Chasing the thermodynamical noise limit in whispering-gallery-mode resonators for ultrastable laser frequency stabilization

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Ultrastable high-spectral-purity lasers have served as the cornerstone behind optical atomic clocks, quantum measurements, precision optical microwave generation, high-resolution optical spectroscopy, and sensing. Hertz-level lasers stabilized to high-finesse Fabry-Pérot cavities are typically used for these studies, which are large and fragile and remain laboratory instruments. There is a clear demand for rugged miniaturized lasers with stabilities comparable to those of bulk lasers. Over the past decade, ultrahigh-⁹ optical whispering-gallery-mode resonators have served as a platform for low-noise microlasers but have not yet reached the stabilities defined by their fundamental noise. Here, we show the noise characteristics of whispering-gallery-mode resonators and demonstrate a resonator-stabilized laser at this limit by compensating the intrinsic thermal expansion, allowing a sub-25 Hz linewidth and a 32 Hz Allan deviation. We also reveal the environmental sensitivities of the resonator at the thermodynamical noise limit and long-term frequency drifts governed by random-walk-noise statistics.
High-precision optical frequency metrology, spectroscopy\(^1\), atomic clocks\(^2\)-\(^3\), optical interferometry\(^4\), ultralow phase noise microwave generation\(^5\), and light detection and ranging\(^6\) benefit from stable and spectrally pure laser oscillators. Such low-frequency noise oscillators can be achieved by stabilizing laser oscillators to a high-quality factor cavity resonance (\(Q = \nu / \Delta \nu\), where \(\nu\) is the resonance frequency and \(\Delta \nu\) is the full-width at half maximum of resonance). When the signal-to-noise ratio (SNR) of the detected laser signal is high enough in the measurement bandwidth (BW), the frequency stability can also be improved, which is scaled by \((Q \times SNR_{BW})^{-1}\). The benchmark ultrahigh-Q resonances in nature are atomic transitions. The precise transitions of trapped atoms have been utilized for improving stability and frequency noise of both microwave and optical oscillators. For instance, the cesium hyperfine transition is the core building block of the well-developed microwave atomic clock and the optical transitions of trapped neutral and ionic atoms are used for modern optical atomic clocks. Although a laser with frequency instability of \(6 \times 10^{-16}\) for 2–8 s integration time has been demonstrated in a cryostat via spectral-hole burning written in the \(\text{Eu}^{3+}\cdot\text{Y}_2\text{SiO}_5\) absorption spectrum\(^7\), making small atomic traps is demanding such that the development of solid-state compact optical references is of immense interest. Laser oscillators with high-finesse optical Fabry-Pérot (FP) cavities have been demonstrated with sub-Hz linewidths and fractional frequency instabilities at the \(10^{-15}\) levels in 1–10 s integration time, which is the noise limit imposed by the unavoidable thermal motion of the cavity’s reflection multilayer coating, by either using ultra-low-expansion material in vacuum or temperature cooling to operate the cavity at the zero thermal expansion point in a cryostat\(^10\)-\(^15\). However, direct miniaturization of such FP mirror cavities to a macroscale is challenging due to the quality of the cavity mirrors such that their applications have largely remained in the laboratory environment.

Over the last decade, ultrahigh-Q whispering-gallery-mode (WGM) resonators\(^16\), \(^17\) have been implemented for developing low-noise microlasers and microcavities\(^18\)-\(^21\), which show the broad transparent window and ultrahigh-Q resonances without sophisticated dielectric mirror coatings and are tolerant to mechanical noise. Since the early studies on the fundamental thermal fluctuations in microspheres by\(^22\), theoretical predictions indicate that the thermodynamically bounded frequency instability of the WGM resonators can be better than \(10^{-13}\) in 1 s integration time\(^23\), \(^24\) if the proper material and stabilization technique are selected. However, in spite of the tremendous progress, the existing WGM resonators still experience large frequency instabilities and long-term frequency drifts\(^25\)-\(^28\) hindering their use in precision metrology and timing applications.

Here, we show the noise characteristics of a WGM resonator at the fundamental thermodynamical noise limit. We employ a thermal-compensation design for reducing the thermal sensitivity of a conventional WGM resonator and reveal the residual environmental sensitivities of the compensated resonator. Subsequently, we suppress the environmental perturbations using an evacuated rigid enclosure and demonstrate that a laser stabilized to the thermal-compensation WGM resonator, working as an optical probe, shows a spectral linewidth of \(<\!25\) Hz and a fractional frequency instability of \(1.67 \times 10^{-13}\) (5.0 \(\times\) \(10^{-12}\)) on the 191 THz carrier at 0.1 (1) s integration time, which is the best among the WGM resonators of the given size and morphology without a stringent ambient temperature control. Furthermore, we confirm that the centre-shifted random walk noise statistics, imposed by the correlation between the WGM resonator temperature change and the ambient temperature or pressure variation, triggers the long-term frequency instability and monotonic frequency drift of the laser stabilized to the resonator at the thermodynamical noise limit.

**Results**

**Thermal-compensation ultrahigh-Q WGM resonator.** The thermal-expansion coefficient of the crystalline MgF\(_2\) is \(<9\) ppm K\(^{-1}\), that is a large value compared with the conventional reference FP cavities possessing \(<0.1\) ppm K\(^{-1}\). To reduce the thermal sensitivity causing the thermo-mechanical fluctuations via expansion of the resonator length, we compensate the thermal sensitivity of a thin MgF\(_2\) WGM resonator. The thermal compensation, sandwiching a WGM resonator with laminated Zerodur as illustrated in the inset of Fig. 1a, is guided by numerical simulations and predicts the significant reduction of the thermal sensitivity (Supplementary Note 1 and Supplementary Fig. 1). The design is applied to a MgF\(_2\) WGM resonator with a 6.9 mm diameter and 100 (25) \(\mu\)m resonator (rim) thickness. The thickness of the Zerodur layer (up and down) used for the MgF\(_2\) WGM resonator is 500 \(\mu\m\). To find the degree of thermal compensation, we take a pair of WGM resonators and mount them on the same temperature-stabilized platform. While two low-noise continuous wave (cw) lasers are locked to the WGM resonances, respectively,
The temperature of one of the WGM resonators is slowly changed and the relative frequency shift of the beatnote between the two stabilized lasers is measured. We assume that all slow changes in the beat frequency are attributed to the thermal expansion of the WGM resonator because the thermorefractive coefficient of crystalline MgF₂ is significantly smaller than the thermal expansion coefficient. The measurement in Fig. 1a shows ~7 times improvement compared with a conventional MgF₂ WGM resonator. Although the enhancement factor is smaller than the value predicted by numerical simulations, attributed to the residual thermal expansion due to the imperfection in the device fabrication, the measurement confirms the validity of our thermal-compensation design. The compensated WGM resonator is packaged into a small form factor prism coupler using a polarization-maintaining single mode fiber with a firmly mounted output tip. The temperature of the WGM resonator is stabilized at 301.2 K by a proportional-integral-derivative (PID) feedback control using the thermoelectric cooler. We then measure the thermal sensitivity again by measuring the beat frequency between a cw laser stabilized to the WGM resonator and a cw laser referenced to the ultrastable FP cavity (Stable Laser Systems) possessing 1 Hz linewidth and 0.1 Hz s⁻¹ drift-rate while the set-temperature of the PID control changes, resulting in 3 ppm K⁻¹. From the measured thermal sensitivity, we calculate the noise limits imposed by the thermo refractive and thermal-expansion sensitivities of the resonator (Supplementary Note 2), which are the two dominant thermal noise sources for WGM resonators, as illustrated in Fig. 1b. After the thermal compensation, the thermal-expansion noise limit is lower than the thermorefractive noise limit near the carrier frequency as shown in Fig. 1b and Supplementary Fig. 2) such that the thermorefractive noise-limited fluctuations can be unveiled. The WGM resonator has the unloaded resonance BW of 26 kHz and the loaded resonator Q is characterized with 4 μs ring-down time corresponding to 2.4 × 10⁹ at 191 THz carrier (Supplementary Note 2 and Supplementary Fig. 2).

Resonant frequency shift due to ambient perturbations. The mechanism behind the resonant frequency shift of a WGM resonator by the ambient perturbation is the interaction between the evanescent wave of the WGM resonator and the air refractive index change caused by temperature and pressure variations. The temperature and pressure are, in principle, coupled quantities connected by the ideal gas law (PV = nRT) in a rigid box, where P is the pressure, V is the volume, n is the number of moles, R is the ideal gas constant, and T is the absolute temperature. Therefore, the ambient temperature stability required for achieving the thermodynamical noise limited fluctuation can be derived from the pressure stability measurement. For this measurement we place the thermal-compensation WGM resonator in a rigid vacuum chamber on vibration isolation pads. A 3 kHz laser operating at 191 THz is stabilized to the compensated WGM resonator using the Pound-Drever-Hall (PDH) locking technique that provides sufficient technical noise suppression and thus enforces the laser frequency to chase the resonance (Methods; Supplementary Note 3 and Supplementary Fig. 3). The resonant frequency shifts induced by pressure changes are measured by counting the beat frequency between the stabilized laser and the FP reference laser at every second. Figure 2a shows the laser frequency shift when the pressure slowly increases (ΔP = 45 mPa s⁻¹) in the vacuum chamber from P₀ = 17 and 25 Pa, respectively. At the given pressure P₀ and the change (ΔP), we measure 13 (9) kHz with P₀ = 17 (25) Pa for a transverse magnetic (TM) mode input. Here, we define that the TM mode of our WGM resonator has the electric field distribution primarily in the radial direction. We also measure the resonant frequency shift at the different pressure increment. While the pressure in the vacuum chamber is increased from 17 Pa with ΔP = 130 mPa s⁻¹, we measure the frequency shift of 36 kHz (Fig. 2b), which shows approximately a linear relationship between the frequency shift and the speed of pressure change. To confirm our measurements, we quantify the impact of the refractive index change of the surrounding air medium on the WGM resonant frequency shift for TM and transverse electric (TE) modes using equations derived from the first order perturbation theory in ref. 30.

\[
\Delta f_{\text{TM}} = -\frac{\Delta n_{\text{air}}}{(n_{\text{TM}} - 1)^{1/2}} \frac{\lambda}{2 \pi r},
\]

(1)

\[
\Delta f_{\text{TE}} = -\frac{\Delta n_{\text{air}}}{(n_{\text{TE}} - 1)^{1/2}} \frac{2 - 1}{2} \frac{\lambda}{2 \pi r},
\]

(2)

\[
\Delta n_{\text{air}} = n_0 |\Delta P/P_0|,
\]

(3)

*Fig. 2* Pressure dependence of the WGM resonance frequency shift. **a** The pressure in the vacuum chamber is slowly increased from 17 Pa (olive) and 25 Pa (red) with 45 mPa s⁻¹. The frequency shift is measured by counting the beat frequency at every second while the laser is stabilized to the WGM resonator by Pound-Dreher-Hall locking. To confirm the frequency shift by the pressure change, the theoretically estimated lines from Eq. (1) for TM mode (dashed line) and Eq. (2) for TE mode (dotted line) are plotted together. The measurement values agree with the theoretical model within the 10% error bars agree with the theoretical model. For comparison, the 10% error zones of the theoretical values are added (gray). **b** The pressure in the vacuum chamber is increased from 17 Pa with 45 mPa s⁻¹ (olive) and 130 mPa s⁻¹ (navy), respectively. The measurements also agree with the theory within the 10% error zone of the theoretical values.
where $r$ is the radius of the WGM resonator, $n_{r e f}$ and $n_{r e f, T}$ are the refractive indices of the resonator host material, $\lambda$ is the wavelength, $\Delta n_{r e f}$ is the change of the refractive index of air and $n_0$ is the residual refractive index of air given by the air itself, that is $\sim 3 \times 10^{-4}$, and therefore, $\Delta n_{r e f} \approx 7.94 \times 10^{-7}$ in these measurements. The calculated results from Eq. 1 for $P_0 = 17 \ (25) \ Pa$ are $\Delta \nu_{T M} = 13.3 \ (9.1) \ kHz \ at \ \Delta P = 45 \ mPa \ s^{-1}$, which agrees well with our measured values of $13 (9) \ kHz$ for a TM mode input as illustrated in Fig. 2a. The theoretical frequency shift from thermorefractive noise of our WGM resonator is estimated by $\Delta \nu_{W G M} \approx 3 \times 10^{-15} \ Hz \ s^{-1}$. The calculated results from Eq. 1 for $\Delta \nu_{W G M} \approx 3 \times 10^{-15} \ Hz \ s^{-1}$ are also illustrated for comparison. The thermorefractive noise shows a $f^{1.5}$ frequency dependence (magenta dashed line) predicted by our theoretical model.

**Frequency noise spectrum of the thermal-compensation WGM resonator.** To minimize the impact of the technical noise from our laboratory environment, the vacuum chamber is evacuated to the pressure of $1.33 \ mPa$ but the chamber temperature is not controlled (see also Supplementary Fig. 4). Figure 3a shows the comparative frequency noise power spectral density (FNPSD) curves of the laser stabilized to the thermal-compensation WGM resonator. For comparison, the theoretically estimated thermodynamical noise limit of the WGM resonator is also plotted. The FNPSD of the free-running laser is shown by the black curve and the noise is substantially suppressed ($30 \ dB$ at $10 \ Hz$) as shown by the olive curve when the laser is stabilized to the WGM resonator. Below $30 \ Hz$ offset frequency, the stabilized laser FNPSD curve falls off as $f^{-1.5}$ implying the thermorefractive noise limit of many thermal modes of the WGM resonator predicted by Matsko et al. (Supplementary Note 4). From $30$ to $100 \ Hz$, the FNPSD curve falls off with $f^{-1}$ implying the impact of flicker noise caused by the residual laser noise and electronic device noise. Two strong peaks originate from $60 \ Hz$ harmonics of the electrical power-line noise. The rising frequency noise above $1 \ kHz$ in the olive curve is due to the poles in the active feedback loops.

The highest spectral purity laser is achieved among the WGM resonators of the given size and morphology via the thermal compensation. The integral linewidth is evaluated from the FNPSD and turns out to be $119 \ Hz$ but when the two $60 \ Hz$ harmonic peaks are removed, it is $<25 \ Hz$ (Supplementary Note 5). To further support our linewidth estimations, we measure the beat signal linewidth (Fig. 3b) in a spectrum analyzer with a resolution BW of $47 \ Hz$ and sweep time of $40 \ ms$ over the $5 \ kHz$ span. Due to the slow frequency drift, the measured line shape shows the asymmetry in the wings but the peak center is nearly symmetric and can be fitted with a Lorentzian lineshape. The resulting full-width at half-maximum linewidth is approximately $100 \ Hz$, matched with the FNPSD measurement including the $60 \ Hz$ noise harmonic peaks.

**Frequency stability of the thermal-compensation WGM resonator.** The resonant frequency instability of the compensated WGM resonator is analyzed by its Allan deviation. We use the FNPSD measurements to evaluate Allan deviations below $0.1 \ s$ averaging time because the frequency error of our counter (Agilent...
thermorefractive-noise limit (blue curve) excluding the random walk noise statistics. The statistically estimated Allan deviation at 0.1 s averaging time has a mean value of 95 Hz and a standard deviation of 68 Hz. To understand this deviation, we analyze the mean and standard deviation of 74 FNPSD measurement traces in the low Fourier frequency regime (1–40 Hz) shown in Fig. 4b. The red line is the mean value and the cyan area is the connected standard deviations along the offset frequency. Both mean frequency noise and standard deviation are diminished along the offset frequency until 20 Hz and reaches the thermorefractive noise limit. However, the residual thermo-mechanical fluctuations still exist near the carrier and change in each measurement causing the standard deviation in Allan deviation measurements. This measurement shows that the WGM resonator has relatively high noise sensitivity at low Fourier offset frequencies and, therefore, further reduction of the thermal sensitivity via the thermal compensation is desirable to reduce the measurement uncertainty.

Random walk noise distribution and long-term frequency instability. The beat frequency at 1 s averaging time is recorded by a frequency counter and the Allan deviations at the longer averaging time are calculated by averaging the 1 s measurement data sets. To check the reproducibility of the stability measurements, we take 10 measurement sets at 1 s averaging time for ~10 min each and analyze them statistically as illustrated in Fig. 4a. The Allan deviations (squares in red) along the averaging time start to deviate from the $n^{0.5}$ curve, which implies a frequency drift. To understand this behavior, we record the temperature of the 10 kΩ thermistor sensor used for detecting the WGM resonator temperature for two and a half hours while the laser is stabilized to the WGM resonator (Fig. 5a). The temperature data is statistically analyzed as shown in Fig. 5b. Due to the digitization of the measured temperature with 10 kΩ thermistor, the resolution is limited such that we can only measure the upper and lower temperature bounds. The inset of Fig. 5a illustrates that the laser frequency shift is $<2$ MHz during this measurement period and, therefore, the WGM resonator temperature is actually stabilized with $<10$ mK instability when the measured thermal-expansion sensitivity of 3 ppm K$^{-1}$ is considered. To illuminate the monotonic frequency drift, we apply the random walk and binomial distribution for 30 sets of the number of upper ($n_u$) and lower bound ($n_d$) temperature data points with the number of samples ($N$) from the continuously measured temperature data set (Supplementary Note 6 and Supplementary Table 1). The different numbers of $N$ are chosen and the average positions ($\bar{m}$) and the standard deviations $(\Delta m)^2$ are calculated for each case. Then we calculate the probability distribution $(P_N(m))$ defined by

$$P_N(m) = \frac{N!}{(N+m)/2!(N-m)/2!} p^{(N+m)/2} q^{(N-m)/2},$$

where $m = n_u - n_d = 2n_u - N$, $p$ ($q$) is the probability that the measured temperature point is at the upper (lower) bound, respectively, and they satisfy the relation, $p + q = 1$. From the measurement data, we deduce the values of $p$ and $q$ and they are 0.45 and 0.55, respectively. Figure 5b shows the probability distribution for $N=100$, $P_{N=100}(m)$. The average position, $\bar{m}$ is not at the centre but at $-10.05$ and the standard deviation, $(\Delta m)^2$ is 9.97 that is approximately $\sqrt{N}$ demonstrating that this measurement follows the random walk distribution. By increasing $N$, the average position shifts monotonically far away from the centre in Fig. 5b inset implying that the measured temperature is monotonically shifted to one of the temperature bounds along the

Figure 4 Fractional frequency instability measurements. a Measured fractional frequency instabilities (FI) of the beat frequency between the Fabry–Pérot (FP) cavity reference laser and the free-running laser (squares in black), and the beat frequency between the reference laser and the laser stabilized to the WGM resonator without evacuation (squares in olive), respectively. The red squares and bars are the mean ($\mu = n^{-1} \sum_{i=1}^{n} F_i$) and standard deviation ($\sigma = (n^{-1} \sum_{i=1}^{n} (F_i - \mu)^2)^{0.5}$) of the frequency instability of the laser stabilized to the resonator calculated from 10 sets of frequency counting data at 1 s averaging time, respectively, measured with a frequency counter where $n$ is the number data set. The red circles and bars are $\mu$ and $\sigma$ of the frequency instability of the laser stabilized to the resonator derived from the 74 frequency noise power spectral density (FNPSD) measurement traces. The magenta area represents the expected Allan deviation bound imposed by random walk frequency noise. b The noise statistics of 74 FNPSD traces. The red curve is the mean value and the cyan area is the connected standard deviations. The blue line is the thermorefractive noise limit. Both mean frequency noise and standard deviation are reduced along the offset frequency and the frequency noise reaches the thermorefractive noise limit at 6 Hz. However, the thermo-mechanical noise, induced by the residual thermal expansion, is still concentrated near the carrier causing the standard deviation of the Allan deviation measurements in a.
increasing integration time, leading to the frequency drift. This frequency shift could be mitigated by improving the resolution of temperature sensing that is currently limited by the 10 kΩ thermistor sensor, because the narrower bound of the measured temperature reduces the range of the frequency drift. This could be realized by implementing the dual-mode temperature compensation technique to the WGM resonator allowing a detection sensitivity of 100 nK. We also note that the monotonic ambient temperature change inferring that the ambient temperature change triggers the feedback to compensate the WGM resonator temperature accordingly. Therefore, an ambient temperature-controlled enclosure might be necessary to enhance the long-term stability.

Discussion
We have shown that the impact of the surrounding medium perturbations is a major problem when the WGM resonator stability approaches the thermodynamical fluctuation limit. The ambient temperature stability to reach the thermorefractive noise limit of our thermal-compensation WGM resonator is 2.7 mK and, therefore, the environment temperature control is necessary to enhance the frequency stability. An evacuated environment provided such temperature stability in the short integration time such that the thermal-compensation WGM resonator showed the \( f^{-1.5} \) frequency noise dependence imposed by thermorefractive noise of the resonator, and allowed the laser line width of <25 Hz and the lowest Allan deviation of 32 Hz in 100 ms integration time, corresponding to the frequency instability of \( 1.67 \times 10^{-13} \) for the 191 THz carrier. The standard deviation of the Allan deviation measurements is attributed to the thermo-mechanical noise concentrated near the carrier originating from the residual thermal expansion noise, which suggests that the enhanced thermal compensation is desirable to improve the stability. In principle, it is possible to achieve an order of magnitude less thermal-expansion sensitivity, which could even more alleviate the required ambient temperature stability. In the longer integration time, the laser stabilized to the WGM resonator experiences the monotonous frequency drift due to the centre-shifted random walk probability distribution of the WGM resonator temperature, which is attributed to the limited temperature sensing accuracy and the correlation of the WGM resonator temperature change with the ambient temperature variation. Hence, we anticipate that the long-term frequency drift could be mitigated by implementing the dual-mode temperature compensation technique and by tightly controlling the environmental temperature. Finally, it is noteworthy that ultrahigh-Q WGM resonators provide enhanced nonlinearity permitting generation of optical frequency combs with low input power, and, therefore, stable microcombs excluding external references are potentially possible via a single WGM resonator, which could advance the microcomb system in size-, weight-, and power-constrained environments.

Methods
Laser stabilization to the thermal-compensation WGM resonator. The 15 mW output power from the self-injection locked laser diode is split into two paths by a 50/50 splitter after an acousto-optic modulator (AOM). One arm is used for stabilizing the laser and the other is reserved for characterizing the noise of the resonator by heterodyne-beating the stabilized laser against a 1 Hz FP cavity reference laser. The piezoelectric transducer (PZT) attached to the laser is used to control the laser frequency. The PZT can be controlled either by a digitized signal on computer interface or by an analog voltage input for frequency modulation to obtain the beat frequency within the photodetector BW (New Focus model 1611). The beatnote is usually generated between 1 and 1.5 GHz and is down-mixed to 50–100 MHz for counting the beat frequency. We place an AOM before the WGM resonator, which assists the frequency stabilization and advances noise suppression in the acoustic offset frequency regime by extending the feedback BW (Supplementary Note 3). We apply the PDH locking technique to stabilize the laser to the WGM resonator. The laser light (1 mW) is phase-modulated by a fiber-coupled electro-optic modulator (5.5 dB loss) and then launched into the resonator. The transmitted light is detected by an internal photodetector in the packaged aluminum box and the detected signal is demodulated with the same microwave source that phase-modulates the laser light at a double balanced frequency mixer (model:ZAD-1+), which produces an error signal. The error signal is optimized by choosing the optimum modulation frequency and the light intensity into the resonator, which are typically ~12 MHz and ~20 μW, respectively. The error signal is split into two branches and fed into commercial high-speed proportional-integral servo controllers (New Focus, LB1005). A slow servo branch acts on the laser PZT and a fast servo branch is used to further suppress acoustic and laser technical noises. The feedback signal from the fast servo controller is fed into a voltage controlled oscillator (VCO). The VCO output is amplified and then applied to the AOM, which shifts the laser frequency to control the frequency noise up to 400 kHz. The contribution of each servo loop is optimized to achieve the lowest noise level.

Data availability. The data that support the plots within this paper and other findings of this research are available from the corresponding author on reasonable request.

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Author contributions
J.L., C.W.W, A.B.M., and L.M. designed the experiment and J.L. performed the experiment and analyzed the stabilization measurements. A.A.S. designed and developed the thermal-compensation WGM resonator along with the package assembly, E.D. made the self-injection locked laser, W.L. assembled the reference WGM resonator, D.E. measured the WGM resonator thermal sensitivity and designed the laser control electronics, and V.I. designed the resonator and laser builds. A.B.M and A.A.S. provided the theory, and all authors helped in the manuscript preparation.

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