

# Modulation index optimization for wavelength modulation spectroscopy

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In this work, the second-harmonic component of wavelength modulation spectroscopy is simulated for  $R(22)$  CO<sub>2</sub> absorption line to investigate the effect of gas temperature and pressure on the modulation index. We found that the optimum value of modulation index, that is 2.2, is not affected by temperature but gas pressure will change the optimized modulation index. Specifically, when the gas pressure decreased to lower pressures of less than 100 mbar, the modulation index is also decreased and tended exponentially to about two. Accordingly, the optimum value of modulation index is recalculated for a range of CO<sub>2</sub> gas pressures to establish a nearly zero pressure deviation in the spectroscopy of very low pressure samples.

Keywords: wavelength modulation spectroscopy, NIR laser spectroscopy, molecular spectroscopy.

## 1. Introduction

Since the invention of tunable diode lasers (TDLs), their attractive characteristics have caused a rapid increase in the detection and quantification of many gaseous compounds in process control of industrial combustions [1], in medical diagnostics [2] and in security demands [3]. Particularly, in the near-infrared (NIR) region of spectrum many NIR-TDL-based gas sensors have been developed and commercially fabricated for quantitative monitoring of many hazardous molecules such as CO, CO<sub>2</sub> and NO<sub>x</sub> in a variety of mixed samples [4, 5]. In order to increase the sensitivity of device down to ppt level, numerous spectroscopic methods have been increasingly grown in the past 25 years to combine with unique properties of TDLs such as narrow line width of ~50 kHz and long-term stability. One example is the quartz enhanced photoacoustics spectroscopy (QEPAS) which is recently used for H<sub>2</sub>S detection based on an erbium-doped fiber amplifier source to improve the detection sensitivity by a factor of ~40 [6]. The capability of rapid tuning has made TDLs as a prime candidate for using in high performance spectroscopic schemes based on modulation methods in the kHz to MHz regime. In this field, wavelength modulation spectroscopy (WMS) has shown a very

robust and powerful technique for detection of low concentration spices with a remarkably increased signal-to-noise ratio (SNR) and sensitivity [7, 8]. By using WMS technique, characteristics of a gas sample are mathematically connected to the harmonic signals made by a lock-in amplifier from modulation frequency at the output. Owing to the technical and optical reasons, the second harmonic component of the modulated signal is of great importance because it provides the most intensive output at the line center and its analysis is simple. Therefore, much of experimental efforts have been directed toward making spectroscopic tools based on using WMS- $2f$  method [9]. And along with the experiment, theoretical investigations have been carried out to fully characterize the WMS- $2f$  method as an efficient means of reducing  $1/f$  noise. An important outcome of these investigations is the line shape function which is affected by the modulation index and depth. In WMS the modulation frequency is taken smaller than the laser line width while the modulation index is large. However, it is found that when the modulation index is set for 2.2, the peak value of  $2f$  line shape is maximized [10, 11]. Based on the above criteria, the performance of  $2f$  signal is further improved by removing the systematical errors imposed on the  $2f$  signal caused by a nonlinear response of laser intensity to the modulating signal. This is accomplished through normalizing  $2f$  signal by  $1f$  signal [12] which was firstly proposed to introduce a calibration-free method in order to obtain a background corrected signal for quantitative measurements in outdoor applications. This method which is named by common WMS- $2f/1f$  is recently developed by a heuristic apodized method in order to make this technique applicable for gas pressures beyond the thin optically condition which can be found in high pressure environments [13]. The performance of apodized WMS- $2f/1f$  is then experimentally realized by using a permeable distributed fiber as a sensing element and a NIR-DFB laser source that was modulated up to  $\sim 3$  kHz and by setting the modulation index at 2.2 in order to trace  $\text{CO}_2$  absorption kept at  $\sim 980$  torr [14]. However, a closer look at the WMS- $2f/1f$  approach shows that when the gas pressure is changed, a little deviation may occur in 2.2 value for  $2f$  signal which affects the peak value of the final signal. This in turn deteriorates the accuracy of WMS- $2f$  method in which the measured gas pressures largely deviate from the real one that was primarily provided by a gauge at the beginning of experiment. Therefore, it seems that the modulation index is a key parameter which must be respected and accurately determined for precise measurement of very low level concentrations.

In this work, the effect of gas pressure and temperature on the modulation index is theoretically studied and numerical simulation is performed for  $R(22)$   $\text{CO}_2$  absorption line centered at  $6363.727 \text{ cm}^{-1}$  [15]. We found that when the gas pressure is decreasingly changed by  $\sim 600$  mbar, the modulation index is accordingly changed and deviates from 2.2 by  $\sim 5\%$ . Such deviation cannot be ignored because it generates a systematic error of  $\sim 2.5\%$  in measuring real gas pressure of 50 mbar through scaling the WMS- $2f$  peak height. This specifically may cause a large error too in determination of very low concentration species in ppt regime.

## 2. Background theory

The theory of WMS method is fully discussed in numerous scientific documents and text books [16, 17]. Here, we confine ourselves to introduce a brief description of this approach in order to provide a systematic discussion. In WMS technique, the injection current of the utilized laser source is modulated by a sinusoidal wave up to a few tens of kHz [13, 14]. Such modulation is embedded by distinctive harmonics which can be discriminated at the detector output. By using a lock-in amplifier, the modulated signal is demodulated and a desirable harmonic will be extracted from the output. From the mathematical point of view,  $1f$  and  $2f$  signals are formulated as

$$X_{1f} = \frac{G\bar{I}_0}{2} \left[ H_1 + i_1 \left( 1 + H_0 + \frac{H_2}{2} \right) \cos(\psi_1) + \frac{i_2}{2} (H_1 + H_3) \cos(\psi_2) \right] \quad (1a)$$

$$X_{2f} = \frac{G\bar{I}_0}{2} \left[ H_2 + \frac{i_1}{2} (H_1 + H_3) \cos(\psi_1) + i_2 \left( 1 + H_0 + \frac{H_4}{2} \right) \cos(\psi_2) \right] \quad (1b)$$

where  $G$  is the electro-optical gain,  $\bar{I}_0$  is the average laser intensity over the modulation period,  $\psi_1$  and  $\psi_2$  are linear and nonlinear relative phase shifts, respectively, which may occur between laser intensity and reference sinusoidal frequency, and  $i_1$  and  $i_2$  are the linear and nonlinear modulation coefficients, respectively, which are connected to the intensity modulation of a laser beam. Here,  $H$  coefficients are the Fourier coefficients of the modulated signal which are

$$H_k (k > 0) = -\frac{1}{\pi} \int_{-\pi}^{\pi} \alpha(v) \cos(k\omega_m t) d(\omega_m t) \quad (2a)$$

$$H_0 = -\frac{1}{2\pi} \int_{-\pi}^{\pi} \alpha(v) d(\omega_m t) \quad (2b)$$

where  $\omega_m = 2\pi f_m$  with  $f_m$  known as the modulation frequency. In order to cancel the effect of  $G$  and  $\bar{I}_0$  and to avoid the losses due to the non-absorption parts of WMS signal, one can construct the calibration-free technique in which  $X_{2f}$  is divided by  $X_{1f}$ . Therefore, by using this approach, the pressure and/or concentration of an absorptive sample can be directly measured from the scaled peak height of WMS- $2f/1f$  signal [12]. However, as can be seen in Fig. 1, for an optically thick sample, the right wing of WMS- $1f$  signal is getting deeper and approaching to zero. This, because of  $2f/1f$  division, resulted in tending the common WMS- $2f/1f$  signal to infinity at the same wing.

As it can be seen, the infinity trend for thick  $\text{CO}_2$  sample clearly occurs in the right wing of common WMS- $2f/1f$  signal due to unavoidable  $2f/1f$  division. Moreover, the peak center is also displaced toward larger wave numbers. In order to solve such ex-

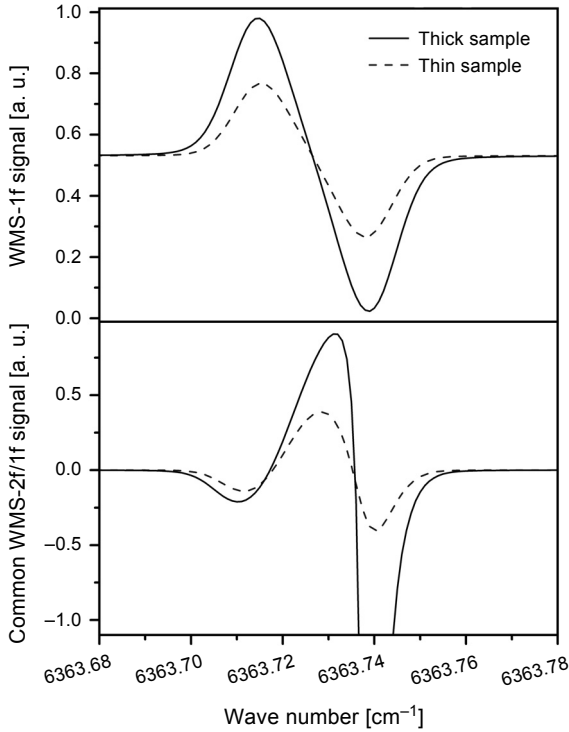


Fig. 1. Simulation results for WMS- $1f$  signal and common WMS- $2f/1f$  to indicate the infinity trend of WMS- $2f/1f$  signal when the pressure of sample increased beyond the optically thin limit. Calculation is performed for  $R(22)$   $\text{CO}_2$  line centered at  $6363.727 \text{ cm}^{-1}$  for gas pressures of 8 and 17 mbar as thin and thick samples, respectively. The line characteristics are inferred from Hitran 2014 [15] and modulation frequency and index are set at 0.3 kHz and 2.2, respectively.

plained problems, the apodized WMS- $2f/1f$  method [13] has been recently suggested to modify  $X_{1f}$  signal as

$$X_{1f}^{\text{apodized}} = \frac{G\bar{I}_0}{2} \left\{ i \cos(\psi_1) + k \left[ H_1 + i_1 \left( H_0 + \frac{H_2}{2} \right) \cos(\psi_1) + \frac{i_2}{2} (H_1 + H_3) \cos(\psi_2) \right] \right\} \quad (3)$$

where  $k$  is an arbitrary positive scaling factor which plays a role of a control parameter to prevent the infinity tendency through symmetrizing  $1f$  signal.

The significance of the apodizing procedure acting on the WMS- $2f/1f$  signal is depicted in Fig. 2.

Clearly, as depicted in the above plot, the disadvantages of the common method, including peak displacement and infinity trend, are modified by the apodizing procedure. Furthermore, it can be seen that the effect of using a larger  $k$  factor appears as a narrowing of the signal width, providing a better resolution in the received signal which has turned the apodized WMS- $2f/1f$  into an exceptional candidate for the iden-

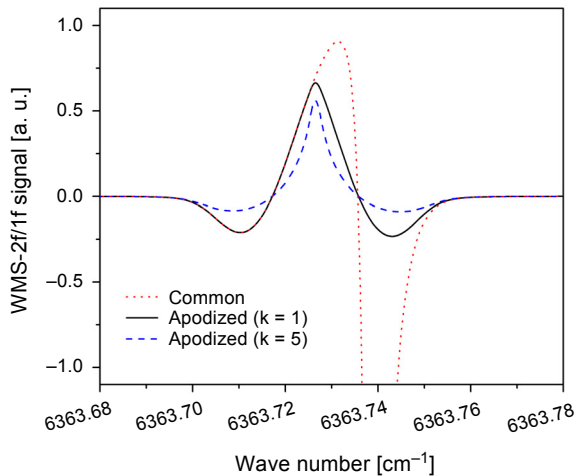


Fig. 2. Significance of the apodized method in making WMS- $2f/1f$  signal for two  $k$  values. Common WMS- $2f/1f$  is brought into the figure for comparison. Simulation is performed for  $R(22)$   $\text{CO}_2$  absorption line centered at  $6363.727 \text{ cm}^{-1}$ . Modulation frequency and index are the same as those used in Fig. 1.

tification of closed lines which are overlapped in the wings. Therefore, it seems that a minor change in the processing of WMS- $2f/1f$  signal generates a major improvement in the final results without a significant need to a substantial change in the experimental setup and spectroscopic circumstances. This goal will be reached by optimizing the effective parameters in mathematical formulation of WMS method such as the modulation frequency and index.

### 3. Modulation index optimization: simulation results

Since the introducing of WMS method, there can be found a numerous theoretical and experimental documents concerning WMS development and its application in spectroscopy of many hazardous and pollutant gaseous species [18, 19]. However, to the best of our knowledge, no distinctive scientific work has been reported to investigate the effect of temperature and pressure on the parameters playing a key role in WMS optimization. Certainly, the modulation index  $m$  is a very important parameter in WMS method. As suggested by REID and LABRIE [10] and LIU *et al.* [11], the peak height of WMS- $2f$  signal will be maximum if  $m$  is equal to 2.2. But, this criterion which has been respected by researchers for a long time, is quite appreciable for working under high pressure conditions. At very low pressure where a fine decrease in WMS- $2f$  peak height is very crucial, particularly in quantitative detections, such a rule is not fully preserved and eventually  $m$  will deviate from 2.2. In order to investigate the effect of gas temperature and pressure on the modulation index, our simulation is performed based on a proposed setup schematically indicated in Fig. 3.

In order to provide a meaningful simulation, the real data associated with an experimental setup reported in our previous work [14] have been used in this study. Therefore,

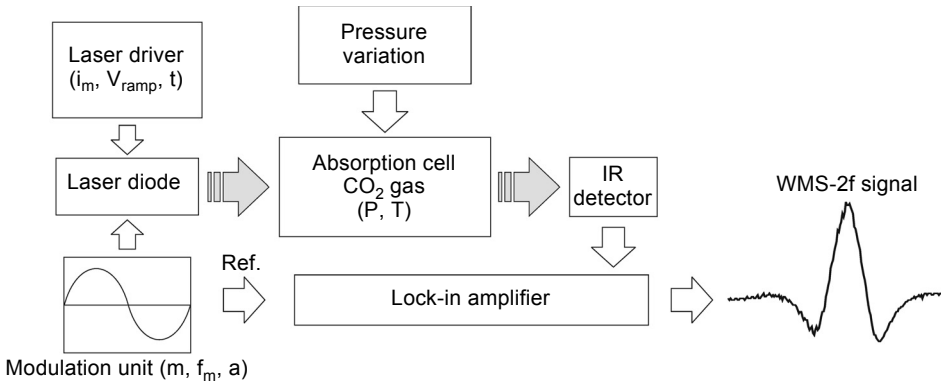


Fig. 3. A diagrammatic apparatus proposed to investigate the effect of temperature and pressure on modulation index.

the laser source is assumed to be a DFB laser operating at central wavelength of  $1.57 \mu\text{m}$  that can be scanned over  $R(22)$   $\text{CO}_2$  line at  $6363.727 \text{ cm}^{-1}$ . Here, an absorption cell is  $3 \text{ m}$  long and the lock-in amplifier can be set at  $1f$  and  $2f$ , alternatively. As can be expected, the variation in  $\text{CO}_2$  temperature and pressure does not affect the center of WMS- $1f$  signal. Therefore, we turn our attention to study the effect of the modulation index on the height of WMS- $2f$  peak. Thus, the optimized value of  $m$  (when it is found) can be applied for the apodized WMS- $2f/1f$  signal, too. Subsequently, our goal is to obtain  $m^{\text{opt}}$  in which the WMS- $2f$  peak height becomes maximum when the  $\text{CO}_2$  characteristics like pressure and temperature are changed.

### 3.1. Effect of temperature on modulation index

We start with the assumption that the absorption cell is filled by a fixed  $\text{CO}_2$  gas pressure of  $650 \text{ mbar}$  and temperature is changed from room temperature up to  $600 \text{ K}$  for four distinctive temperatures. By the variation of temperature in the described range we calculated the peak height of WMS- $2f$  signal when the modulation index is changed in an appropriate range around  $2.2$ . The results of this simulation are indicated in Fig. 4.

It is confirmed by the figure that temperature variation does not significantly affect the modulation index being deviated from its optimum value of  $2.2$  even though the temperature is drifted by about  $300 \text{ K}$ . However, as can be seen at  $m \sim 2.2$ , WMS- $2f$  peak height reaches the maximum regardless of gas temperature. This could be expected because such large temperature deviation will impose an extra bandwidth of less than  $0.005 \text{ cm}^{-1}$  on  $R(22)$   $\text{CO}_2$  line width which, compared to the pressure broadening, is negligible. Therefore, it can be anticipated that the gas pressure may have more contribution to the deviation of  $m$  from the contracted value of  $2.2$ .

### 3.2. Effect of pressure on modulation index

In order to prove our claim discussed above, we change the  $\text{CO}_2$  pressure for specified values while the gas temperature is kept at room temperature. In this case, the Voigt

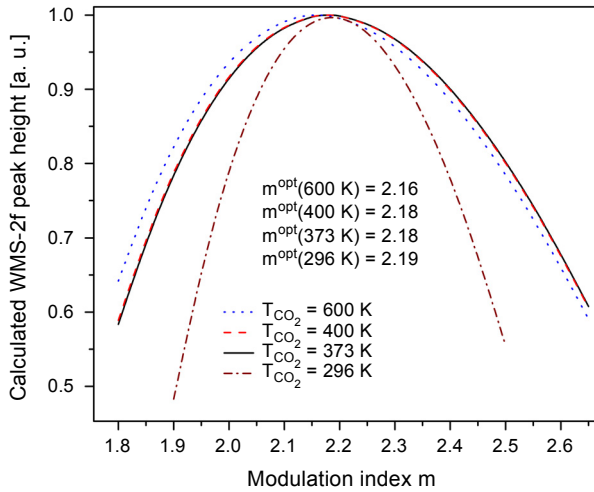


Fig. 4. Variation of WMS-2f peak height with modulation index for four distinctive gas temperatures. Simulation is performed for R(22) CO<sub>2</sub> absorption line centered at 6363.727 cm<sup>-1</sup>. Gas pressure is taken fixed at 650 mbar and modulation frequency is assumed 300 Hz.

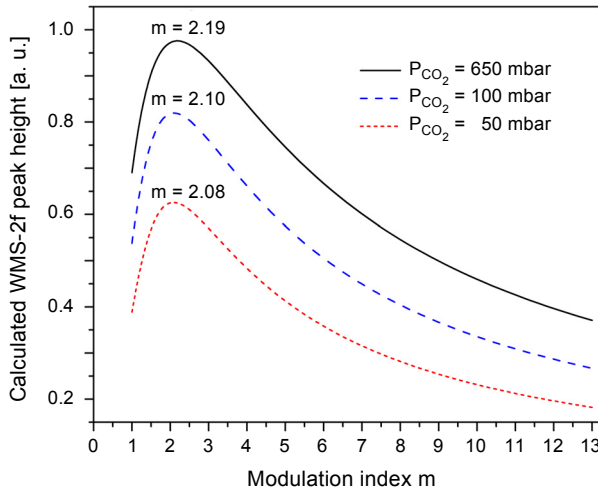


Fig. 5. Variation of WMS-2f peak height with modulation index for three CO<sub>2</sub> pressures. Simulation is performed at room temperature for R(22) CO<sub>2</sub> absorption line centered at 6363.727 cm<sup>-1</sup>. Modulation frequency is assumed 300 Hz.

broadening is mostly affected by the Lorentzian line shape rather than the Doppler broadening. In Fig. 5 results of this investigation are illustrated.

As it can be seen for high pressure sample of 650 mbar, the maximum of WMS-2f intensity occurs at  $m = 2.19$ , as expected. When the CO<sub>2</sub> pressure is decreased toward a thin optically limit to 50 mbar, the maximum of WMS-2f intensity is obtained at  $m = 2.08$ . However, compared to the case of  $m = 2.19$ , about 40% decrease observed

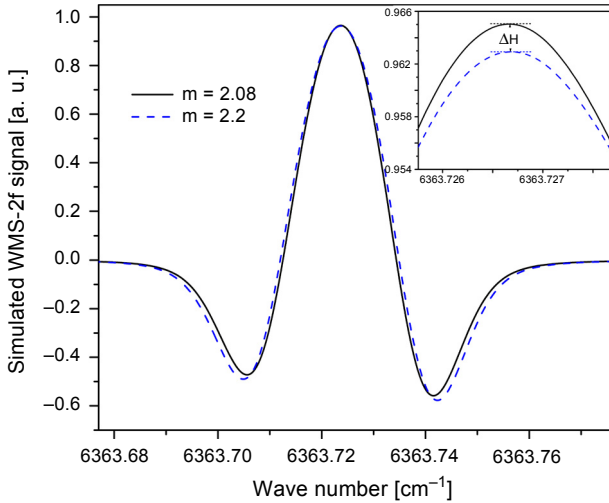


Fig. 6. Effect of two modulation indices on WMS- $2f$  signal for gas pressure of 50 mbar. Simulation is performed for  $R(22)$   $\text{CO}_2$  line and modulation frequency of 300 Hz. Inset shows the magnified version of plot around line center to indicate the difference in peak heights that is specified by  $\Delta H$ .

in the maximum of WMS- $2f$  signal is clearly due to the decrease in pressure. This confirms that the modulation index is coupled with the pressure and therefore  $m = 2.2$  can be used just as a special case for optically thick pressure. In order to indicate the significance of altering the modulation index in the variation of peak intensity, in Fig. 6 the WMS- $2f$  signal is simulated for  $\text{CO}_2$  pressure of 50 mbar for two modulation indices of 2.20 and 2.08, respectively.

Clearly, as shown in the inset at  $\text{CO}_2$  pressure of 50 mbar and even lower, using  $m = 2.2$  does not always lead to a greater WMS- $2f$  peak height. However, we found that by changing  $m$  from 2.2 to 2.08, the peak height difference  $\Delta H$  is increased by about 0.2%. At a first glance it seems not too significant but when facing very low pressure samples, such deviation causes a non-negligible error in pressure and eventually in concentration determination. This can be followed by noting the fact that the resultant WMS- $2f$  signal is proportional to the real pressure of  $\text{CO}_2$  gas that is primarily inserted into the absorption cell. Thus, it has been clearly experienced that what is measured is different from the reality. Accordingly, we define [7]

$$P_f = \frac{H_f}{H_{\text{ref}}} P_{\text{ref}} \quad (4)$$

as the unknown pressure that can be measured by scaling the peak height of its own WMS- $2f$  trace, that is  $H_f$ . Such scaling can be performed by helping  $P_{\text{ref}}$  as the certain value of a reference pressure that similarly generates a WMS- $2f$  peak height of  $H_{\text{ref}}$  in the output. In Fig. 7 four described parameters are schematically indicated.



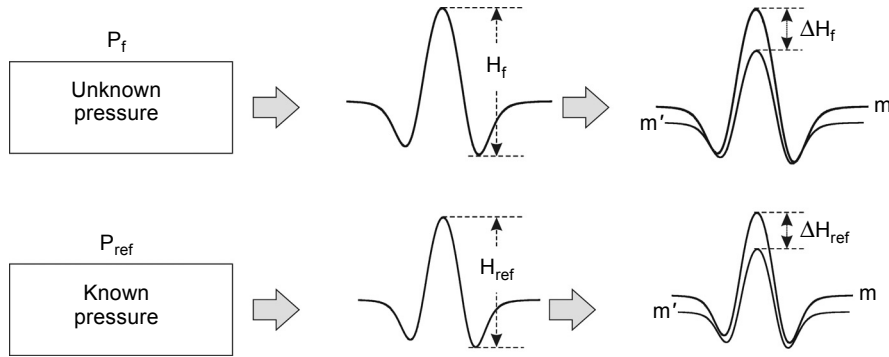


Fig. 7. Schematic indication to describe four parameters introduced in Eq. (4) leading to obtain an optimum value for modulation index. The  $\Delta H_f$  and  $\Delta H_{ref}$  are obtained due to different values of modulation index as  $m$  and  $m'$  used for demodulating the final WMS-2f signal.

Obtaining the pressure deviation is achieved by

$$\Delta P = \Delta\left(\frac{H_f}{H_{ref}} P_{ref}\right) = \frac{P_{ref}}{H_{ref}} \Delta H_f + \frac{H_f}{H_{ref}} \Delta P_{ref} + H_f P_{ref} \Delta\left(\frac{1}{H_{ref}}\right) \quad (5)$$

where we introduce  $\Delta P$  as the deviation of measured  $\text{CO}_2$  pressure from the real one which is linearly related to  $\Delta H_f$ . As clearly shown in Fig. 7, both  $\Delta H_f$  and  $\Delta H_{ref}$  are the functions of modulation index  $m$  and appeared because of incorrect selection of  $m^{opt}$ . However, as shown in Fig. 5, for high pressures close to atmospheric, the deviation of  $m$  from 2.2 does not generate a significant effect on WMS-2f peak

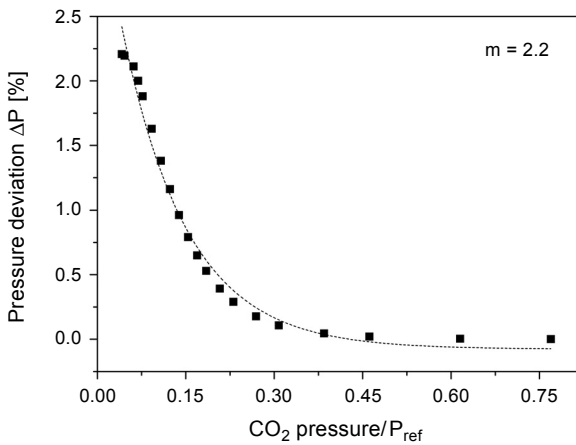


Fig. 8. Variation of pressure deviation  $\Delta P$  with a wide range of  $\text{CO}_2$  pressures/ $P_{ref}$  while modulation index is fixed at 2.2. Simulation is performed for  $R(22)$   $\text{CO}_2$  absorption line centered at  $6363.727 \text{ cm}^{-1}$  and modulation frequency of 300 Hz. The  $P_{ref}$  is fixed at 650 mbar and dotted line indicates a mathematical fit to better trace the trend of variation.

height and, therefore, we can assume that  $\Delta H_{\text{ref}}^{-1} \approx 0$ . This in turn resulted in putting  $\Delta P_{\text{ref}} \approx 0$ . Thus, Eq. (5) simplifies to

$$\Delta P \approx \frac{P_{\text{ref}}}{H_{\text{ref}}} \Delta H_f \quad (6)$$

In Figure 8 the pressure deviation  $\Delta P$  is calculated for a wide range of optically thin and thick CO<sub>2</sub> pressures relative to  $P_{\text{ref}}$ , while the modulation index is fixed at 2.2.

As it can be seen from Fig. 9, by increasing the pressure toward 500 mbar, pressure deviation is significantly decreased with an exponential trend. At lower pressures, where the Doppler broadening is a dominant mechanism,  $\Delta P$  is increased. Particularly, at CO<sub>2</sub> pressure of 25.5 mbar, greater pressure deviation of 2.21% is obtained. Therefore, it is desirable to obtain an optimum value of the modulation index in which the  $\Delta P$  approaches to zero. This is investigated within a wide range of CO<sub>2</sub> pressures from 50 to 750 mbar. Obtained results have been illustrated in Fig. 9.

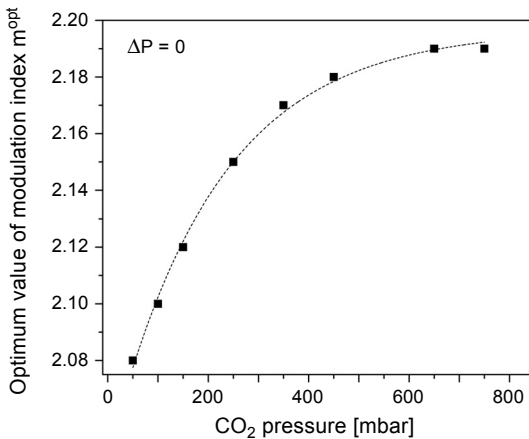


Fig. 9. Calculation of optimum modulation index for a wide range of CO<sub>2</sub> pressures in which the  $\Delta P$  approached to zero. Required parameters for simulation are the same as those used in Fig. 8. Dotted line indicates a mathematical fit to follow the trend of variation.

Apparently, it is confirmed by the plot that for obtaining the lowest deviation of nearly zero for each corresponding gas pressure, the modulation index has to be adjusted at an optimum value that differs from 2.2.

#### 4. Conclusion

In the present work we reported on the optimization of WMS method with respect to the modulation index. This investigation is based on the study of the effect of temperature and pressure on the value of the modulation index which has been found by REID and LABRIE [10] and LIU *et al.* [11] to be 2.2 for optimum operation. The study is performed by the simulation of WMS-2f signal for R(22) CO<sub>2</sub> absorption line centered at

6363.727  $\text{cm}^{-1}$ . It is found that variation in gas temperature does not significantly alter the modulation index from 2.2. On the contrary, we found that when the gas pressure is changed and decreased to lower pressures, the maximum WMS-2f intensity will occur at different value of 2.2. For example, at  $\text{CO}_2$  pressure of about 50 mbar, the highest WMS-2f signal is obtained at a modulation index of 2.08 and a peak height deviation  $\Delta H$  of about 0.2% was introduced. Simulation results confirmed that at sufficiently high pressures close to the atmosphere,  $m = 2.2$  is still valid and can be held as the optimum value. A pressure deviation  $\Delta P$  is introduced and mathematically formulated to search for the optimum value of  $m$  at particularly lower  $\text{CO}_2$  pressures. Calculations indicated that at the presence of the Doppler broadening only,  $\Delta P$  is increased to  $\sim 2.21\%$  for a  $\text{CO}_2$  pressure of 25.5 mbar. Eventually, by making  $\Delta P$  close to zero, the optimum value of  $m$  is obtained within a wide range of  $\text{CO}_2$  pressures. As a result, we further emphasize that the obtained  $m^{\text{opt}}$  can be used in processing the common and apodized WMS-2f/1f for making a precise measurement of gas pressure with the accuracy better than 2.21%.

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