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CO Laser Stabilization Using the Optogalvanic Lamb-Dip

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Abstract. Frequency stabilization of the CO laser using a CO Lamb-dip is achieved in the range from 5.0–6.3 μm. The CO saturation signal is obtained from a low-pressure discharge in absorption and is detected using optogalvanic detection. The frequency stability and reproducibility has been verified to be better than 100 kHz; this is an improvement of more than one order of magnitude compared with locking techniques using CO laser gain profiles.

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The CO laser operates at fixed frequencies in the mid infrared from 36 to 62 THz (8.2 to 4.8 μm) [1]. Several hundred rotational-vibrational laser transitions from 40 to 59 THz (7.5 to 5.0 μm) oscillate in sealed-off operation using different isotopic species. Although the CO laser is not continuously tunable, it serves as a relatively powerful coherent source for spectroscopic applications, for example, in laser-magnetic-resonance spectroscopy [2]. The CO laser could serve as an important frequency standard for mixing experiments. However, until now, the CO laser lacked a reliable and accurate stabilization method. Previously, CO lasers had been stabilized to the tops of the Doppler broadened gain curves, and yielded an accuracy of 5 MHz at best. In 1973 Freed and Haus reported the observation of sub-Doppler saturation (Lamb dips) on 16 CO laser lines [3]. They examined velocity changing collisions and measured pressure broadening and saturation intensities in gas mixtures typical of sealed-off CO lasers. The homogenous linewidth of the infrared transitions of the CO molecule is determined by pressure broadening. In order to detect the Lamb dip in a CO plasma it is essential to operate the discharge in the pressure region below 2 Torr. A CO laser operating at this low pressure results in an output power of less than 20 mW and in an enormous reduction in the number of laser lines. In order to combine the typical characteristics of the CO laser (i.e., output powers of more than 1 W on the strongest laser lines and a large number of lasing transitions) with saturation stabilization on Lamb dips, we have combined an ordinary CO laser resonator with a low-pressure intracavity absorption cell. Unlike the CO_2 stabilization scheme [4], the energy levels in CO belonging to lasing transitions are not populated at room temperature and must be excited with a discharge. Excitation by means of a radio-frequency discharge was abandoned because of the critical coupling between plasma and oscillator: in contrast, a quiet dc discharge was maintained even at low pressures.

Absorption in the low-pressure cell is difficult to detect in the output power of the laser, even with an intracavity arrangement. However, we were able to detected the Lamb dips easily by observing changes in the low-pressure discharge impedance by means of the optogalvanic method. A very good signal-to-noise ratio was obtained, and the optogalvanic signal permitted the stabilization of the laser using both first and third derivative techniques. By heterodyning two CO lasers, both stabilized in this way, we measured the reproducibility of each laser frequency to be better than 100 kHz.

Similar to the CO_2 fluorescence stabilization technique, we also observed the $\Delta v = 2$ fluorescence emitted

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from the low-pressure discharge. It is possible to detect Lamb dips on this radiation; however, the *S/N* ratio using the optogalvanic method is more than an order of magnitude larger.

Experimental Setup

Figure 1 shows the diagram of our CO laser system. The laser resonator consists of a gold coated end-mirror and either a 150 line/mm or a 300 line/mm grating. The radiation is coupled out via the zeroth order of the grating. The sealed-off laser cell is made either of a 6 mm i.d. quartz tube for waveguide operation or a 13 mm i.d. Pyrex tube in open structure configuration. The waveguide laser provides better mode purity (using the mode selector described in [5]); while the open-structure laser utilizes more grating lines and, consequently, has better wavelength selectivity; thus it lases on more lines. The discharge is cooled to about −90°C by means of a specially designed refrigeration circuit described in [2].

The detection of Lamb dips in the CO plasma requires a low-pressure discharge because of the large pressure broadening of 7.3 MHz/Torr [3]. Saturation intensities of CO transitions have been measured to be 2–6 W/cm^2 at 1.5 Torr [3]. In order to saturate the transitions even with weak laser lines, the low-pressure cell is placed inside the cavity.

In an initial experiment we attempted to observe Lamb dips on the laser output power. This could be achieved only on a very few laser lines. It was necessary to decrease the laser gain intentionally in order to get a sufficient fractional absorption from the low-pressure cell. But under normal lasing conditions the Lamb dip could not be detected on the laser power. In order to obtain the saturation signal independently from the laser output power we used the optogalvanic effect (the change of the discharge impedance) in a low-pressure cell. The process responsible for the optogalvanic effect in CO is very complex and is not yet understood in detail [6]. In our experiment, the change in impedance produces a change of the voltage across the discharge. This signal is directly coupled via a high voltage capacitor (plus a protection circuit) into a lock-in amplifier. In order to observe the optogalvanic effect, a quiet low-pressure discharge must be used because all fluctuations of the plasma result in noise on the optogalvanic Lamb dip. To provide these characteristics, the construction of a specially designed low-pressure discharge cell was essential.

The low-pressure stabilization cell consists of a Pyrex tube of 12 mm i.d. and 80 cm in length with a cooling jacket. Cooling the discharge to −80°C reduces the discharge noise and improves the S/N ratio of the optogalvanic signal by an order of magnitude. A nickel tube, 30 cm in length and 2.7 cm in i.d., with a

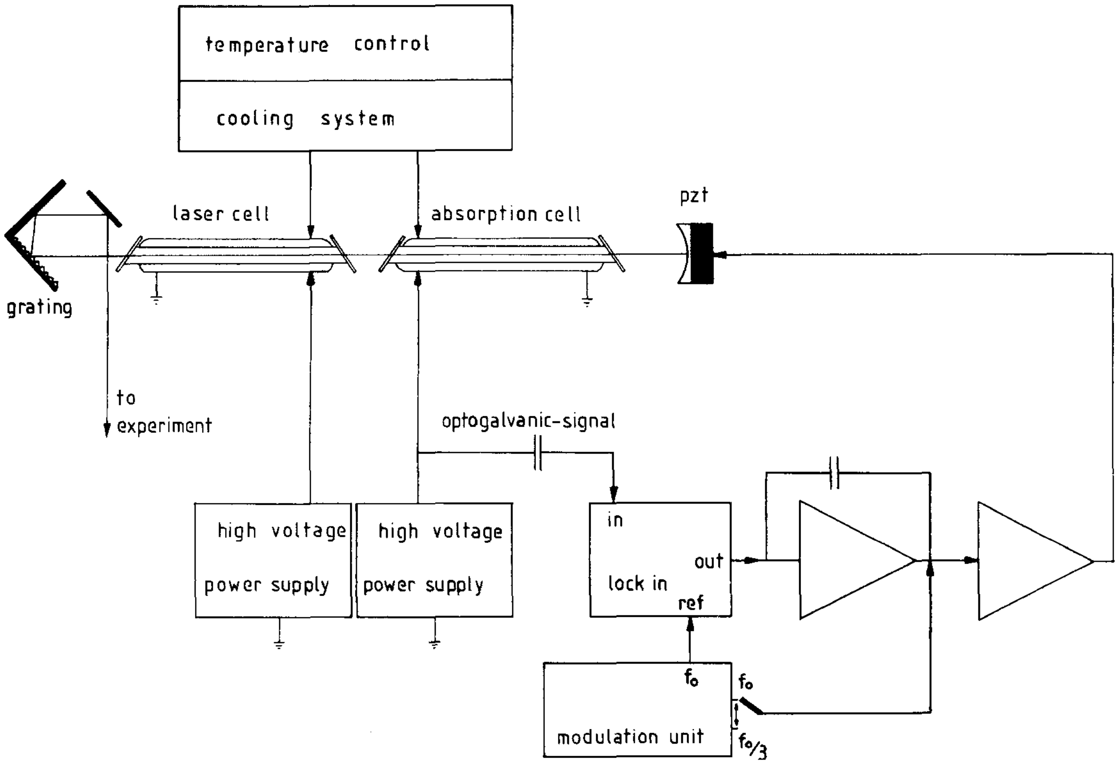

Fig. 1. The laser system. The gain cell is operated at pressures of about 12 Torr. The working pressure in the Lamb dip cell is 1.3 Torr

purity of better than 99.5% was used for the cathode. The surface has been electrochemically cleaned by ac-etching in 1.5 normal sulfuric acid. The hemispherical anode with 15 mm diameter is also made of pure nickel, but with a purity of better than 99.9%. Carbon deposits on this electrode can occur after several days of operation and must be removed in order to keep the discharge quiet. The gas mixture used in this sealed-off cell is different from the ordinary CO laser gas mixture. Best results were obtained with a mixture of Xe, CO, and N_2 in the ratios 3:2:2. With this mixture, stable discharge conditions could be achieved in the pressure range from 1.0 to 1.5 Torr. One day operation is possible without refilling the cell.

Results

The optogalvanic effect yields saturation signals on at least 170 laser lines from each isotope from the $v=6\rightarrow5$ vibrational band to the $v=21\rightarrow20$ vibrational band (5.0 to 6.3 μm). Under the present discharge conditions, higher vibrational levels do not yield an optogalvanic signal.

In Fig. 2 several saturation signals are shown. Lacking an adequate modulation technique in the intracavity setup, a non-"derivative" line shape could not be obtained. In order to make the line shape of the other signals more evident, a reconstructed trace is shown in Fig. 2a. The $1f$ signal in Fig. 2b was taken using a modulation amplitude of 10 MHz (peak-to-peak) and at a frequency of 420 Hz. The time constant of the lock-in amplifier was 50 ms.

The good S/N ratio of the $1f$ signal suggested the possibility of using third derivative locking techniques in order to avoid frequency shifts due to pressure shifts in the laser and possibly asymmetric gain curves. The optimum modulation width (peak-to-peak) for $1f$ detection is known to be 0.7 times the line width of the Lamb dip [7] and it increases to 1.6 times the Lamb dip linewidth for $3f$ detection.

Figure 2c represents the $3f$ signal taken with a modulation width of 20 MHz (peak-to-peak). The frequency of the modulation was adjusted to 140 Hz, one third of the detection and reference frequency for the lock-in amplifier which was at a frequency of 420 Hz. The signal strength decreased by about a factor of 2 going from $1f$ to $3f$ detection as expected from theoretical considerations [7]. The time constant of the lock-in amplifier was again 50 ms. The examination of the error signal when the laser is locked (Fig. 2c) indicates a relative stability of better than 90 kHz.

All optogalvanic signals improve with increasing discharge current, Fig. 3 illustrates this behavior. Below 10 mA the fluctuations of the output voltage

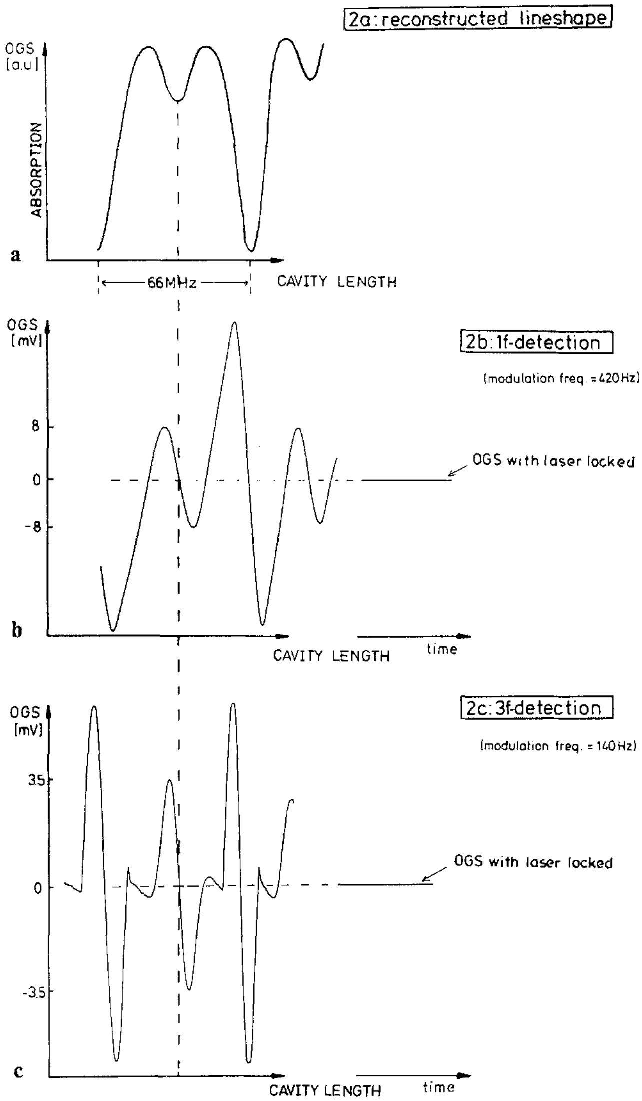

Fig. 2a–c. Optogalvanic signals (OGS) at 1.3 Torr as a function of resonator length. (a) Reconstructed non-"derivative" lineshape, (b) 1f detection: lock-in time constant 50 ms, (c) 3f detection: lock-in time constant 50 ms

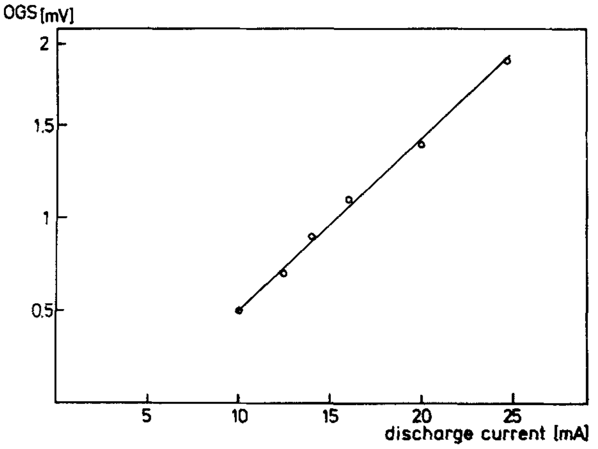

Fig. 3. Optogalvanic signal amplitude (peak-to-peak) as a function of discharge current

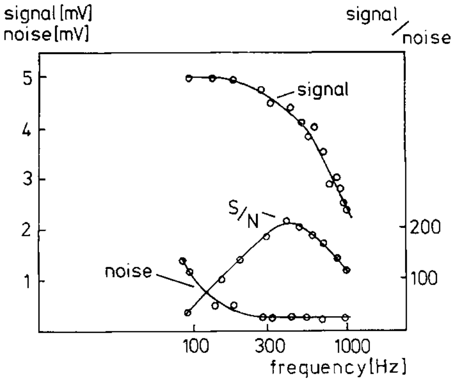

Fig. 4. Signal, noise, and signal-to-noise ratio of the optogalvanic signal as a function of modulation frequency taken at 25 mA discharge current

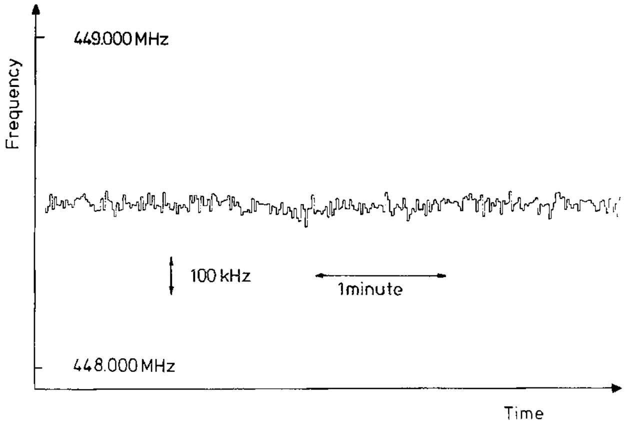

Fig. 5. Recorder trace of the frequency measurements between two different CO laser lines. Each measurement was taken with 1 s averaging time

increase drastically and do not permit the locking of the laser. Currents in excess of 25 mA could not be attained with the power supply.

The dependence of the optogalvanic signal on the modulation frequency is shown in Fig. 4. Although the noise on the signal remains almost constant above 200 Hz, the signal to noise exhibits a peak near 450 Hz; the process which leads to this low cut-off frequency must still be investigated. For optimum S/N ratio, we adjusted the detection frequency to 420 Hz and, correspondingly, the modulation frequency to 140 Hz for the third derivative technique.

Frequency Stability and Reproducibility

In order to better test the absolute frequency stability of the laser, we measured the beat frequency between two identical Lamb-dip stabilized CO lasers by heterodyning them on a metal-insulator-metal (MIM) diode. Reliable results could not be obtained with two ~~identical laser lines as there was injection locking due~~ to optical feedback. The smallest frequency difference for equal gas fillings in each laser that could be obtained was 40 GHz: this was determined by the resolving power of the intracavity grating in each laser system. This difference frequency was too high for the present available signal processing unit, and we would have had to radiate the MIM diode with microwave to generate a low difference frequency. This would have increased the mixing order and decreased the signal-to-noise ratio.

We overcame this problem by using two different isotopic species in the two laser systems. The following pair of laser lines brings the beat note below 500 MHz:

$$^{12}C^{16}O: P(10)_{14-13}\,53.039032(10)\,\mathrm{THz},$$

$$^{12}C^{18}O: P(14)_{12-11}\,53.038585(10)\,\mathrm{THz}.$$

These frequencies were calculated from Dunham coefficients published in Guelachvili et al. [8] who measured the frequencies of the Doppler-limited CO transitions.

A four minute trace of the frequency difference is shown in Fig. 5, the measurements were taken with one second averaging. With $3f$ detection and lock, the measured beat frequency showed a long term drift of 140 kHz in 5 h. Assuming both lasers were independent from each other this indicates a $140/\sqrt{2}$ kHz or 100 kHz drift in each laser. With 1f detection in both lasers the corresponding long term drift was about 800 kHz per each laser in several hours.

With 3f locking, the beat note between the cells was 448.430(70) MHz at 1.3 Torr pressure. This is in good agreement with the measurements of Guelachvili et al. 447(15) MHz using their Doppler-limited results [8]. This beat frequency could not be changed by relocking, by exchanging the isotopes between the lasers, by exchanging the electronic detection and locking circuitry, nor by changing the discharge currents of the low-pressure cells.

A pressure shift of about −300 kHz/Torr for each laser was measured for each of these two laser lines. This pressure shift produces a systematic shift of several MHz in the frequencies of CO lasers which operate at discharge pressures of 10–20 Torr and are stabilized on top of their gain curves.

Conclusions

The CO laser can be Lamb-dip stabilized, preserving power and spectral bandwidth, by means of a low-pressure absorption cell using the optogalvanic effect to detect the saturation signal. The difference-frequency between two stabilized lasers oscillating on different isotopes was measured indicating a reproducibility of each laser frequency of better than

100 kHz. Precise measurements of the absolute laser frequencies and detailed examination of their pressure dependence by using CO_2 frequency standards are in progress.

CO frequencies that can be used as stable and reproducible references will open up new possibilities for high resolution ir spectroscopy and precision frequency measurements. Until now, the CO laser was used as a transfer oscillator and the frequencies had to be measured simultaneously by frequency mixing with CO_2 lasers [9, 10]. It will now be possible to use the CO lines as accurate references themselves. A first sum-frequency mixing experiment with two CO lasers synthesizing frequencies in the 113 THz region is in preparation. Heterodyning a color center laser oscillating near 113 THz (2.6 μm) with the two CO lasers will provide accurate frequency data for high resolution Doppler-free spectroscopy.

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