Optical Absorption Spectrometry using Laser Amplitude Modulation

Jong H. Chow, Andrew R. Wade
Conor Mow-Lowry, David S. Rabeling
Ian C.M. Littler and David E. McClelland
Centre for Gravitational Physics
Research School of Physics and Engineering
Australian National University
Canberra, ACT 0200, Australia
Email: jong.chow@anu.edu.au

Malcolm B. Gray
Length, Time and Optical Standards
National Measurement Institute
Bradfield Road, West Lindfield, NSW 2070, Australia

***Abstract*— We present a new technique for cavity enhanced optical absorption spectrometry, employing a radio-frequency amplitude modulated laser to interrogate an optical cavity. It is capable of sensing molecular absorption at the fundamental laser shot noise limit, and is able to perform quantum noise limited gas sensing at atmospheric pressure. We implement this technique, for the first time, in a free space cavity, and demonstrate that the demodulated error signal can be used to measure losses in a resonator.**

I. Introduction

The field of spectroscopy was revolutionised with the advent of lasers, which provide a high purity light source to facilitate a dramatic improvement in measurement precision. Today, optical absorption spectrometry employing high performance lasers is an active branch of research and development within the broader family of spectroscopy [1], and finds important applications in biochemical sensing and environmental monitoring. For high resolution detection of molecular absorption the compact size, low cost, and suitability for field deployment of optical spectrometers are significant advantages over large scale mass spectrometers. One of the main challenges in high precision optical absorption spectrometry is to attain ever lower detection thresholds, with the fundamental quantum limit of laser shot noise as the ultimate goal.

To reach the quantum limit of molecular absorption measurement sensitivity, two key factors are required: amplification of the minute absorption effects; and mitigation of technical noise which may pollute the detected signal [2]. Optical cavities are used as transducers to amplify the absorption signal by increasing the effective interaction length with the sample. To reduce the instrument technical noise in a cavity enhanced system, it is important to utilise a stable laser source, while employing active frequency locking to avoid the effects of mechanical noise.

The most common methodology for absorption spectrometry that meets these requirements is the cavity-ring-down spectroscopy (CRDS) technique. It typically employs high finesse cavities with premium quality mirrors. There are several variants to CRDS, but most involve some form of high speed shutter for the input laser beam, with the absorption metrology derived from the cavity output ring-down time after the shutter is closed. Of these, the experiment by Spence et al. [3] is at the forefront, reaching a sensitivity of $3.7 \times 10^{-10}/\sqrt{\text{Hz}}$ by interrogating a weak CO_2 transition at 1064 nm using a cavity with a finesse of 12,000.

The standard bearer for high sensitivity cavity enhanced absorption spectrometry is the NICE-OHMS technique [4], which employs radio-frequency (RF) modulation of the interrogating laser. The absorption signal is derived from differential attenuation and phase rotation of phase modulation sidebands, which relies on narrow absorption transitions in low-pressure gases to achieve quantum noise limited sensitivity in the order of $1 \times 10^{-13}/\sqrt{\text{Hz}}$.

We present a new technique for cavity enhanced optical absorption spectrometry, employing a radio-frequency amplitude modulated laser to interrogate an optical cavity. The demodulated signal gives a direct measure of the coupling condition of a resonator, and provides a readout of intra-cavity losses or absorption. This technique is capable of sensing molecular absorption at the fundamental laser shot noise limit, and is able to perform quantum noise limited gas sensing at atmospheric pressure. However, this technique is useful for spectrometric sensing in both gaseous and liquid phase samples. In isotopic ratio measurements of molecules such as CO_2 and H_2O, it promises significant sensitivity improvement when compared with cavity ring-down spectroscopy and traditional mass spectrometry. We implement this technique, for the first time, in a free space cavity, and demonstrate that the demodulated error signal can be used to measure losses in a resonator.

II. Amplitude Modulation and the Cavity Coupling Condition

Consider a simple two mirror cavity composed of lossless mirrors with a lossy sample in between, interrogated by an amplitude modulated laser as shown in Fig. 1, where R_1 and R_2 are the reflectivities for the input and output couplers respectivity. We assume a sample loss or absorption coefficient

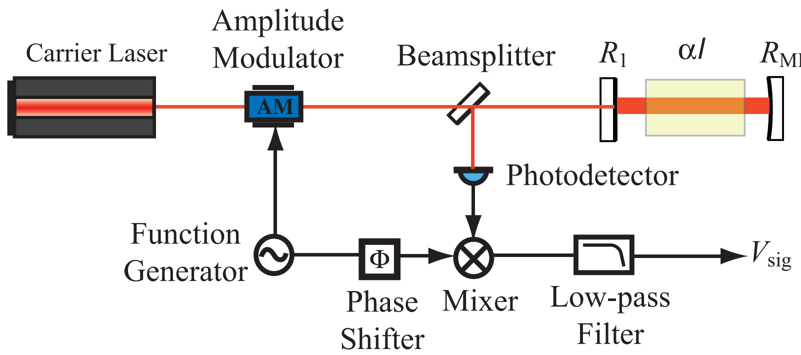
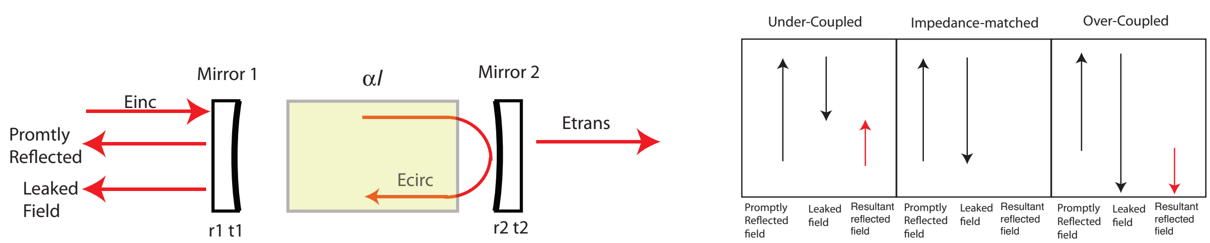


Fig. 1. A two-mirror resonant cavity interrogated by an amplitude modulated laser.



(a) The optical fields in a linear cavity.

(b) The phasors representing optical fields from a cavity in reflection.

Fig. 2. Schematic to illustrate the three coupling conditions of a linear optical cavity.

α, with sample length of l. The cavity coupling conditions can be categorized as [5]

$$r_1 > r_2 e^{-\alpha l} \quad \text{Under coupled} \tag{1}$$

$$r_1 < r_2 e^{-\alpha l} \quad \text{Over coupled} \tag{2}$$

$$r_1 = r_2 e^{-\alpha l} \quad \text{Impedance matched} \tag{3}$$

A linear cavity is impedance matched when the promptly reflected field is completely cancelled by the leaked circulating field from within the cavity. On resonance the leakage field from a cavity is π radians out of phase with the promptly reflected light. The three possible impedance conditions are illustrated schematically in Fig. 2, where Fig. 2(a) shows the respective optical fields of a linear cavity, while Fig. 2(b) illustrates the reflected light as summation of phasors.

We assume that the modulation frequency is well outside the resonance full-width half-maximum as illustrated by Fig. 3. We further assume that the laser carrier is kept on resonance by active feedback control using a technique such as the Pound-Drever-Hall frequency locking [6]. The reflected light is received by a photodetector, whose electronic signal is then demodulated with a local oscillator. Following the derivations as described in detail in Ref. [7], we obtain the demodulated error signal voltage for a lossless cavity:

$$V_{\text{sig}} \approx \rho\beta P_{\text{opt}} R_{\text{pd}} \frac{r_1 - r_2 e^{-\alpha l}}{1 - r_1 r_2 e^{-\alpha l}}, \tag{4}$$

where ρ is the photodetector responsivity (Amps/Watt); β is the modulation depth, such that $\beta \ll 1$; P_{opt} is the incident laser power; and $r_1 = \sqrt{R_1}$, $r_2 = \sqrt{R_2}$ are the amplitude reflection coefficients of the two cavity mirrors.

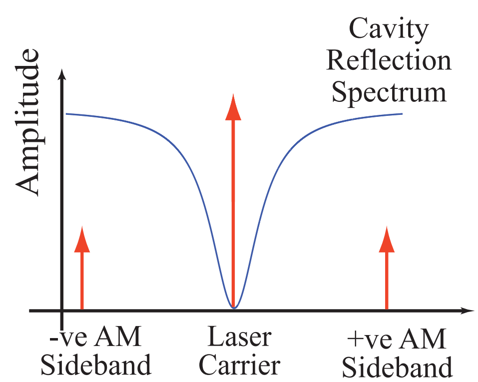


Fig. 3. A two-mirror resonant cavity interrogated by an amplitude modulated laser, where the sidebands are outside of the full-width half-maximum.

Near the impedance matching condition, $r_1 \approx r_2 e^{-\alpha l}$, and Eqn. 4 can be approximated by

$$\begin{aligned} V_{\text{sig}} &\approx \rho\beta P_{\text{opt}} R_{\text{pd}} \frac{r_1 - r_2 e^{-\alpha l}}{1 - r_1^2} \\ \Delta V_{\text{sig}} &\propto \Delta\alpha l. \end{aligned} \tag{5}$$

Equation 5 implies that the magnitude and sign of the demodulated AM voltage is a direct measure of the coupling condition of the cavity on resonance, with its zero crossing occurring when the cavity is exactly impedance matched. This error signal can be used both as a readout for the change in intra-cavity loss or absorption, and as a feedback signal in a control loop to actively impedance match the cavity, provided there is a mechanism to vary the reflectivity of one of the mirrors.

We note that this technique works only if the laser carrier is resonant with the optical cavity. In practice, this is typically facilitated by an active feedback control loop, such as the Pound-Drever-Hall technique [6], to frequency lock the laser to a resonant mode of the cavity.

III. The Variable Loss Cavity

To test our amplitude modulation technique on a free space cavity, we used a linear cavity with variable intra-cavity loss. This loss was introduced by a knife edge mounted on an adjustable screw. The design and measured specifications are summarized in Fig. 4 and Table I [8]. By adjusting the screw, we were able to operate the cavity in the three coupling regimes described by Eqns. 1-3.

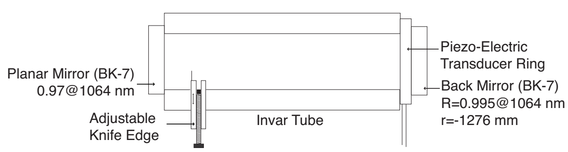


Fig. 4. A variable loss cavity, with spacer constructed with low thermal expansion invar. The intra-cavity loss was introduced by a screw mounted knife edge.

IV. The Experimental AM Error Signals

We obtained the experimental AM error signals by modulating the laser as shown in Fig. 1, and observing the demodulated voltage signal V_{sig} while scanning the the frequency of the laser across resonance. The amplitude modulator was modulated sinusoidally at 80 MHz with a function generator, which also acts as the local oscillator during demodulation. The output from the mixer is low-pass filtered to yield the low frequency variations in cavity loss or absorption. The frequency of the laser was scanned via its piezo electric transducer frequency tuner. Three coupling conditions were investigated: over coupled, under coupled, and impedance matched, by varying the intra-cavity loss via the screw mounted knife edge.

The experimental AM error signals for the variable loss cavity are summarized in Fig. 5. The red and blue traces are for the cases of over coupled and under coupled cavities, respectively, while the green trace is for the case of an impedance matched cavity. Figure 5a shows the reflected laser power as measured with a photodetector and observed with an oscilloscope when the laser frequency was scanned across resonance. For the case of minimum reflected power on resonance, we expect the cavity to be impedance matched. We note that although the reflected carrier power should be zero on resonance when impedance matched, the measured reflected intensity is not zero. This is due to the optical power in the radio frequency sidebands.

The AM error signal traces in Fig. 5b each exhibits three dips. The central dip occurs when the laser carrier is resonant, while the two satellite dips occurs when the sidebands become resonant. The region in the error signals of interest to spectroscopy is when the carrier is on resonance. It can be seen that the error signal is zero on resonance when the cavity is impedance matched, negative when under coupled, and positive when over coupled. This polarity dependence on the coupling condition is consistent with the predictions of Eqn. 5.

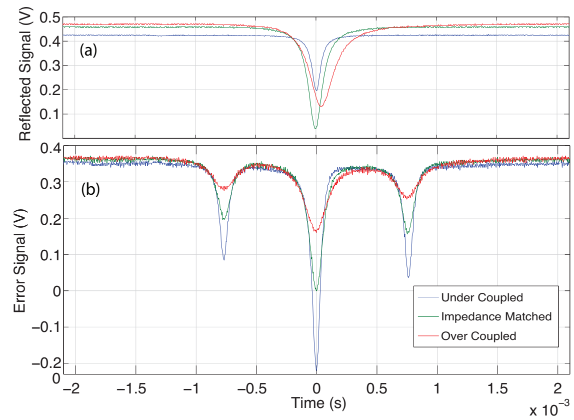


Fig. 5. Impedance readout error signal for the three cases of under-coupled, over-coupled, and impedance matched, with reflected signal curves

V. Impedance Matching Readouts

The complete schematic of the experiment, with both Pound-Drever-Hall frequency locking, as well as impedance matching readout, is illustrated in Fig. 6. The Pound-Drever-Hall technique [6] is an active feedback control loop, which uses phase modulation interferometry to keep the laser resonant with the cavity. The error signal from the frequency locking loop was used by a feedback servo to provide a control signal to the frequency tuning piezo electric transducer of the cavity, thereby compensating for any frequency excursions between the laser and the cavity, and tracks any frequency jitters and drifts in the laser.

With the cavity frequency locked to the laser, and the AM interferometric readout turned on, we observed the AM error signal on resonance as we slowly adjusted the screw mounted knife-edge, as we continuously varied the intra-cavity loss. The experimental result is displayed in Fig. 7, where the green trace is the reflected optical power from the cavity as measured with a photodetector and recorded with a digital oscilloscope. The blue trace is the demodulated AM error signal. As the knife edge was inserted into the cavity, the intra-cavity losses increased. Hence the cavity changed from under coupled to over coupled. We see that when the cavity is impedance matched, where we observet a minima in reflected power, the AM error signal crossed zero, in accordance with Eqn. 5.

TABLE I

SUMMARY OF CAVITY PROPERTIES

Cavity Property	Design Value	Measured Value
Mirror 1 Intensity Reflectivity	$R_1 = 0.97$	$R_1 = 0.9716 \pm 0.0005$
Mirror 1 Intensity Loss Factor	$L_1 = 0$	$L_1 = 0.0008 \pm 0.0005$
Mirror 2 Intensity Reflectivity	$R_2 = 0.995$	$R_2 = 0.9933 \pm 0.0005$
Mirror 2 Intensity Loss Factor	$L_2 = 0$	$L_2 = 0.0004 \pm 0.0005$
Free Spectral Range	1.5 GHz	1.500 ± 0.0005 GHz
HWHM (ν_F)	4.23 MHz	4.24 ± 0.05MHz
Finesse ($\mathcal{F}$)	177	177 ± 1
Transverse Mode Spacing ($\delta\nu$)	136 MHz	140 ± 10MHz
Rayleigh Range	$z_0 = 343$ mm	-
Waist Radius	$W_0 = 341\mu m$	-

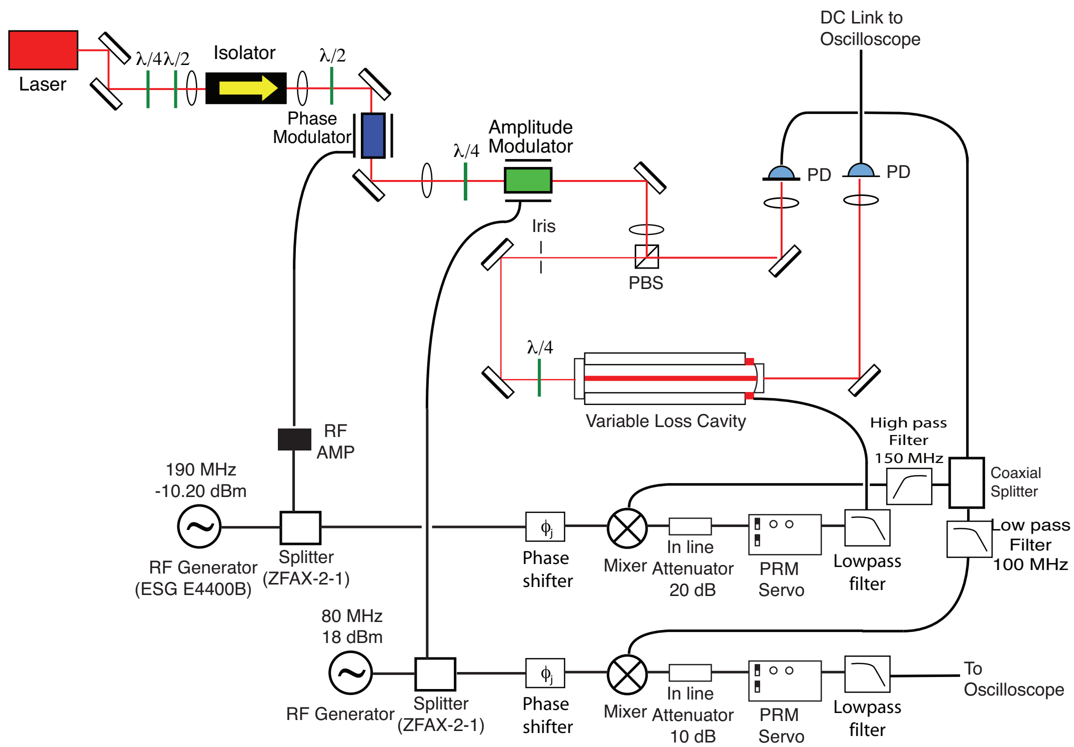


Fig. 6. Modified experimental setup to measure the impedance response function.

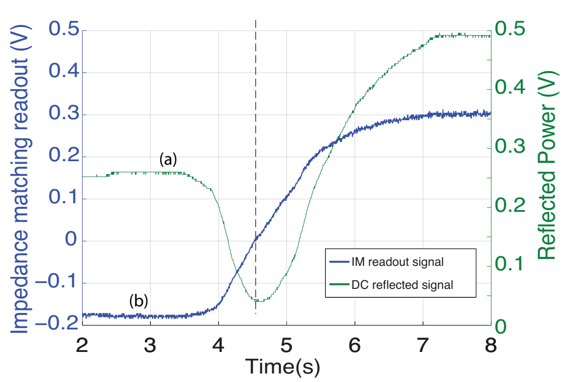


Fig. 7. Experimental AM error signal while the laser is resonant. Trace (a) is the reflected power from the cavity, while Trace B is the corresponding AM error signal. The cavity was adjusted from under coupled, through impedance matched, to over coupled.

VI. CONCLUDING REMARKS

We have presented a cavity enhanced amplitude modulation technique for detecting intra-cavity losses and absorption. We have demonstrated this technique in a free space cavity for the first time, by introducing intra-cavity losses with an adjustable knife edge. We have shown that the AM error signal voltage depends on the cavity losses. Its zero crossing corresponds to an impedance matched cavity, while the voltage polarity depends on whether the cavity is over or under coupled. This technique, therefore, can be used to extract loss or absorption signals with the demodulated AM error signal in free space cavities.

When compared with cavity ring down spectroscopy, this technique is capable of reaching the limit of laser shot noise [7], as it is a continuous-wave measurement and the noise bandwidth can be arbitrarily small, thus significantly improving sensitivity. However, similar to cavity ring down spectroscopy, but in contrast to NICE-OHMS, the AM technique can work with either narrowband or broadband losses or absorption transitions. This opens up a wide range of potential applications including: pressure broadened gases, liquids, and high clarity solids such as ultra-pure fused silica.

Finally, since V_{sig} is a measure of the cavity coupling condition, it can also be used by a servo to implement an active impedance matching feedback control loop by actuating on mirror R_1 in Fig. 1. The feedback loop then corrects for any changes in absorption to keep the cavity impedance matched throughout the measurement duration. When the cavity is impedance matched, the minimum detectable shot-noise limited absorption reaches its lowest value. Hence, with active impedance matching the sensitivity is always optimum.

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CONTACTS

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