Characterization of an external cavity diode laser based ring cavity NICE-OHMS system


Department of Chemistry, Physical and Theoretical Chemistry Laboratory, University of Oxford, South Parks Road, Oxford OX1 3QZ, UK
*gus.hancock@chem.ox.ac.uk

Abstract: The performance of an external cavity diode laser based noise immune cavity enhanced optical heterodyne molecular spectrometer is presented. To reduce the noise on the signal a ring cavity and a circuit to remove residual amplitude modulation on the pre-cavity laser radiation was implemented. We demonstrate a sensitivity of $4 \times 10^{-11} \text{ cm}^{-1} \text{ Hz}^{-1/2}$ using a cavity with a finesse of 2600 on a Doppler-broadened transition of CH$_4$ at 6610.063 cm$^{-1}$.

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References and links


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1. Introduction

Noise-immune cavity-enhanced optical heterodyne molecular spectroscopy (NICE-OHMS) combines cavity enhanced spectroscopy with frequency modulation spectroscopy (FMS). This results in an ultrasensitive technique for which an absorption sensitivity of $1 \times 10^{-14}$ cm$^{-1}$ has been demonstrated [1]. The operating principle behind the NICE-OHMS technique is to use FM techniques to detect molecular absorption and the associated dispersion occurring inside an optical cavity, by modulating the injected radiation at the same frequency as the cavity free spectral range (FSR) (or multiples thereof). With cavity enhanced absorption, small variations in the laser frequency with respect to a cavity mode lead to fluctuations in the amplitude and phase of the transmitted light. In NICE-OHMS the fluctuations in the transmitted carrier laser frequency are mirrored by fluctuations on the sideband frequencies, which are transmitted by adjacent longitudinal cavity modes. Accordingly, the transmitted light is still an FM triplet with minimal amplitude modulation. This allows the overall noise level to approach the fundamental noise level of the laser.

Although a sensitivity of $1 \times 10^{-14}$ cm$^{-1}$ close to the shot noise limit has been demonstrated, subsequently NICE-OHMS has been used to perform Doppler and pressure broadened [2–8] and sub-Doppler spectroscopy [9–12], resulting in sensitivities in the $10^{-10}$-$10^{-11}$ cm$^{-1}$ range. Only a few NICE-OHMS setups have used an external cavity diode laser (ECDL) [5–7,9], primarily because they are more difficult to lock than the very low noise solid state and fiber lasers used in other experiments. However, ECDLs have the advantage that they are relatively cheap, compact, easily tunable and controllable devices, and available over a wide wavelength range allowing the observation of a wide range of molecular species. The application of NICE-OHMS to both Doppler and pressure broadened samples, where wavelength scans are performed over 100s of MHz, is limited by fluctuations in the level of residual amplitude modulation (RAM) and the fringes associated with etalons formed between or within optical components [4,7]. Reflections between the optical cavity mirrors and other optics, such as detectors and fibers, and reflections within single optical components, such as electro-optic modulators, fibers, and cavity mirrors, can be difficult to minimise.

In this paper, we present an external cavity diode laser based NICE-OMHS spectrometer with a ring cavity and a RAM reduction circuit to improve sensitivity. The ring cavity, in a bow-tie configuration, reduces the effect of etalons, while also eliminating any unwanted feedback effects resulting from the direct reflection associated with a linear cavity. In most NICE-OHMS experiments, the spectrometer is used for Doppler-free spectroscopy in which counter propagating beams within a linear cavity are required. Etalon fringes arising from the cavity mirrors occur because of the precise on axis alignment. However, for scanning over pressure broadened line shapes, saturation of the transitions becomes less likely as collisions within the gas increase the homogeneous width of the transition and hence greatly increase saturation intensities. Therefore, the counter-propagating beams are no longer required and alternative cavities can be used. Assuming that we have reduced the cavity etalons with the ring cavity arrangement, the remaining etalons are likely to be caused by the optical components before the optical cavity. In NICE-OHMS, etalon fringes are detected because they convert a fraction of the frequency modulation to fringe-induced amplitude modulation (FIAM), a form of RAM. Like RAM, produced as a side effect of modulation, this is present in the radiation before the cavity. Thus we use a second detector to monitor the laser radiation before it is incident on the cavity and to record the combined level of RAM and FIAM. We demonstrate a method of using this pre-cavity FMS signal to improve the sensitivity of the
NICE-OHMS apparatus by the use of a RAM reduction circuit on the pre-cavity laser radiation. This signal can be processed and used as an error signal for the purposeful production of RAM with opposite sign, thus providing a stable background signal. The RAM produced by the RAM reduction circuit matches the dispersion phase of the FM and NICE-OHMS signals. Thus, the demodulation phase of both the pre-cavity FMS signal and the NICE-OHMS signal were set to dispersion.

2. Experimental details

A schematic of the experimental setup is shown in Fig. 1. The light source was an external cavity diode laser (ECDL) (Sacher Lasertechnik LION, linewidth < 2 MHz) tunable in the range 1480 – 1540 nm with an output power of 3.2 mW providing a mode-hop free tuning range of 56 GHz using current coupling. The laser light was first directed through an optical isolator to prevent optical feedback. Two beam shaping lenses were employed to mode match the light to the TEM$_{00}$ mode of the cavity. Following these lenses two electro-optic modulators (EOMs) were situated: EOM1 (Nova Phase Electro-Optic Phase Modulator (EO-PM-NR-C3)) to modulate the light for locking of the laser frequency to a TEM$_{00}$ mode of the cavity, and EOM2 (Quantum Technologies (TWAP 10-1350-1650nm)) for the modulation at the cavity free spectral range and post cavity FM detection. To isolate the alignment of the cavity from adjustments to the laser and EOMs, as well as providing a clean laser beam, predominantly Gaussian shaped, the light was fed through a single mode polarization preserving fiber (OZ Optics). The resulting power incident on the cavity was 1 mW. The wavelength of the laser was monitored with a wavemeter (Burleigh WA-1000) and a frequency scale for the data was defined by monitoring the laser output with a spectrum analyser (10 GHz Melles Griot 1300-1500 nm, adjusted for 0.81 GHz).

The ring cavity was formed by four high reflectivity mirrors (1 inch diameter, a radius of curvature of r = 1 m) in a bow tie configuration with a half-round trip, $L$, of 137 cm and a cavity FSR of 109.5 MHz. The specified reflectivity of the input and output coupling mirrors is 0.9994 (Newport 10CV00SR.70T), and that of the other two mirrors is 0.9997 (10CV00SR.70F). The two highest reflectivity mirrors were mounted on piezoelectric transducers (PZTs): PZT1 (PiezoMechanik HPSt 150/20-15/55) was used for scanning the cavity length and PZT2 (Physik Instrumente GmbH P-622.10L) was used for the faster modulation of the cavity length for wavelength modulation spectroscopy (WMS). The vacuum chamber housing the ring cavity was evacuated to 10$^{-4}$ Torr before it was filled with a
given pressure of CH$_4$, monitored with a capacitance manometer (MKS instruments 722A11TBA2FA, 0-10 Torr). To avoid etalons, the wedged windows on the housing were mounted at a slight angle and antireflection coated for 1050 to 1600 nm.

For the locking of the laser frequency to a cavity TEM$_{00}$ mode and the FM frequency to twice the cavity FSR, three linked frequencies were generated at 25, 219, and 244 MHz. Two voltage-controlled oscillators (VCO) (Mini-Circuits JTOS-300) provided 219 and 244 MHz signals, which were mixed and filtered to produce the 25 MHz difference frequency. The laser frequency was locked to a TEM$_{00}$ mode of the cavity using the Pound-Drever-Hall (PDH) method [13], with EOM1 used to produce sidebands at ± 25 MHz on the laser frequency. The laser light reflected from the cavity was directed onto a fast photodetector (PD1) (New Focus 1611) and amplified (Mini-Circuits ZFL-500LN), and demodulated at 25 MHz using a double balance mixer (Mini-Circuits ZAD-1) to generate the PDH error signal. This was processed by a fast (MHz) locking circuit with a bandwidth of ~2 MHz, and fed back to the laser injection current for primary locking of the laser frequency to the cavity. A slow locking circuit follows the change in cavity length and generates a signal which is fed back to the laser PZT input for the ECDL. The bandwidth when the laser is locked is necessarily better than ~50 kHz.

With the laser locked to the cavity, the cavity length was changed such that the laser frequency was scanned up to 12 GHz by applying a sinusoidal voltage ramp, at a frequency of 1 Hz, to PZT1. A feed forward loop was implemented in the locking scheme to reduce the load placed on the locking electronics and increase the scan rate for which mode-hop free tuning range of the laser was maintained. The transmission of the cavity was monitored by a fast photodetector (PD2) (New Focus 1611) with a bandwidth of 1 GHz. The dc output of the detector was recorded in a locked cavity enhanced absorption spectroscopy (CEAS) experiment with a digital oscilloscope (Lecroy 9304) and stored on a computer to determine the cavity finesse. For the determination of the cavity finesse 8.1 Torr of methane was introduced into the cavity and the methane spectrum for an unassigned CH$_4$ transition at 6595.90 cm$^{-1}$ with a line strength $S$ of 1.115 x 10$^{-25}$ cm$^2$ cm$^{-1}$ was recorded with 20 averages. A mean mirror reflectivity $R$ of 0.9988 ± 0.0001, and thus a finesse of 2600 ± 200, was calculated from the area ($A$) under the locked CEAS data using $1-R = S$c$L/A$, where $c$ is the concentration in cm$^{-3}$ and $L$ is the half round trip length of the cavity.

To perform $fm$-NICE-OHMS, EOM 2 frequency modulates the laser light at $f_{FM} \sim$219 MHz in order to generate sidebands which are ± 2 FSRs of the cavity for FM detection. However, the FSR of the cavity changes slightly during a frequency scan over an absorption as the length of the cavity changes. The noise-immune aspect of NICE-OHMS requires a good match between the modulation frequency and FSR of the cavity. To lock the modulation frequency to the FSR we used the DeVoe and Brewer method [14]. A fraction of the amplified signal from PD1 is split off (Mini-Circuits ZFSC-2-1-S) and demodulated with a double balance mixer (Mini-Circuits ZAD-1) at 244 MHz (the sum of the cavity FSR and the PDH locking frequency) to produce an error signal for locking the FM frequency to the cavity FSR. This error signal was fed into a proportional-integral locking circuit to adjust the tuning voltage on the VCO and thus the modulation frequency of EOM2 to match twice the free spectral range.

To produce the $fm$-NICE-OHMS signal, the ac output of PD2 was amplified (Mini-Circuits ZKI-1R5 and ZFL500HLN), high-pass filtered (Mini-Circuits SHP-175), before being demodulated at the $f_{FM}$ with a double balance mixer (Mini-Circuits ZDM-1W-S). This signal was passed through a 100 Hz low-pass filter and recorded using the same oscilloscope and data acquisition system. Changing the demodulation phase allowed either the absorption or dispersion FMS signal to be acquired.

The signal at this stage is affected by RAM noise resulting from imperfections in the EOMs and etalons before the cavity. In order to remove the RAM, an electronic circuit on the FMS dispersion signal was implemented based on the method described by Wong et al. [15]. The RAM on the laser radiation before the cavity was monitored by using a beam splitter to direct 8% of the laser radiation onto the fast photodetector PD3. The ac output of PD3 was
amplified (Mini-Circuits ZKI-1R5 and ZFL1000LN), and high-pass filtered (Mini-Circuits SHP-175), before being demodulated at $f_{FM}$ with a double balance mixer (Mini-Circuits ZDM-1W-S). The RAM detected in this way provided an error signal to stabilize the level of RAM prior to the cavity, by using EOM2 to create amplitude modulation (AM) with the opposite sign. For this, two polarizers, with their angle set at 10 degrees from vertical, were positioned at either side of EOM2. A bias voltage applied to the EOM produced AM at the same phase as the FMS dispersion signal, and feedback of the processed error signal controlled the RAM level at this phase. By stabilizing the RAM observed before the cavity, a flatter, less noisy baseline on the $fm$-NICE-OHMS dispersion signal can be achieved, thus increasing the sensitivity.

With this more stable $fm$-NICE-OHMS dispersion signal, the detection sensitivity was further improved by applying an additional modulation to the cavity in the form of WMS. The cavity length and, via the locking circuit, the laser frequency, was dithered using PZT2 at a low frequency of 60 Hz with a frequency excursion of 212 MHz. To produce this $wm$-NICE-OHMS signal, the $fm$-NICE-OHMS signal was demodulated at 60 Hz using a lock-in amplifier (EG&G 7265 DSP). Demodulation of the signal occurred with a time constant of 50 ms, corresponding to a bandwidth of 3.2 Hz. As the RAM removal circuit worked only at the dispersion phase of the signal, all of the $wm$-NICE-OHMS signals recorded are dispersion signals. Again, the processed signals were recorded with the oscilloscope and passed to a computer for analysis.

3. Results and discussion

![Fig. 2. – (a) $fm$-NICE-OHMS signal with the RAM reduction circuit on and off for a 0.2 Torr sample of CH$_4$ at 6610.063 cm$^{-1}$ (traces offset for clarity), (b) $wm$-NICE-OHMS signal for a 0.04 Torr sample of CH$_4$ probing the same line.](image)

To illustrate the effect of the RAM reduction circuit on the $fm$-NICE-OHMS dispersion signal, we recorded a spectrum of CH$_4$ for an unassigned transition at 6610.063 cm$^{-1}$ [16,17]. The resulting dispersion signals with RAM reduction circuit on or off are presented in Fig. 2a. The signal-to-noise (S/N) ratio with the RAM circuit off is 85 (where the noise is defined as the standard deviation of the noise on the baseline). When the RAM reduction circuit is in operation, a noticeable reduction in noise on the dispersion signal can be observed. The S/N ratio improves to 177. This demonstrates the usefulness and benefits of the RAM reduction circuit as noise which is present on the $fm$-NICE-OHMS signal would be carried forward in the $wm$-NICE-OHMS signal, and would manifest itself as a limitation to the final sensitivity obtained. The effect of the RAM circuit improves the S/N ratio to an even greater extent when operating the ECDL at different wavelengths where etalons and noise sources are more significant. We note that despite the RAM reduction circuit an offset in the baseline signal is observed. This offset is relatively stable during the measuring period and is a result of an equipment based limitation in the range of our feedback to the EOM. To overcome this, an
offset voltage is added to the error signal to stabilize the RAM to a non-zero value; any further persistent offset may arise from changes in laser characteristics across a scan.

Having demonstrated the benefits of the RAM reduction circuit in operation, we now move to the full \textit{wm-}NICE-OHMS setup and present the first results of the technique using both a ring cavity and a RAM reduction circuit. Figure 2b shows the \textit{wm-}NICE-OHMS dispersion signal for 0.04 Torr of CH$_4$ recorded for the transition at 6610.063 cm$^{-1}$ with a line strength, \( S \), of 7.56 x 10$^{-25}$ cm$^2$ cm$^{-1}$. This NICE-OHMS signal was acquired again by scanning over 8 GHz at a scan rate of 0.26 Hz. The signal was acquired with 6 averages over an acquisition time of 10 seconds. We found that this was the optimal acquisition time to improve the S/N ratio limited by longer term laser drift problems. The S/N ratio of the signal is around 1600 which gives a minimum detectable absorption \( \alpha_{\min} \) (under optimal conditions) of 3 x 10$^{-11}$ cm$^{-1}$, highlighting significant improvements over the typical \textit{fm-}NICE-OHMS sensitivities obtained with this instrument of \(~10^{-10}\) cm$^{-1}$. The noise equivalent bandwidth reduced sensitivity is 4 x 10$^{-11}$ cm$^{-1}$ Hz$^{1/2}$, determined using \( \alpha_{\min}(BW) = \alpha_{\min} \sqrt{2\pi n} \), where \( \tau \) is the time constant on the lock-in amplifier and \( n \) is number of averages. This sensitivity is comparable with sensitivities obtained with other NICE-OHMS setups whilst having a lower finesse [18], and has the added advantage of operating over wider range of wavelengths, offering the opportunity to access larger regions of the spectrum. The theoretical minimum detectable absorption [19] (i.e. the shot-noise limit) of the \textit{wm-}NICE-OHMS setup is given by

\[
(\alpha L)_{\min} = \frac{\pi}{2F} \left( \frac{2e}{\eta P_0} \right)^{1/2} \frac{\sqrt{2}}{J_0(\beta) J_1(\beta)},
\]

where \( F \) is the cavity finesse, \( e \) is the electron charge, \( \eta \) is the responsivity of the photodetector in A/W, and \( P_0 \) is the power incident on the photodetector in the absence of absorbing media, \( J_0(\beta) \) and \( J_1(\beta) \) are the nth-order Bessel functions and \( \beta \) is the modulation index. Here, \( \eta = 1 \) A/W, \( F = 2600 \), \( P_0 \sim 0.3 \) mW (cavity transmission of 30%), \( \beta = 0.55 \), which for a cavity of length 137 cm, gives a minimum detectable absorption corresponding to \( \alpha_{\min} = 8.9 \times 10^{-13} \) cm$^{-1}$ Hz$^{-1/2}$. Thus the sensitivity reported here is a factor 45 above the shot-noise limit. The remaining baseline noise is composed of random background fluctuations and they are onerous to eliminate entirely.

4. Conclusions

We have presented the performance of an external cavity diode laser based noise immune cavity enhanced optical heterodyne molecular spectrometer (NICE-OHMS) with a ring cavity by measurements on a weak Doppler-broadened transition of CH$_4$ at 6610.063 cm$^{-1}$. The ECDL has the advantage of operating over wider range of wavelengths, offering the opportunity to access larger regions of the spectrum. We have demonstrated that the use of a circuit to remove residual amplitude modulation on the pre-cavity laser radiation improves the sensitivity of \textit{fm-}NICE-OHMS in the dispersion phase by a factor of 2, resulting in a sensitivity of 1.3 x 10$^{-9}$ cm$^{-1}$. We achieved a sensitivity limit of 3 x 10$^{-11}$ cm$^{-1}$ for \textit{wm-}NICE-OHMS. The sensitivity reported here has been attained with a relatively low finesse of the 2600. As such, the sensitivity achieved may be improved with a higher finesse cavity with the use of higher reflectivity mirrors.

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