zero crossing temperature control is as simple as the one used with ULE cavities.

5. Conclusion

We have developed a CW laser stabilized to a low thermal expansion ceramic cavity which presents a lower frequency drift than ultralow-expansion glass (ULE) cavities. This cavity consisted of a low thermal expansion “NEXCERA” ceramic spacer and high reflection mirrors made of a ULE substrate, which were mounted onto the spacer by optical contact.
bonding. A ULE cavity, identical to the ceramic one, apart from the spacer material, was also developed. The two cavities, which were produced in tandem, had finesses of approximately 300,000. The zero crossing temperature was approximately 26.5 °C for both of them. Although the ceramic cavity was about five times more sensitive to thermal changes than the ULE cavity, the zero crossing temperature could be regulated with the same temperature controls used for the ULE cavity. The linewidth was approximately 1 Hz, and the Allan deviation was at $10^{-15}$ level for averaging times between 1 s and 100 s, as observed through the beat note between the CW lasers stabilized to the ceramic and ULE cavities.

We have measured the absolute frequency of the CW lasers stabilized to the ceramic and ULE cavities over the course of one year using an Yb fiber optical frequency comb locked to a GPS-linked Rb frequency clock. The frequency drift of the CW laser stabilized to the ceramic cavity was 200 kHz/year, whereas that of the ULE cavity was 900 kHz/year. The trend of the plotted drifts seemed to be in good agreement with the fitted exponential curves, and the derived time constant was smaller for the ceramic cavity than it was for the ULE one. The absolute frequency drift obtained from the time differential of the exponential fits was 4.9 mHz/s for the ceramic cavity and 23 mHz/s for the ULE one after one year of ageing. Three years from now, it is projected that the absolute frequency drift rate of the ceramic cavity stabilized CW laser may reach 1 mHz/sec ($\sim 10^{-10}$ in one year), which is lower than the ones achieved so far with ULE cavities.

Such a performance might be useful for multiple applications with stringent demands on the long-term stability, such as astro-comb calibrated spectrographs used in observatories.

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