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基于双正弦复合调制技术的高精度氨气浓度检测方法

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摘　要:针对传统可调谐二极管激光吸收光谱技术中锯齿波扫描导致的基线拟合误差与噪声敏感性问题,提出采用双正弦复合调制的二次谐波检测方法。该方法融合低频正弦扫描与高频正弦调制,规避了基线拟合误差,同时提升系统抗噪能力,实现痕量氨气(NH_3)的高精度检测。实验表明,双正弦复合调制方式相较于传统低频锯齿扫描叠加高频正弦调制,可以优化系统的信噪比,浓度与二次谐波幅值的线性拟合度提升至0.997 8,检测灵敏度提升至0.053 ppm。实验验证了双正弦复合调制在痕量气体浓度检测中的可行性与稳定性,为高鲁棒性、高精度及低成本的NH_3工业排放实时监测提供了新思路与技术路径。

关键词:TDLAS-WMS技术;波长调制光谱;二次谐波;双正弦调制;痕量气体浓度检测
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0　引言

可调谐二极管激光吸收光谱技术(Tunable Diode Laser Absorption Spectroscopy,TDLAS)因其高灵敏度、高精度特性,已成为痕量气体检测的重要手段[1-3]。波长调制光谱技术通过高频调制激光频率提取二次谐波信号,有效抑制低频噪声,显著提升了气体浓度的检测精度,并在实际中应用[4-5]。然而,波长调制技术(Wavelength Modulation Technology,WMS)技术常采用锯齿波扫描方案来恢复吸收线形[6-7],在实际应用中面临两大关键挑战:其一,锯齿波的非线性特性导致频域能量分散,因为其固定的高频分量,系统对于带宽具有较高的要求。调制后产生的多边带信号在气体吸收过程中因高频分量的过度衰减而大幅削弱有效信号强度,故需要对调制频率进行有效限制[8];其二,锯齿波扫描导致低频噪声及环境干扰(如机械振动)混叠,导致锁相放大器带宽内噪声积分范围扩大,信噪比(Signal-to-Noise Ratio,SNR)难以满足高动态响应工业场景的需求。近年来研究者通过改进数据处理的算法或引入参考光路监测激光光强变化等方法缓解了上述问题[9-12]。但参考光路法需增加分束器与额外探测器,这将引入光路不对称误差;算法优化法仅事后补偿谐波非线性衰减,无法抑制信号采集阶段的噪声混叠,在强振动环境下性能急剧退化。另外,锯齿波调制的固有限制如基线拟合精度有限[13](例如对于1%的吸收特征,基线拟合中的1%误差会导致直接吸收光谱

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(Direct Absorption Spectroscopy,DAS)中产生100%的峰值测量误差[14])、谐波幅值非线性衰减及抗干扰能力不足等特性,仍是制约痕量气体检测精度进一步提升的瓶颈。

针对上述问题,本文提出一种对激光进行双正弦复合调制的改进方法,核心是通过频率为f_s的低频扫描正弦波与频率为f_m的高频调制正弦波的复合作用,实现对激光频率的精准控制。利用正弦波的连续平滑特性消除时域非线性导致的基线漂移,并且可将信号能量集中于频率为$2f_m$的所需谐波边带附近,规避$1/f$等噪声主导的低频干扰,实验根据窄带锁放大器原理计算分析,对谐波信号进行提取与分析处理[15]。通过进行对比实验,对在不同扫描方式下,经过NH_3吸收后的光强信号进行采集及分析处理[16],并对计算得出的相应气体浓度值进行数据分析对比,进而验证正弦波扫描在气体浓度测量方面的优越性。

1 基本原理

1.1 Beer-Lambert定律

当特定频率的激光穿过待测气体分子时,激光部分能量会被气体分子吸收,导致出射光强发生衰减,衰减程度符合Beer-Lambert定律[17],如式(1)所示。

$$I(v)=I_0(v)\exp\left[-\alpha(v)CL\right] \tag{1}$$

式中,$I_0(v)$为激光入射光强,$I(v)$为穿过待测气体后透射光强,$\alpha(v)$为待测气体的分子吸收系数,C为待测气体浓度,L为待测气体吸收光程。

1.2 波长调制技术

波长调制光谱技术是通过对激光控制器注入正弦信号来对激光波长进行调制,使吸收信号转化为谐波分量同时也调制了激光光强[18],经调制过后的光强可表示为

$$I(t)=I_0(1+n\sin\omega_m t) \tag{2}$$

式中ω_m为调制频率,$\omega_m=2\pi f_m$,n为光强调制系数。结合Beer-Lanbert定律,在标准大气压下,以洛伦兹吸收线型为例[19],可以得出光强信号与调制系数、调制频率的关系为

$$I(t)=I_0\left[1+n\sin\omega_m t-\frac{\alpha_0 CL}{1+\left(\frac{v_f}{\Delta_v}\right)^2\sin^2\omega_m t}\right] \tag{3}$$

式中,α_0为吸收截面,v_f为波长调制幅度,Δ_v为吸收线半峰全宽。光强$I(t)$中包含不同频率的谐波分量,将其进行傅里叶级数展开,可得一次谐波分量系数和二次谐波分量系数分别为

$$I_{1f}=nI_0 \tag{4}$$

$$I_{2f}=-k\alpha_0 LCI_0 \tag{5}$$

式中,k为与v_f、Δ_v相关的常数值。由式(4)、(5)可得,一次谐波I_{1f}受初始光强和调频系数所影响,二次谐波I_{2f}与气体的浓度C相关。但由于一次谐波频率较低,易受光源强度波动、基线漂移及低频噪声影响,导致其幅值不稳定。二次谐波频率较高,通过锁相放大器可有效滤除低频干扰,且对吸收线中心频率更敏感,鲁棒性更强。因此,二次谐波不仅从理论分析严格关联浓度,且在工程中兼具抗干扰、高灵敏和强鲁棒性,是实际系统的解调气体浓度首选依据。为避免光强的影响,本文的对比实验均使用一次谐波中心值I_{1f}对二次谐波峰值I_{2f}进行归一化处理,该方法得到的归一化二次谐波信号只与待测气体本身有关,并与气体浓度成正比关系。

1.3 频域变换理论与噪声功率谱密度分析

对于系统扫描信号和调制信号,在相同参数下正弦波的频谱能量集中于单一主边带(如f_s+f_m),而锯齿波因其周期性非连续性,其傅里叶级数包含多个高次谐波分量(如nf_s+f_m)。这种能量分散特性导致锯齿波各边带幅值随谐波次数n递减(幅值衰减为$1/n$)。此外,气体吸收系数$\alpha(f)$通常随频率递增,导致锯齿波高频边带因吸收增强而显著衰减;而正弦波主边带频率固定且较低,吸收损耗较小。通过实验验证,正弦波调制的二次谐波幅值在动态检测中表现出更强的鲁棒性。

对于传统锯齿波扫描加正弦波调制的方式，其多边带信号的衰减因子为：$g_1=\mathrm{e}^{-\alpha(nf_s+f_m)L}$。由于高频分量$(nf_s+f_m)$的吸收系数$\alpha$更大，边带信号衰减严重，尤其对于高次谐波$(n\geqslant 2)$，衰减幅度呈指数级增加。而对于正弦波扫描加正弦波调制的方式，单一主边带的衰减因子：$g_2=\mathrm{e}^{-\alpha(f_s+f_m)L}$，由于能量集中于单一主边带，所以可有效地避免高频分量的过度衰减。

在痕量气体检测中，噪声特性对检测精度有着至关重要的影响。锁相放大器的噪声功率谱密度通常表现为$S_{\mathrm{noise}}(f)=S_0\cdot\frac{1}{f^{\gamma}}$，$S_0$为基准噪声功率谱密度，$\gamma$为噪声特性指数。对于常见的$1/f$噪声，$\gamma=1$；而对于白噪声，$\gamma=0$。在锯齿波扫描、正弦波调制方式下，由于锯齿波的频谱包含基波和多谐波分量，调制后会产生多个边带。每个边带都会引入相应的噪声，噪声积分需覆盖所有边带频带范围。这导致总噪声功率显著增加。相比之下，双正弦波调制将信号能量集中于单一主边带，频带宽度较窄。因此，其噪声积分范围也相应减小，噪声功率大幅降低。具体来说，正弦波扫描使得主边带频带宽度远小于所有锯齿波调制产生多个边带频带宽度之和，目标信号噪声功率也显著降低。这种窄带噪声特性使得双正弦调制在实际应用中能够更有效地抑制环境干扰，提高检测系统的稳定性和可靠性。可以看出双正弦调制在频域噪声抑制方面的显著优势，为高精度痕量气体检测提供了更优的技术方案。

2　实验与讨论

2.1　实验装置设置

本实验系统基于可调谐二极管激光吸收光谱-波长调制光谱技术（TDLAS-WMS）搭建，如图1所示，主要由计算机、数据采集卡（NI-USB6363）输出激光驱动信号并采集经气体吸收后的信号、分布式反馈激光器（DFB）、激光器驱动器（LCM-6000）、石英气体池（气室长100 cm、内径3 cm，出入气口长3 cm、内径5 mm）、光电探测器（THORLABS-DET10D2）、配气仪、真空泵以及基于LabVIEW的信号处理平台组成。实验中，上位机通过数据采集卡输出驱动信号控制DFB激光器，利用注入电流调制实现激光波长动态扫描，完整覆盖NH_3在1 512 nm附近的基频吸收线；激光经准直后通过气体吸收池，透射光由光电探测器转换为电信号，经数据采集卡数字化传输至上位机后，通过LabVIEW平台进行锁相放大计算处理，提取二次谐波分量$(2f_m)$，并结合Beer-Lambert定律反演NH_3气体浓度。实验均在常压（(101.3±1.0) kPa）及室温（(296±1.0) K）条件下完成，以抑制环境波动对测量精度的影响。

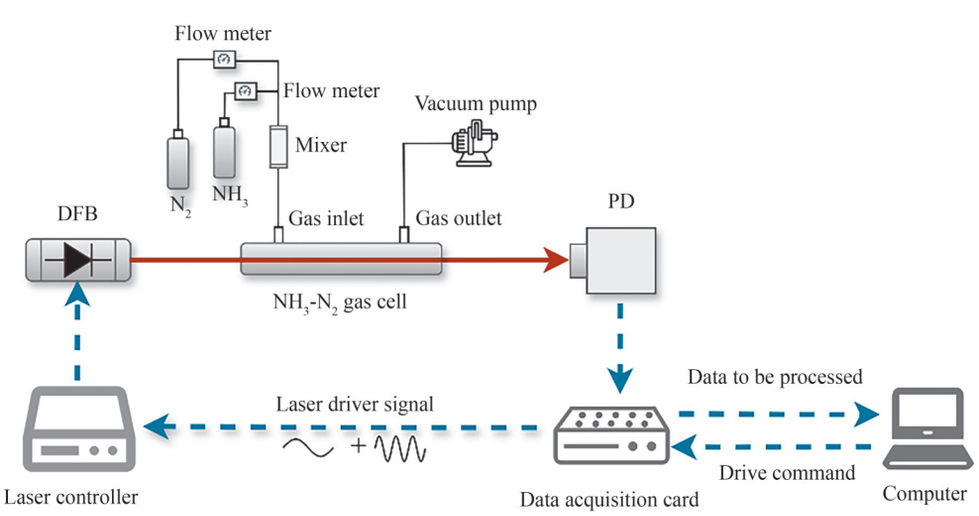


图1　实验装置图

Fig.1　Experimental device diagram

实验分两组进行，为了更直观地对比两组实验结果的差异，激光分别用相同频率$f_s=10$ Hz的锯齿波和正弦波扫描，并设置调制频率均是$f_m=10$ kHz。为避免扫描范围不足，无法捕获线型函数的中心附近的光谱特征区域进而导致的信号失真和基线漂移，实验根据NH_3吸收半宽（FWHM≈0.13 $\mathrm{cm^{-1}}$），设定激光器出

射激光波长调制范围。实验使用质量流量控制器将300 ppm浓度的NH_3标准气体与99.999%浓度的N_2按照一系列比例进行混合，得到一系列梯度痕量NH_3标准气体，将配置的分别充入气体池中，对配置的275 ppm、250 ppm、225 ppm、200 ppm、175 ppm、150 ppm、120 ppm、100 ppm共计8种浓度测试气体进行测量实验。

2.2 调制参数标定

为优化WMS的检测灵敏度，需对调制深度进行精确设定。根据波长调制光谱理论，二次谐波信号的幅值取决于调制系数m（即调制幅度与气体吸收线半宽之比）。对于洛伦兹线型（Lorentzian profile）的吸收谱线，理论推导与数值模拟均表明，当频率调制范围约为气体吸收线半高全宽的2.2倍时（2.2×FWHM），二次谐波信号的幅值达到最大值[20-22]。此时系统具有最高的信噪比（SNR），能够最有效地提取微弱的吸收信号。因此，实验设定激光频率调制范围覆盖气体吸收线宽度的2.2倍，以达到最佳的调制效果。基于HITRAN数据库，NH_3在波长1 512 nm处的吸收线半高宽（FWHM）为0.13 cm^{-1}。为精确确定波长WMS的调制电压范围，需标定激光频率扫描与调制深度的关系。因空间腔标准具的FSR过大，测得的激光扫频范围误差较大，故实验先通过空间腔标准具标定光纤标准具的FSR，本实验通过空间腔标准具（自由光谱范围（FSR）=9.37 GHz）与光纤标准具的联合标定，实现调制参数的优化。参数标定实验光路如图2所示：使用锯齿波扫描信号驱动激光器，出激光经分束器分为两路，一路通过空间腔标准具，另一路通过光纤标准具（FSR待标定）。两路信号由光电探测器同步采集，结合扫描电压信号（0～1.8 V）分析频率响应特性，并确定调制参数。

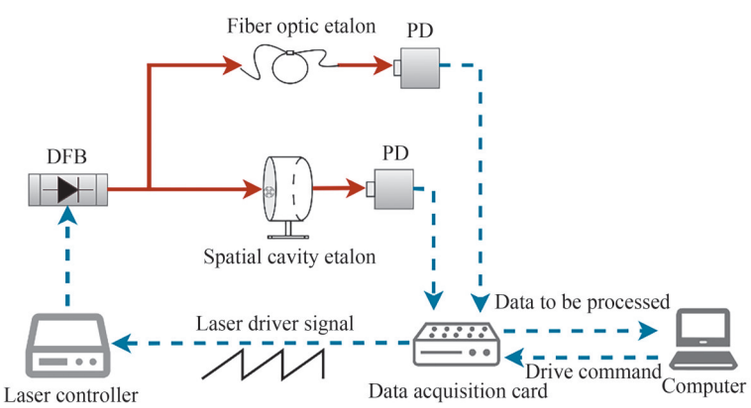


图2 调制参数标定光路示意图

Fig.2 Schematic diagram of modulation parameter calibration optical path

测得6个空间腔FSR间隔对应73个光纤标准具峰（如图3），计算可得到实验所用光纤标准具FSR为

$$\mathrm{FSR}_{\mathrm{fiber}}=\frac{9.37\ \mathrm{GHz}\times 6}{73}\approx 0.77\ \mathrm{GHz} \tag{6}$$

根据目标处NH_3吸收线半高宽（FWHM=0.13 cm^{-1}），目标调制带宽需覆盖2.2×FWHM（对应8.58 GHz），

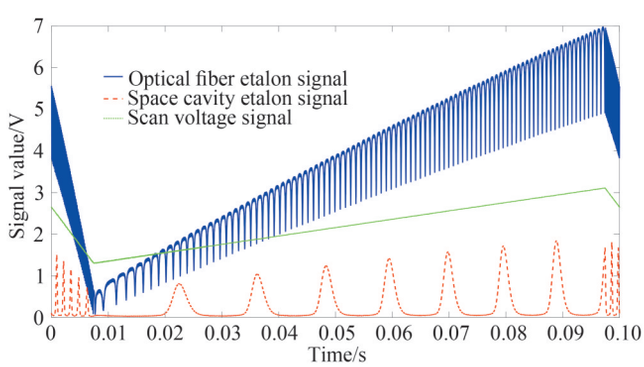


图3 标准具信号对比

Fig.3 Comparison of etalon signals

光纤标准具的FSR为0.77 GHz，所需FSR数量$N=\left(\frac{8.58\ \mathrm{GHz}}{0.77\ \mathrm{GHz}}\right)\approx 11.14$个。调整调制电压的幅值，使得其能够覆盖11.14个左右的峰，得到最佳调制电压幅度在0.16 V左右。

3 数据处理结果分析

3.1 二次谐波幅值及线性度分析

在测试不同浓度气体前，首先以99.999% 的高纯度N_2对气体池进行3 min冲扫，消除残留气体的干扰，随后将待测气体按浓度梯度充入气体池。实验采用正弦波调制方式对激光进行扫描，通过锁相放大器提取二次谐波信号，并且为消除光强对实验结果来的影响，实验对二次谐波信号与一次谐波信号进行幅值相除处理，得到归一化二次谐波峰峰值$2_f/1_f$，结果如图4所示。

对实验梯度氨气浓度与对应归一化二次谐波峰峰值进行线性拟合，关系如图5所示。使用正弦波扫描得到归一化二次谐波峰值与NH3浓度的函数关系为：$\mathrm{Amp}_1=0.0018C+0.0424$，线性拟合度$R_1^2=0.9978$，在相同实验条件下改用锯齿波扫描模式，其二次谐波信号经相同处理后，梯度氨气浓度与对应归一化二次谐波峰峰值拟合关系为$\mathrm{Amp}_2=0.0019C-0.0034$，线性拟合度$R_2^2=0.9919$。实验结果表明：相较于使用锯齿波对激光进行扫描的方式，正弦波扫描系统提高了标定曲线的线性度与准确性。

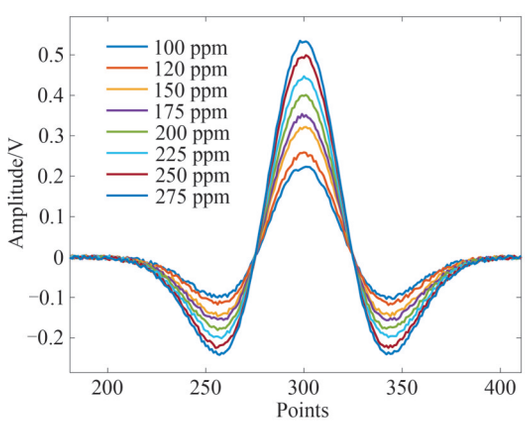


图4 NH_3的二次谐波信号

Fig.4 Second harmonic signal of NH_3

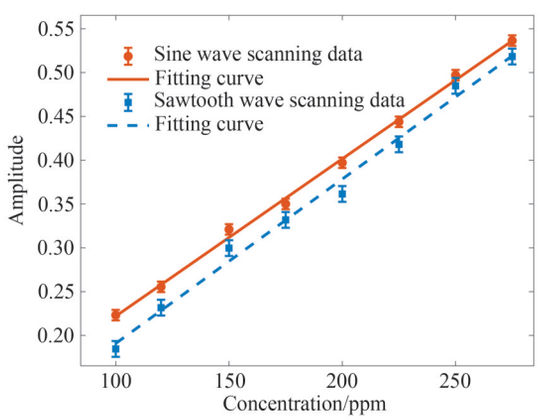


图5 二次谐波峰值与氨气浓度的关系

Fig. 5 Relationship between second harmonic peak and NH_3 concentration

3.2 系统稳定性与检测灵敏度对比分析

实验对配置的150 ppm浓度的NH_3，每0.1 s记录每一次输出的浓度数据，持续记录900 s，记录结果如图6所示。

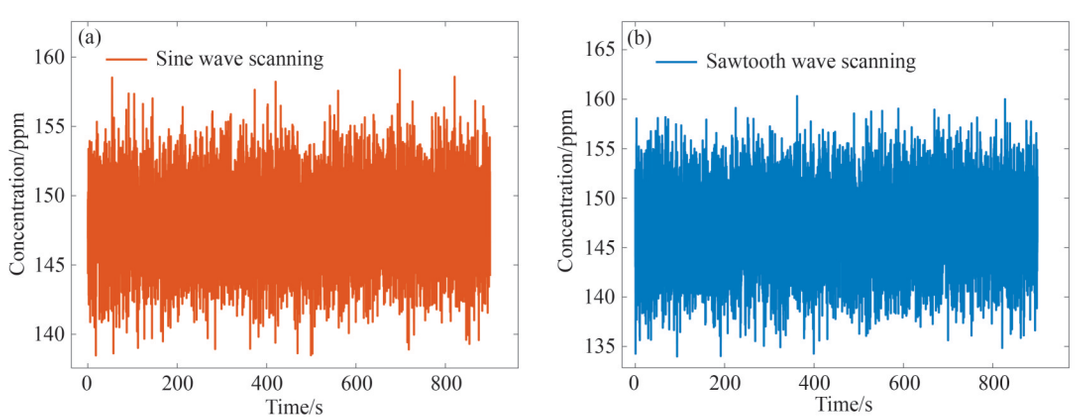


图6 浓度测量结果随时间分布图。(a)正弦波扫描；(b)锯齿波扫描

Fig.6 Time distribution of concentration measurement results. (a)Sine wave scanning; (b) Sawtooth wave scanning

对900 s内150 ppm NH_3的浓度检测数据进一步分析，其数据分布直方图和概率分布的拟合曲线如图7所示。由拟合曲线可知，测量的浓度数据类似于高斯分布，由此计算出双正弦调制结果的半峰半宽（HWHM）为3.28 ppm，即系统的测量精度优于2.1%；锯齿波扫描结果的半峰半宽为4.62 ppm，系统的测量精度优于3.08%。由概率分布结果验证在多次测量同一稳定气体浓度时，正弦波扫描的方式得到的浓度值更为集中，波动较小。这表明双正弦波调制系统具有更高的测量精度、更好的重复性和更强的稳定性。

对900 s内的实验所得到的浓度数据进行Allan方差计算以评估系统稳定性，得到结果如图8所示。

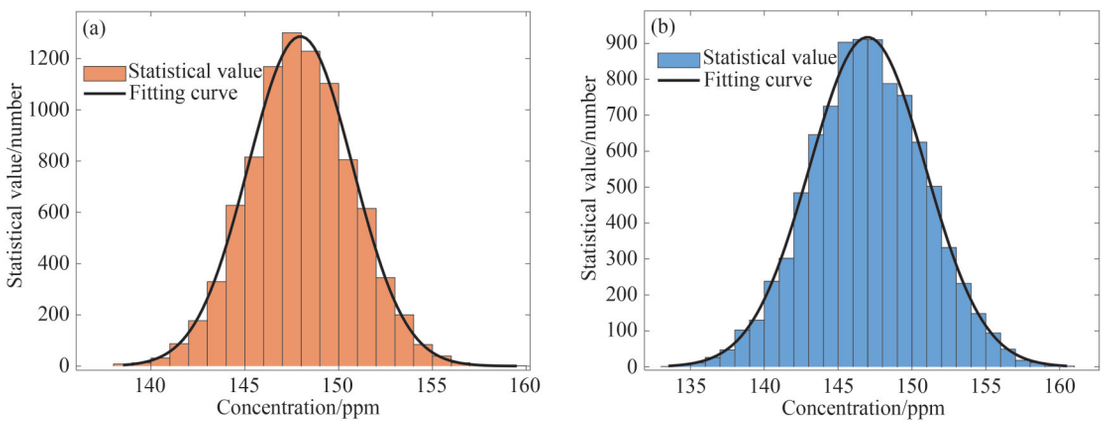


图7 浓度测量结果直方图及拟合曲线。(a)正弦波扫描；(b)锯齿波扫描

Fig.7 Histogram and fitting curve of concentration measurement results. (a)Sine wave scanning; (b) Sawtooth wave scanning

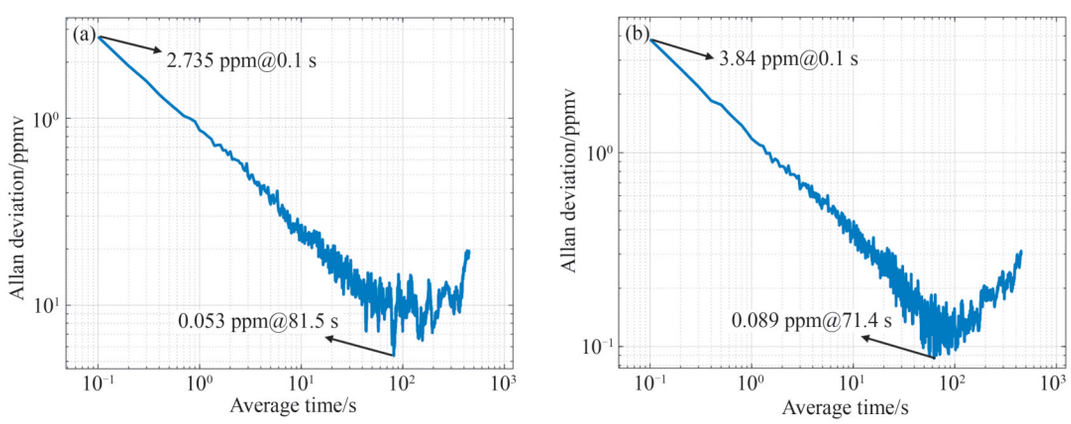


图8 不同扫描方式下系统Allan方差评估结果。(a)正弦波扫描；(b)锯齿波扫描

Fig.8 Allan variance evaluation results of the system under different scanning modes.(a)Sine wave scanning; (b) Sawtooth wave scanning

由结果可知对于锯齿波扫描加正弦波调制的方式在积分时间为0.1 s时得到Allan方差为3.84 ppm，当积分时间增加至71.4 s时检测灵敏度为0.089 ppm；对于双正弦调制方法，在积分时间为0.1 s时得到Allan方差为2.735 ppm，相较于锯齿波扫描的方式，约降低28.8%，当积分时间至81.5 s时对应的检测灵敏度降至为0.053 ppm，相较于锯齿波扫描的方式，检测下限降低40.5%；结果表明，正弦波扫描弦波调制的激光器驱动方式，可大幅提升了系统的稳定性和低浓度氨气检测灵敏度。

文献[23]用近红外激光器，采用波长调制光谱技术测量20 cm混合气体池中的氨气浓度，当积分时间为0.1 s时得到Allan方差为0.3 ppm，当积分时间增加至441 s时检测灵敏度为0.04 ppm。文献[24]使用波长高频扫描技术和多次反射池技术测量氨气浓度，可在24.32 m光程下系统检测限为0.157 mg/m^3，测量精度优于2.86%。与文献的测量结果及精度相比，本文在较短的吸收池内可达到更优的检测灵敏度、更快地响应时间，与更优的测量精度。未来的研究可将继续聚焦探索于系统的工程化与智能化。一方面，基于FPGA等嵌入式平台实现双正弦调制解调系统的全数字化与集成化，以降低成本并提升便携性；另一方面，结合如

神经网络等先进信号处理算法与温压补偿模型,解决复杂工业现场中的不同噪声干扰与环境波动问题,从而为痕量气体的高精度实时监测提供更完善的技术解决方案

4 结论

鉴于传统锯齿波扫描提取二次谐波的局限性,本文提出基于双正弦复合调制的二次谐波检测方法,通过将低频正弦扫描与高频正弦调制协同作用,实现了NH_3吸收线中心频率的连续覆盖及二次谐波信号的高效提取。实验结果表明,在相同硬件系统的条件下,相较于传统低频锯齿扫描加高频正弦波调制,该方法有效抑制低频噪声与宽带干扰,提高了检测系统的信噪比,减少了二次谐波峰值信号的波动,避免了传统锯齿波多谐波边带导致的高频分量吸收衰减,使得浓度与谐波幅值线性拟合度提高至0.099 78。通过Allan方差分析,系统检测下限在更短的积分时间内出现明显降低,检测灵敏度达到0.053 ppm,显著增强了检测灵敏度与动态响应能力。研究结果可为工业排放场景中NH_3的高精度实时监测提供了新的思路与技术路径。

但仍存在一些潜在的局限性与挑战。首先,该方法对激光器的调制响应带宽提出了更高的要求,尤其是在高频调制部分,这可能限制了某些带宽较窄的激光器的应用。其次,在极高浓度的NH_3或存在多种强干扰气体共存的复杂工业排放场景中,吸收信号的非线性效应和多组分交叉干扰可能更为显著,本方法的性能可能会受到影响。此外,如需要更精密的调制信号生成和信号处理等系统复杂性的增加,也可能对成本和维护提出更高要求。针对上述挑战,未来的研究将着重于调制参数的改变、引入更优的滤波算法以应对复杂噪声干扰、引入长光池来提升检测极限等优化路径。进一步开发更集成、低成本的检测系统,拓宽其在工业及环境监测领域的应用。

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High Precision Ammonia Concentration Detection Method Based on Double Sinusoidal Compound Modulation Technology

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Abstract: Ammonia (NH_3) is a fundamental chemical agent extensively utilized in industrial synthesis, agricultural fertilizer production, and large-scale refrigeration systems. However, its leakage poses severe threats to both human physiological health and the atmospheric environment, necessitating the development of high-precision, real-time concentration monitoring techniques. Currently, Tunable Diode Laser Absorption Spectroscopy (TDLAS) combined with Wavelength Modulation Spectroscopy (WMS) is the preferred method for trace gas detection due to its high selectivity and sensitivity. Nevertheless, traditional TDLAS-WMS systems typically employ a low-frequency sawtooth wave for wavelength scanning. This conventional approach faces two critical technical bottlenecks: first, the inherent non-linearity and discontinuous nature of the sawtooth waveform lead to spectral energy dispersion and significant baseline fitting inaccuracies; second, the sawtooth scanning often overlaps with low-frequency environmental noise and mechanical vibrations, which expands the noise integration bandwidth of the

lock-in amplifier and consequently limits the Signal-to-Noise Ratio (SNR) in complex industrial scenarios.

To overcome these limitations, this paper proposes an optimized detection method based on dual-sinusoidal compound modulation for second-harmonic signal extraction. The core innovation lies in the simultaneous application of a low-frequency sinusoidal scanning signal (f_s) and a high-frequency sinusoidal modulation signal (f_m) to achieve precise control of the laser frequency. Unlike the abrupt transitions of a sawtooth wave, the smooth and continuous nature of the dual-sinusoidal waveform effectively eliminates time-domain non-linearities, thereby suppressing baseline drifts at the source. Theoretically, this compound modulation concentrates the signal energy more efficiently near the specific harmonic sidebands ($2f_m$), which significantly reduces the bandwidth requirements for the detection system and enhances its immunity to $1/f$ noise. By utilizing narrow-band lock-in amplification to extract the normalized second-harmonic ($2f/1f$) signal, the proposed method ensures a more robust and stable representation of the gas absorption line shape, providing a theoretical foundation for higher detection accuracy and lower cost.

The experimental validation was conducted using a 1 512 nm Distributed Feedback (DFB) laser and a 100 cm quartz gas cell under controlled laboratory conditions. A comprehensive platform based on a high-speed data acquisition card (NI-USB6363) and LabVIEW was established to compare the performance of the dual-sinusoidal method against the traditional sawtooth-plus-sine modulation. Precision calibration was performed using a combination of spatial cavity and fiber etalons to optimize the modulation depth. The experimental results demonstrate a substantial performance enhancement: the linear fitting degree (R^2) between NH_3 concentration and the second-harmonic amplitude improved from 0.991 9 to 0.997 8. Quantitatively, the detection sensitivity was enhanced from 0.089 ppm to 0.053 ppm, representing a significant improvement of approximately 40.5%. Furthermore, Allan variance analysis confirms that the system achieves its optimum detection limit within a shorter integration time of 81.5 seconds, exhibiting superior long-term stability and a more concentrated Gaussian distribution of measurement data compared to the traditional scanning mode. These results quantitatively confirm that the dual-sine approach ensures higher repeatability, lower detection limits, and faster convergence to optimal precision.

In conclusion, this research successfully demonstrates that dual-sinusoidal compound modulation is a highly feasible and stable approach for high-precision trace gas detection. By circumventing the intrinsic flaws of sawtooth scanning-specifically baseline fitting errors and high sensitivity to low-frequency noise—this method provides a new technical pathway for the robust, real-time monitoring of NH_3 industrial emissions. The findings verify that the optimized system can maintain high accuracy and stability even in environments with significant physical disturbances. Future research will focus on further refining the modulation parameters and integrating advanced digital signal processing algorithms, such as adaptive filtering or artificial neural networks, to address multi-component gas interference in even more complex scenarios. Additionally, the development of integrated, FPGA-based hardware platforms and the incorporation of multipass cells will be explored to further enhance the detection limits and promote the widespread application of this technology in portable environmental monitoring and industrial safety systems.

Key words: TDLAS-WMS technology; Wavelength modulation spectroscopy; Second harmonic; Double sine modulation; Trace gas concentration detection

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