Atmospheric observations of multiple molecular species using ultra-high-resolution external cavity quantum cascade laser heterodyne radiometry

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Received March 1, 2011; revised April 22, 2011; accepted April 24, 2011;
posted April 26, 2011 (Doc. ID 143411); published May 20, 2011

We demonstrate a widely tunable laser heterodyne radiometer operating in the thermal IR during an atmospheric observation campaign in the solar occultation viewing mode. An external cavity quantum cascade laser tunable within a range of 1120 to 1238 cm^{-1} is used as the local oscillator (LO) of the instrument. Ultra-high-resolution (60 MHz or 0.002 cm^{-1}) transmission spectroscopy of several atmospheric species (water vapor, ozone, nitrous oxide, methane, and dichlorodifluoromethane) has been demonstrated within four precisely selected molecule-specific narrow spectral windows (~1 cm^{-1}). Atmospheric transmission lines within each selected window were fully resolved through mode-hop-free continuous tuning of the LO frequency. Comparison measurements were made simultaneously with a high-resolution Fourier transform spectrometer to demonstrate the advantages of the laser heterodyne system for atmospheric sounding at high spectral and spatial resolutions.

OCIS codes: 300.6310, 140.5965, 280.4991.

Since the pioneering work by Menzies on thermal IR laser heterodyne radiometers (LHRs) for atmospheric sounding, LHR capabilities have been partly determined by the performance of the laser source providing the local oscillator (LO) [1]. Initially, carbon dioxide gas lasers were used despite the lack of frequency tuning flexibility. Lead salt diode lasers offer greater mid-IR frequency choice and continuous tuning, but they lack reliability and optical power [2]. They also require cryogenic cooling. The advent of room-temperature distributed feedback quantum cascade lasers (DFB-QCLs) has made available high-quality mid-IR sources well suited for the role of a frequency tunable optical LO. With the DFB-QCL, continuous tuning (~10 cm^{-1}) within the 8–12 μm atmospheric window, allowed the development of LHR with a spectral coverage suitable for optimal probing of one or two atmospheric molecules [3].

Recent advances on external-cavity (EC) QCL systems have dramatically increased the frequency range accessible with cw laser sources. Tuning ranges of ~200 cm^{-1} have been demonstrated [4], and coverage of ~400 cm^{-1} is within reach via the development of broad-gain QCL structures [5]. An LHR with an EC-QCL LO has been demonstrated in the laboratory to operate over a range greater than 100 cm^{-1} [6].

This Letter reports the demonstration of multispecies atmospheric sounding performed with a widely tunable (1120–1238 cm^{-1}) EC-QC-LHR. Atmospheric transmission spectra at ultrahigh spectral resolution (60 MHz) have been recorded in the solar occultation viewing mode. Five atmospheric molecular constituents have been investigated: ozone (O_3), nitrous oxide (N_2O), methane (CH_4), water vapor (H_2O), and dichlorodifluoromethane (CCl_2F_2). Experimental spectra show that line shape information without contribution from the instrument line shape function is obtained for atmospheric molecules with vertical mixing ratio profiles that are uniformly (CH_4, N_2O) and nonuniformly (O_3, CCl_2F_2, H_2O) distributed. Simultaneous measurements with a high-resolution Fourier transform spectrometer (FTS) have been conducted for comparison, which demonstrates the unique capabilities offered by EC-QC-LHR where sensitivity, ultranarrow field of view (FoV), and ultrahigh spectral resolution are simultaneously desirable.

The optical layout of the experimental arrangement is shown in Fig. 1. Solar radiation is captured by an external solar tracker and directed into the laboratory. The FoV is narrow (~5.2 arcmin) and corresponds to observation of an area of one thirty-eighth of the solar disk surface. At the entrance of the EC-QC-LHR, an optical filter transmits wavelengths longer than 7 μm while the reflected part is directed to a broadband (800–1750 nm), large-area, germanium detector (D1), which monitors the solar intensity. The >7 μm solar radiation is amplitude modulated by a chopper at ~1.8 kHz, directed to the 25R/75T beam splitter and superimposed with the LO beam from the EC-QCL (see [4]). The combined beams are imaged

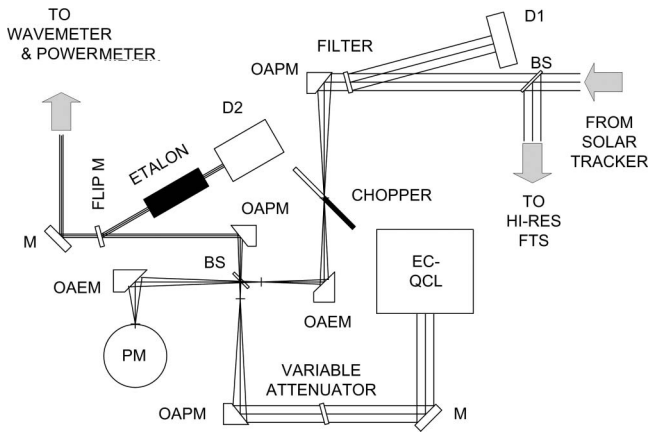

Fig. 1. Optical layout of the EC-QC-LHR: M, mirror; BS, beam splitter; D1, solar intensity detector; D2, mid-IR detector; OAEM, off-axis elliptical mirror; OAPM, off-axis parabolic mirror; PM, photomixer.

onto a high-speed (>1 GHz) mercury cadmium telluride photomixer (PM). The remaining 75% of the LO power is used for frequency calibration. A flip mirror directs this part of the LO radiation to either a wavemeter for absolute frequency determination, or to a 1 in. long germanium etalon and a mid-IR detector (D2) for relative frequency calibration while the LO frequency is tuned. The DC output signal from the PM provides LO power monitoring, while the AC signal contains the spectral information carried by the heterodyne signal at radio frequencies. This signal is amplified and filtered by a 10–40 MHz bandpass filter before being detected by a Schottky diode. The diode output voltage is fed into a DSP lock-in amplifier to be demodulated at the chopping frequency.

The EC-QC-LHR operating range is determined by the tuning range of the EC-QCL (1120–1238 cm^{-1}). The corresponding synthetic atmospheric transmission spectra at ground level and zenithal elevation are shown in Fig. 2. Calculations were made with the Reference Forward Model using standard midlatitude atmospheric profiles of temperature, pressure, and molecular volume mixing ratios as geophysical inputs [7]. The HITRAN database provided the spectral line information. Within the available spectral range, five molecules, H_2O, O_3, N_2O, CH_4, and CCl_2F_2, exhibit significant optical depths.

Four narrow (~1 cm^{-1}) spectral windows have been identified, in which one or more absorbers can be investigated at close to Doppler resolution. Window 1 (1129.8–1130.4 cm^{-1}) contains intense O_3 absorption lines. Window 2 (1160.6–1161.6 cm^{-1}) contains absorption from CCl_2F_2, N_2O, and O_3. Window 3 (1193.3–1194.3 cm^{-1}) combines strong H_2O and N_2O absorption features. Lastly, window 4 (1216.2–1216.9 cm^{-1}) covers methane absorption signatures. While these windows were selected for the demonstration, any comparable window within the 118 cm^{-1} wide laser tuning range is readily selectable.

The tuning principles of the EC-QCL source have been extensively described [4,6]. Briefly, the EC-QCL provides coarse tuning across the entire laser gain spectrum

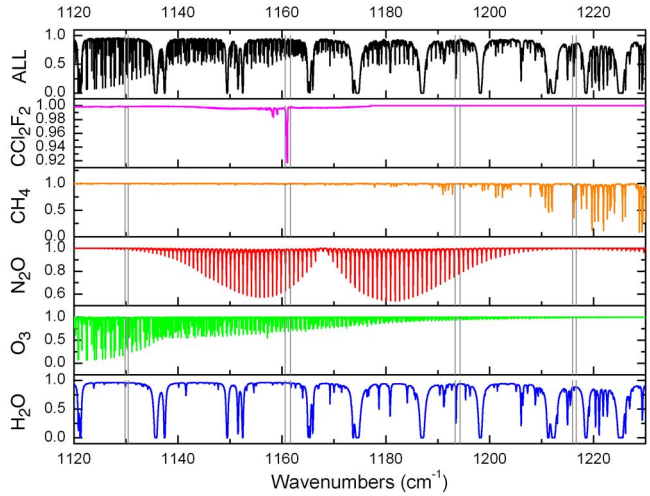

Fig. 2. (Color online) Synthetic atmospheric transmission spectra of the significant absorbers present within the EC-QC-LHR tuning range. Vertical lines indicate the narrow, molecule-specific, spectral windows selected.

through the adjustment of the external grating angle. If not controlled, mode hops occur typically at intervals equal to a laser chip cavity free spectral range of ~0.6 cm^{-1}. By simultaneously varying the QCL injection current, the EC length, and the grating angle via piezoelectric actuators, mode-hop-free tuning can be achieved over the microwindow of interest. As the LO is continuously tuned, the heterodyne signal, solar intensity, LO power, and transmission of the germanium etalon are all simultaneously recorded for data postprocessing.

The mode-hop-free continuous LO tuning was carried out within the four narrow windows selected. The instrument was located at –1.31° longitude, 51.57° latitude, and 128 m altitude. The LO power was adjusted with a variable attenuator to 100–400 μW during a frequency scan. The data sampling time interval was set to 2.5 times the lock-in amplifier integration time, which also sets the total acquisition time. Table 1 provides experimental conditions for the heterodyne spectra of Fig. 3, where θ, τ, and Tt are the solar elevation angle, the lock-in amplifier integration time, and the total measurement time, respectively. The panel above each spectrum in Fig. 3 shows the corresponding part of the synthetic spectra of Fig. 2, so that intense lines can be identified.

Spectra in Fig. 3 show that the EC-QC-LHR with 60 MHz resolution can resolve actual molecular absorption line shapes in the atmosphere, which are invaluable for obtaining information about altitudinal distribution of constituents. The baseline modulation appearing in the experimental spectra is related to the LO power modulation occurring during a mode-hop-free frequency sweep. Solar intensity variation may also contribute though to a much smaller extent, unless clouds cross the FoV, which was not the case during these measurements. The heterodyne detection performance was evaluated using a parameter, ρ, that indicates the ratio of the actual detection limit with that of an ideal shot-noise-limited LHR and can be expressed as

$$\rho = \frac{\eta \cdot \Delta \cdot \sqrt{B \cdot \tau}}{\exp\left(\frac{h \cdot \nu}{k \cdot T}\right) - 1} \cdot \frac{1}{\mathrm{SNR}_m}, \tag{1}$$

where η is the heterodyne efficiency of the PM (33 to 27% depending on frequency), Δ is the transmission of the incoming radiation up to the PM (52%), B is the LHR spectral resolution (60 MHz, set by an RF filter at the output of the PM), τ is the integration time, h is the Planck constant, ν is the optical frequency, k is the Boltzmann constant, T is the equivalent blackbody temperature of the Sun (5780 K), and SNR_m is the signal-to-noise ratio calculated from the experimental spectra. SNR_m was estimated by isolating a part of the spectra without

Table 1. Experimental Parameters for the Spectra Shown in Fig. 3

	Date	Time	θ (deg)	τ (ms)	Tt (s)
1	18 Jan. 2011	10:17 am	13.4	200	276
2	18 Jan. 2011	1:39 pm	15.6	100	126
3	18 Jan. 2011	10:36 am	14.7	200	251
4	28 Jan. 2011	12:54 pm	19.8	50	52

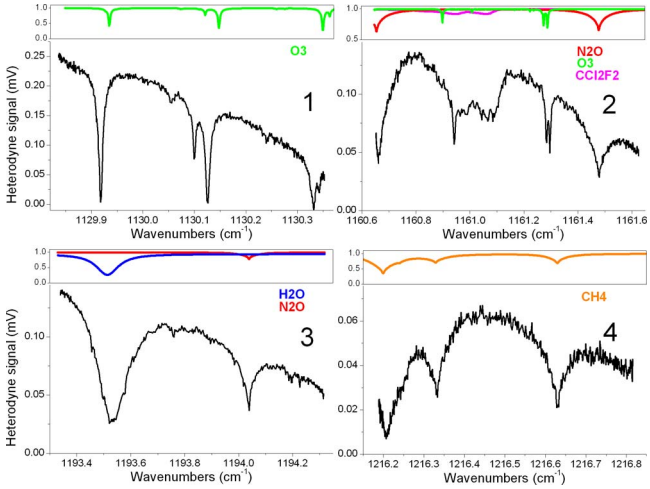

Fig. 3. (Color online) Raw heterodyne atmospheric spectra corresponding to the four spectral narrow windows shown in Fig. 2. The upper panel of each spectrum shows calculated zenithal spectra for line identification. For clarity, weakly contributing molecules have been omitted in the calculation (e.g., in spectrum 3, small features over the H_2O and N_2O lines are ozone absorption lines).

structured absorption features and calculating the ratio of the standard deviation to the mean of the dataset after baseline correction. For spectra 1 to 4, ρ values were 25.6, 25.4, 23.0, and 22.3, corresponding to noise equivalent absorption of 1.4%, 2.2%, 1.6%, and 3.9%, respectively. For comparison, ρ values obtained in the laboratory were ~7, primarily limited by a small amount of excess noise from the laser. The degradation is likely to be related to atmospheric scattering and phase front distortions, as a highly turbid atmosphere can reduce performance by more than 2 orders of magnitude [8].

To qualitatively compare the performance of the EC-QC-LHR to a well-established remote sounding instrument, atmospheric transmission spectra have been recorded using a Bruker IFS 125HR FTS, which occupies $4 \times 2\,m^2$ of laboratory space and is capable of up to 50 MHz resolution. The experimental conditions were identical to those of the EC-QC-LHR, except the amount of solar radiation reaching the FTS was halved due to insertion of a 50/50 beam splitter sampling the solar beam. Using a KBr beamsplitter and a liquid-nitrogen-cooled mercury cadmium telluride detector, the 3 dB spectral coverage of the FTS is 800 to 4000 cm^{-1}. Figure 4 shows subsets of the spectra recorded for different resolutions and scan averaging. An average of 10 scans at 60 MHz resolution, identical to that of the EC-QC-LHR, takes 2 h and exhibits mainly noise. The spectrum recorded at 600 MHz resolution with an average of 60 scans provides exploitable data. Total measurement time was 72 min, and the atmospheric absorption lines are far from being fully resolved. The FTS does offer a much wider spectral coverage than the EC-QC-LHR; however, when the FoV is narrow (meaning a high spatial resolution), it does not provide sufficient sensitivity at ultrahigh spectral resolution. Hence, the EC-QC-LHR offers the prospect

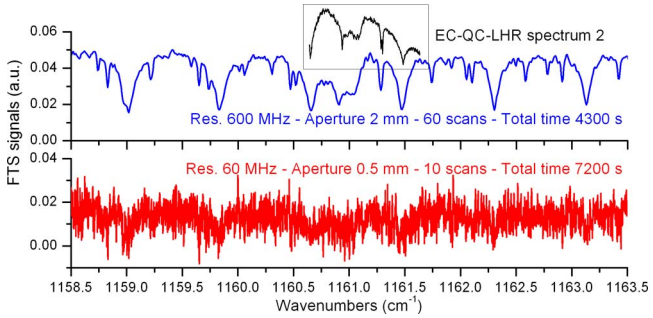

Fig. 4. (Color online) Subset of atmospheric FTS spectra recorded in various conditions of resolution and scan averaging. Inset shows the EC-QC-LHR spectrum 2 from Fig. 3.

for significantly more precise altitudinal distribution measurements at high spatial resolution.

To conclude, multispecies atmospheric observations in the mid-IR using an EC-QC-LHR has been demonstrated. The data show that line-shape-resolved spectral features of five atmospheric molecules could be observed by targeting specific narrow windows within the 118 cm^{-1} operating range of the instrument. These observations required only minutes of total acquisition time with both very high spectral and spatial resolutions. Comparison with experimental data obtained with FTS have shown that when high spectral resolution, narrow FoV, and short measurement times are required, an EC-QC-LHR focusing on specific narrow spectral windows provides far superior spectral information that may lead to enhanced knowledge on the state of the atmosphere. A large number of atmospheric spectra have been recorded during this observation campaign, which are currently being analyzed using inversion methods to retrieve atmospheric profiles of molecular concentrations.

The authors acknowledge Wayne Robins and the UK Natural Environment Research Council (NERC) Molecular Spectroscopy Facility at the Rutherford Appleton Laboratory for their support. This work was supported through NERC research grant NE/H002383/1, by the U.S. National Science Foundation (NSF) Engineering Research Center Mid-InfraRed Technologies for Health and the Environment (MIRTHE), and the NSF Graduate Research Fellowship DGE-0646086.

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