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DETECTION OF GAS CONCENTRATION BY CORRELATION SPECTROSCOPY USING A MULTI-WAVELENGTH FIBER LASER

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Abstract—A correlation spectroscopy (COSPEC) based on a multi-wavelength fiber laser is first proposed for the detection of gas concentration. The lasing wavelengths are selected to match several characteristic absorption peaks of the gas under test, and the gas concentration is easily measured by correlating it with the reference gas. The present method is immune from the instability of the light source and the influence of other gases. The concentration measurement of C_2H_2 is demonstrated in the experiment in its near-infrared dominant absorption region. The technique has prospects for simultaneous detection of multiple gases, and the measurement of mixed gases of C_2H_2 and CO_2 is also analyzed.

1. INTRODUCTION

Gas concentration measurements have gained more and more attention in the field of industrial process control and environmental pollution monitoring, etc.. Many methods for gas detection have been developed. Laser technologies [1–3], which have significant applications in all kinds of lines, are also used for the detection of gases. Using the Beer-Lambert law, gas concentration can be easily measured with a single-mode tunable laser which scans in a continuous wavelength range including some absorption peaks of the target gas. When using

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a tunable diode laser, it has a common gas measurement technique called TDLAS (tunable diode laser absorption spectroscopy) [4–7]. Its measurement precision is limited by the unstableness of a laser mode and the presence of interfering gases. Although some more stable tunable laser can be developed, the cost will also increase.

Correlation spectroscopy (COSPEC) is a particularly simple and powerful technique for fast and selective measurements yielding moderate sensitivity. COSPEC is immune to fluctuations of the light source and the disturbance of other gases. Two typical COSPEC methods, namely, gas filter correlation (GFC) [7, 8] and interferometric correlation (INC) [7], have been proposed. Both methods used differential measurements to reject various kinds of interference. However, according to their correlation principles, broadband light source must be used, and the wavelength modulation spectroscopy (WMS) [9] technique can not be employed. Consequently the sensitivity is limited to enhance further.

Multimode diode laser (MDL)-based COSPEC [10–13] is a correlation method based on temporal correlation between absorption signals from an external gas of unknown concentration and a reference gas of known concentration. The technique combines the sensitivity of WMS technique and the robustness of the correlation method. Furthermore, the spectrum of MDL can cover the absorption lines of two gases [13] with overlapping absorption lines, and thus MDL-based COSPEC can measure their concentrations simultaneously. However, concerning the general performance of MDL (the lasing spectrum is much broader than gas absorption lines), the sensitivity in [13] is only 200 ppm. Both the problem of the mode-hopping of the MDL and the low-efficiency problem of the data processing (by the operator's experience) lead to a low efficiency of MDL-based COSPEC.

To solve these problems, multi-wavelength fiber laser-based COSPEC is first proposed in the present paper. A multi-wavelength fiber laser in the near-infrared with narrow spectral lines, which can be designed to match absorption lines of target gases well, is used to enhance the sensitivity and the selectivity, and the Unscrambler software [14–16] is applied to improve the efficiency of the signal processing. Furthermore, because of the wide wavelength tunable range of multi-wavelength fiber laser, the present technique can measure multi-gas simultaneously even if the absorption peaks of the gases are relatively far away from each other. For a multi-gas measurement, the wavelengths of the fiber laser can be easily tuned to the absorption peaks of the target gases by employing the fiber Bragg gratings (FBGs) [17, 18]. Gas simultaneous detection is required on diverse occasions, e.g., in steel industry, the concentrations of CO and

CO_2 in exhaust are directly related to the quality of products. A simultaneous measurement could establish a calibrated standard more precisely.

2. MEASUREMENT PRINCIPLE

The principle of multi-wavelength fiber laser based COSPEC is basically the same as the one with broadband light source [19, 20] or MDL [10–13]. The main differences are their optical configurations and signal processing methods. The light beams are split into two parts and coupled into a reference cell and a sample cell (or open field). When the wavelengths of the fiber laser are tuned over absorption peaks of a gas, the absorbance of each wavelength follows the Beer-Lambert law. Owning to the time correlation between the signals of the reference and sample cells, the interfering gas signals and other background noises can be easily eliminated. When the gas sample is optically thin (i.e., the absorbance is far less than 1), the absorption signal is linearly proportional to the gas concentration. Consequently, the target gas concentration can be deduced from the magnitude ratio between the absorption signals of the two optical paths.

As for multi-wavelength fiber laser-based COSPEC, neither the precise knowledge about the gas absorption peaks nor the stability of the light source is necessary. Since the wavelengths of the fiber laser are selectable in a large range and tunable, they can measure many kinds of gases and have a promising prospect in simultaneous multi-gas detection.

3. EXPERIMENT AND RESULTS

The schematic configuration of for our multi-wavelength fiber laser-based COSPEC experiment is shown in Fig. 1. The upper part shows the schematic diagram of our multi-wavelength fiber laser. A semiconductor optical amplifier (SOA; the gain medium for amplification is semiconductor; OptoSci Ltd, LDR250CKE), which is the initial light source of the fiber laser, together with an erbium-doped fiber amplifier (EDFA; the output power range is from -5 dBm to 24 dBm), provides a hybrid gain for the fiber laser. Because of the limited gain spectrum of SOA and EDFA, the four lasing wavelengths of the fiber laser selected by four FBGs [21] are limited within C band from 1520 nm to 1570 nm. As the reflection peaks of FBGs can be adjusted by stress, they are tautened manually in the experiment in order to scan the wavelengths of the fiber laser. In order to obtain four lasing wavelengths with approximately equal intensity, three variable

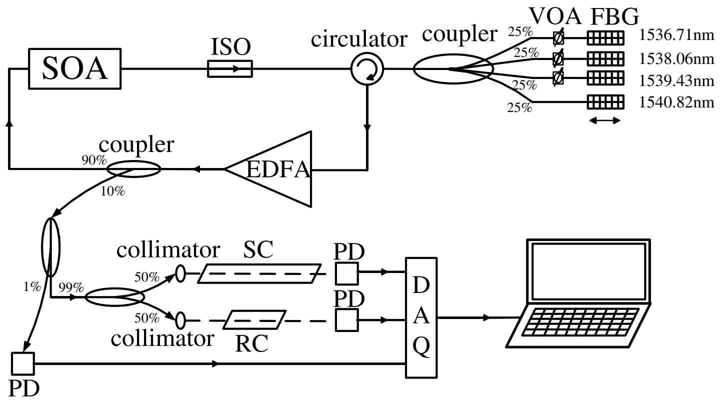

Figure 1. The schematic diagram of our multi-wavelength fiber laser-based COSPEC. SOA: semiconductor optical amplifier; ISO: optical isolator; VOA: variable optical attenuator; FBG: fiber Bragg grating; EDFA: erbium-doped fiber amplifier; SC: sample cell; RC: reference cell; PD: photodetector; DAQ: data acquisition card.

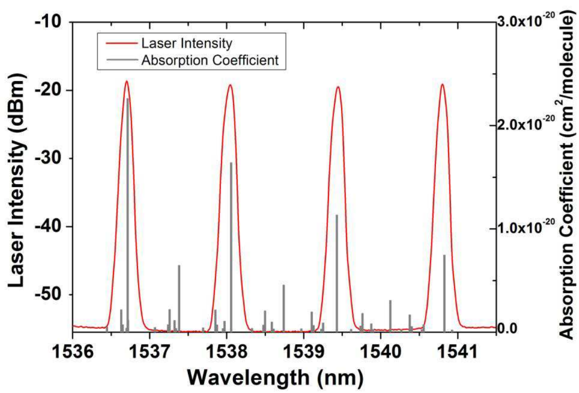

Figure 2. The spectrum of 10% power multi-wavelength fiber laser (red line) detected by an optical spectrum analyzer (OSA) before the experiment. Absorption coefficient of C_2H_2 (gray line, calculated with HITRAN 2004) is also shown from 1536 nm to 1541 nm.

optical attenuators (VOAs) are used before the FBGs. The power from the 10% arm of the coupler is about 1 mW.

The spectrum of the multi-wavelength laser measured by an optical spectrum analyzer (OSA; Ando, AQ6317) with a resolution of 0.01 nm is shown in Fig. 2 together with the absorption lines of

C_2H_2. The wavelengths of the laser in accordance with some dominant absorption peaks of C_2H_2 are 1536.71 nm, 1538.06 nm, 1539.43 nm and 1540.82 nm. The extinction ratios of four wavelengths are over 35 dB and the full width at half-maximum (FWHM) of each wavelength is about 0.1 nm. 1% power of the laser output is received by a photodetector (PD) (Thorlabs PDA400) to monitor the fluctuation of the laser intensity. The residual power is split into two equal parts by a 3-dB coupler. One part passes through a 37-cm-long sample cell containing C_2H_2 under test, and the other passes through a 23-cm-long reference cell filled with well-calibrated 2% ($\pm 0.1\%$) C_2H_2. The transmitted laser beams are simultaneously detected by two photodetectors (Thorlabs PDA50B). Three channels of electronic signals are collected by a 16-bit data acquisition card (DAQ) (National Instrument NI cRIO-9215) and transmitted to a personal computer for data recording and processing.

According to the Beer-Lambert law, when the wavelengths were tuned by tautening FBGs manually and randomly, the absorption signals from two optical paths varied simultaneously. The signals from the reference cell with C_2H_2 of 2% concentration and the sample cell with C_2H_2 of about 1% concentration normalized with the signal of the PD (Thorlabs PDA400) and their cell lengths were shown in Fig. 3. When the wavelengths are tuned, the sample-to-reference magnitude ratio related to the concentration ratio of C_2H_2 will not change. As various disturbances (from interfering gases, abnormal fluctuation of laser intensity) to the magnitude ratios are inevitable, we selected only correlated pairs to calculate the concentration of C_2H_2. Since the peak-

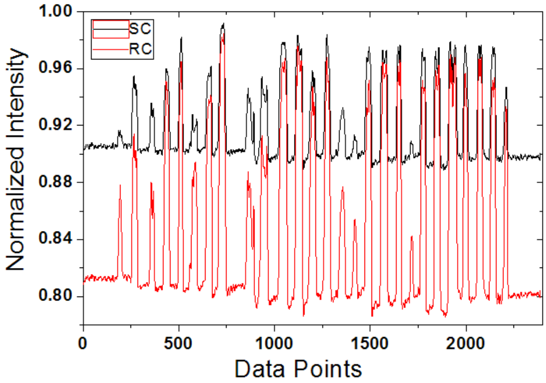

Figure 3. Normalized signals of the two optical paths [reference cell (RC) and sample cell (SC)] are recorded simultaneously when the wavelengths of the fiber laser were tuned.

points are dominant in the acquired data, the measuring process will not take long even the sampling rate of DAQ was set at a low level. In our experiment, we set the sampling rate of DAQ at 100 Hz, and acquired 500 points for the measurement.

Instead of using some Priori method [10–13], we used a convenient and reliable signal processing method with the Unscrambler software [14–16] to exclude the disturbances and select the correlated pairs. Initially 1–3 correlated pairs were selected manually from the data in Fig. 3 to set up a model through Principal Component Analysis (PCA) [22] processing. According to the model, the rest of the data was analyzed with Soft Independent Modeling of Class Analogy (SIMCA) [23] processing to exclude wrong data when sifting correlated pairs. After the processing of the Unscrambler, linear fitting [$R^2 = 0.993$ (regression coefficient; the value is closer to 1 if the linearity is better)] for the two channel signals (normalized) was obtained and shown in Fig. 4. The slope of the fitting line is the concentration ratio of the gases in the sample and reference cells after normalizing their cell lengths. As the slope of the fitting line is 0.49327, the concentration of C_2H_2 in the sample cell is thus determined to be 0.987%.

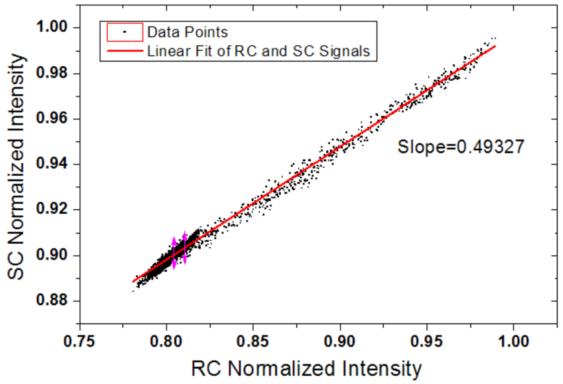

Figure 4. Correlated data points (normalized intensities) of the two optical paths [reference cell (RC) and sample cell (SC)] and a linear fitting of the data points.

In order to evaluate the accuracy and linearity of the measurement, various concentrations of C_2H_2 were measured and compared with the calculated ones. Fig. 5 shows scatter plot of the measured concentrations of C_2H_2 versus the calculated values in the range of 1%–2%. The linearity ($R^2 = 0.986$) between the measured and calculated concentrations was obtained.

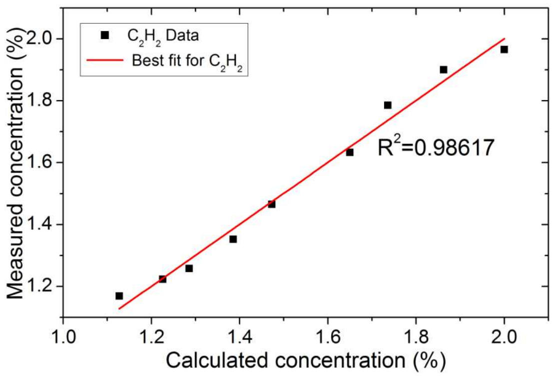

Figure 5. Scatter plot of the measured concentrations of C_2H_2 versus the calculated values. Best linear fitting for these data is also shown.

We also simulate a case of two mixed gases (C_2H_2 and CO_2) with a MATLAB software. The data of the laser spectrum (used in our experiment) and the absorption coefficients of C_2H_2 and CO_2 (HITRAN 2004 [24]) from 1536 nm to 1541 nm were used in our simulation. Under the same temperature and atmosphere pressure conditions, the sample cell is filled with 0.01% C_2H_2 and 20% CO_2, (the rest is air) one reference cell is filled with 0.1% C_2H_2 (the rest is air), and the other reference one is filled with 100% CO_2. The two gases have quite different absorption coefficients and consequently their reference concentration differ greatly. The three gas cells are all 3-m long (could be achieved through multi-pass cell; such a long cell is required for measuring C_2H_2 of small concentration; unfortunately we only have gas cells of short lengths in our lab and thus only simulation results are given in this paragraph). With a regular swing of ± 0.3 nm for each wavelength, the three absorption signals were shown in Fig. 6(a). The concentrations of C_2H_2 and CO_2 in the sample cell versus the values in the reference cells after the correlation processing were shown in Figs. 6(b) and (c), respectively. The C_2H_2 fitting curve is linear ($y = 0.10814x + 0.89186$; y is SC normalized intensity and x is RC normalized intensity), and its slope is 0.10814. Thus, the deduced concentration of C_2H_2 in the sample cell is 0.0108%. Likewise, as the slope of CO_2 fitting line is 0.21689, the deduced concentration of CO_2 in the sample cell is 21.689%. The excellent linear fitting and the simulation results showed that simultaneous measurement of multiple gases is possible.

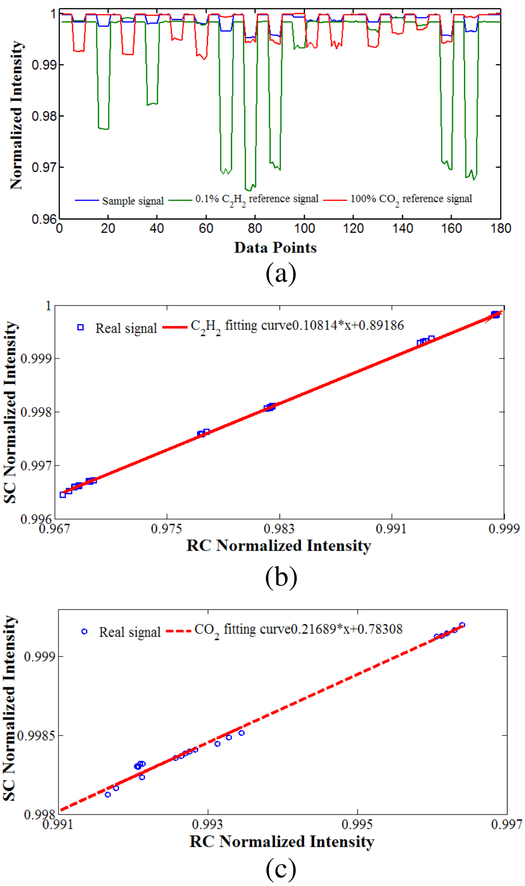

Figure 6. (a) Absorption signals in three gas cells; (b) C_2H_2 concentration in sample versus reference cells and the best linear fit of them; (c) CO_2 concentration in sample versus reference cells and the best linear fit of them.

4. DISCUSSION AND CONCLUSION

In summary, the capability of multi-wavelength fiber laser-based COSPEC has been clearly demonstrated in the present paper. It is very convenient to change FBGs of the fiber laser for different gas measurements. Multi-wavelength fiber laser-based COSPEC has a good sensitivity (absorptions 10^{-3}) for the concentration measurement without any SNR enhancement techniques. In the future the manual adjustment of FBGs can be replaced with automatic adjustment with piezoelectric ceramics (PZT), and the results can be improved further if wavelength modulation spectroscopy (WMS) [25] or cavity enhancement technique (like a multi-pass cell) is utilized.

We have also shown the potential advantages of the multi-wavelength fiber laser-based COSPEC for simultaneous multi-gas measurement. With frequency modulating-demodulating, only one detector suffices. Since the wavelengths of the fiber laser can be selected in the range from 1520 nm to 1570 nm, this method overcomes the limited wavelength range of the MDL and can even measure a mixture of multiple gases whose absorption lines are relatively far away from each other. It can also be used to measure one gas concentration with the over-determined four-wavelength absorbance signals, which is more stable and accurate than a conventional method using only one signal. Therefore, the present method is quite useful for measuring one target gas with interfering gases or simultaneously detecting multiple gases.

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REFERENCES

1. Liu, S. C., Z. W. Yin, L. Zhang, X. F. Chen, L. Gao, and J. C. Cheng, "Dual-wavelength fbg laser sensor based on photonic generation of radio frequency demodulation technique," *Journal of Electromagnetic Waves and Applications*, Vol. 23, No. 16, 2177–2185, 2009.
2. Fu, X., C. Cui, and S. C. Chan, "Optically injected semiconductor laser for photonic microwave frequency mixing in radio-over-fiber," *Journal of Electromagnetic Waves and Applications*, Vol. 24, No. 7, 849–860, 2010.
3. Yang, B., X. F. Jin, X. M. Zhang, H. Chi, and S. L. Zheng, "Photonic generation of 60 GHz millimeter-wave by frequency quadrupling based on a mode-locking soa fiber ring laser with a low modulation depth MZM," *Journal of Electromagnetic Waves and Applications*, Vol. 24, No. 13, 1773–1782, 2010.
4. Bergamaschi, P., M. Schupp, and G. W. Harris, "High-precision direct measurements of 13CH4/12CH4 and 12CH3D/12CH4 ratios in atmospheric methane sources by means of a long-path tunable

diode laser absorption spectrometer," *Appl. Optics*, Vol. 33, 7704–7716, 1994.

5. Roller, C., K. Namjou, J. D. Jeffers, M. Camp, A. Mock, P. J. McCann, and J. Grego, "Nitric oxide breath testing by tunable-diode laser absorption spectroscopy: Application in monitoring respiratory inflammation," *Appl. Optics*, Vol. 41, 6018–6029, 2002.
6. Guan, Z., M. Lewander, and S. Svanberg, "Quasi zero-background tunable diode laser absorption spectroscopy employing a balanced Michelson interferometer," *Opt. Express*, Vol. 16, 21714–21720, 2008.
7. Galais, A., G. Fortunato, and P. Chavel, "Gas concentration measurement by spectral correlation: Rejection of interferent species," *Appl. Optics*, Vol. 24, 2127–2134, 1985.
8. Sandsten, J., H. Edner, and S. Svanberg, "Gas imaging by infrared gas-correlation spectrometry," *Opt. Lett.*, Vol. 21, 1945–1947, 1996.
9. Reid, J., J. Shewchun, B. K. Garside, and E. A. Ballik, "High sensitivity pollution detection employing tunable diode lasers," *Appl. Optics*, Vol. 17, 300–307, 1978.
10. Somesfalean, G., M. Sjöholm, L. Persson, H. Gao, T. Svensson, and S. Svanberg, "Temporal correlation scheme for spectroscopic gas analysis using multimode diode lasers," *Appl. Phys. Lett.*, Vol. 86, 184102, 2005.
11. Lou, X. T., G. Somesfalean, F. Xu, Y. G. Zhang, and Z. G. Zhang, "Gas sensing by tunable multimode diode laser using correlation spectroscopy," *Appl. Phys. B*, Vol. 93, 671–676, 2008.
12. Lou, X. T., G. Somesfalean, B. Chen, and Z. G. Zhang, "Oxygen measurement by multimode diode lasers employing gas correlation spectroscopy," *Appl. Optics*, Vol. 48, 990–997, 2009.
13. Zhang, Z. G., X. T. Lou, G. Somesfalean, B. Chen, Y. G. Zhang, H. Wang, S. Wu, and Y. Qin, "Simultaneous detection of multiple gas species by correlation spectroscopy using a multi-mode diode laser," *Opt. Lett.*, Vol. 35, 1749–1751, 2010.
14. Macho, S., R. Boqué, M. S. Larrechi, and F. X. Rius, "Multivariate determination of several compositional parameters related to the content of hydrocarbon in naphtha by MIR spectroscopy," *Analyst*, Vol. 124, 1827–1831, 1999.
15. Forina, M., S. Lanteri, M. C. Cerrato Oliveros, and C. Pizarro Millan, "Selection of useful predictors in multivariate calibration," *Anal. Bioanal. Chem.*, Vol. 380, 397–418, 2004.

16. Barton, F. E., J. D. Bargeron, G. R. Gamble, D. L. Mcalister, and E. Hequet, "Analysis of Sticky Cotton by Near-Infrared Spectroscopy," *Appl. Spectrosc.*, Vol. 59, 1388–1392, 2005.
17. Liau, J.-J., N.-H. Sun, S.-C. Lin, R.-Y. Ro, J.-S. Chiang, C.-L. Pan, and H.-W. Chang, "A new look at numerical analysis of uniform fiber bragg gratings using coupled mode theory," *Progress In Electromagnetics Research*, Vol. 93, 385–401, 2009.
18. Chen, B., S. L. Zheng, X. M. Zhang, X. F. Jin, and H. Chi, "Simultaneously realizing PM-IM conversion and efficiency improvement of fiber-optic links using FBG," *Journal of Electromagnetic Waves and Applications*, Vol. 23, No. 2–3, 161–170, 2009.
19. Sandsten, J., P. Wiebring, H. Edner, and S. Svanberg, "Realtime radiation gas-correlation imaging employing thermal background," *Opt. Express*, Vol. 6, 92–103, 2000.
20. Dakin, J. P., M. J. Gunning, P. Chambers, and Z. J. Xin, "Detection of gases by correlation spectroscopy," *Sens. Actuators. B*, Vol. 90, 124–131, 2003.
21. Ahmad, H., A. H. Sulaiman, S. Shahi, and S. W. Harun, "SOA-based multi-wavelength laser using fiber Bragg gratings," *Laser Phys.*, Vol. 19, 1002–1005, 2009.
22. http://www.camo.com/resources/principal-component-analysis.html.
23. http://www.camo.com/resources/simca.html.
24. http://www.cfa.harvard.edu/HITRAN/.
25. Chan, Y. K., M. Y. Chua, and V. C. Koo, "Sidelobes reduction using simple two and tri-stages non linear frequency modulation (NLFM)," *Progress In Electromagnetics Research*, Vol. 98, 33–52, 2009.