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Two stage laser stabilization using external cavities: Application to a $10 \ \mu m \ CO_2$ laser locked to an OsO₄ transition

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ABSTRACT A cw carbon dioxide laser, operating on the 10 μ m R(0) transition (28.832 THz), was stabilized by locking its frequency to an evacuated Fabry–Pérot cavity. The Fabry–Pérot cavity was in turn stabilized to the saturated absorption resonance of the Q(15) line of ¹⁸⁸OsO₄, contained in a second Fabry–Pérot cavity. Cesium-clock referenced, frequency chain measurements of the resultant line frequency are presented together with detailed studies of the line shift sensitivities to various operating parameters.

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1 Introduction

The desire to stabilize the frequency of the $10 \,\mu m$ R(0) transition of a cw CO₂ laser arose a few years ago in our laboratory, where this frequency was used to bridge the frequency gap between an iodine-stabilized HeNe laser $(I_2/HeNe)$ and the NRC strontium ion frequency standard [1]. The difference frequency between the 474 THz (633 nm) HeNe laser frequency and the 28.8 THz (10.4 μ m) CO₂ laser frequency was generated by angle tuned phase matching in a AgGaS₂ crystal and then heterodyned against the strontium ion frequency standard at 445 THz [2]. The CO₂ laser frequency was simultaneously measured by the NRC infrared frequency chain [3]. Given the wide use of the 474 THz radiation together with the high level of accuracy afforded by the 445 THz strontium single ion standard, it was felt that the R(0) transition could be further exploited as an effective frequency "tie-point" in the infrared region of the spectrum. This would enable intercomparison between the I2/HeNe standard and the ion standard by all optical means. Moreover, the referencing of 474 THz and 445 THz frequencies to a femtosecond laser based optical frequency comb in the visible [4] would allow an alternative means to measure infrared wavelengths in the region of the R(0) transition. For these reasons, studies have been devoted to the development of the midinfrared standard to produce reproducibilities well below that of the 474 THz standard (< 500 Hz). The goal of the present investigation was to increase the confidence and reduce the uncertainty in the Q(15) line frequency of the v_3 band of ¹⁸⁸OsO₄ which has been used to provide long term stability and accuracy for the laser operating on the R(0) transition.

In previous work [5], the CO_2 laser stabilization scheme used a single Fabry-Pérot (F-P) cavity, which was filled with OsO_4 at low pressure. Such a technique has been shown by a number of groups [6-8] to yield excellent reproducibilities and accuracies for the high quality saturated absorption reference transitions in OsO₄. This is due to the long effective path lengths and the well behaved Gaussian beam structure with constant power density which interacts with the gas phase molecules. In our previous experiments, the resonance frequency of the cavity was modulated via an internal piezoelectric transducer (PZT) and stabilized to the input laser frequency. The laser frequency itself was modulated at another (higher) frequency and locked to one of the OsO4 saturation lines. This scheme resulted in a reproducibility of the Q(15)line of 188 OsO₄ of about ± 1 kHz with a relatively large dependence on the operating parameters. The results from our previous investigation [5] suggested that the free running laser frequency noise was significantly larger than the power/pressure broadened OsO₄ line. The distortions in the line-shape dominated by the short-term (< 1 s) laser frequency fluctuations were considered a significant factor contributing to the high shift sensitivities encountered. It was suspected that an independent pre-stabilization of the laser, in which the probe laser linewidth would be a negligible contribution to the total observed linewidth, would lead to a smaller uncertainty in the lock point to OsO_4 . At first, it appeared that a frequency prestabilization of the laser could be simply achieved by locking the laser to the OsO4 filled F-P cavity rather than locking the F-P cavity to the laser, as had been done. However, the relatively broad and smooth transmission peak of the gas filled F–P cavity is distorted when the cavity is tuned to one of the OsO₄ saturation features. This brings instability to the lock point of the laser, particularly for a strong absorber like OsO₄. To avoid this problem, a two-cavity scheme was used.

2 Experimental arrangement

Figure 1 shows the experimental layout. The CO_2 laser consisted of a 2-m long, 12-mm bore, sealed-off dis-

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to frequency chain



FIGURE 1 Schematic diagram for the experimental arrangement: PAS-1 and PAS-3 (Stanford Research SR510) and PAS-2 (Stanford Research SR830) are phase sensitive amplifiers. DET-1 and DET-2 are liquid N_2 cooled HgCdTe detectors. Cavity-1 and cavity-2 are Fabry–Pérot cavities. EOM is an electro-optic modulator (97 kHz). Items marked as "V" are notch filters set to attenuate 1.4 kHz. Objects marked "AT" are a set of attenuators

charge tube which produced a continuous output power of 1.2 W at the 10.4 μ m R(0) line. The discharge tube possessed NaCl Brewster windows and was held within a 2.4 m long optical cavity consisting of a high reflector of 5.5 m radius of curvature mounted on a piezo-electric translator (PZT) and a diffraction grating of 150 lines/mm having a diffraction efficiency of 0.95. Although the optical path between the reflectors and gain tube was closed to prevent air drafts, the present design of the optical cavity precluded the hermetic sealing of the internal optical path. The main portion ($\sim 1 \text{ W}$) of the beam power of the CO_2 laser was sent to the NRC frequency chain [3] for a cesium-clock referenced frequency measurement of the Q(15) line of the ν_3 band of ¹⁸⁸OsO₄. This power was also sufficient to be used in applications for the difference frequency generation with the HeNe laser [2]. A fraction $(\sim 200 \text{ mW})$ of the main beam was sent through a Faraday isolator and then split into two parts. One beam ($\sim 10 \text{ mW}$) was used for locking the laser frequency to an evacuated Fabry-Pérot cavity (cavity-1), while the other beam was used to detect the OsO₄ saturation features with help of a second F–P cavity (cavity-2).

Cavity-1 was made of a 310-mm-long Invar tube terminated by two identical mirrors (concave 5 m radius, 98% reflectivity), which resulted in a resonance with a full width at half maximum (FWHM) of 1.6 MHz. Both mirrors were mounted onto piezo-electric transducers for length tuning and modulation. The cavity rested on Viton rubber rings for vibrational damping inside an evacuated stainless steel tank. Two windows made of BaF2 mounted onto the vacuum tank permitted input and output of the IR radiation. Beam-1 was frequency modulated by a commercial electro-optic-modulator (EOM) [10]. The EOM was made of a 50-mm-long CdTe single crystal. Modulation frequencies for this material should be lower than 100 kHz or higher than 1 MHz due to phonon absorption of the rf radiation in the crystal's lattice. After measurements indicated that most of our laser frequency noise was below 1 kHz, a modulation frequency of 97 kHz was chosen which was commensurate with the maximum operating frequencies of our phase-sensitive amplifiers. The signal from a stable frequency generator was resonantly amplified to achieve a voltage of 2.2 kV across the CdTe crystal, which produced two side bands each with 20% of the power of the carrier frequency. The side bands at ± 97 kHz from the carrier frequency are within the FWHM of cavity-1 which enabled detection of the 97 kHz frequency in transmission rather than in reflection as normally used with the Pound-Drever-Hall technique [9]. The 97 kHz signal from the detector (DET-1) was resonantly amplified and phase-sensitively detected (PSA-1). The output signal from the PSA-1 was then integrated and amplified for the frequency lock of the laser to the cavity. The fastest available time constant of the PSA-1 system limited the overall bandwidth of the servo loop to ~ 6.8 kHz.

In order to lock the cavity-stabilized laser to the OsO₄ lines a modulation frequency of 1.4 kHz was applied to one of the PZT's in cavity-1. This modulation, with an adjustable amplitude and phase, was also applied directly to the PZT of the laser through a "feed forward" arrangement to facilitate a tight lock of the laser to the cavity. A modulation depth of 100 kHz (peak to peak) was chosen for our standard conditions. The modulation depth could be varied through the applied modulation voltage at the rate of $0.9 \text{ kHz/mV} (\pm 0.1)$ as determined from heterodyne measurements with the NRC infrared frequency chain (see Sect. 3). Since the modulation depth played an important role in the determination of the spectral width of the OsO4 saturation features, a second measurement method was used to verify its calibration. A dc voltage was applied to move the laser frequency between the two saturation features Q(14) and Q(15) of OsO₄, which by previous absolute frequency measurements had been determined to be 3.543 MHz [5]. This dc voltage induced a displacement of 1.18 kHz/mV. As expected from the hysteresis of the PZT material, the value is slightly larger than that determined from the modulation measurements, but the result contributes confidence to the heterodyne measurements. To prevent leakage of the 1.4 kHz modulation (used for locking to the OsO4 lines), a 20-dB notch filter at 1.4 kHz was employed before the input to the PSA-1 amplifier. The filter reduced the 1.4 kHz signal at the PSA-1 input to a level that would not interfere with the phase-sensitive detection of the 97 kHz signal used in the laser frequency stabilization.

The performance of the laser's lock to the cavity-1 was tested with help of the second cavity (cavity-2), which was evacuated and tuned to the side of its resonance peak. Figure 2 shows the 1.4 kHz modulation signal with a modulation depth of 90 kHz (peak to peak) to provide a calibration scale for the vertical axis. When the modulation was switched off, some

FIGURE 2 Observed frequency excursion of the 1.4 kHz modulated laser frequency while locked to cavity-1. Cavity-2 was tuned to the side of the transmission peak to observe the modulation depth of 90 kHz. Centre trace: When the 1.4 kHz modulation is switched off a residual laser frequency noise of ~ 3.2 kHz remains. The laser power noise was negligible

short-term frequency noise of the laser remained, as shown by the center trace. The contribution of the laser power noise was negligible, as was tested by tuning cavity-2 to the peak of its transmission. The contribution of residual high voltage noise on the PZT of cavity-2 was calculated to be below 100 Hz. The average (1σ) laser frequency noise measured 3.2 kHz for averaging times of < 0.1 s as opposed to about 50 kHz or more when the laser was free running [5]. The 1.6 MHz wide FWHM resonance width of cavity-1 provided a large capture range for the laser frequency combined with a good signal to noise ratio, so that the servo lock would hold over time periods of several hours. This was a significant improvement over the previous arrangement where the broad laser line was locked directly to the narrow spectral width (HWHM ~ 40 kHz) of the saturated absorption of Q(15) line [5].

Cavity-2 was made of six 0.9-m-long, 12-mm thick Invar plates, which were screwed together to form a rigid hexagonal spacer. This spacer then rested on four small supports inside a vacuum-tight stainless-steel tank with BaF2 windows. Identical concave mirrors with 50-m radius and 98.8% reflectivity mounted on piezo-electric transducers formed the optical cavity. The FWHM of the evacuated cavity-2 measured 550 kHz and increased to 800 kHz when filled with OsO4 at a pressure of 0.066 Pa (0.5 mTorr). For the undistorted detection of the saturated absorption signal, the transmission resonance of cavity-2 must be centered on the laser frequency. A 1-f demodulated servo lock of the cavity accomplished the necessary stabilization to the input laser frequency. A modulation voltage at 11.5 kHz was applied to one of the PZT's of cavity-2, which resulted in a modulation depth of the resonance frequency of 20 kHz (peak to peak). With a servo loop bandwidth of 1 kHz (determined by the PSA-3 servo), the cavity remained centered on the average laser frequency. A resonance amplifier tuned to 11.5 kHz and located between DET-2 and the PSA-3 system prevented leakage of the 1.4 kHz signal from the laser beam to the PSA-3 servo. Under the chosen standard conditions, the power of the beam before cavity-2 was 18 μ W and 4.28 μ W after the cavity at an OsO₄ pressure of 0.066 Pa (0.50 mTorr). From the known mirror reflectivity, an intra-cavity power of $355 \,\mu\text{W}$ is calculated for our standard conditions.

After the laser was locked to cavity-1 and cavity-2 was locked to the laser frequency, the third servo loop was used to lock cavity-1 to the saturation absorption features of the Q(15) or Q(14) resonances of OsO₄. For this purpose, the laser frequency was slowly tuned (via cavity-1) until one of the saturation resonances was observed at the input of PSA-2 and the loop could be closed. Servo stabilization onto the saturated absorption line was performed using demodulation at the third harmonic of the applied 1.4 kHz frequency. A notch filter set at 1.4 kHz prevented overloading of the input of the phase sensitive amplifier PSA-2. Detector-2 was a liquid nitrogen cooled HgCdTe detector ($D^* = 17 \times 10^{-9}$) which was capacitively coupled to a transimpedance amplifier. The active detector area $(1.5 \text{ mm} \times 1.5 \text{ mm})$ was larger than the focused beam size to avoid a frequency shift from partial beam obstruction [6]. The time constant in the servo loop stabilizing to the OsO₄ resonance was 30 ms, a compromise between minimizing the servo noise carried into cavity-1 and the tightness of the lock to the saturation feature. Figure 3 shows an overview of the spectrum taken at a pressure of 0.27 Pa (2 mTorr) and a power of 1.8 mW in front of cavity-2. The small secondary dispersion shaped features characteristic of a 3-f signal are missing due to the small modulation depth (FWHM = 100 kHz) applied. Apart from the strong saturation features, Q(14) and Q(15), a number of weaker saturated

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a pressure of 0.27 Pa (2 mtorr) and a laser power of 1.8 mW in front of cavity-2. The frequency span Q(15)-Q(14) = 3.543 MHz and the absolute frequency of Q(15) were used for scaling the x-axis



lines are recorded, which were not observed before [5] and may be the result of the improved laser spectral brightness. In order to calculate the spectral width of the O(15) saturated absorption line, a number of such spectra were taken at our standard operating pressure (0.066 Pa) and power (355 μ W) as a function of the modulation depth of the laser frequency. The peak-to-peak width of the saturation feature was measured relative to the frequency spacing (3542.6 kHz) between the Q(14) and Q(15) lines. The resultant plot of the measured peak-to-peak width of the saturation feature as a function of the applied modulation depth was fitted to the theoretical width for the 3rd harmonic of a Lorentzian line [11]. The best fit lead to a HWHM of 40 kHz for Q(15) under the chosen standard conditions. The 40 kHz line width includes a transit time broadening of 7 kHz (beam radius 4.05 mm), some contributions from the pressure broadening and a main component due to power broadening. The minimum power level used was limited by the sensitivity of our detector, which was external to the cavity. It is interesting to note that the reported line width is about twice as wide as that reported for other OsO_4 lines at the same pressure [6, 7].

3 Results and discussion

Direct absolute frequency measurements of the OsO₄ stabilized laser at 29 THz were obtained using the NRC frequency chain. A detailed description of the system has been published elsewhere [2, 3]. Briefly, the chain is formed using two microwave oscillators and four CO₂ lasers as the primary radiation sources. Tungsten-nickel, metal-insulatormetal (MIM) point-contact diodes are used to produce harmonics of the laser and microwave radiations and produce measurable heterodyne beats. By phase locking the lasers using the observed beat signals, it is possible to phase-lock the entire chain in a self-consistent way to a reference 5-MHz signal provided by a NRC hydrogen maser referenced to a Cs atomic clock. The R(0) radiation frequency is measured by a heterodyne beat with the chain CO_2 lasers operating at $v_{\rm B} = 32\,185\,080\,\text{MHz}$, and $v_{\rm C} = 31\,093\,485\,\text{MHz}$ and a microwave source $V = 60\,670\,\text{MHz}$ on a MIM point contact diode. A heterodyne beat frequency of $(3\nu_{\rm C} - 2\nu_{\rm B} - V -$ R(0) + AOM = 17648 MHz was obtained where AOM is the frequency (approx. 40 MHz) of an acousto-optic modulator used in the isolation of the R(0) laser from reflections from the point contact diode. The 17 648 MHz heterodyne beat signal was mixed down to 1548 MHz using a Cs-clock referenced local oscillator at 16100 MHz. A tracking oscillator was phase locked to the resulting heterodyne signal of 20 dB S/N in a 100 kHz bandwidth. The tracking oscillator signal was then counted and stored on a computer for later analysis.

Figure 4 shows the results of the laser frequency measurements of the Q(15) line under our standard conditions (modulation depth = 100 kHz, intra-cavity power = 355 μ W, gas pressure = 0.066 Pa) with the NRC infrared frequency chain on three days spanning a period of two months in 2003. An average absolute frequency of 28 832 016 698 563 Hz was found for the lock point of Q(15) with a standard deviation (1 σ) of 150 Hz. This value includes a correction factor of -2.6×10^{-13} , which arose from the offset of our hydrogen maser referenced to the Cs clock ensemble realizing the

FIGURE 4 Summary of all frequency measurements of Q(15) uncorrected for the hydrogen maser offset. A corrected mean value of 28 832 016 698 563 Hz was obtained with a 1σ standard uncertainty of 150 Hz

SI second. The majority of the measurement runs were approximately 3 min in duration and resulted in a statistical uncertainty of ± 10 Hz to ± 20 Hz. For each measurement the lock point of Q(15) was re-adjusted and all offsets from zero were checked. The data from Feb. 27th and Mar. 13th scatter over a larger frequency range than those from the Apr. 4th, 2003. Most of this scatter is believed to arise from evacuating and then refilling the OsO₄ cavity-2 to the same nominal pressure (0.066 Pa). A precision membrane gauge [12] was mounted at one end of the vacuum enclosure for cavity-2. After every new OsO₄ gas fill into the evacuated cavity-2, the pressure gauge indicated a monotonic decrease in pressure for about two hours. On Apr. 4th , cavity-2 was pre-evacuated for several days and the frequency measurements were done between 4 and 8 h after the OsO₄ fill, when the pressure had stabilized and required no adding of OsO4. The measured frequencies scatter less, but they cluster about at a value of 28832016698430 Hz (± 20 Hz), which is lower than the average value. Only the last two data points on Apr. 4th were taken with a fresh gas fill at the standard pressure (0.066 Pa), and are clearly outside of the cluster and in closer agreement with the previously found average frequency. These results suggest that over a longer time, the partial OsO₄ pressure in the path of the beam is changing possibly due to a small air leak. After a lengthy evacuation to ensure that there was no adsorbed OsO₄ remaining in the chamber, the air leak rate of cavity-2 measured 0.002 Pa/hr [0.015 mTorr/hr]. After 8 hours, this would amount to a 24% reduction in the partial pressure of OsO_4 in the chamber, which could account for the observed frequency difference.

Figure 5 shows the frequency shift of the lock point of Q(15) as a function of the OsO₄ pressure. The frequency of



FIGURE 5 Frequency of the lock point of Q(15) as a function of the OsO, pressure in cavity-2

the Q(15) line in these studies was measured with the NRC infrared frequency chain. The modulation depth (100 kHz) and intra-cavity power (355 μ W) of the laser beam were kept constant during this experiment. Therefore, the pressure range over which a signal could be detected was limited to the range around our standard pressure (66 mPa) and resulted in a pressure shift of 8 kHz/Pa. The pressure dependence is in the same direction but larger than that found for other OsO₄ lines [6]. Our pressure shift includes the power dependent saturation parameter and intra cavity power changes as opposed to the "true" pressure shift of an almost undisturbed absorption line given in [6].

Figure 6 shows the frequency dependence of the lock point of Q(15) on the 1.4 kHz modulation depth of the laser frequency. A slope of $-1.35 \pm 0.1 \,\text{Hz/kHz}$ was observed for the Q(15) line at the quoted standard conditions. The frequency shift is relatively small, an indication that the saturated absorption line was reasonably symmetric and the transmission peak of cavity-2 was well centered to the laser frequency. The repeatability of the modulation depth is approximately ± 10 kHz, so its contribution to the uncertainty in the absolute frequency of Q(15) would be quite small. However, for a cavity with such a high finesse as used here, a slight offset in the lock point of the cavity transmission peak relative to the laser frequency was observed to cause a significant shift in the measured Q(15) frequency. If the cavity (HWHM = 400 kHz) was not centered to the laser frequency while probing the saturated absorption line (HWHM = 40 kHz) the OsO₄ Lorentzian line shape became distorted and the apparent centre shifted away from the true line centre. This effect was measured with help of the NRC infrared frequency chain over an offset range of ± 40 kHz of the cavity relative to the laser frequency. The absolute frequency of Q(15) shifted linearly at the rate of +160 Hz for +1 kHz offset of the cavity. To minimize a systematic error in the Q(15) frequency due to this effect, the 1-f lock of cavity-2 to the laser frequency was periodically tested for symmetry of the locking signal and for the absence of electronic offsets. Although this effect plays a critical role in realizing an accu**FIGURE 6** Frequency of the lock point of Q(15) as a function of the peakto-peak modulation depth of the 1.4 kHz modulation of the laser frequency

rate and reproducible value for the stabilized laser frequency, to our knowledge, it has not been mentioned in previous thorough investigations of a CO_2/OsO_4 standards.

As a last major parameter, the influence of the laser power on the lock point of the Q(15) resonance was investigated. For this purpose, a set of calibrated attenuation filters was used in front of cavity-2. The 0.025 mm thick Mylar filters did not alter the beam direction or mode shape but were quite coarse in their attenuation (T = 39%). For finer control of the power, a set of ZnSe interference filters, which might have caused some small misalignment of the beam, was used as well. The 1.4 kHz modulation depth of 100 kHz and the OsO₄ pressure of 0.066 Pa (0.5 mTorr) were kept constant for this experiment. The laser power controls the signal-to-noise of the lock of cavity-2 to the laser as well as the lock of cavity-1 to the OsO₄ line. Therefore, the range over which the power could be varied was limited from $6 \,\mu\text{W}$ to $18 \,\mu\text{W}$ in front of cavity-2 $(100 \,\mu\text{W} \text{ to } 350 \,\mu\text{W} \text{ inside the cavity})$. For each power level used, the power after the cavity was measured and used to calculate the intra-cavity power with help of the mirror's reflectivity. Figure 7 shows the results. An average slope of -2.8 ± 0.8 Hz/µW was observed for the data obtained. The scatter of the data points may be the result of small misalignments of the beam, but the general strong dependence of the Q(15) lock point on the power is most likely caused by the high power inside the cavity. Compared to other CO_2/OsO_4 standards [6-8], the power level used was about three times higher with a power density of $14 \,\mu W/mm^2$ for our beam radius of 4.05 mm. However, the incident power was necessary to obtain a sufficient signal-to-noise ratio at the detector, which was operating near the photo-conductive noise limited regime.





FIGURE 7 Frequency of the lock point of Q(15) as a function of the laser power inside the OsO₄ filled cavity-2

4 Summary and conclusion

Under our standard conditions, a pressure of 0.066 Pa (0.5 mTorr), a power of $355 \,\mu\text{W}$ inside the cavity and a modulation depth of 100 kHz (at 1.4 kHz mod. freq.) the lock point of Q(15) was:

$$Q(15)$$
 ¹⁸⁸OsO₄ $f = 28\,832\,016\,698\,563\,\text{Hz}$ (±150 Hz)

This value is 837 Hz lower than our previously reported frequency for Q(15) [5], but well within the expected range for the lower pressure used here, and the (1σ) uncertainty has been reduced by a factor of three. Extrapolation to zero pressure, zero power and zero modulation amplitude (from Figs. 5, 6 and 7) gives a value of 28 832 016 699 184 Hz with a combined estimated uncertainty of \pm 500 Hz, mainly due to the uncertainty in the power extrapolation.

Two measurements of the absolute frequency of Q(14) on different days agreed to within 100 Hz, but the uncertainty given below was increased to reflect the low number of data. For our standard conditions, the frequency of Q(14) was:

$$Q(14)$$
 ¹⁸⁸OsO₄ $f = 28\,832\,013\,155\,972\,\text{Hz}$ (±300 Hz).

The frequencies reported here were directly measured with help of the NRC infrared frequency chain, which is phase locked to NRC's hydrogen maser and referenced to the atomic time realization of the SI second. The uncertainties given are dominated by the reproducibility and resetability of the CO_2/OsO_4 system itself and not by the infrared chain. With the present configuration, the instrumental uncertainties are well below the requirements for a difference frequency bridge between an iodine stabilized HeNe laser and NRC's strontium ion standard.

In its application as a high quality reference absorber in the mid-infrared, the Q(15) line appears to possess relatively higher lineshift sensitivities to pressure and power than some of the best lines reported by workers [6] investigating other OsO₄ reference transitions. Although the present system is considered to provide a good quality reference in the IR spectrum, further improvements will probably require investigation as to the origins of the shifts. Ideally, future configurations would take advantage of using lower intracavity powers within the absorption cell. This could be accomplished with lower reflectivity cavity optics within the absorption cell whereby the intracavity power is lowered while still maintaining sufficient optical power for the detector. With lower power levels, the shift sensitivities are expected to be similar to those reported for other reference lines in OsO₄. For the Q(15) line, the observed sensitivity to pressure may be due to its proximity to other [Fig. 3] unidentified lines causing some line shape asymmetry [8]. It may, thus, be advantageous to select an OsO4 feature that is well away from adjacent strong resonances. Surveys of the spectral region within the R(0) tuning range have shown strong single resonances located about 41 MHz and 104 MHz above the R(0) transition center frequency. These lines could be accessed by frequency shifting the CO₂ laser light with acousto-optic modulators. It is expected that future work may investigate the advantages of using these reference lines as high quality frequency references in the infrared spectrum.

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