

Absolute frequency measurement of a CO₂/OsO₄ stabilized laser at 28.8 THz

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Received: 21 August 2000/Revised version: 15 January 2001/Published online: 30 March 2001 – © Springer-Verlag 2001

Abstract. A cw carbon dioxide laser operating on the 10 μm R(0)₁ transition (28.832 THz) was frequency stabilized by a servo lock to the saturated absorption dip of the Q(15) transition of ¹⁸⁸OsO₄. The laser frequency was measured with a cesium-clock-based frequency chain. In addition, the absorption line frequencies Q(14) of ¹⁸⁸OsO₄ and sQ(4,3) of ¹⁵NH₃, were measured relative to the frequency of Q(15).

PACS: 42.62.Eh; 42.62.F; 33.20.Ea

Secondary frequency standards in the infrared have played an important role in bridging the frequency gap between the cesium-clock microwave standard and optical frequency standards [1, 2]. Although, with the recent arrival of the femto-second laser optical-comb generator, this frequency gap can potentially be spanned in a single step [3], an infrared frequency standard at 10 μm might still be more convenient to use in some applications. In our laboratory, the frequency gap between the HeNe laser standard at 474 THz (633 nm) [4] and the strontium ion frequency standard at 445 THz (674 nm) [5], was bridged by a CO₂ laser frequency at 28.8 THz. This was achieved via difference frequency generation of the 474-THz HeNe and 28.8-THz CO₂ laser radiations in a AgGaS₂ crystal which synthesized 445-THz light [6]. The 10- μm R(0)₁ line of a CO₂ laser falls at a convenient frequency to bridge the gap between the two standards, leaving only a small difference frequency of ~ 1 GHz to be counted. The absolute frequency of our strontium ion frequency standard was measured in previous work with an uncertainty of ± 200 Hz [5]. This provided the means to measure the absolute frequency of the HeNe laser standard with an uncertainty much better than ± 12 kHz, which is the uncertainty currently recommended by the Comité International des Poids et Mesures (CIPM) [4]. This in turn required the development of a frequency-stable CO₂ laser operating at R(0)₁ and the accurate measurement of its frequency.

The frequencies of most of the {00⁰1 – [10⁰0, 02⁰0]} 10- μm and 9- μm CO₂ laser transitions were measured via the classical saturated 4.3- μm CO₂ fluorescence technique [7]

and related to the absolute frequency measurement of a few of these CO₂ laser lines [8, 9]. However, the R(0)₁ line centre was not determined directly because the weak 4.3- μm fluorescence of R(0)₁ did not suffice to accurately lock the laser frequency to the line centre. The weak 4.3- μm fluorescence was confirmed by our own experiments, which also showed no measurable absorption of R(0)₁ in a 6-m-long CO₂ filled absorber cell. Therefore, a search was undertaken for a saturable absorber within the tuning range of the R(0)₁ line to which the laser could be locked.

In the last decade, extensive research has been performed by the group from the LPTF/BNM in France on the CO₂ laser/OsO₄ absorber system. This has resulted in a calibrated infrared frequency grid of OsO₄ transitions in near coincidence with CO₂ laser lines with a frequency reproducibility of 2×10^{-12} [10, 11]. The frequencies of the R(10), R(12) and P(14) CO₂ laser lines, locked to transitions in OsO₄, were measured against a cesium atomic frequency standard. Several other OsO₄/CO₂ coincidences were then measured relative to these absolute frequencies. Parallel to this work, the group from the Université Paris-Nord developed a similar CO₂ laser/OsO₄ frequency grid [12–14]. In addition to providing one of the best frequency reference points in the infrared, the group used their OsO₄ frequency grid to measure some CO₂ laser line centres using saturated absorption in low-pressure CO₂ instead of the 4.3- μm fluorescence technique [15, 16]. However, the CO₂ laser R(0)₁ line was not measured by either of the two groups in France.

In this work, the absolute frequency of the R(0)₁ CO₂ laser line stabilized on OsO₄ is measured. The R(0)₁ laser line has very low gain and is generally only observed with laser discharge tubes of more than 1.5 m length. Two saturable absorbers, osmium-tetra-oxide (OsO₄) in its natural mixed isotopic composition and isotopic ammonia (¹⁵NH₃), were investigated for the purpose of stabilizing the frequency of the R(0)₁ CO₂ laser. L. Ricci et al. [17] had already measured the frequencies of seven ro-vibrational transitions in the ν_3 band of ¹⁸⁸OsO₄ in coincidence with the R(0)₁ CO₂ laser line. These frequencies were measured with an estimated uncertainty of ± 30 kHz relative to a second CO₂ laser which was stabilized on a CO₂ reference line using the 4.3- μm

fluorescence technique. This work was extremely useful for the identification of the OsO_4 spectrum, but the measurement uncertainty had to be reduced for our application. We report here a direct Cs-clock-based frequency-chain measurement of the $Q(15)$ transition in $^{188}\text{OsO}_4$ and relative frequency measurements of the transitions $Q(14)$ in $^{188}\text{OsO}_4$ and $sQ(4,3)$ in $^{15}\text{NH}_3$. We had observed the coincidence of the strong $sQ(4,3)$ absorption line of $^{15}\text{NH}_3$ with the $R(0)_1$ laser line earlier [18], but had not measured its frequency prior to this work.

1 Experimental

Figure 1 shows the experimental layout. The CO_2 laser used in the present experiments was originally designed for laser lines with low gain [19]. Its 2-m-long narrow-bore discharge tube produced an output power of 2 W at the $R(0)_1$ line under sealed-off operating conditions. However, the free-running short-term linewidth was, at best, only 20 kHz as measured with the infrared frequency chain of the National Research Council (NRC) [20]. This frequency noise appeared as a result of the narrow bore of the discharge tube, and the use of Brewster windows and air gaps in the cavity.

About half of the CO_2 laser power was sent via a set of mirrors to the NRC infrared frequency chain, located in another room. The frequency chain consisted of four CO_2 lasers and two microwave oscillators which were all phase locked to a reference signal provided by a hydrogen maser. The hydrogen maser was in turn referenced to an ensemble of three NRC cesium clocks. Details of the chain can be obtained from [6, 20]. The frequency of the $R(0)_1$ CO_2 laser was measured by mixing it on a tungsten–nickel metal–insulator–metal (MIM) point-contact diode with the outputs from two CO_2 lasers and a microwave oscillator from the chain [6]. A beat frequency of $3C - 2B - V - R(0) + AOM = 17\,648$ MHz

was observed. Here $C = 31\,093\,485$ MHz is a CO_2 laser running on the $R(28)$ transition of $^{13}\text{C}^{16}\text{O}_2$, $B = 32\,185\,080$ MHz is a CO_2 laser running on the $P(14)$ transition of $^{12}\text{C}^{18}\text{O}_2$, $V = 60\,670$ MHz is a chain klystron frequency, and AOM is the frequency (approx. 40 MHz) of an acousto-optic modulator used for isolation of the $R(0)_1$ laser. The beat signal was mixed down to 1548 MHz with a Cs-referenced local oscillator at 16 100 MHz. Typical beat signal-to-noise ratios at 1548 MHz of 15–20 dB were observed in a 100 kHz bandwidth with frequency excursions lying in the range 10–20 kHz. A tracking oscillator was phase-locked to the beat signal and its frequency counted. Measurements exceeding 1000 s duration were obtained with no evidence of chain cycle slips during such periods [6].

As the frequency lock of the laser to the saturation dip of the absorber is crucial for this experiment, a scheme was adopted which was already well tested at the LPTF/BNM [10] and the Université Paris-Nord in France [15]. A small fraction of the laser power was sent through a Faraday isolator and a set of calibrated attenuators, then through a telescope to mode match the beam into a Fabry–Perot (F–P) cavity. The absorbing gas was contained in the F–P cavity, which effectively increased the optical path length in the gas over a simple two-way cell, and ensured a well-defined symmetrical saturating and probing Gaussian beam. The cavity was made of rigid Invar blocks of 1-m length and fitted with identical concave mirrors of 50-m radius on each end, resulting in a standing wave with an average Gaussian mode radius of $\omega = 4.0$ mm. Each mirror had a reflectivity of 98.75% at $10.39\ \mu\text{m}$ wavelength. At resonance the full-width at half-maximum (FWHM) of the transmission peaks was 550 kHz, in agreement with the value calculated from the reflectivity and absorption of the mirrors. The laser power in front of the cavity was typically $60\ \mu\text{W}$, and after the cavity $20\ \mu\text{W}$, due to the mirror's absorption and residual beam mismatch. The power inside the evacuated cavity was calculated to be 1.6 mW by using the mirror reflectivity. The F–P cavity was placed inside a vacuum-tight stainless-steel tank with NaCl windows at each end. An ion pump (Varian, Vacion, 8 l/s) was used for the final evacuation before the tank was filled with the absorber gas. The gas pressure was measured with a capacitance manometer [MKS, Baratron, 6.65 Pa (50 mtorr) full scale].

Both cavity mirrors were mounted on tube-type piezoelectric transducers (PZT) [21]. A 15-kHz modulation was applied to the PZT of the input mirror causing the cavity resonance frequency to deviate sinusoidally from the fixed laser frequency by a modulation depth of ± 150 kHz. The resulting power change in the transmitted beam was detected by a HgCdTe detector, mounted behind the cavity. The signal from the HgCdTe detector was amplified by a transimpedance amplifier (which amplifies the detector's current as opposed to the voltage across a fixed load resistor), and then sent through a resonance filter ($Q = 10$) to a commercial phase-sensitive amplifier (PSA), referenced to the 15-kHz modulation frequency. A $1/f$ correction signal was applied to the PZT of the output mirror to lock the peak of the cavity resonance to the laser frequency. A slow servo loop (bandwidth ~ 30 Hz) was used to avoid transferring the intrinsic laser noise (and frequency modulation) to the more stable cavity. As the laser was slowly tuned over its power profile (~ 50 MHz), the cavity followed the laser frequency.

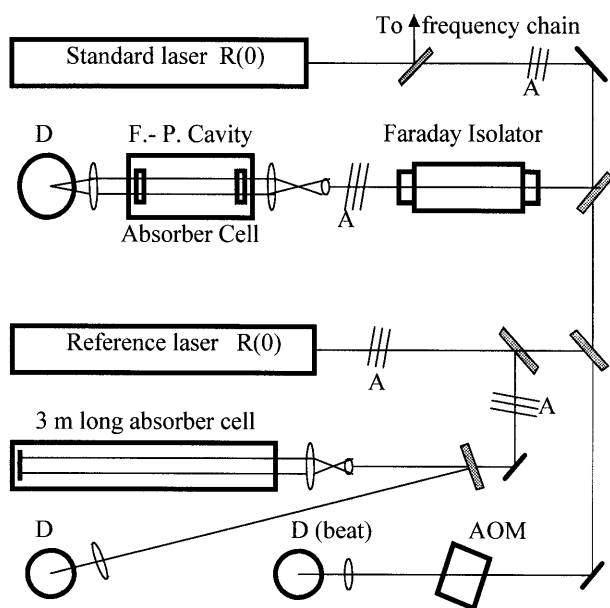


Fig. 1. Schematic diagram of the experimental arrangement: D, HgCdTe detector; A, calibrated attenuators; AOM, acousto-optic modulator used as an optical isolator

In order to detect and lock to the saturation dips, the laser frequency itself must be modulated. A modulation frequency of 1.4 kHz with a 300 kHz peak-to-peak deviation of the laser frequency was employed. The modulation frequency was chosen to be high enough so that the cavity-to-laser servo lock would not respond to the modulation frequency, and low enough so that the third harmonic of the 1.4-kHz signal (at 4.2 kHz) and the 15-kHz signal, as detected by the same infrared detector, could easily be separated. For the detection of the third harmonic signal, the output of the transimpedance amplifier was filtered by two notch filters tuned to 1.4 kHz and 15 kHz. The signal was then sent to a commercial PSA, referenced to a 4.2-kHz signal. The reference signal was generated by phase locking a digital harmonic generator to the 1.4-kHz modulation frequency. The output of the PSA was amplified and fed back to the laser's PZT.

Initial characterization of the CO₂ laser/OsO₄ system was done by heterodyning against a second CO₂ laser system, which is shown in the lower part of Fig. 1. The reference laser system employed a 3-m-long absorber cell, which was double passed by retro-reflecting the laser beam. The Gaussian beam radius was 6 mm at the front and 4 mm at the flat mirror at the end of the cell. The reference laser was frequency modulated at 1.5 kHz and locked to the Q(14) transition of OsO₄ using the 3*f* harmonic detection scheme. The Q(14) centre frequency is about 3.54 MHz below the Q(15) frequency, but still within the tuning range of the R(0)₁ line of the CO₂ laser. The beat signal (3.54 MHz) between the two lasers, as obtained from a HgCdTe detector with a flat-band amplifier (30 dB) was sufficiently strong to be directly counted. To avoid optical feedback from the detector to the lasers, an acousto-optic modulator (AOM) served as an isolator. The feedback from the other optical elements was sufficiently reduced by attenuators. From the beat signal, as observed with a spectrum analyzer, the laser's frequency modulation depth and its free running linewidth were determined. The day-to-day frequency reproducibility of the reference laser system was sensitive to beam alignment and was generally worse than that of the F-P-locked laser system. However, after a sufficient warm-up time, the beat signal between the two lasers was, for a short time, reproducible enough to allow the relative frequency measurements reported below.

2 Spectroscopic measurements with OsO₄

Figure 2 shows part of the OsO₄ spectrum as obtained with the F-P cavity, gas filled with OsO₄ at a pressure of 0.27 Pa, and locked to the laser frequency. The demodulated output signal from the phase-sensitive amplifier was recorded with a digital storage oscilloscope, and is shown as a function of the frequency modulated and slowly scanned laser frequency. The R(0)₁ line centre is located at 28 832 026 219.5 kHz [9], just outside of the spectrum shown in Fig. 2. The central strong saturation dip located 9.52 MHz below the R(0)₁ line centre was identified as the Q(15)A₂⁽²⁾ transition of the ν₃ band of ¹⁸⁸OsO₄, following the assignment of [17]. Because of its strength and proximity to the R(0)₁ line centre, this absorption feature was chosen for stabilizing the CO₂ laser frequency. From the pressure broadening given in [10] and the OsO₄ pressure of 0.27 Pa employed, the width (FWHM) of the saturation dips was expected to be ~ 40 kHz, but the

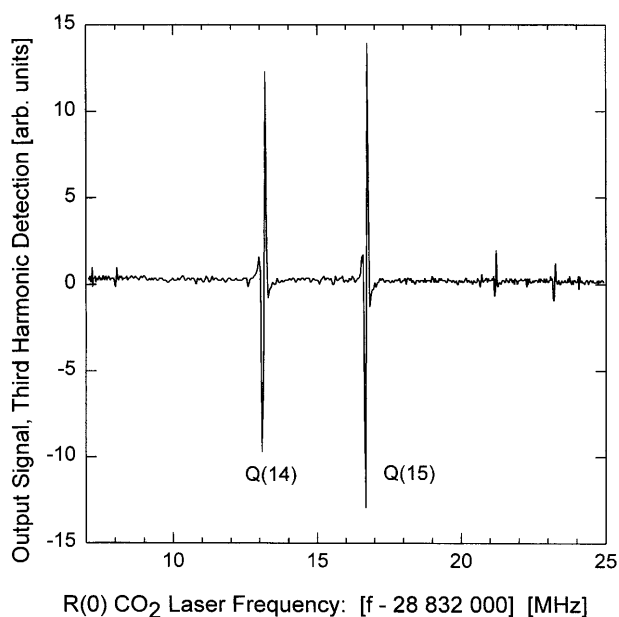


Fig. 2. Spectrum of OsO₄ (demodulated third-derivative) on the low-frequency side of the R(0)₁ transition obtained with the Fabry–Perot cavity at an OsO₄ pressure of 0.27 Pa (2 mtorr)

saturation dips shown in Fig. 2 are ~ 120 kHz wide. Reducing the laser power and the applied modulation depth of 300 kHz did not reduce the line width for scans using time constants of the order of 1 s. However, in a separate experiment, the gas-filled F-P cavity was manually centered to the saturation dip, while the laser frequency was rapidly (at 1.4 kHz) scanned over the FWHM of the F-P. A narrow, symmetric saturation dip, located on top of the wide F-P resonance was observed with an X–Y oscilloscope. The observed width (FWHM) of the saturation dip was ~ 1/20 of the width of the gas-filled F-P resonance, which amounts to ~ 42 kHz, as expected from the pressure broadening. It seems that the large line widths observed in Fig. 2 appear as a result of the inherent free-running laser frequency and power noise averaged over the time (~ 4 s) it takes to scan over the saturation dip. The small offset on the left side of the scan from true zero is caused by the frequency-dependent laser power profile, since the Q(15) saturation dip is located on the low-frequency side of the R(0)₁ line centre.

The OsO₄ absorption changed the transmission and resonance width of the Fabry–Perot cavity. At our standard operating pressure of 0.27 Pa (2 mtorr) and a laser power of 60 μW in front of the cavity, the cavity's FWHM increased from 550 kHz (vacuum) to 850 kHz. The laser power after the cavity decreased from 20 μW (vacuum) to 2 μW, resulting in an intra-cavity laser power of 160 μW. The gain, phase and time constant of the servo loops were set for the gas-filled cavity, but had to be adjusted when large power and pressure changes were studied. The effect of changing parameters of the electronic circuitry on the frequency of the CO₂ laser locked to the Q(15) line was studied while the reference CO₂ laser was locked to Q(14). The largest effect on the lock point of the Q(15) transition was caused by the depth of the applied laser modulation. Figure 3a shows the frequency of the lock point as a function of the modulation depth (peak-to-peak deviation of the laser carrier frequency) at our standard

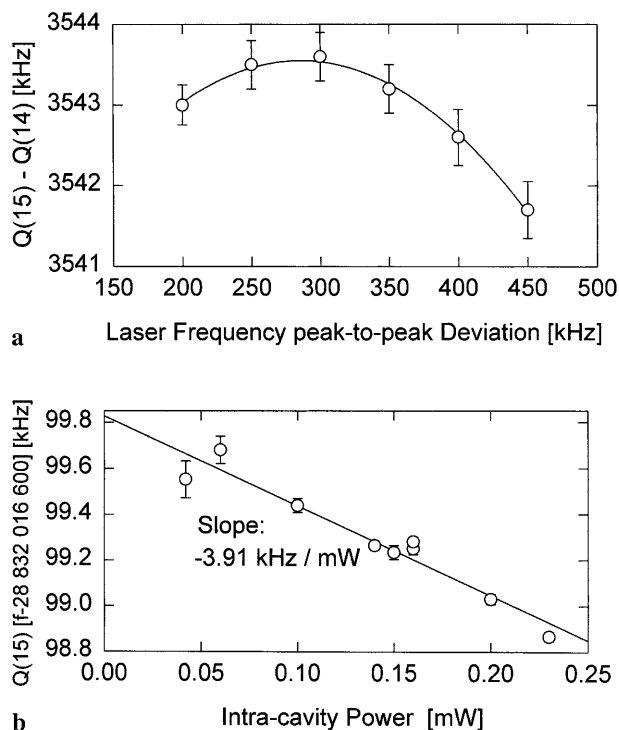


Fig. 3. a : The beat frequency $Q(15) - Q(14)$ as a function of the peak-to-peak frequency deviation of the standard laser. The standard laser was locked to $Q(15)$ with a modulation frequency of 1.4 kHz, and the modulation amplitude was varied for this study. The reference laser was locked to $Q(14)$. **b :** The frequency of the lock point of $Q(15)$ as a function of the laser power inside the F-P cavity at a constant pressure of 0.27 Pa (2 mtorr)

pressure and laser power. The frequency of the lock point first increased and then decreased with increasing modulation depth. It was not possible to reduce the modulation depth much below 200 kHz without losing the lock point. Therefore, to obtain a reliable lock for several hours as required by our experiment and to minimize the observed dependence on modulation depth, a peak-to-peak deviation of 300 kHz was chosen as a standard operating condition.

Figure 3b shows the power dependence of the lock point on $Q(15)$ as measured with the help of the NRC infrared frequency chain. In this experiment, the OsO_4 pressure was held constant at 0.27 Pa and the laser power was measured in front of the F-P cavity. In order to account for the varying degrees of saturation, the intra-cavity power was calculated from a separate series of measurements of the transmitted power of the OsO_4 -filled cavity versus the incident power. A linear fit applied to the measurements in Fig. 3b shows a decrease of the $Q(15)$ frequency with increasing power at a rate of -3.9 kHz/mW . The observed sensitivity of the lock point of $Q(15)$ to the laser power is much larger than the sensitivities reported for most other OsO_4 lines from the infrared frequency grid [10]. This result, combined with the sensitivity to the laser's modulation depth could be caused by an unresolved absorption line doublet. However, in view of the 42-kHz OsO_4 line width observed with the X - Y oscilloscope above, it is much more likely that the relatively large modulation depth used here induced a residual amplitude modulation which shifted the lock point. The shifts in the lock point could also be the result of a slightly asymmetric superposition of the saturation dip over the F-P resonance.

Figure 4 shows the pressure dependence of the lock point of $Q(15)$ over a wide range of pressures. These measurements were done by using the second CO_2 laser system as a reference due to the time-consuming evacuation and re-filling of the F-P cell and the difficulty in operating the infrared chain over extended time periods. The frequencies of the lock points of $Q(15)$ in Fig. 4 were calculated from the measured difference frequencies $Q(15) - Q(14)$, and the absolute value of $Q(14)$, which was later measured with the chain. At a constant laser power of $60 \mu\text{W}$ in front of the cavity, the frequency of $Q(15)$ first increased and then decreased with increasing pressure. This unexpected behavior can be explained by considering the large intra-cavity power changes which occur when the gas pressure is changed, while the incident power stays constant. The probe power inside the cavity for OsO_4 pressures of 0.13 Pa, 0.27 Pa and 0.39 Pa was calculated from the measured power after the cavity at each pressure, and is shown above the x -axis. If the measured frequencies are corrected for a constant intra-cavity power of 0.16 mW by using the power shift of -3.9 kHz/mW from Fig. 3b, the values shown by the open circles are obtained. For pressures below -0.25 Pa the pressure shift becomes very small, as observed with other OsO_4 lines [10]. Figure 4 shows that at the chosen standard pressure of 0.27 Pa, a small pressure dependence already exists which increases to -36 kHz/Pa for higher pressures. This was confirmed by two chain measurements of the lock point of $Q(15)$. As a result of a very slight air leak in the absorber cell, the total gas pressure inside the F-P cavity increased by 0.02 Pa/h . The measured frequencies thus changed with time and, if interpreted as a pressure shift, an average slope of -30 kHz/Pa was found, in reasonable agreement with the measurement above.

Frequency measurements of the lock point of $Q(15)$ under the chosen standard conditions were performed over several

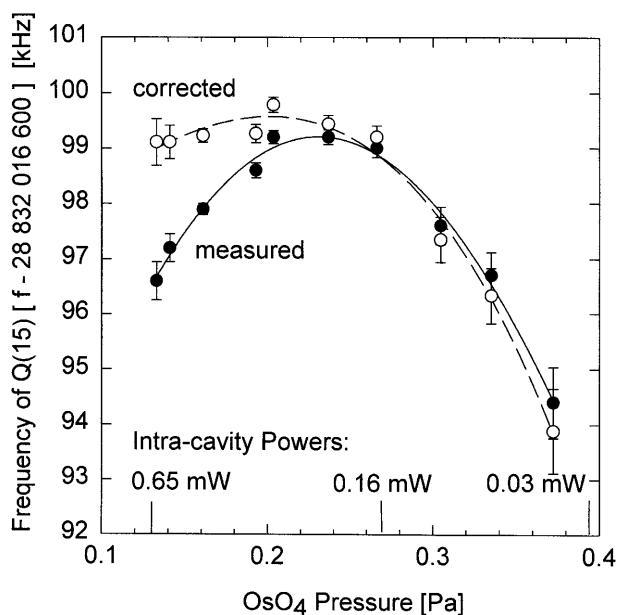


Fig. 4. Frequency of the lock point of the $Q(15)$ transition as a function of the OsO_4 pressure. A constant incident laser power of $60 \mu\text{W}$ was used in the measurement. The measured frequencies are shown by the filled circles, and the values corrected for a constant laser power of 0.16 mW inside the Fabry-Perot cavity are shown by the open circles. At higher pressures, a slope of approximately -36 kHz/Pa is found

days by heterodyning with the NRC infrared chain. The aim of these measurements was to obtain a frequency value for the lock point, with a low enough uncertainty that future calibrations of the HeNe/I₂ standard could be done without simultaneous operation of the frequency chain. Figure 5a shows the Allan deviation of the R(0)₁ laser frequency under standard operating conditions with a 0.16-mW intra-cavity power, an OsO₄ pressure of 0.27 Pa and a peak-to-peak laser frequency deviation of 300 kHz. For an averaging time of $\tau = 300$ s, the Allan deviation dropped to 10 Hz. A fit of the measured values follows closely a $\tau^{-1/2}$ dependence, as expected for white noise. The noise level of 10 Hz corresponds approximately to the residual changes in the OsO₄ pressure and laser power under our standard operating conditions.

Figure 5b shows the results of frequency measurements of Q(15) with the NRC infrared chain on 5 days spanning a period of 4 months in the year 2000. An average absolute frequency of 28 832 016 699.4 kHz for the lock point of Q(15) was obtained with a standard deviation (1σ) of 0.5 kHz at our operating conditions. The maximum day-to-day variation was 1.6 kHz, caused by the resetability of the operating parameters. Although the reproducibility is worse than anticipated from previous work with other CO₂/OsO₄ systems [10, 12, 15], it is sufficient for the present application of calibrating the HeNe/I₂ laser system. For spectroscopic purposes, the “undisturbed” centre frequency of the Q(15) line of ¹⁸⁸OsO₄ was estimated:

Extrapolation to zero power for Q(15) results in a value of 28 832 016 699.8 kHz (± 0.8 kHz), and further extrapolation to zero pressure gives 28 832 016 700.5 kHz (± 2 kHz) at a modulation depth (peak-to-peak) of 300 kHz.

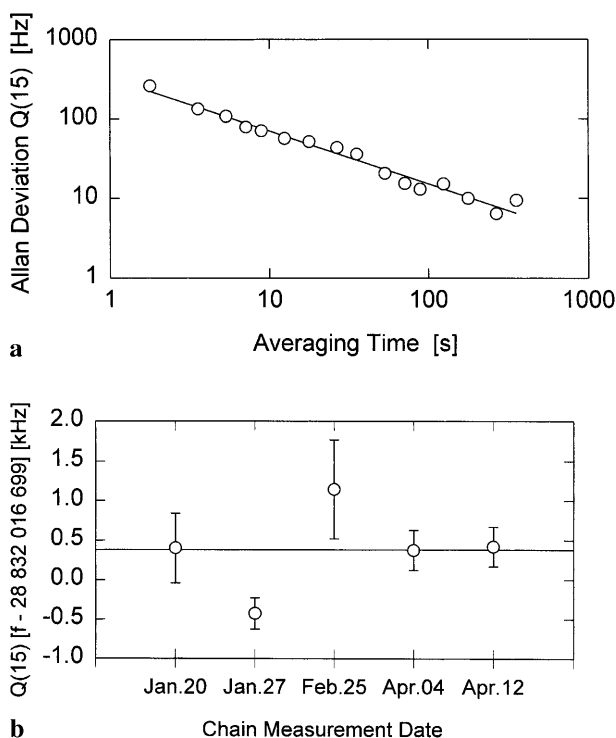


Fig. 5. a : Allan deviation of the R(0)₁ laser frequency locked to Q(15) in ¹⁸⁸OsO₄. **b** : Summary of the frequency measurements of Q(15) with the NRC infrared chain. A mean value of 28 832 016 699.4 kHz (± 0.5 kHz) was obtained

Once the absolute frequency of Q(15) was measured, the frequency of the Q(14) transition became available from the measured beat frequency between Q(15) and Q(14). However, for Q(14) the 3-m absorber cell was used, whereas for Q(15) the F–P cell was employed. There was always a systematic difference of a few kHz in the frequency of the locking point between the two techniques, which depended on the operating conditions of both systems. The difference was measured by observing the beat signal between both laser systems when operated on the same transition, and then applied to correct the frequency of Q(14). These relative measurements of Q(14) were supplemented by one absolute frequency chain measurement, resulting in a combined value of $Q(14) = 28\,832\,013\,156.7 \pm 0.8$ kHz under our standard operating conditions and use of the F–P cell. The sensitivity of Q(14) to laser power, laser-frequency deviation and OsO₄ pressure was not as extensively studied, but appeared similar to that of Q(15).

3 Spectroscopic measurements with ¹⁵NH₃

¹⁵NH₃ does not have a nuclear quadrupole moment (as opposed to normal ¹⁴NH₃) so that the saturation dip has no quadrupole hyperfine structure. The *s*Q(4,3) transition is the only absorber line of ¹⁵NH₃ within the tuning range of the R(0)₁ CO₂ laser. With its convenient location of 4.28 MHz above the R(0)₁ line centre and a very strong unsaturated absorption coefficient of $3.8\text{ m}^{-1}(\text{Pa})^{-1}$ it initially appeared attractive as a reference line for the CO₂ laser. Although the width of the saturation dip of ¹⁵NH₃ (120 kHz) was similar to that of OsO₄, the day-to-day frequency reproducibility of the line centre was found to be worse than that for OsO₄. This may have been caused by inadequate control of the very low pressure, which was required because of the strong absorption. Also, the loss of pressure due to adsorption of ¹⁵NH₃ at the walls of the stainless-steel tank was very noticeable and required frequent adjustments of the pressure.

Figure 6a shows the power dependence of the lock point for *s*Q(4,3) at a ¹⁵NH₃ pressure of 0.013 Pa. These measurements were done by locking the reference CO₂ laser to the *s*Q(4,3) line of ¹⁵NH₃ and heterodyning against the standard CO₂ laser locked to Q(15) of OsO₄ in the F–P cell. From the measured beat frequencies and the absolute frequency of Q(15), the frequency of *s*Q(4,3) as shown on the y-axis in Fig. 6a is derived. The probe laser power, as varied by a set of calibrated absorbing filters, was measured at the entrance of the 3-m-long absorber cell. At the ¹⁵NH₃ pressure of 0.013 Pa chosen for this experiment, the saturating and probing beams were approximately equal, at the lowest power the return beam had still 40% of the incident power. A slope of 0.35 kHz/mW was obtained for the power shift shown in Fig. 6a. Thus the *s*Q(4,3) transition frequency is less sensitive to power changes than the Q(15) OsO₄ transition. However, this metrologically useful property is offset by the very strong pressure dependence of the *s*Q(4,3) lock point.

Figure 6b shows the pressure dependence of *s*Q(4,3). A constant laser power of 5.6 mW at the entrance of the 3-m-long absorber cell was used. The saturating and probing beam powers became very different with increasing pressure; at 0.09 Pa only 5% of the incident power was measured at the exit of the cell. Thus the measured pressure dependence

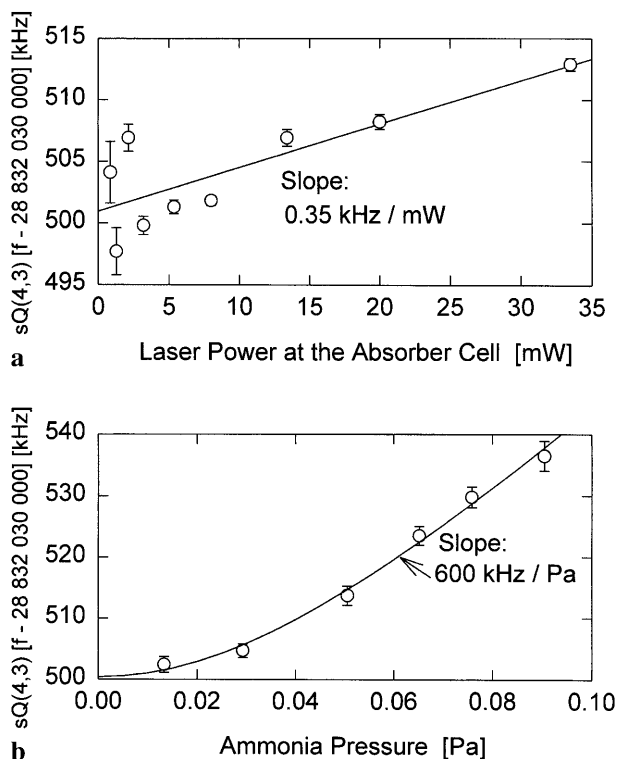


Fig. 6. a : Frequency of the sQ(4,3) lock point as a function of the incident laser power. The 3-m-long absorber cell was used at a constant $^{15}\text{NH}_3$ pressure of 0.013 Pa. **b :** Frequency of the sQ(4,3) lock point transition as a function of the $^{15}\text{NH}_3$ pressure in the 3-m-long absorber cell at a constant incident laser power of 5.6 mW

involves some power dependence. The strong absorption terminated the pressure range over which the laser could be locked at this power level. By increasing the incident laser power, the pressure range was extended. In either case, a very large pressure dependence of approximately 600 kHz/Pa was found for pressures above 0.04 Pa. Figure 6b also shows that the pressure shift minimizes for the lowest pressure at which the laser could still be locked. At a pressure of 0.013 Pa, an incident laser power of 5.6 mW, and a peak-to-peak laser frequency deviation of 300 kHz, the lock point of the sQ(4,3) frequency was 28 832 030 502 kHz (± 3 kHz). The uncertainty includes the estimated systematic difference between the two laser systems and the statistical errors of the beat frequency measurements.

4 Summary and conclusion

The frequencies of two OsO_4 transitions were measured in saturated absorption relative to NRC's cesium primary frequency standard. The probe laser was a cw line-tunable CO_2 laser operating on the $\text{R}(0)_1$ transition. For (normal isotope mixture) OsO_4 at a pressure of 0.27 Pa (2 mtorr), a probe laser power of 0.16 mW inside the F-P cavity (power density of 0.62 mW/cm^2), and a modulation depth (peak-to-peak) of 300 kHz the frequencies of the lock points were:

$$Q(15)^{188}\text{OsO}_4 f = 28\,832\,016\,699.4 \text{ kHz} (\pm 0.5 \text{ kHz})$$

$$Q(14)^{188}\text{OsO}_4 f = 28\,832\,013\,156.7 \text{ kHz} (\pm 0.8 \text{ kHz})$$

Extrapolation to zero pressure and power was only studied for Q(15) and resulted in:

$$Q(15)^{188}\text{OsO}_4 f = 28\,832\,016\,700.5 \text{ kHz} (\pm 2 \text{ kHz})$$

Both absorption lines were chosen for their convenient proximity to the line centre of the $\text{R}(0)_1$ CO_2 laser transition but showed a larger dependence on the laser power and modulation depth than expected, which clearly limited the day-to-day reproducibility of the measurement. It appears that the laser frequency noise is ultimately responsible for these effects and a different laser design could improve the results.

For isotopic ammonia $^{15}\text{NH}_3$ at a pressure of 1.7 mPa, a probe laser power of 5.6 mW (14 mW/cm^2), and a modulation depth (peak-to-peak) of 300 kHz, the frequency of the sQ(4,3) lock point was:

$$sQ(4,3)^{15}\text{NH}_3 f = 28\,832\,030\,502 \text{ kHz} (\pm 3 \text{ kHz})$$

Extrapolation to zero power and pressure gave:

$$sQ(4,3)^{15}\text{NH}_3 f = 28\,832\,030\,500 \text{ kHz} (\pm 5 \text{ kHz})$$

The ammonia molecule's relatively high pressure shift sensitivity and the problem of gas adsorption on the cell walls make this reference line more difficult to implement reliably than transitions in OsO_4 . Again a laser source with less frequency noise would result in an increased signal-to-noise ratio of the saturation dips and improve the tightness of the lock point. The system already can reliably provide a known spanning frequency to connect the 474-THz standard to the 445-THz single ion reference at levels which are equivalent to the limiting reproducibilities of the iodine stabilized HeNe laser standard. Given the worldwide use of the 474-THz standard in precision measurement and in realizing the metre in metrology labs, together with the central location of the 28.8-THz $\text{R}(0)_1$ transition in the $10\text{-}\mu\text{m}$ CO_2 laser band, the current absolute frequency measurements should be of continued utility in future precision measurements.

Acknowledgements. We would like to acknowledge the important support provided by J.-S. Boulanger and S. Cundy in supplying and monitoring the Cs-based reference frequency signal. The invaluable assistance of R. Pelletier and B. Hoger in the development and construction of the electronic laser servo systems is gratefully appreciated. We are greatly indebted to the referee for suggesting an explanation for the observed asymmetry in the OsO_4 saturation dip.

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