Characterization of the long-term dimensional stability of a NEXCERA block using the optical resonator technique

Chang Jian Kwong, Michael G. Hansen, Jun Sugawara, and Stephan Schiller

1. Institut für Experimentalphysik, Heinrich-Heine-Universität Düsseldorf, Düsseldorf, Germany
2. Technical Management Department, Krosaki Harima Corporation, Kitakyushu-City 806-8586, Japan

Email: Step.Schiller@hhu.de

Abstract

NEXCERA is a machinable and highly polishable ceramic with attractive properties for use in precision instruments, in particular because its coefficient of thermal expansion exhibits a zero crossing at room temperature. We performed an accurate measurement of the long-term drift of the length of a 12 cm long NEXCERA block by using it as a spacer of a high-finesse optical cavity. At room temperature, we found a fractional length drift rate $L^{-1}d\Delta L/dt = -1.74 \times 10^{-8} \text{ yr}^{-1}$. 
I. INTRODUCTION

Materials with ultra-low thermal expansion and high long-term dimensional stability are highly desirable in the field of precision instruments and metrology. Two well-known examples are ULE (ultra-low expansion glass) from Corning, Inc. and Zerodur from Schott AG. Both ULE and Zerodur have a small coefficient of thermal expansion $\alpha$, quoted as $0 \pm 0.03 \times 10^{-6}/K$ in the range 278 K - 308 K [2], and $0 \pm 0.006 \times 10^{-6}/K$ in the range 273 K - 323 K [1], respectively. The long-term dimensional stability of materials has been the subject of many studies, e.g. [4, 9]. For ULE, values of the fractional length drift rate $L^{-1}d\Delta L/dt \approx -2.5 \times 10^{-8}$ yr$^{-1}$ have been reported [15], as well as significantly lower ones, $-0.4 \times 10^{-8}$ yr$^{-1}$ [12]. Zerodur has a relatively large dimensional instability, $-0.1 \times 10^{-6}$ to $-0.2 \times 10^{-6}$ yr$^{-1}$, [13, 15], making it less suitable for applications requiring extreme long-term stability.

A relatively recent ultra-low thermal expansion ceramic, NEXCERA, developed by Krosaki Harima (Japan), is also a material of interest in instrumentation. NEXCERA can be produced with a thermal expansion coefficient $|\alpha| < 0.03 \times 10^{-6}$/K at 23°C. The Young’s modulus, 140 GPa, is higher than that of both ULE (67.6 GPa) and Zerodur (90.3 GPa). NEXCERA’s bulk density, 2.58 g/cm$^3$, is comparable to Zerodur (2.53 g/cm$^3$) but moderately higher than ULE’s (2.21 g/cm$^3$).

The first characterization of the dimensional stability of NEXCERA was recently reported by Takahashi [14]. Length measurements of line scales made from NEXCERA showed that the ceramic has good long-term dimensional stability. The fractional length change $\delta L/L$ over a time interval of 13 months was determined as $(1.1 \pm 1.1) \times 10^{-8}$ (2$\sigma$ error) [14], which is consistent with zero length change. On fundamental grounds and for extremely demanding applications it is desirable to determine the dimensional stability of NEXCERA more precisely. This was the purpose of the present work.

An extremely sensitive method for determining the long-term dimensional stability of a material consists of manufacturing the material into a spacer for an optical cavity. One of the mode frequencies of such a cavity is then measured repeatedly against an atomic frequency standard. For ULE, such measurements have been done in many laboratories world-wide. The lowest long-term linear drift rates $L^{-1}d\Delta L/dt \approx 5 \times 10^{-17}$ s$^{-1}$ (0.15 $\times 10^{-8}$ yr$^{-1}$) have recently been found [6, 11]. Other measurements of ULE long-term linear drift include $L^{-1}d\Delta L/dt = 2.04 \times 10^{-16}$ s$^{-1}$ (0.64 $\times 10^{-8}$ yr$^{-1}$) [3] and $L^{-1}d\Delta L/dt = 1.45 \times 10^{-16}$ s$^{-1}$ (0.44 $\times 10^{-8}$ yr$^{-1}$)
A measurement of the long-term linear drift rate of a Zerodur optical resonator has shown $L^{-1}d\Delta L/dt = 3.05 \times 10^{-15} \text{s}^{-1} (9 \times 10^{-8} \text{yr}^{-1})$, measured over 2 years.

A NEXCERA optical resonator has been realised by Hosaka et. al. It consisted of a 75 mm long cylindrical NEXCERA spacer and a pair of ULE mirror substrates. The temperature at which the coefficient of thermal expansion of the resonator is zero (zero-CTE temperature), was found to be at $T_0 = 16.4 \pm 0.1 \degree\text{C}$ [7]. As an upper limit of the drift rate, $L^{-1}|d\Delta L/dt| < 1.2 \times 10^{-7} \text{yr}^{-1}$ was given. This corresponds to a drift of the optical frequency of 1 Hz/s at 1.064 µm wavelength.

In this work, we present the first accurate measurement of the long-term drift of a NEXCERA N118C optical resonator, which is found to be non-zero.

This paper is structured as follows: the experimental setup is presented in Sec. II. The characterization of zero-CTE temperature and the characterization of the long-term drift of the NEXCERA resonator are discussed in Sec. III. We draw conclusions in Sec. IV.

II. EXPERIMENTAL SETUP

The NEXCERA sample we used (sample number N118C) was sintered in December 2015 and machined in March 2016. The resonator consists of a NEXCERA spacer of biconical shape with a central rim, see Fig. 1 (a). The length is 120 mm, the conical angle is $20\degree$. The diameter at the endfaces and the rim are 30 mm and 90 mm respectively. A 10 mm diameter center bore allows the confined light to propagate in vacuum. Evacuation occurs via a 4 mm diameter transverse pumping hole.

Two ULE concave mirror substrates with 0.5 m radius of curvature, having a high reflectivity coating for 1064 nm, are optically contacted to the cavity spacer in-house. The contacting was done on 21 December 2016. The resonator linewidth was measured to be 20 kHz (FWHM) (finesse
of $62 \times 10^3$). Fig. 1(b) and 2 illustrate the mounting of the NEXCERA resonator within a custom-made vacuum chamber. The NEXCERA cavity is placed vertically on three stainless steel supports with viton cylinders (approximately 5 mm long and 1.5 mm in diameter) placed in-between the supports and the resonator. These provide thermal insulation and mechanical damping. The steel supports are fixed to a base plate. Thermoelectric elements glued onto the bottom side of the base plate, together with a thermistor for temperature measurement, allow active stabilization of the temperature by a PID controller. The resonator and base plate are placed inside a polished aluminium heat shield which provides for further thermal insulation by reflecting radiative heat from the environment.

Figure 2. Schematic diagram of the NEXCERA resonator mounted within its vacuum chamber.
Fig. 3 shows the optical setup. The frequency of a particular $\text{TEM}_{00}$ mode of the resonator is interrogated by a Nd:YAG reference laser (frequency $f_{\text{laser}}$) which is stabilized in frequency to an independent ultra-stable ULE reference cavity [5]. With a fibre splitter, the light from the reference laser is split into two arms. One arm directs the light to an erbium-doped fibre-based frequency comb referenced to a hydrogen maser, while the other arm sends the light to the NEXCERA cavity. The laser’s frequency, as measured with the frequency comb, is determined from

$$f_{\text{laser}} = n f_{\text{rep}} \pm f_{\text{beat}} \pm f_{\text{offset}},$$  \hspace{1cm} (1)$$

where $f_{\text{rep}}$ is the frequency comb repetition rate, $f_{\text{beat}}$ is the frequency of the beat note between the selected mode of the frequency comb and the Nd:YAG laser, $f_{\text{offset}}$ is the comb carrier envelope offset frequency and $n$ is the number of the frequency comb’s mode. In our apparatus, $f_{\text{rep}} \approx 250 \text{MHz}$, $f_{\text{offset}} = 20 \text{ MHz}$, $f_{\text{beat}} = 50 \text{ MHz}$, $n \approx 1.126 \times 10^6$. The sign of $f_{\text{offset}}$ is determined by a particular setting of the locking electronics. The frequency of the laser is first coarsely measured with a wavemeter and the measured value, $f_{\text{laser}, w}$ is used as a starting point for the more precise
measurement with the frequency comb. The correct sign for $f_{\text{beat}}$ in Eqn. (1) is chosen so that the measured value of $f_{\text{laser}}$ is closest to $f_{\text{laser},w}$. With $f_{\text{beat}}$ fixed at 50 MHz, the change in the frequency comb’s repetition rate due to drifts of the reference laser $f_{\text{laser}}$ can be determined with high precision. $f_{\text{laser}}$, as measured by the comb, is then averaged over the 15 s duration of each frequency scan, resulting in the average laser frequency, $f_{\text{laser,av}}$ corresponding to each frequency scan. The averaging time of 15 s is chosen because it corresponds to the duration of one frequency scan of the NEXCERA cavity as explained below. Since the drift of the frequency-stabilized Nd:YAG laser is small ($\frac{df_{\text{laser}}}{dt} = 0.055 \text{ Hz/s}$), its frequency does not change significantly during the averaging time.

The other part of the laser light that is transmitted through the NEXCERA cavity is split by a polarising beam splitter (PBS) and sent to a camera and a photodetector (PD) for measurement. A half-wave plate ($\lambda/2$) is used for maximisation of the photodetector signal. Using a waveguide electro-optic modulator (EOM), we generate two sidebands on the laser wave before the light is sent to the NEXCERA cavity. Due to frequency bandwidth limitations of the RF sources used, the RF signal driving the EOM (frequency $f_{\text{EOM}}$) is produced by mixing an RF signal having constant frequency ($f_{\text{syn}} = 607.663 \text{ MHz}$) with an RF signal (frequency $f_{\text{DDS}} \approx 60 \text{ MHz}$) from a direct digital synthesizer (DDS), $f_{\text{EOM}} = f_{\text{syn}} - f_{\text{DDS}}$. By varying $f_{\text{DDS}}$, one of the laser sidebands can be scanned across the resonator’s mode frequency. Thus, the resonator’s frequency can be determined, as well as its linewidth. The synthesizer and the DDS are both referenced to the maser.

The wave exiting from the EOM is coupled into a TEM$_{00}$ mode of the resonator. The transmitted light is split by a polarising beam splitter (PBS) and sent to a camera and a photodetector (PD) for measurement. A half-wave plate ($\lambda/2$) is used for maximization of the photodetector signal.

A computer controls the DDS and acquires the photodetector (PD) signal. The employed DDS frequency scan has a span of 100 kHz and a step size of 1 kHz with a dwell time of 150 ms. This amounts to 15 s duration per line scan. Such frequency scans are typically repeated for approximately 1 hour in the measurement of resonator’s frequency. The optical frequency measurement of the laser using the frequency comb is performed in parallel to the interrogation.

The determination of the Nd:YAG laser frequency and of the EOM drive frequency at which the resonator mode is maximally excited, $f_{\text{EOM, res}}$, allows us to determine the absolute resonator frequency $f_{\text{res}} = f_{\text{laser,av}} \pm f_{\text{EOM, res}}$. The sign is chosen according to which of the two sidebands interrogates the resonator.
III. RESULTS AND DISCUSSION

A. Transmission signal measurement

An example of a frequency scan over the resonance is shown in Fig. 4. A Lorentzian fit yields the sideband frequency for achieving resonance, $f_{\text{EOM, res}}$.

![Graph showing transmission signal measurement](image)

Figure 4. A resonator transmission signal recorded during a laser frequency scan. Blue: the transmission signal measured with PD. Red: Lorentzian fit to the data.

B. Characterization of thermal expansion

In order to achieve an accurate characterization of the resonator’s long-term frequency drift, we first determine its thermal time constant. The resonator was initially set to 20°C. After the set temperature was changed to 19°C, the resonator’s frequency was repeatedly measured at intervals of 10 minutes. The result is shown in Fig. 5. The thermal time constant is determined to be $2 \times 10^4$ s $\approx 5.6$ hours, by fitting an exponential decay curve to the data.
Figure 5. Determination of thermal time constant from the reaction of the resonator frequency to a 1 K temperature change of the set point. The EOM RF frequency change, $\Delta f_{EOM} = f_{EOM} - f_{EOM,0}$. Blue: data. Red: exponential decay fit.

Since the thermal settling follows an exponential decay, we decided to perform measurements always 8 hours after the temperature change to allow for two measurements per workday and find the zero-CTE point within a reasonable time. The resonator frequencies $f_{res}(T)$ at temperatures $T$ between 16°C and 29°C were measured at intervals of 1 K or smaller, in order to obtain a precise determination of the zero-CTE temperature. Here, measurement of only $f_{EOM}$ is sufficient because the change in the resonator’s frequency due to change in temperature is much larger than any drifts in the laser frequency. Fig. 6 shows the data. A quadratic fit to the data was performed, and the zero-CTE temperature was determined from the turning point of the fit function.

The zero-CTE temperature is found to be at $T_0 = 22.9 \pm 0.2 \degree C$. Both our measurement and the result by Hosaka et. al. [7], 16.4°C, yield values near room temperature.
Figure 6. NEXCERA resonator frequency at temperatures between 16°C and 29°C. Blue: measurement data. Red: a quadratic fit to the data.

Figure 7. Normalised resonator frequency change around its zero-CTE temperature, $T_0$. Blue: experimental data. Red: quadratic fit to data. Inset: thermal expansion coefficient, $\alpha$ as a function of temperature.

From Fig. 7 a quadratic function is fitted to the experimental data. The thermal expansion coefficient, $\alpha$, i.e. the derivative of the fitted function is plotted inset. Our CTE temperature derivative, $d\alpha(T_0)/dT = (7.58 \pm 0.08) \times 10^{-9}$ K$^{-2}$, is approximately 2 times larger than the value of Hosaka et al., $(3.86 \pm 0.03) \times 10^{-9}$ K$^{-2}$.

The NEXCERA resonator temperature was maintained at its zero-CTE temperature value throughout the characterization of its long-term frequency drift.
C. Absolute frequency measurements

For the characterization of long-term frequency drift, the cavity interrogations and Nd:YAG laser frequency measurements were performed once per weekday. Laser frequency measurements and sideband frequency $f_{EOM,\text{res}}$ were both averaged over approximately 30 minutes, and the resonator frequency $f_{\text{res}}$ was computed from these time-averaged frequencies. The frequency change was then given by the difference between the resonator frequency relative to the initial frequency $f_{\text{res}}(0)$, i.e. $\Delta f_{\text{res}}(t) = f_{\text{res}}(t) - f_{\text{res}}(0)$. The initial frequency measurement was performed on 25 April 2017, 125 days after optical contacting.

Fig. 8 shows the resonator frequency change plotted against time. The frequency measurement was carried out over a period of approximately 1608 hours, i.e. 67 days, as shown in Fig. 8. The data points, plotted in green, exhibit a temporary offset of 125 kHz which could be due to external mechanical perturbations on the experimental setup.

![Figure 8. Long-term frequency drift of the NEXCERA resonator. The red line is a linear fit to the blue data points. The green data points are ignored.](image)

From linear fit to the data, we find $d\Delta f_{\text{res}}/dt = (0.155 \pm 0.001)$ Hz/s. The fit to the data was done excluding the green data points. These rates correspond to fractional frequency drift rates $f_{\text{res}}(0)^{-1}d\Delta f_{\text{res}}/dt = (5.50 \pm 0.04) \times 10^{-16}$/s. The error is statistical $1\sigma$ standard error; systematic errors due to the frequency measurement technique are negligible.
IV. DISCUSSION AND CONCLUSION

We have observed, and precisely measured, the drift of the frequency of a resonator comprising a NEXCERA spacer and a pair of high finesse ULE mirrors. We have found a nonzero drift rate. If we neglect a possible contribution of the optical contacts between the ULE mirror substrates and the NEXCERA spacer, we can assign this frequency drift to an equal but opposite length drift rate

\[ L^{-1} \frac{dL}{dt} = -f_{\text{res}}(0)^{-1} \frac{df_{\text{res}}}{dt}. \]

Thus, we find a length contraction,

\[ L^{-1} \frac{d\Delta L}{dt} = (-1.74 \pm 0.01) \times 10^{-8} \text{ yr}^{-1} \text{ (interval II)}. \]

This rate is of the same sign and comparable in magnitude to that of ULE resonators. Indeed, the drift rate of our own ULE-resonator stabilized Nd:YAG laser (see Fig. 3) amounted to

\[ f_{\text{laser}}(0)^{-1} \frac{df_{\text{laser}}}{dt} = (0.625 \pm 0.003) \times 10^{-8} \text{ yr}^{-1}. \]

The error of our NEXCERA drift rate measurement is approximately \(0.419 \times 10^{-8} \text{ yr}^{-1}\), 50 times lower compared to the measurement in ref. [14]. To ensure that the long-term frequency drift measurement was not affected by the quality of the optical contacts, we tested them visually and mechanically after completion of the measurements. No conspicuous features were found.

Upon the completion of this work, an independent study on NEXCERA N117B has been published [8], where a significantly lower drift, 4.9 mHz/s was measured at the wavelength of 1064 nm. The NEXCERA N118C, used in this work, and the N117B have similar chemical compositions and microstructures but are sintered in different atmospheres, i.e. the N117B is sintered in Argon atmosphere while N118C is sintered in air. In view of the differences in long-term drift between the materials, it is of interest to characterize more samples of these materials, in order to determine whether the observed drift rate difference is indeed a reproducible property.

Despite the difference in the measurement results of the long-term drifts for optical resonators made from different type of NEXCERA ceramic, NEXCERA, nevertheless, remains a promising material for applications in the field of laser frequency stabilization. For such applications, first proposed by Hosaka et al. [7], NEXCERA has the potential advantages of larger ratio of Young’s modulus to density, hence potentially lower acceleration sensitivity than ULE. Also, it can be produced in larger sizes (up to 1 m), potentially opening an avenue to lower thermal noise by increasing the resonator’s length. The larger CTE temperature derivative compared to ULE can be compensated by advanced passive and active temperature stabilization.

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