Wavelength modulation spectroscopy based on quasi-continuous-wave diode lasers

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A modified wavelength modulation spectroscopy (WMS) based on the self-heating effect of the tunable diode laser when driven in quasi-continuous-wave (QCW) mode is investigated. A near-infrared distributed feedback (DFB) diode laser working at the QCW mode is employed as the QCW light source, and CO_2 is selected as the target gas. The characteristic of the QCW second harmonic (2f) line profile is analyzed through a comparison with that of the traditional CW WMS with the same system. A noise-equivalent absorbance of 3.2×10^{-5} $Hz^{-1/2}$ for CO_2 at 1.58 μm is obtained with 18-m optical path. The QCW WMS lowers the dependence on lasers and expands selectivity, thus verifying the feasibility of the method.

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Wavelength modulation spectroscopy (WMS) is a widely used technique for trace gas detection using tunable diode lasers[1−3]. Theoretically, WMS has an improvement of 3–5 orders of magnitude in sensitivity compared with direct absorption spectroscopy, which is due to a higher signal-to-noise ratio (SNR) and reduced sensitivity to the baseline[1]. Commonly, WMS requires diode lasers continuously working at room temperature and generating single-mode emissions. However, in some wavelength regions that are limited by the semiconductor materials and technique levels, continuous current supply can cause over-heating of the diodes that must be cooled at very low temperatures. Diode lasers can only be operated at room temperature in pule or quasi-continuous-wave (QCW) mode[4−6]. QCW diode lasers are attractive light sources for trace-gas sensing, because they have easy-operating conditions and large wavelength coverage that can potentially reduce the system cost and expand the detectable species.

At present, trace gas detections in laser absorption spectroscopy using pulse or QCW lasers are mainly centered on mid-infrared (MIR), where many thermoelectrically cooled quantum-cascade lasers (QCLs) can operate at room temperature in the pulse or QCW mode[4−9]. In these studies, the emitting wavelengths of the lasers were tuned through absorption lines by slowly swept temperature or by sub-threshold current ramp whose heating effect caused the wavelength shift[4,5,9]. Gated integrators have mostly been employed in these systems to collect pulsed signals from the photodiodes, which means that the final acquired signals are direct absorption signals[4,5]. Another pulsed-laser-based absorption method relies on the linear frequency chirp obtained from QCL, which is brought about by self-heating of the active region when relatively long (>100 ns) current pulses are applied[6,10−12]. WMS has been applied to pulsed QCLs in Refs. [9, 13], wherein a high frequency sinusoidal dither is superimposed onto the sub-threshold current ramp or onto the long duration current pulse to implement higher-frequency temperature modulation.

The previous work on pulsed-QCL-based spectroscopy mainly adopts direct absorption spectroscopy and takes advantage of the relatively stronger absorptions in the fundamental vibrational bands. However, direct absorption spectroscopy distinguishes weak absorption signals from large intensity backgrounds, because these may be easily interfered by intensity fluctuations and low frequency noises, except for sufficient high scanning speed.

So far, a few low-cost diode lasers in many wavelength ranges have been utilized. These are easy to operate, although they need pulsed or QCW current supplying at room temperature. Thus, it is important to develop a simple, easy to operate, and highly sensitive wavelength modulation spectroscopy based on pulsed or QCW lasers. Continuous-wave (CW) lasers can also work at the QCW mode to increase the peak power and improve thermostability and sensitivity, thus increasing immunity against harsh environment conditions, such as high temperature or long path field applications.

In this letter, we investigated a modified QCW-based WMS different from the previous work. A near-infrared (NIR) distributed feedback (DFB) diode laser working at the QCW mode was employed, and CO_2 was selected as the target gas in this letter.

The QCW modulation signal consists of two fundamental waves, namely, ramp and square (Fig. 1), wherein the voltage ramp with a given direct current (DC) offset is generated from a function generator first, and then the ramp is electrically chopped by a higher frequency QCW signal. The QCW modulation voltage signal is converted to current signal through a laser diode (LD) driver described below. The amplitude of the ramp can induce a wavelength shift of the laser to cover a complete absorption line. In addition, when the diode laser is driven by a pulsed or QCW current, a frequency shift (or wavelength shift) occurs during the excitation due to the self-heating of the active region of the laser chip[6,10]. The frequency varies linearly with pulsed time when relatively short duration current is used[6]. Therefore, combined with phase-sensitive detection technology (e.g., lock-in amplifier), a new modified QCW WMS is achieved, wherein the ramp current sweeps the wavelength and the QCW current modulates it.

When the diode laser is driven by a short duration

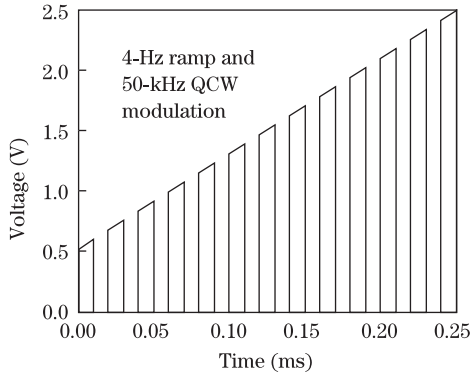

Fig. 1. QCW modulation signal.

QCW current (10 μs in this letter), the frequency shifts in a linear manner. Thus, the instantaneous output frequency of the diode laser at the moment t can be expressed as

$$v(t) = (v_c + mt) \cdot \mathrm{sqr}(ft), \tag{1}$$

where v_c is the emitting frequency of the laser related to the intensity of the square wave driving current, m is the frequency tuning coefficient caused by the heating effect in the active region of the laser chip, f is the frequency of the square wave signal, and $\mathrm{sqr}(ft)$ is the unit square wave signal given as

$$\mathrm{sqr}(ft) = \begin{cases} 1, & -\dfrac{1}{4f} \leqslant t < \dfrac{1}{4f} \\ 0, & -\dfrac{1}{2f} \leqslant t < -\dfrac{1}{4f}, \quad \dfrac{1}{4f} \leqslant t < \dfrac{1}{2f} \end{cases}. \tag{2}$$

The light intensity of the laser after gas absorption can be expressed as

$$I(v) = I_0(v) \exp[-\sigma_0 \overline{\chi}(v) NL], \tag{3}$$

where $I_0(v)$ stands for the light intensity before being absorbed, σ_0 is the absorption cross-section at the absorption line center, $\overline{\chi}(v)$ refers to the line shape function of an absorption, N is gas concentration, and L is optical absorbing length. Under the weak absorption conditions:

$$I(v) = I_0(v)[1 - \sigma_0 \overline{\chi}(v) NL]. \tag{4}$$

The laser frequency v_c varies slowly with ramp current, so does the laser output power $I_0(v)$. Here, we ignore the influence of the variable laser power, that is, $I_0(v) \approx I_0$.

Under atmospheric conditions, the spectral lines are broadened predominantly by pressure, so the absorption line can be expressed by the Lorentzian lineshape function given as

$$\begin{aligned} \overline{\chi}_{\mathrm{L}}(v) &= \frac{(\Delta v_{\mathrm{L}})^2}{(v_0 - v)^2 + (\Delta v_{\mathrm{L}})^2} \\ &= \frac{(\Delta v_{\mathrm{L}})^2}{[v_0 - (v_c + mt) \cdot \mathrm{sqr}(ft)]^2 + (\Delta v_{\mathrm{L}})^2}, \end{aligned} \tag{5}$$

where v_0 is the center frequency of optical transition, and Δv_{L} is the half-width at half-maximum of the Lorentzian lineshape function.

The light signal in Eq. (4) can be expressed by the Fourier series, and the even component[13] of the nth Fourier coefficient of the detector signal can be acquired, and is given by

$$\begin{aligned} A_{L,n}^{\mathrm{even}}(v_c, m) &= I_0 \sigma_0 NLf(2 - \delta_{n0}) \\ &\quad \int_{-1/(2f)}^{1/(2f)} \overline{\chi}_L(v) \cos(2\pi nft) \mathrm{d}t \\ &= I_0 \sigma_0 NLf(2 - \delta_{n0}) \int_{-1/(2f)}^{1/(2f)} \\ &\quad \frac{(\Delta v_L)^2}{[v_0 - (v_c + mt) \cdot \mathrm{sqr}(ft)]^2 + (\Delta v_L)^2} \\ &\quad \cos(2\pi nft) \mathrm{d}t, \end{aligned} \tag{6}$$

where δ_{n0} is the Kronecker delta.

The nth harmonic of absorption signal expressed in Eq. (6) can be obtained experimentally by a lock-in amplifier with the reference frequency selected on the n times of the square wave. Detailed theoretical description of the method used here is discussed in subsequent articles.

The schematic diagram of the QCW WMS system is shown in Fig. 2. The QCW modulation signal was generated using a function generator (Fluke284) and a homemade interface circuit, which was then converted to current signal through a LD controller (ILX LDC3908) to drive the diode laser. A NIR DFB diode laser (Furukawa: FRL15DDR0-A31), which worked under the QCW mode, was used to detect the absorption line of CO_2 at 1 579.574 nm in the experiment. The emitting light of the diode laser was first coupled into a multi-pass gas cell and absorbed by the analyte. Afterwards, it was transmitted to a photodiode and converted to an electrical signal. After conducting amplification using a pre-amplifier, the detector signal was fed to a lock-in amplifier, the output of which was the nth harmonic of the detector signal when selecting the reference frequency on the n times of the square wave. A data-acquisition-card (NI PCI-4474) equipped in a personal computer was employed to collect and convert the lock-in output to digital signals for further processing and analysis.

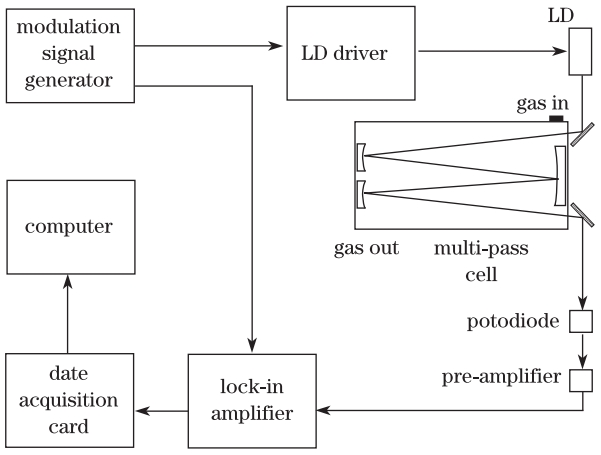

Fig. 2. Schematic diagram of the QCW tunable diode laser absorption spectroscopy system.

In the application of WMS, various harmonics with very narrow bandwidth are achieved at high frequency, using phase sensitive detections (usually a lock-in amplifier). In addition, the second harmonic (2f) is widely used in the measurement of gas concentration and temperature, because of its relatively higher SNR compared with direct absorption signal or other orders of harmonics and its peak value at the absorption line center. In the experiment, the 2nd multi-frequency of the QCW signal was selected as the reference frequency of the lock-in amplifier, and the 2f spectrum of CO_2 with 99.9% concentration was obtained under an 18-m optical length (Fig. 3). Figure 3 shows the subplot as the computed 2f spectrogram from Eq. (6) with line-width-normalized modulation amplitude of 2. As can be seen, the right sideband of the measured QCW 2f signal has a relatively flat feature and small amplitude compared with the computed 2f signal. This is because different frequency shift ranges during the square current are produced by varied square intensities along with the current ramp—a process which is known as parasitic amplitude modulation. Typically, a larger frequency shift range occurs at the higher square wave current intensity.

The 2f spectra of 5% CO_2 measured by the CW WMS and QCW WMS under an 18-m optical length are respectively shown in Fig. 4. Comparing the two spectra, the 2f of the CW WMS has a narrower line width and distinct side bands, whereas that of QCW WMS has a wider line width and an indistinct right side band due to parasitic amplitude modulation at higher pulsed current. Fringes are observed in the measurement (Fig. 4(a)), which are caused by the interference between partially reflected beams of the laser light[14].

The 2f spectra of several lower concentrations of CO_2 were also investigated using QCW WMS in our experiment. The results are shown in Fig. 5. The concentrations between 5% and 1% of CO_2 were acquired from a standard gas using a homemade gas divider. As shown in Fig. 5, the right side bands of the 2f are becoming indistinct along with the decreasing concentrations due to parasitic amplitude modulation; moreover, large absorption signals at high concentrations are relatively less affected by parasitic amplitude modulation (Fig. 3).

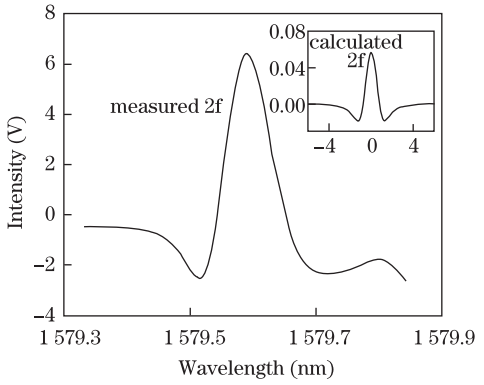

Fig. 3. Measured the 2f spectrum of 99.9% CO_2; inset shows the theoretical the 2f spectrum.

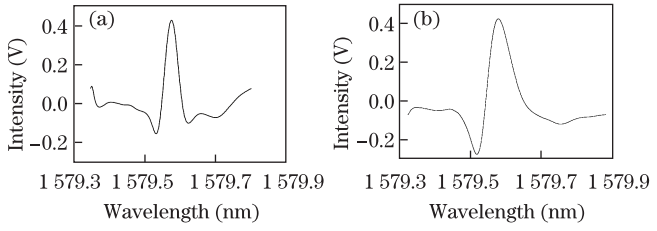

Fig. 4. 2f spectra of CO_2 with 5% concentration detected with (a) CW WMS and (b) QCW WMS.

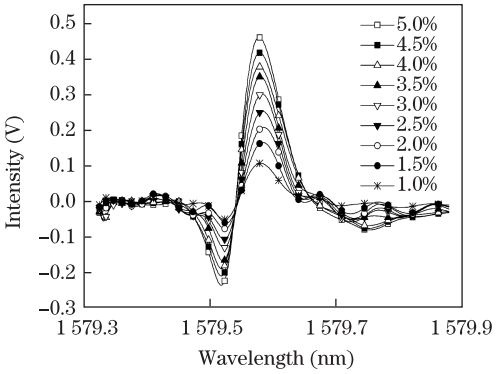

Fig. 5. 2f spectra of CO_2 with different concentrations.

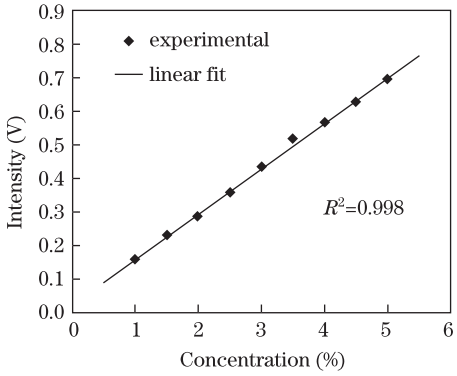

Fig. 6. Linear relationship between the 2f signal intensities and gas concentrations.

The linear relationships between the 2f signal intensities shown in Fig. 5 and the concentrations are depicted in Fig. 6, wherein a linear fitting result of $R^2 = 0.998$ is shown. A noise-equivalent absorbance of 3.2×10^{-5} $Hz^{-1/2}$ has been achieved for CO_2 with an 18-m path length and 5-s integration time. Certainly, by increasing the path length to several hundred meters or by employing an open optical path, our system can detect the CO_2 in ambient air or with even lower concentrations. Thus, the QCW WMS system investigated in this letter shows a similar performance to continuous WMS, making it suitable for use in trace gas detections.

In conclusion, a new modified WMS based on the QCW mode operation is investigated. The wavelength modulation is performed by the intra-pulse wavelength tuning due to the temperature effect in the laser chip when driven by a pulsed or QCW current. The wavelength scanning is implemented by a current ramp. A noise-equivalent absorbance of 3.2×10^{-5} $Hz^{-1/2}$ is achieved for CO_2, which corresponds with the result of continuous WMS with the same system. We demonstrate the feasibility and performance of gas-sensing applications using

the modified QCW WMS. In short, QCW WMS lowers the dependence on lasers and expanded the selectable laser sources, thus extending the detectable trace gas species and reducing system cost. Further study on the theoretical analysis and stabilization and SNR improvement of the system shall be performed in the subsequent work.

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