Optimization of calibration-free wavelength modulation spectroscopy technique for gas parameter measurement

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Abstract: The residual amplitude modulation (RAM) method, which is the simplest wavelength modulation spectroscopy (WMS) method for calibration-free measurement of gas concentration and pressure, can be optimized by selecting a specific modulation frequency of the laser diode. If the laser diode is modulated at its phase quadrature frequency ($f_{\pi/2}$) at which the phase difference, ψ , between the intensity modulation (IM) and frequency modulation (FM) is 90°, the lineshape–bearing signal of interest is maximized for a given modulation index (m), instead of being scaled by $sin(\psi)$. The 1650nm laser diode used in this study has a $f_{\pi/2}$ of 100kHz which is an order of magnitude lower than values reported in the literature and well within the range of low-cost electronics. This aspect of optimizing the RAM method has not yet been reported in the literature perhaps because the RAM method is a fairly recent development. This paper demonstrates time-varying calibration-free measurement of methane concentration and pressure with the laser diode modulated at its phase quadrature frequency. The time resolution of measurements is currently 10s which is suitable for many industrial processes. The time resolution can be improved by using dedicated data acquisition and processing hardware.
Keywords [RAM, phase-quadrature, calibration-free, WMS]

1. Introduction

Tunable diode laser spectroscopy (TDLS) has emerged as the firm favorite in applications that require rapid, non-invasive, non-destructive, high-sensitivity and high-specificity gas detection and measurement of gas parameters such as concentration, pressure and temperature [1-6]. TDLS involves tuning the emission wavelength of a narrow-linewidth diode laser across one or more rotational-vibrational absorption lines of a gas and measuring the relative transmission. A variant of the basic TDLS approach is wavelength modulation spectroscopy (WMS), in which the laser wavelength is modulated through a high-frequency sinusoid superimposed on a low-frequency ramp. The resultant spectral modulation of the absorption line results in the generation of signal components at various harmonics (1f, 2f etc) of the modulation frequency (f_m) and a lock-in amplifier (LIA) is used for phase-sensitive harmonic detection of any given harmonic. The detected harmonic component can be related back to the absorption profile of the target species to infer gas parameters with a higher sensitivity than that achievable with direct detection. WMS is classified as 1f- and 2f-WMS based on the order of the harmonic of the modulation frequency used to infer gas parameters [7-12]. The problem of periodic instrument calibration was a long-standing challenge in TDLS that has now been well and truly overcome [13-15]. This has significantly enhanced the utility of TDLS for measurement of gas parameters for industrial process control where safety is critical and long down-times for maintenance are unacceptable.

The first of the two distinct calibration-free approaches comprises two closely related methods namely the residual amplitude modulation (RAM) method and the phasor decomposition (PD) method [13, 14]. Both of these techniques use 1f WMS to recover the absolute absorption lineshape but in slightly different ways. The gas parameters are extracted by fitting a simulated line shape to the experimental trace in both cases, with concentration and pressure as the two fitting parameters. Both methods have been demonstrated to be robust. The second calibration-free method is a 2f WMS method that uses normalization of the information-bearing 2f signal by the 1f signal to eliminate the effects of variations in laser intensity [15] due to variable system throughput. Although the RAM method is the simplest of all the calibration-free approaches, it has the limitation of low signal level for low values of the phase difference, ψ , between the intensity modulation (IM) and frequency modulation (FM). This value is typically not known before one purchases a laser and one will not be able to extract the full utility of the RAM method unless one exploits the frequency-dependence of ψ . This paper shows that by modulating the laser diode at a frequency $f_{\pi/2}$ at which ψ is 90°, the RAM signal can be maximized and the RAM method optimized for accurate lineshape recovery. This step of optimization brings the RAM method on par with other calibration free detection methods. For the laser used in this study $f_{\pi/2}$ is about 100kHz, which is way below typical values of 1.1MHz reported in the literature [16]. This makes this approach economical to implement with traditional laser driver and detection electronics whose cost rises sharply as the operating frequency increases.

2. The residual amplitude modulation (RAM) method

The RAM and PD methods are both based on the 1*f* WMS signal components as given by a simplified Taylor series expansion of the spectrally modulated transmission profile [13, 14]. The limitations of the Taylor series formulation have been addressed by others [16], but this simple picture has been used in this paper for the sake of clarity. For low values of m and for lasers with predominantly linear laser IM, the first three relevant terms are given by,

$$I_{1f} = \Delta I(\lambda_C)\left[1-\alpha(\lambda_C)\cdot C\cdot L\right]\cos(\omega_m t)+\ldots \\ I(\lambda_C)\frac{d\alpha(\lambda)}{d\lambda}\cdot C\cdot L\cos(\omega_m t+\psi) \quad (1)$$

Here C is the gas concentration and L is the interaction length. The first term is an undesirable concentration-independent background RAM signal that arises due to the laser IM that is synchronous with the FM. The second term is the concentration-dependent RAM term that carries information about the gas parameters (pressure and temperature) through the absorption coefficient $\alpha(\lambda_c)$. The last term is the concentration-dependent 1st derivative term (in the low modulation index approximation), also known as the IM-FM) term. Conventional 1*f* WMS was based on the recovery of this term and calibration was therefore a necessary step because the signal is dependent on the intensity which may vary depending on the coupling losses. The introduction of the 1*f* RAM method has changed this scenario entirely. In the RAM method the LIA measurement axis is aligned orthogonal to the third term thereby eliminating it. A projection of the RAM signal (second term) given by $I_{1f} = \Delta I(\lambda_c)\left[1-\alpha(\lambda_c)\cdot C\cdot L\right]\sin\psi$ is recovered. The user needs to be aware that the signal strength in the RAM method is scaled by the factor $sin(\psi)$ and hence only a projection of the full RAM signal is recovered. For lasers with a small value of ψ (depends on choice of f_m as well), the SNR may be too low for meaningful measurements to be made. The PD method gets around this problem by recovering the full RAM signal and is therefore not explicitly dependent on ψ. However, unlike the RAM method which uses the signal along only one of the LIA axes, it is necessary to use the signals recovered on both the LIA axes along with the value of ψ to recover the full absorption line. It would be advantageous to eliminate these additional steps for hardware implementation of the data acquisition and processing, more so for applications which need real-time measurements. In the PD method the LIA axes are oriented in such a way so that one of the axes is at phase quadrature to RAM signal and has a component $\left[I(\lambda_C)\frac{d\alpha(\lambda)}{d\lambda}CL\right]\sin\psi$ along it. However for low concentrations and low values of ψ, proper orientation of the LIA axes becomes difficult. An error in calculation of ψ causes an error in the extraction of concentration and pressure. This paper demonstrates experimental results of time-varying measurements of concentration and pressure using the RAM method at the phase-quadrature point. At this frequency, the RAM signal comprising the first and second terms in Eq 1 are maximized for any given value of m. For operation at $f_{\pi/2}$ therefore the RAM method becomes equivalent to the PD method. In both these methods there is an absorption-independent background RAM signal due to the laser IM. The level of this background signal can be significantly high for lasers with low value of current tuning coefficient and for operation at a high m-value (to maximize the 1*f* or 2*f* signal) particularly for high-pressures. The background RAM limits detection sensitivity but can be eliminated by RAM nulling [17], to enable low concentrations to be measured.

3. Experimental set-up and characterization of resonator and $f_{\pi/2}$

Figure 1 shows the experimental setup used for these studies. The output wavelength of a 1650nm edge-emitting laser (Toptica Photonics LD-1665-0010-DFB-1) which is temperature-stabilized using Thorlabs TED200C, is tuned with a current controller (Thorlabs LDC220C). A high-frequency sinusoid superimposed on a low frequency ramp signal generated by the arbitrary function generator (Tektronix AFG3022B), is used to modulate the laser. The output of the laser is divided into two parts using a 3-dB coupler (Thorlabs 10202A-50-APC). One part of the light is passed through a collimator (Thorlabs 50-1550-FC/APC) and then through the gas cell. The light is detected by a photo-detector (Thorlabs PDA10DT-EC) and the 1*f* WMS signals are recovered with the LIA (HF2LI, Zurich Instruments). The output from the LIA is captured by the digital oscilloscope (Tektronix TDS3054C). The electronic and data acquisition systems are controlled by a LabVIEW program. The other part of the laser is coupled to a fiber resonator with a free spectral range (FSR) of 0.2095GHz. The resonator is required for wavelength referencing of the time-indexed data captured by oscilloscope.

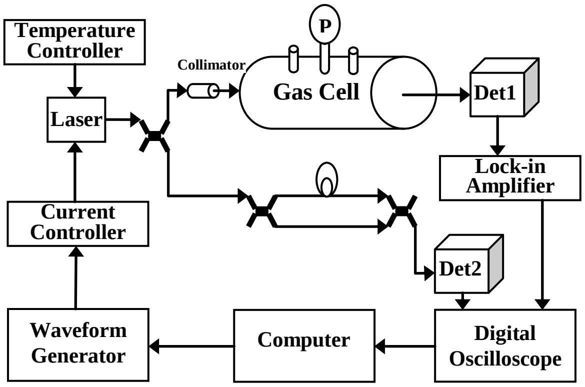

Fig. 1. Experimental setup up for obtaining absorption line and resonator output simultaneously to determine the concentration and pressure of the gas

Figure 2 shows the variation of ψ with f_m. The value of ψ was measured using the PD method [14]. The central result for this paper is that the phase quadrature point is attained at a very modest modulation frequency of about 100kHz. This is an order of magnitude lower than the frequency of 1.1 MHz reported by [16]. If the laser is modulated at $f_{\pi/2}$, the LIA can be phase-tuned to simultaneously obtain the full RAM signal along one axis and the full AM-FM signal along the other axis as shown in Fig. 3. This maximizes the recovered RAM signal for a given m-value. The concentration-dependent RAM signal increases with m and in fact, exceeds the FM signal strength for high m-values [16].

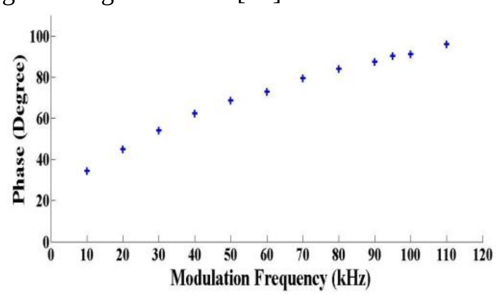

Fig. 2. Phase shift between IM and FM increases with increase in modulation frequency and reaches 90° at 100 kHz

4. Implementation of RAM nulling at $f_{\pi/2}$

The RAM signal recovered here has a high background which can be readily eliminated, as shown in the inset of Fig. 3, by implementing RAM nulling technique [18]. In optical RAM nulling, the laser output is split in to two parts, one part interacts with the gas while the other is passed through a fiber delay line that introduces a phase difference of π between the IM components on the two arms. When these two parts are recombined at the detector the background RAM (term 1 in Eq 1) is eliminated because the anti-phase IM components cancel, leaving the concentration-dependent RAM signal (term 2 in Eq 1) appears on a zero background. This signal can now be selectively amplified. It is important to use a fiber delay line that is much longer than the coherence length of the laser to avoid stable optical interference fringes. The path length, given by $\Delta L = c/2nf_m$, turns out to be 1km for f_m equal to 100kHz, which is much greater than typical coherence lengths that are on the order of a few hundred meters. For $f_{\pi/2}$ on the order of 1MHz, the path length reduces drastically to 100m. Optical interference fringes then become an issue and the simplicity of the method is lost. The low value of $f_{\pi/2}$ is therefore very useful because it permits one to use a manageable fiber length to implement RAM nulling and that is also long enough to avoid optical interference fringes.

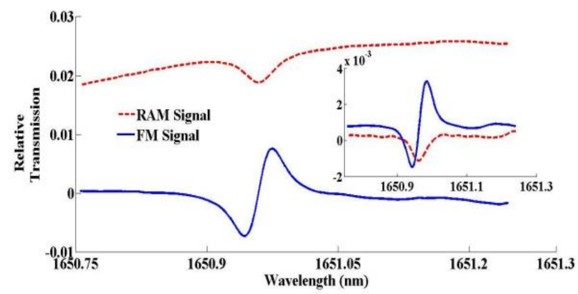

Fig. 3. AM-FM and RAM signals at the $f_{\pi/2}$ of 100kHz with inset showing the RAM nulled signals

5. Dynamic measurement of methane at $f_{\pi/2}$

Time-varying measurements of methane concentration and pressure were carried out with the laser modulated at $f_{\pi/2}$ = 100 kHz. Each acquired signal was normalized by a baseline generated from the spectral wings and the relative transmission profile was fitted with a Voigt profile simulated using spectral parameters from the HITRAN database [18]. The concentration and pressure of the simulated lineshape are varied iteratively to obtain minimum mean square error between the simulated and experimental lineshapes. Figure 4(a) and 4(b) show measurements of 1.2%, and 0.1% methane.

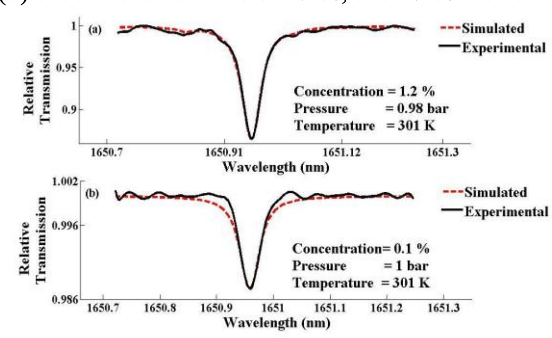

Fig. 4. Experimental and simulated lineshapes of (a) 1.2%, (b) 0.1% methane samples when operating at $f_{\pi/2}$

In order to obtain a varying concentration of methane, the gas cell was initially evacuated and flushed with nitrogen. A 10% methane balanced with nitrogen was flowed in for a short duration to increase the methane concentration in the gas cell. Then methane supply was then stopped and 99.9% nitrogen was flowed in the gas cell to decrease the methane concentration. In order to maintain a constant pressure of about 1 bar in the gas cell, one of the outlets was kept open throughout the process. The experimental lineshapes obtained at these time instants have been shown in Fig. 5. For the sake of clarity experimental lineshapes only at selected time instances have been shown. The variation of the mole fraction is shown in the inset of Fig. 5.

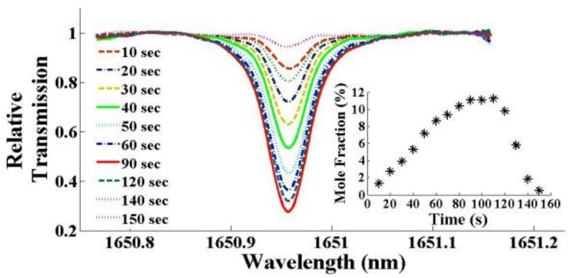

Fig. 5. Methane measurements at different time instants with a time resolution of 10 seconds

The time resolution of measurement of about 10s is clearly more than adequate for many industrial processes that do not change very rapidly. This can be improved further by using a dedicated a data acquisition system and a dedicated processor.

6. References

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