Improved Gas Concentration Model Accuracy in Wavelength Modulation Spectroscopy

ABSTRACT

Researchers from China have developed a new concentration inversion model for wavelength modulation spectroscopy (WMS) techniques in tunable diode laser absorption spectroscopy (TDLAS) applications. Traditional models do not take into account variations of the modulation depth in the second harmonic signal of the laser, thereby increasing the error of the measurement and decreasing the accuracy of the system. Through simulations and controlled experiments measuring CO gas concentrations, researchers achieve root-mean-square error (RMSE) of the gas concentration measurements of 5.468×10^{-5} with relative error of < 0.37% using an improved model of concentration inversion with modulation depth as a variable parameter. This model produces almost an order of magnitude better RMSE and almost five times better relative error, making this applicable in fields such as industrial and environmental gas monitoring.

INTRODUCTION / TDLAS

With a wide variety of methods to measure gas concentrations in many industrial and environmental applications, Tunable Diode Laser Absorption Spectroscopy (TDLAS) is a popular technique that not only achieves gas concentration measurements with high accuracy but can measure temperature, flow velocity, pressure, and other parameters important to gas monitoring.[1] **Figure 1** shows a thermal power plant which is suited to the energy and power applications of TDLAS.[2] As TDLAS is based on absorption spectroscopy, it can benefit from the unique characteristics and nature of molecular spectroscopy. This gives this technique the ability to measure quantity and quality parameters of a gas substance. By using low-power semiconductor lasers that are tunable and narrow-linewidth, the monitoring systems can be lower-cost, real-time, time-resolved, and long-term in-situ systems.[2]

Figure 1. Thermal Power Plant - One of the many applications of TDLAS

Laser absorption spectroscopy is built upon the properties of the Beer-Lambert Law, in the most basic form, relating the absorption of light and the concentration of a particular gas. Other parameters are present in the Beer-Lambert law as well: transmitted light intensity, total pressure, spectral absorption coefficient, absorption path-length, and absorption line-strength among others. The comparison of the light intensities before and after propagation or absorption through a medium is the basis of absorption spectroscopy. The use of laser diodes provides possibilities of fixed or scanned wavelengths. By scanning the laser over a range of wavelengths, multiple absorption feature profiles can be constructed with a single laser source.[2] Scanning the laser diode is accomplished through injection current or temperature. Careful changes to either of these parameters can offer promising benefits to the experiment.

To detect the signal after transmission through the medium, the absorption spectroscopy sensor can be used with two different techniques: Direct Absorption Spectroscopy (DAS) or Wavelength Modulation Spectroscopy (WMS).

DAS uses the remaining transmitted light to infer gas properties from the Beer-Lambert Law. The issue with this technique, however, is relating the transmitted light to the initial light before the absorption. Added components can help to determine the baseline of the laser, but these can add cost, time, and even error from additional optical systems. Scanning the laser can help, but noise from the environment and laser baseline fluctuation can make this technique difficult while uncertainties increase.[2]

WMS offers a solution to some of these problems by adding frequency modulation to the laser diode injection current.

WAVELENGTH MODULATION

WMS reduces the influence of noise and achieves a high detection accuracy by the use of both low and high frequency signals. The lower frequency signal scans the laser wavelength in a range close to the center wavelength of the laser, as is common in many spectroscopy applications. The higher frequency signal, however, modulates the signal from the laser. Custom tuning of these signals can enhance the signal-to-noise ratio. This is commonly achieved through the injection current or laser driver of the system.

To determine gas concentration and other parameters, a photodetector detects the light that passes through the gas. A lock-in amplifier (LIA) detects the harmonic signal of the transmitted laser intensity. The concentration is inversed using the peak value of the second harmonic signal.[1] Many different parameters can affect the second harmonic lineshape shown in **Figure 2**.

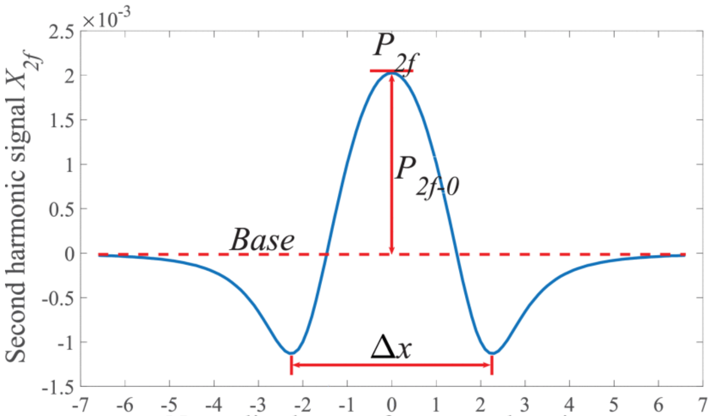

Figure 2. Schematic diagram for second harmonic signal.[1]

Environmental parameters and the modulation parameters both affect the lineshape of the second harmonic signal, and related to both of these parameters is the modulation depth. Modulation depth is defined as the ratio of the modulation amplitude to the mean value as seen in **Equation 1**.[1]

$$m = \frac{a}{\Delta v_c / 2} \tag{1}$$

where *m* is the modulation depth, *a* is the frequency modulation amplitude, and Δv_C is the full width at half maximum (FWHM) or the mean value of the gas absorption feature. Modulation depth can bring errors to the concentration inversion results, and it can limit applications of the WMS technique.[1]

Determining the proper modulation depth and modulation parameters is crucial to WMS, and many researchers have simulated and experimented to find the optimal modulation values. However, when the gas sample is unknown, the modulation depth can not be calculated from parameters such as pressure and temperature. Finding a relationship between the modulation depth and the valley spacing of the second harmonic signal is vital to calculating and calibrating the modulation depth for enhanced measurements with greater signal-to-noise ratio.

METHOD

Researchers from the Chinese Academy of Sciences and the University of Chinese Academy of Sciences have developed an improved concentration inversion model using the relationship between the modulation depth and the valley spacing of the second harmonic signal. Here the calibrated modulation depth is taken as a parameter in the improved model.

To create a new model, researchers started with the Beer-Lambert law and the transmittance coefficient in **Equation 2**.[1]

$$\tau(v) = 1 - PS(T)\,\phi(v)\,CL \tag{2}$$

where $\tau(v)$ is the transmittance coefficient, P is the total pressure of the mixed gas (atm), $S(T)$ is the line strength of the gas absorption feature ($cm^{-2}\cdot atm^{-1}$) at the temperature T (Kelvin), $\phi(v)$ is the lineshape function of the gas absorption feature (cm), C is the concentration of the gas (volume fraction), and L is the effective optical path length (cm).

Through Fourier expansions and many substitutions, important expressions are found in the following equations:

$$m = b_1 \times \Delta x + b_2 \tag{3}$$

where Δx is the valley spacing (seen in **Figure 2**), and b_1 and b_2 are different constants according to the actual measurement condition and are calibrated in advance during the measurement. This linear relationship is a result of the theoretical scatter plot of the valley spacing versus modulation depth in **Figure 3**.

$$k_1(m) = c_1 \times m^3 + c_2 \times m^2 + c_3 \times m + c_4 \tag{4}$$

where k_1(m) is the coefficient expression of the inversion model. A theoretical scatter plot of $k_1(m)$ versus modulation depth can be seen in **Figure 4**. Here the coefficient is not taken as a constant, hence the 3rd degree polynomial expression with respect to the modulation depth.

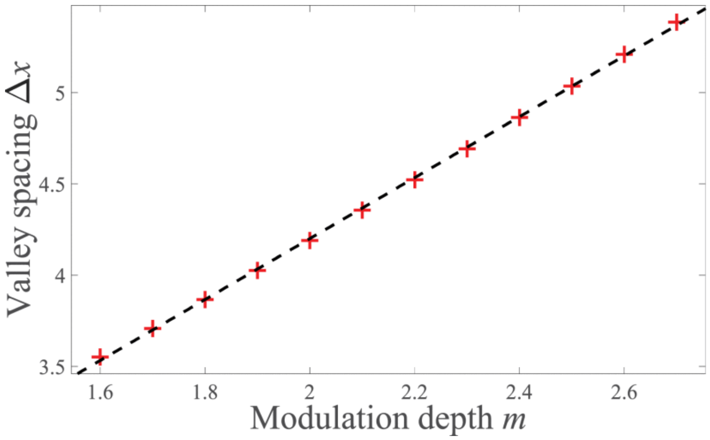

Figure 3. Theoretical scatter plot of valley spacing Δ*x* versus modulation depth *m*.[1]

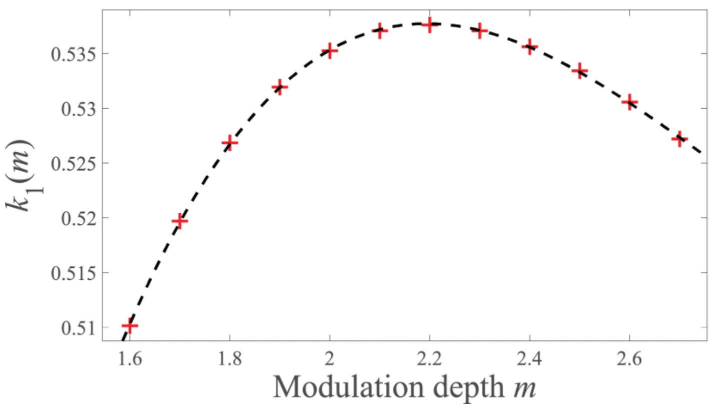

Figure 4. Theoretical scatter plot of $k_1(m)$ versus modulation depth *m*.[1]

With **Equation 4**, the improved concentration inversion model is as follows:

$$C = \frac{P_{2f-0}}{c_1 \times m^3 + c_2 \times m^2 + c_3 \times m + c_4} \quad \textbf{(5)}$$

where $P_{2f\text{-}0}$ is the maximum of the peak value of the second harmonic signal seen in **Figure 2**.

By calibrating and calculating the modulation depth, instead of using a same constant value, error can be reduced while improving gas concentration measurement accuracy.

Researchers used two methods to test and prove this new concentration inversion model for WMS: a simulation system in Matlab and an experimental setup with CO gas measurements.

The simulation system used a sawtooth wave with frequency of 5 Hz to scan the signal and a sine wave with frequency of 2000 Hz to modulate the simulated laser signal. Intensity modulation of the nonlinear laser and the reference signal is 4000 Hz. The simulation start time was 0s, and the end time was 0.2s. The parameters in the setup are as follows:

ν_0 = 6383.09 cm^{-1}	$\Delta\nu$ = ±0.45 cm^{-1}
$\bar{I}_0$ = 13.26 mW	L = 100 cm
P = 1 atm	T = 296 K
$S(T)$ = 5.011 x 10^{-4} cm^{-2}·atm^{-1}	
$\Delta\nu_c$ = 0.135 cm^{-1}	

where ν_0 is the central frequency (cm^{-1}), $\Delta\nu$ is the scanning range (cm^{-1}), and $\bar{I}_0$ is the average laser intensity driven by the scanning signal.

In the simulation, the frequency modulation amplitude *a* and concentration *C* can be varied to obtain the corresponding second harmonic signal and calculate modulation depth, $P_{2f\text{-}0}$, and valley spacing Δx.

To experimentally test this improved model, CO gas with a fixed volume fraction of 2.5% was used to perform TDLAS. A 10 mW Distributed Feedback laser diode was used with center wavelength around 1566 nm. The laser was driven and cooled by Wavelength Electronics' LDTC0520 laser driver and temperature controller. The emitted laser light passed through a stainless steel gas cell with calcium fluoride windows. The transmitted light was detected by a Ge photodetector that has a peak response of around 1550 nm. This is connected to the LIA which integrates the functions of signal generation and lock-in amplification.[1] The signal is then recorded, ensuring the second harmonic signal result. This setup can be seen in **Figure 5**.

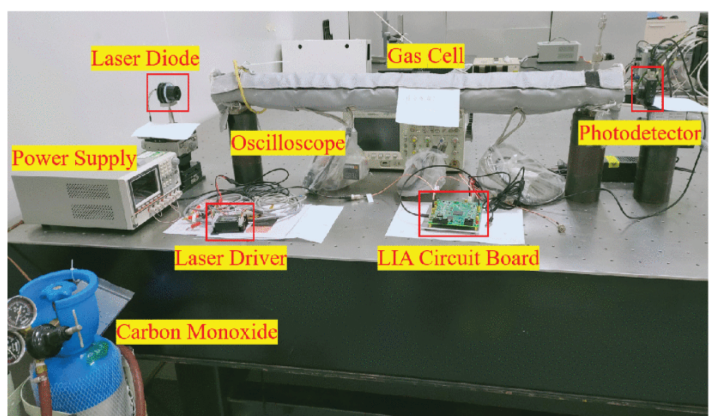

Figure 5. Photo of the experimental setup.[1]

To scan and modulate the laser, a 5 Hz sawtooth wave scans the laser wavelength near the absorption line of CO gas at 1566.64 nm, and a 4000 Hz sinewave modulation signal modulates the laser wavelength. The LIA has a frequency of 8000 Hz to amplify the second harmonic signals.

RESULTS

Simulation Results

The results of the simulation of the improved concentration inversion model were compared to another simulation of the traditional concentration inversion model. The scatter plots of Δx versus m and $k_1(m)$ versus m are consistent with the theoretical plots shown in Figure 3 and Figure 4. Using Equation 3 and Equation 5, the parameters can be fitted as seen below:

$$m = 0.605 \times \Delta x - 0.558 \quad (6)$$

$$C = \frac{P_{2f-0}}{9.783 \times 10^{-3} m^3 - 0.1144 m^2 + 0.3665 m + 0.06627} \quad (7)$$

The scatter plot of inversed values versus true values of the modulation depth is shown in Figure 6.

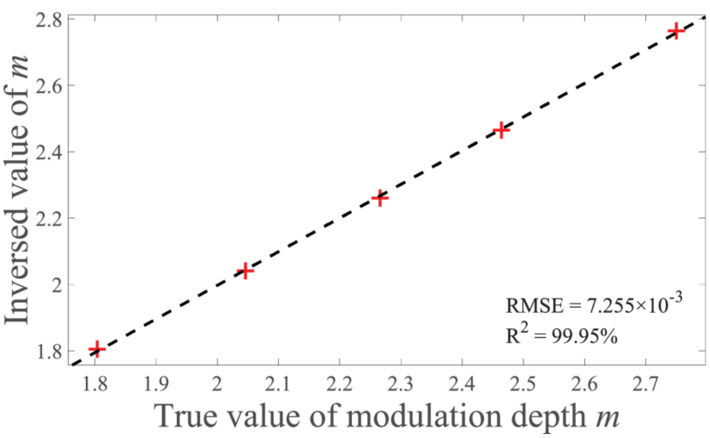

Figure 6. Scatter plot of inversed values versus true values of modulation depth *m* in simulations.[1]

To compare these expressions and values to those of the traditional concentration inversion model, the root-mean-square error (RMSE) is used. For the improved model the fitted RMSE of modulation depth is 5.563×10^{-3}, and R^2 is 99.97%. The calculated RMSE of concentration C is 5.766×10^{-5}, and R^2 is 1.[1]

The calculated RMSE of the traditional model concentration C is 3.179×10^{-4}, and R^2 is 99.96%. This shows that the RMSE of the new improved model is almost an order of magnitude lower than the traditional model, and the absolute value of relative error of the improved model (<0.35%) is almost five times lower than that of the traditional model (<1.54%).[1] The relative error can be seen in Figure 7.

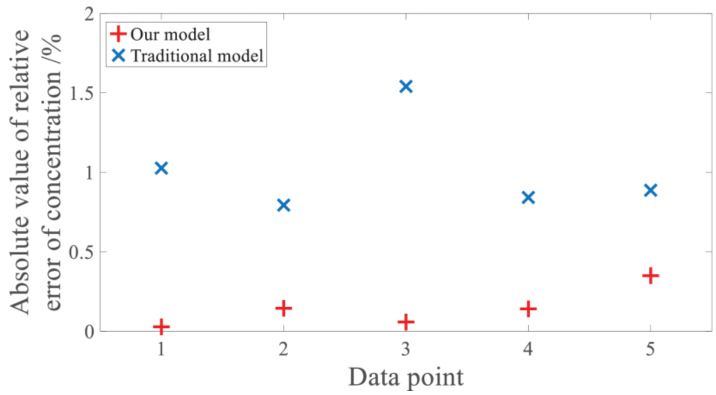

Figure 7. Scatter plot of the absolute value of relative error of inversed concentration *C* in simulations.[1]

Experiment Results

The same methods were used, as in the simulations, to obtain and calculate the parameter fitting dataset and scatter plots. Expressions obtained are below.

$$m = 0.5991 \times \Delta x - 0.5178 \quad (8)$$

$$C = \frac{P_{2f-0}}{-0.5884 \times m^3 + 2.261 \times m^2 - 1.375 \times m + 18.53} \quad (9)$$

Scatter plots of both the inversed values compared to the true values of m and the relative error of both models can be seen in Figure 8 and Figure 9, respectively.

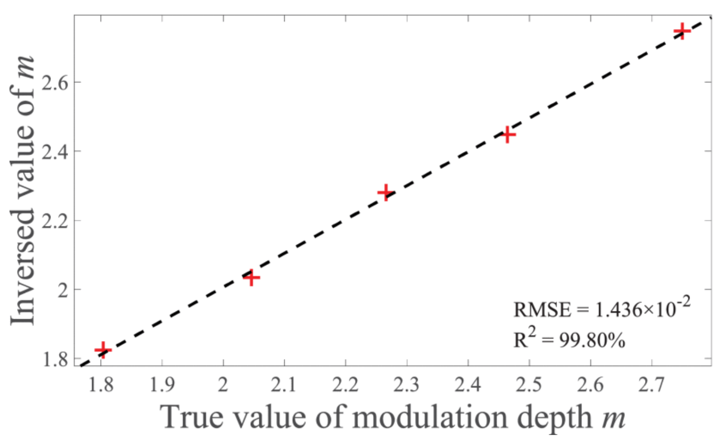

Figure 8. Scatter plot of inversed values versus true values of modulation depth *m* in experiments.[1]

The RMSE between the inversed values and the true values of the CO concentration using the improved model is 5.468×10^{-5} while the traditional model RMSE is 2.178×10^{-4}. Relative error for the improved model is <0.37%, and the relative error for the traditional model is <1.70%. Once again, the experimental data and results show almost an order of magnitude improvement with the

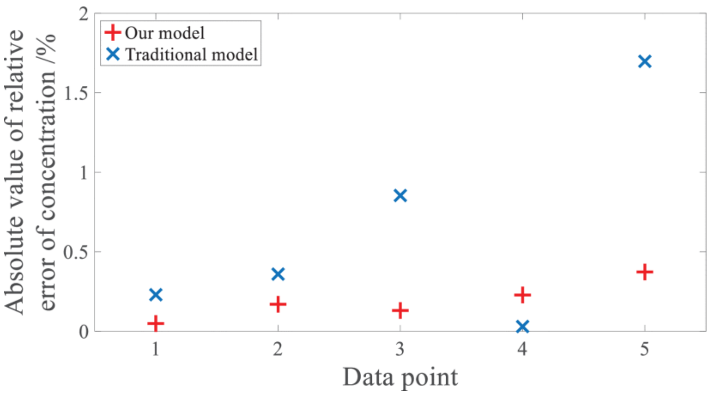

Figure 9. Scatter plot of the absolute value of relative error of inversed concentration *C* in experiments.[1]

new model compared to the traditional model. Relative error is also improved (with the exception of one data point) to a lower and more consistent value.

The experimental results are consistent with theoretical predictions and simulation results, verifying the effectiveness of modulation depth calibration using the valley spacing of the second harmonic signals. This concentration inversion model improves the accuracy of gas detection using WMS in TDLAS experiments. By including the modulation depth as a parameter in the concentration inversion model, this method is immune to variations in the modulation depth and lowers error in the measurements. Further experiments will provide more evidence of verification in outside applications such as industrial and environmental gas control on monitoring.[1]

WAVELENGTH'S ROLE

Finding gas concentrations in industrial and environmental applications requires high precision, high accuracy, and stable temperature control of the laser diode used for the TDLAS. Wavelength Electronics' laser drivers and temperature controllers enable the sensitive measurements and experiments for concentration inversion models based on second harmonic valley spacing, useful for gas detection in the commercial world.

The stability of the laser diode in gas detection is critical, both in current stability of the injection current and the temperature control. Wavelength's low noise, high stability laser diode driver and temperature controller, the LDTC0520, can precisely deliver up to 500 mA to the laser. Many laser safety features include current limit circuity, slow start, and brownout protection. This module was sent a modulation signal to modulate the output current to the laser, simplifying steps for WMS. Scanning and modulating the laser diode was achieved using the LDTC0520 laser driver.

For stable output power and accurate wavelength scanning with repeatable results, researchers used the temperature controller in the LDTC0520. This controller is able to maintain the laser temperature with stability better than 0.005ºC for 1 hour on ambient temperature. It can provide up to ±2.2 A of current to a thermoelectric with both heating and cooling current limits. The PI control loop offers maximum stability while maintaining efficiency. The benefits of the temperature controller can also help narrow linewidth and can ensure the wavelength does not have any unwanted fluctuations. The laser diode's life and performance are improved by tighter and more precise control of the laser heatsink temperature.

With the combined temperature controller and laser diode driver, the LDTC0520 module is compact and easy to use with controls and indicators on-board for simple plug-and-play operation. The LDTC0520 combines the proprietary FL500 laser diode and ultra-stable WTC3243 into one compact module for trouble-free operation. With dimensions of 2.9 x 2.35 x 1.08 inches, the LDTC0520 can fit into most portable or benchtop equipment for gas detection in the lab or out in the field.

REFERENCES

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2. Wang, Z., Fu, P., Chao, X., "Laser Absorption Sensing Systems: Challenges, Modeling, and Design Optimization," *Appl. Sci*. **9** (2019), 2723, https://doi.org/10.3390/app9132723

USEFUL LINKS

- LDTC0520 **Product Page**

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PRODUCTS USED
LDTC0520

KEYWORDS
TDLAS, WMS, spectroscopy, gas concentration, valley spacing, second harmonic, modulation depth, laser diode, laser driver, temperature controller, LDTC0520

REVISION HISTORY

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REVISION	DATE	NOTES
A	January 2022	Initial Release