FTS Based on MIR Supercontinuum Sources for Trace Gas Detection

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Abstract: Mid-infrared supercontinuum sources can provide a broad spectral bandwidth in the molecular fingerprint region, making them very attractive for multispecies trace gas detection. We demonstrate our recent developments in combining these sources with FTS.

1. Introduction

The mid-infrared (MIR) wavelength range contains the strongest molecular absorption features of many interesting gas species, each demonstrating a unique absorption pattern. Therefore, broadband molecular spectroscopy in the MIR range allows for sensitive multispecies detection. Traditionally, thermal sources were used to provide a broadband coverage in the MIR for molecular spectroscopy. Despite their broadband coverage and smooth power spectral density (PSD), these sources suffer from low brightness and lack of directionality, which makes it harder to provide a high interaction length between the light and the gas sample. A viable alternative can be supercontinuum sources (SCs), which are extending their reach from the near-infrared (NIR) range further towards MIR, demonstrating high brightness and broad spectral coverage within a spatially coherent (directional) beam [1]. However, SC generation is usually associated with high relative intensity noise (RIN), due to noise amplification in the nonlinear broadening process [2,3]. Although recent advancement in NIR SC generation using all-normal dispersion photonic crystal fibers [4] and cascading-based MIR SC sources [5] show reduced levels of RIN, noise reduction/cancelation techniques are still needed to achieve high sensitivity and low detection limits in SC-based spectroscopy. We have recently demonstrated SC-based absorption spectroscopy for multispecies (trace) gas detection in combination with a multipass cell (MPC), using a direct dispersive MIR spectrometer [6] and synchronous detection [7] as well as by up-converting the MIR beam into NIR and using a dispersive NIR spectrometer [8]. Both of these techniques demonstrate fast (tens of ms measurement time) and sensitive (sub-ppm sensitivity for several gas species in 1 s averaging time) detection, however, they have limited spectral resolution and spectral coverage. Therefore, they cannot benefit from all of the advantages of the SC source. On the other hand, Fourier transform spectroscopy (FTS) can provide both of these benefits while combined with SC sources. Here, we explore the combination of an FTS, a multipass cell, and an MIR SC for trace gas detection in different applications.

2. Combining FTS with high repetition rate SC sources

The spatial coherence of the SC enables us to construct a more compact FTS compared to regular FTIRs. The size of the FTS can be effectively reduced to a shoe-box scale, while still providing a high spectral resolution (e.g. sub 1 GHz/0.033 cm^{-1}) [9]. The high repetition rate of the novel SC sources [10] enables fast scanning of the FTS with measurement times of ~1 s for a spectral resolution of 3 GHz which is usually sufficient for multispecies gas detection at atmospheric pressure with a negligible instrument line-shape function. In addition, one can use synchronous detection in the FTS by lock-in amplification to the repetition rate of the SC source to reduce 1/f noise. However, in practice, this method of noise reduction is not very effective and is limited to the pulse-to-pulse instability of the SC source [9]. Balanced-detection in the FTS is a better choice which can be implemented by using two detectors in the Michelson interferometer to detect the two out-of-phase interferograms, providing much more efficient noise reduction [9]. This method is potentially capable of canceling the entire RIN noise of the SC and providing a detector noise limited sensitivity. Utilizing a 31.2 m MPC, we can obtain sub-ppm detection limits for a long list of species after one minute of averaging [9]. The special coherence of the beam makes it possible to open up new applications for in-situ monitoring of gas concentrations. We have used this system to monitor plasma-based gas conversion directly inside a discharge tube or via an MPC [11]. The SC beam can travel through the long discharge tube (length 1.80 m, internal diameter of 1.5 cm) without any interaction with the tube walls. Figure 1 demonstrates the measured spectrum of reaction products of (a) a 50/50% CO_2/N_2 discharge and (b) a 70/30% CH_4/CO_2 discharge. The retrieved concentrations of the detected species are also noted in the figure.

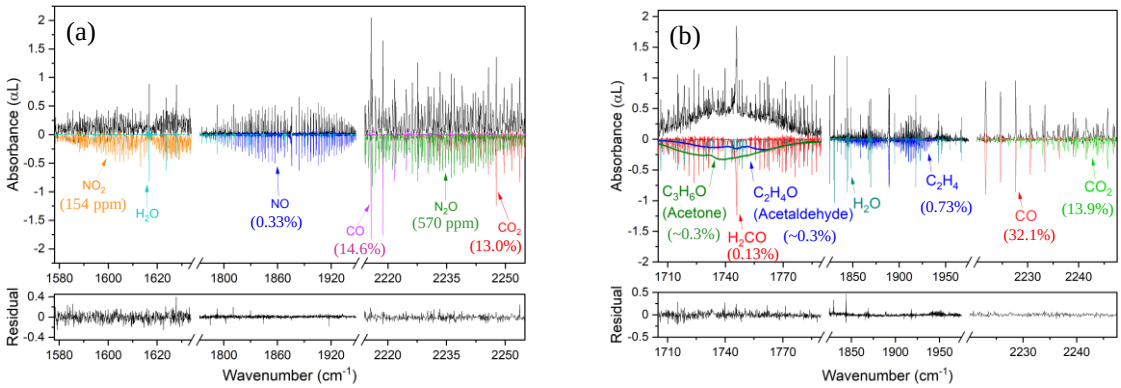

Fig. 1. Measured spectrum (in black) of reaction products of (a) a 50/50% CO_2/N_2 discharge and (b) a 70/30% CH_4/CO_2 discharge and the fitted model spectra (in colors, inverted) based on the HITRAN/PNNL/GEISA databases. The residuals of the fits are shown in the lower panel.

The special coherence of the beam also facilitates free-space spectroscopy. Figure 2 demonstrates the measured spectrum (measurement time 0.4 s, resolution 10 GHz) over the head-space of an ethanol container outside of the lab in (a) and the retrieved ethanol and water concentrations over time in (b) with a sampling rate of 2.5 Hz. The SC beam is transmitted above the container towards a retroreflector and is aligned back to the FTS, which is positioned in a transportable unit, next to the SC source. The influence of the wind turbulence can be clearly seen in the retrieved ethanol concentration, while the water concentration is stable in the measurement path.

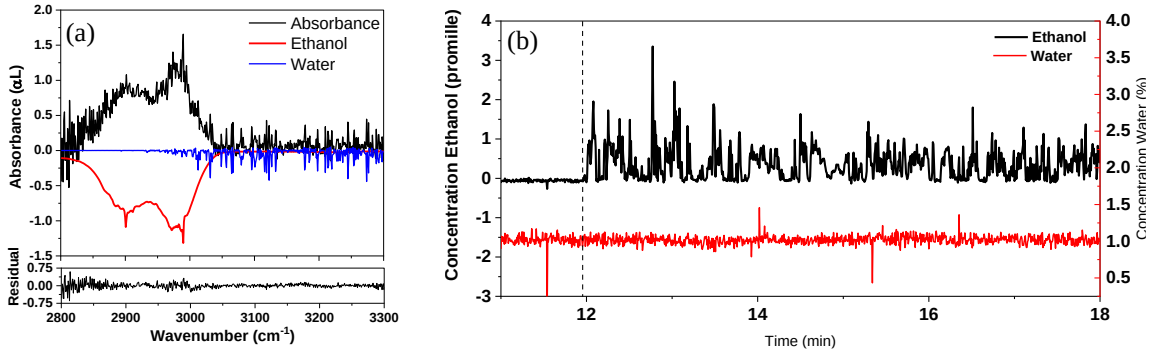

Fig. 2. (a) Measured spectrum (in black) over the head-space of an ethanol container and the fitted model spectra (in colors, inverted) based on the HITRAN/PNNL databases. (b) The retrieved ethanol and water concentrations over time.

3. References

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