# Multi-frequency Modulation Method for Optical Interference Suppression in TDLAS System

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**Abstract:** Online accurate detection of residual oxygen content in encapsulated medicine bottles is critical to ensure the integrity of the seal of sterile preparations. Tunable diode laser absorption spectroscopy (TDLAS) technology, as a new type of spectral detection technology, has become the main method for gas concentration detection with advantages of high detection sensitivity and resolution. However, in actual production, there are various forms of noises in TDLAS system. Existing studies mostly focus on solving the interference and random noise in the surrounding environment while neglecting the optical interference fringe noise. Furthermore, general processing methods designed for optical interference are always more complicated having weak timeliness and poor system robustness. In this paper, we rationally explore the internal mechanism of optical interference and innovatively apply the multi-frequency modulation to the TDLAS system. Specifically, this methodology is successfully verified with encapsulated medicine vials on the actual oxygen concentration detection platform. Consequently, the multi-frequency modulation method with optimized parameters produces competitive results, with 2.96 standard deviation and 36.979dB SNR, which achieves better static stability and higher dynamic accuracy.

Key Words: Tunable diode laser absorption spectroscopy (TDLAS), multi-frequency modulation, optical interference

# 1 Introduction

Sealing integrity detection of sterile preparation is the basic technology in the pharmaceutical industry. Considering the content of air components and the detectability of gas, oxygen concentration is mostly used as the standard to measure the sealing quality of encapsulated medicine bottles [1]. Therefore, accurate detection of residual oxygen content in penicillin bottles is essential to ensure the sealing integrity, drug quality and safety [2]. According to the Lambert-Beer law, nowadays TDLAS [3] has been widely used in the field of trace gas detection (e.g., temperature, pressure, concentration and flow velocity) [4, 5, 6]. To further address the noise-sensitivity problem of direct absorption spectroscopy (DAS), a scheme of wavelength modulation spectroscopy (WMS) was proposed [7, 8] to transform the broadband signal measurement to narrow bands. Currently, wavelength-modulation-based tunable diode laser absorption spectroscopy (TDLAS/WMS) [9] is enjoying popularity due to its detection ability of gas concentration [10, 11].

However, different from other application scenarios, oxygen concentration detection in the encapsulated medicine bottles must take the effect of optical interference brought by glass walls, principally optical fringe noise, into consideration. In real world, optical fringe noise essentially affects the final detection accuracy. Aiming at suppressing the interference of optical fringes, four research approaches are mainly adopted at present: (1) Separate optical path, and take one of the paths as reference to subtract and eliminate interference fringes in the same part of optical path [12, 13], however, which is infeasible for different parts; (2) By adjusting optical elements to change optical path, the etalon signal is directly transformed into a nearly random signal, and then return to the algorithm level for processing [14].

However, this method increases the complexity of system and greatly limits the adaptation between elements and system; (3) Coating the element with antireflection film can eliminate inference from root [15], but there still exists the element limitation; (4) Take the disturbed signal as processing object, such as wavelet de-noising [16], empirical mode decomposition (EMD) algorithm [17], electromagnetic wavelet transform with adaptive Savitzky– Golay filtering (EWT-ASG) [18] method. Unfortunately, they increase complexity and reduce instantaneity of system, and fail to meet the requirements of on-line detection.

Recently, a new idea of seeking interference suppression methods from the perspective of signal modulation has emerged. The multi-frequency modulation method is to add a low-frequency jitter signal with small amplitude to the original modulation signal [19], which has been tentatively applied to the open-environment concentration measurement of CH4 [20] and other fields, so as to realize the gas concentration detection with both rapidity and accuracy. Through theoretical derivation, model simulation and experimental verification, this paper explores the inherent mechanism of optical interference fringes noise in TDLAS system, and innovatively applies multi-frequency modulation method to the online detection system of residual oxygen in cylinder based on TDLAS technology, which can effectively suppress the complex interference fringes noise and baseline drift, greatly improve the SNR and stability of TDLAS system. After the principle verification on the TDLAS optical platform, the parameteroptimized multi-frequency modulation method has obtained the static stability (i.e., standard deviation is 2.96) and dynamic accuracy (i.e., SNR is 36.979dB) in the practical application. In addition, with few parameters needed to be adjusted, our method also shows strong system robustness, it is no need to make adjustment once the hardware platform of the detection system is determined.

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The rest of this paper is organized as follows. Section 2 illustrates the interference fringe suppression method based on multi-frequency modulation, including the study on the mechanism of optical interference generation and parameter-optimized multi-frequency modulation method. Section 3 introduces the experimental setup details on TDLAS system. Section 4 elaborates our relative parameter configurations and experimental results. Finally, Section 5 concludes this paper.

# 2 Interference Fringe Suppression Method Based on Multi-frequency Modulation

### 2.1 Study on the Formation Mechanism of Interference Fringe

A beam of laser with frequency v is reflected multiple times on the surfaces of two optical devices, then the simplified transmission function T is:

$$T(v) = 1 - \frac{F}{2} [1 - \cos \psi(v)]$$
(1)

Where  $\psi(v) = 4\pi v l$  is the normalized free spectral range, *l* is the length of etalon and *F* is the fineness coefficient of etalon.

When the laser center frequency is  $v_1$ , the average output light intensity is  $I_0$ , the light intensity modulation amplitude is  $i_0$ , according to Beer-Lambert law, the modulation frequency v is higher, and the modulation amplitude m is smaller ( $m \ll 1$ ), nonlinear intensity modulation and phase difference can be ignored. The Lorentz line type is selected as the line type function  $\varphi(v)$  of the gas to be measured. S(T)is the gas line intensity at temperature T. L is the optical path length of the laser in the gas to be measured, c is the concentration. Define A =  $1-PcLS\varphi(v_1+m\sin\omega t)$ , B =  $[T(v_1+m\sin\omega t)][I_0+i_0\sin\omega t]$ , then the absorption light intensity of the gas to be measured can be simplified as: I(t)= AB.

For harmonic analysis, take the Fourier transform of I(t) to get n<sup>th</sup> order Fourier component as follows:

$$H_{n} = \frac{2}{\pi} \int_{0}^{\pi} I(t) \cos(nwt) dwt$$
  
$$= \frac{2}{\pi} \int_{0}^{\pi} B \cos(nwt) dwt$$
  
$$- \frac{2PcLS}{\pi} \int_{0}^{\pi} B\varphi(v_{1} + m\sin\omega t) \cos(nwt) dwt$$
  
(2)

The term before the minus sign is called the background signal and denoted as  $S_{BG}^{n}$ . Extract main component of the second harmonic component in  $S_{BG}^{n}$ , according to Jacobi-Ingres identity and Euler formula, define M=2lm, then the primary and second harmonics of  $S_{BG}^{n}$  are respectively:

$$BG_{1f} = i_0 (1 - F / 2) - I_0 F \sin(4\pi l v_1) J_1(2\pi M) \quad (3)$$

$$BG_{2f} = I_0 F \cos(4\pi l v_1) J_2(2\pi M)$$
(4)

Where  $BG_{2f}$  reflects the interference fringe information of the system. It is clear that  $BG_{2f}$  is determined by the laser center frequency  $v_l$ , the standard length l, and the modulation amplitude m of the laser frequency, while the laser center frequency  $v_l$  is in turn affected by the laser temperature and the average value of control current. Therefore, finally, the main factors affecting the background interference fringe include the length of the standard tool l, the laser frequency modulation amplitude m, the laser temperature  $T_L$ , and the average value of the control current I.

#### 2.2 Parameter-optimized Multi-frequency Modulation Method

Multi-frequency modulation method is to superimpose a low-frequency jitter modulation signal with a small amplitude on top of the original high-frequency sinusoidal modulation signal. Without considering the nonlinear intensity modulation and phase difference, the laser output intensity and frequency are respectively:

$$I(t) = I_0 + I_1 \cos(\omega_1 t) + I_2 \cos(\omega_2 t)$$
 (5)

$$v(t) = v_1 + m_1 \cos \omega_1 t + m_2 \cos \omega_2 t \tag{6}$$

Where,  $I_0$  is the average output light intensity,  $I_1$  and  $\omega_1$  are the light stress amplitude and angular frequency of the base modulation signal, and  $I_2$  and  $\omega_2$  are the light stress amplitude and angular frequency of the dither modulation signal.  $v_1$  is the center frequency of the laser output,  $m_1$  is the frequency modulation amplitude of the base modulation signal, and  $m_2$  is the frequency modulation amplitude of the dither modulation signal. Besides,  $M_1=2lm_1$  and  $M_2=2lm_2$  were defined to extract the main components of the second harmonic in the background signal as follows:

$$\frac{I_0 F}{2} \cos\left(4\pi l v_1 + 2\pi M_1 \cos \omega_1 t + 2\pi M_2 \cos \omega_2 t\right)$$
(7)

According to the Jacobi-Ingres identity, define

$$C = \varepsilon_s (-1)^{r+s} \cos(2s\omega_2 t) J_{2s} (2\pi M_2)$$
  
$$D = J_{2s+1} (2\pi M_2) \cos((2s+1)\omega_2 t)$$

Hence, the above equation can be expanded to:

$$\frac{I_0 F}{2} \cos(4\pi l v_1) \left[ \sum_{r,s=0}^{\infty} \varepsilon_r C J_{2r} (2\pi M_1) \cos(2r\omega_1 t) -4 \sum_{r,s=0}^{\infty} J_{2r+1} D (2\pi M_1) \cos((2r+1)\omega_1 t) \right] + \frac{I_0 F}{2} \sin(4\pi l v_1) \left[ \sum_{r,s=0}^{\infty} 2C J_{2r+1} (2\pi M_1) \cos((2r+1)\omega_1 t) + \sum_{r,s=0}^{\infty} 2\varepsilon_r (-1)^{r+s} J_{2r} D (2\pi M_1) \cos(2r\omega_1 t) \right]$$
(8)

In this paper, we take r=1, s=0, the terms of higher-order Bessel function is ignored to make an approximation:

$$I_0 F \cos(4\pi l v_1) J_2(2\pi M_1) J_0(2\pi M_2)$$
(9)

Obviously, interference fringe noise can be reduced by adjusting the fundamental frequency modulation amplitude



Fig. 1: The variation trend of the amplitude of each cosine term in the second harmonics of the background interference fringes with the increase of  $M_2$ 



Fig. 2: The variation of the second harmonic amplitude of the background interference fringe signal with  $M_2$ 

 $m_1$  so that  $M_1$  can reach any zero point of  $J_2$ , but  $m_1$  will also affect the size of the second harmonic absorption signal. Therefore, a more effective method is to introduce an additional jitter modulation signal independent of the basic modulation signal. By adjusting  $m_1$  to maximize the second harmonic signal, and then adjusting  $m_2$  to make  $M_2$  reach any zero point of  $J_0$ , the influence of interference fringe is reduced. It can be seen from Fig.1 that the second harmonic of background interference fringes can be completely expressed by formula (9) only when  $M_2$  is small. Therefore, in order to meet  $J_0(4\pi lm_2)=0$ , we theoretically select the first zero point 0.383 in Fig.2 for the second harmonic signal amplitude of background interference fringes at other zero points is no longer 0. However, in practical application, there is a certain discrepancy between the experimental optimal value and theoretical optimal value, so it is necessary to adjust within a certain range including the theoretical optimal value to obtain the optimal parameters based on the minimum measured second harmonic noise level.

When the multi-frequency modulation method is applied to TDLAS system, the actual parameters that can be directly adjusted are the basic modulation current amplitude  $i_1$ , dither current amplitude  $i_2$  and dither current frequency  $f_2$ . Through fine-tuning  $i_1$ , the position of the second harmonic absorption peak will coincide with the interference fringe envelope minimum value point. Taking the standard deviation (STD) of 500 second harmonic peak data as the evaluation standard,  $i_1$  and  $i_2$  are repeatedly adjusted to minimize the noise level of the second harmonic peak, so as to eliminate the interference of system and environmental factors fluctuations to the greatest extent.

# 3 Experimental Setup on TDLAS System

Based on the above theory, this paper built the principal verification platform for online oxygen concentration detection in encapsulated medicine bottles based on TDLAS technology, as shown in Fig. 3, signal generator, lock-in



Fig. 3: TDLAS principle verification platform



Fig. 4: Application test site

amplifier, laser temperature controller, oscilloscope and other basic digital signal equipment are included. In addition, a vertical-cavity surface-emitting laser (VCSEL) diode (Laser component, single-mode VCSEL, 760 nm, TO5 footprint) with a free-running output power of 0.3 mW was selected as the laser source. The laser temperature and exciting current were strictly controlled by a laser controller (Thorlabs, VITC002). The operation temperature of the laser diode is adaptively controlled at 21.20°C, the center frequency scanning around the spectral lines is around 760.885 nm for covering the scope of oxygen absorption spectral lines.

After the TDLAS principle verification, in order to measure the interference suppression effect of the parameter-optimized multi-frequency modulation method, we carried out the residual oxygen detection of packaged medicine bottles on site. The detailed size of penicillin vial used is 7 ml volume, 22 mm bottom diameter, and 22 mm bottle height. By a simple oxygen-nitrogen mixing method, we prepared three groups of bottles with oxygen concentrations of 0%, 5% and 21%, respectively. Application tests was conducted on the actual lamp inspection line. The measured results are described in Section 4.

# 4 Experimental Results and Discussion

A series of verification experiments were carried out, and the specific parameters were set as follows: The open optical path length is 10cm, the laser frequency-current tuning coefficient is (0.6cm-1cm)/mA, the laser temperature is 30°C, the standard length is 5.2cm, the sawtooth wave scanning frequency is 25Hz, the sawtooth wave scanning range is [0.5mA, 2mA], and after the parameter optimization method described in Section 2.2, as shown in Fig. 5, the



Fig. 5: Variation of standard deviation of second harmonic peak data with  $i_2$  and  $f_2$ 



Fig. 6: Data of 50min second harmonic peak under different modulation methods

amplitude of the underlying modulated current  $i_2$  is 0.17mA, the basic modulation current frequency  $f_2$  is 2500Hz.

# 4.1 Static Stability Measurement

A standard bottle with 5% oxygen concentration was selected as the measurement sample. The multi-frequency modulation method with optimized parameters and singlefrequency modulation method were compared under continuous long measurement for nearly 50min. The amplitude of single-frequency modulation current was 0.17mA, and the frequency was 2500Hz. The basic modulation current parameters of multi-frequency modulation are the same as those of single-frequency modulation, and the dither modulation current amplitude is  $62 \mu A$ , the frequency is 500Hz. Fig. 6 shows the second harmonic peak data of the two methods. The standard deviation of the second harmonic peak data under multi frequency modulation is 2.96, while that under single frequency modulation is as high as 32.55. It is obvious that the multi frequency modulation method after parameter optimization has obvious improvement effect on interference fringe suppression.

### 4.2 Dynamic Accuracy Measurement

Comparing the multi-frequency modulation system with the single-frequency modulation system, Fig. 7 shows the background interference fringe 2f signal under the condition of no absorption gas. The mean value of background interference fringe noise is 5.07, the variance is 2.85, and the variance to mean ratio is 56.2%; while under multifrequency modulation, the mean value of background interference fringe noise is 0.084, the variance is 0.0048, and the variance to mean ratio is 5.7%, which is significantly lower than the former and restrains the baseline drift at the same time. As for the absorption condition, Fig. 8 shows that the second harmonic of the absorbed signal in the singlefrequency system has been distorted and baseline drift compared with the ideal case (no interference fringe), and



Fig. 7: Comparison of background interference fringes with no absorption



Fig. 8: Comparison diagram of the second harmonic of the absorbed signal

the SNR is 3.122dB; after multi-frequency modulation, a high fitting degree can be obtained, and the SNR reaches 36.979dB, which is an order of magnitude higher and greatly improves the measurement accuracy of system. In addition, the dynamic positive detection rates of 0% and 5% oxygen concentration packaged bottles on the high-speed lamp inspection line can reach 99.87% and 93.56%, which proves that our multi-frequency modulation method has excellent performance in TDLAS-based detection system.

# 5 Conclusion

This paper mainly studies parameter-optimized multifrequency modulation method applying in on-line detection for residual oxygen in encapsulated medicine bottles based on TDLAS technology, so as to provide a reliable solution to the complex optical interference problem of TDLAS system. From the perspective of signal modulation, by exploring the internal mechanism of optical interference fringes, we found how the parameter changes affect detection accuracy, and proposed to use multi frequency modulation method to suppress the interference noise of TDLAS system, which was verified by experiments in the TDLAS-based online detection system for residual oxygen in the cylinder. Under the condition of fewer adjusting parameters and the robustness of the parameters with the system hardware platform, the static stability (i.e., standard deviation is 2.96) and dynamic accuracy (i.e., SNR is 36.979dB) were obtained.

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