# Active coherent laser spectrometer for remote detection and identification of chemicals

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### ABSTRACT

Currently, there exists a capability gap for the remote detection and identification of threat chemicals. We report here on the development of an Active Coherent Laser Spectrometer (ACLaS) operating in the thermal infrared and capable of multi-species stand-off detection of chemicals at sub ppm.m levels. A bench top prototype of the instrument has been developed using distributed feedback mid-infrared quantum cascade lasers as spectroscopic sources. The instrument provides active eye-safe illumination of a topographic target and subsequent spectroscopic analysis through optical heterodyne detection of the diffuse backscattered field. Chemical selectivity is provided by the combination of the narrow laser spectral bandwidth (typically < 2 MHz) and frequency tunability that allows the recording of the full absorption spectrum of any species within the instrument line of sight. Stand-off detection at distances up to 12 m has been demonstrated on light molecules such as  $H_2O$ ,  $CH_4$  and  $N_2O$ . A physical model of the stand-off detection scenario including ro-vibrational molecular absorption parameters was used in conjunction with a fitting algorithm to retrieve quantitative mixing ratio information on multiple absorbers.

Keywords: lasers, explosives, threat chemicals, stand-off detection, QCL's, heterodyne

# **1. INTRODUCTION**

At the current time, no instrument exists that can meet the stringent requirements for remote detection and quantification of a wide variety of threat chemicals including explosives, toxic gases and chemical warfare agents. The requirements include simultaneous multi-species detection, high detection sensitivity (ppb level), working distances of 50 m or more, rapid response times (seconds to minutes), laser eye-safe operation and a compact, cost effective and easily deployable design. The most promising developments which meet some of these requirements involve the active transmission of laser radiation towards a distant target. Spectral analysis of the backscattered radiation allows the identification and quantification of any chemical species between the instrument and the target. However, the extremely rapid fall-off in collected backscattered signal with target distance has required the use of higher power sources which breach laser emission safety limits to achieve the required sensitivity levels [1].

Coherent or heterodyne mixing [2, 3] offers a number of potential advantages over direct detection methods. These include high sensitivity (ideally limited only by the fundamental laser shot noise on the detector), high spectral resolution (~2 MHz or  $10^{-4}$  cm<sup>-1</sup> corresponding to a resolving power of  $10^{7}$  at 1000 cm<sup>-1</sup>) and inherent high spatial resolution (typically centimeters squared at 50 m distance). Operation in the mid-infrared spectral region (2-20 µm) allows the use of compact high power Quantum Cascade Lasers (QCL's) which can be continuously tuned through spectral absorption features. Absorption cross sections in this spectral region are orders of magnitude greater than those found in the near-infrared (1-2 µm). Laser intensity exposure limits for mid-infrared radiation are also far less stringent than for the near infrared. For a continuous wave laser operating at longer wavelengths than 1.4 µm the upper limit for designation as an eyesafe Class 1 laser is 100 mWcm<sup>-2</sup>. QCL's typically have continuous power outputs of 10-500 mW which allows eyesafe operation with relatively small beam diameters (4-25 mm).

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Figure 1. Left, schematic of a remote detection scenario using the Active Coherent Laser Spectrometer. Right, schematic of the optical system for the Active Coherent Laser Spectrometer.

A typical stand-off detection scenario underpinning the Active Coherent Laser Spectrometer (ACLaS) development is shown in the left hand panel of Figure 1. A plume of chemical vapour emanating from a suspicious item of luggage is present at a distance from the active instrument. Infrared laser radiation is transmitted from the instrument and passes through the gas plume. The intensity of the transmitted radiation is well within laser safety exposure limits and poses no threat to human health. The transmitted radiation is backscattered from a nearby topographic target (for instance a wall) or by atmospheric aerosols. For an ideal Lambertian diffuse reflector, the backscattered radiation is spread evenly across a spherical surface. In the non-ideal case, the diffuse emission lobe is likely to peak in the direction defined by specular reflection conditions. The small fraction of the backscattered radiation falling within the instrument field of view is collected by the instrument and detected by heterodyne mixing with a local oscillator field which is coherent with the illumination field. The right hand panel of Figure 1 shows a schematic of the principles of the optical system used within the instrument. A single QCL laser provides both the active probe beam and the local oscillator required for heterodyne mixing via a beamsplitter. The returned backscattered radiation is combined with the local oscillator at a second beamsplitter and focused onto a fast photodiode. The resulting AC output of the detector (shifted into the radiofrequency domain) is filtered and detected by commercially available components. The filter bandwidth can be matched to the laser bandwidth (~ 1 MHz) for optimum performance. Wavelength tuning of the laser provides a way to resolve the spectral signature of the chemical vapour in the line of sight. Spectral fitting of experimental traces using reference absorption spectra provides both identification and quantification of the chemicals (both benign and potentially malignant) present in the plume.

On the basis of these principles, an Active Coherent Laser Spectrometer (ACLaS) operating in the mid-infrared spectral region based on QCL's has been developed to demonstrate the potential of the concept. Initial experimental results using a prototype instrument at distances up to 12 m are presented. The output of the instrument is analyzed through an instrumental model which returns the molecular mixing ratios with an uncertainty attached to their measurements.

# 2. METHODOLOGY

Figure 2 shows a schematic of the optical design of the active stand-off spectrometer that also includes modules dedicated to calibration and diagnostics. The optical components were constructed on an optical breadboard (0.75 x 0.75 m) which is mounted on a portable workstation which can be moved to any location. As far as possible, reflective optics were used to minimize optical feedback and to allow wavelength flexibility. A single distributed feedback quantum cascade laser (DFB-QCL) operating between 1274 and 1281 cm<sup>-1</sup> (7.85-7.81  $\mu$ m) with a maximum continuous power

output of 50 mW provided both the transmitted probe beam and the local oscillator (LO) beam. The highly spatially divergent output of the QCL was collimated with a black diamond aspheric lens (focal length = 4 mm).



Figure 2. Optical design of the Active Coherent Laser Spectrometer. Black traces are the local oscillator beam, green traces are the calibration beam and red traces are the active probe beam transmitted towards a distant target. Backscattered radiation is shown in blue. M indicates flat, gold coated mirrors, FM indicates flip mounted gold mirrors, OAPM are off-axis (90°) parabolic mirrors and BS are beamsplitters.

A ZnSe beamplitter (BS1, 10 % transmission) separated off the probe and LO beams. The LO beam was frequency shifted (by 76 MHz) relative to the probe beam by a Germanium acousto-optical modulator (AOM) with a conversion efficiency of 75 %. The remaining unshifted radiation was passed through a Germanium etalon (free spectral range of 480 MHz) and focused onto a slow Mercury-Cadmium-Tellurium photodiode (VIGO) to provide relative spectral calibration during wavelength scans. The probe beam was expanded by a pair of OAPM's and transmitted towards a distant target. The light intensity was well within the laser safety limits for a Class 1 laser in the mid-infrared region (< 100 mWcm<sup>-2</sup>). Diffuse backscattered radiation from the target was collected by an OAPM and co-imaged onto a fast MCT photodiode with the LO beam using a second beamsplitter (BS2, 10 % transmission). The image size of the LO beam was matched to the detector size (50 µm) by magnification through a pair of OAPM's. A hot blackbody (1300 K) was used to characterize the heterodyne mixing performance of the detector using a flip-mounted mirror. The AC output of the photomixer was filtered by an RF filter centered at 76 MHz with a bandwidth (3 dB level) of 0.8 MHz and detected by a Schottky diode. The signal was modulated by a mechanical chopper. A lock-in amplifier was used to demodulate the signal. The DC output was used as the input to an active feedback loop which maintained a constant level of LO power during a wavelength scan of the QCL by adjustment of a wire-grid polarizer on a motorized rotation stage. The outputs of the lock-in amplifier and both DC signals were read by a data-acquisition card using Labview software. An aperture in the incoming signal beam before OAPM6 showed the heterodyne field-of view to be 16.4 mm (95 % signal transmitted aperture size) for a 25 mm diameter optic.

## 3. RESULTS

#### 3.1 Forward model

Figure 3 shows a schematic of the forward model used to describe the Active Coherent Laser Spectrometer and its measurement principles. The forward model consists of three subsections; the detection scenario which includes the properties of the target and the atmosphere through which the radiation propagates, receiver properties which includes atmospheric turbulence and the physical optics of the instrument and finally, the receiver model which includes all potential sources of noise in the instrument. The model developed is extremely flexible and can be readily extended to include scattering by atmospheric aerosols or by solid, diffusely scattering targets.



Figure 3. Schematic outline of the forward model which describes the active coherent laser spectrometer.

For the specific case of a solid, diffusely scattering target as shown in Figure 1, the backscattered radiation received by the instrument ( $P_{BS}(\nu,L)$ ) can be approximated by Equation 1 where  $P_0(\nu)$  is the transmitted power from the instrument,  $\rho(\nu)$  is the diffuse reflectivity of the target,  $A_R$  is the area of the receiver aperture of the instrument, K is the collection efficiency of the optics within the instrument, L is the instrument to target distance and  $\alpha(z, \nu)$  is the absorption coefficient of the atmosphere which is given by Equation 2 where  $n_j(z)$  is the concentration of a particular species and  $\sigma_{j,abs}(\nu)$  is the wavelength dependent absorption cross section of the species j. Scattering by atmospheric aerosols between the instrument and the solid target is ignored.

$$P_{BS}(\nu,L) = P_0(\nu) \cdot \rho(\nu) \cdot \frac{A_R \cdot K}{4\pi L^2} \cdot exp\left[-2 \cdot \int_0^L \alpha(z,\nu) \cdot dz\right]$$
(1)

$$\alpha(z, v) = \sum_{j} n_{j}(z) \cdot \sigma_{j,ext}(v)$$
(2)

The wavelength (v) and distance (z) terms in Equations 1 and 2 allow the backscattered radiation to be calculated at any wavelength and for any atmospheric profile between the instrument and the target. For a homogenous atmosphere (constant concentration of all species) Equation 3 can be determined where  $\alpha(v)$  is the wavelength dependant absorption coefficient of the atmosphere.

$$P_{BS}(\nu,L) = P_0(\nu) \cdot \rho(\nu) \cdot \frac{A_R \cdot K}{4\pi L^2} \cdot exp[-2 \cdot \alpha(\nu) \cdot L] \quad (3)$$

The performance of the active instrument is given by Equation 4 where  $\langle i^2_{Het} \rangle$  indicates the time averaged heterodyne signal and  $\langle i^2_1 \rangle$ ,  $\langle i^2_2 \rangle$ ,  $\langle i^2_3 \rangle$ , etc. represent all the potential sources of noise in the experiment which includes shot noise, thermal background noise, detector (Johnson) noise, laser excess noise and speckle noise.

$$\frac{s}{N} = \frac{\langle i_{het}^2 \rangle}{\langle i_1^2 \rangle + \langle i_2^2 \rangle + \langle i_3^2 \rangle + \cdots}$$
(4)

The use of a single laser to provide both probe and local oscillator beams combined with a fixed frequency AOM means that the heterodyne beat frequency is constant regardless of the wavelength of the laser. This allows a narrow bandwidth RF filter (0.8 MHz) to be used which minimizes thermal background noise, laser excess noise and detector noise. Laser excess noise is further minimized by tuning the AOM frequency to a region free of strong noise features. The dominant noise sources are given in Equations 5 (local oscillator shot noise) and 6 (speckle noise) where  $\eta_q$  is the heterodyne efficiency, G is the preamplifier gain, e is the electron charge,  $P_{LO}$  is the local oscillator power, B is the double sided bandwidth, h is Planck's constant, v is the laser frequency and M is the speckle diversity parameter.

$$\langle i_N^2 \rangle = 2 \cdot \eta_q \cdot \frac{(G \cdot e)^2}{hv} \cdot P_{LO} \cdot B \tag{5}$$

$$\langle i_S^2 \rangle = \frac{\langle i_{het}^2 \rangle}{\sqrt{M}}$$
 (6)

The shot noise is the result of the random arrival of photons at the detector surface and represents the fundamental detection limit for coherent detection. For realistic detection scenarios the model suggests detection limits of less than 1 ppb for a gas plume of 1 m length for typical mid-infrared absorption cross-sections. However, speckle noise is an unavoidable consequence of coherent illumination of a surface which has a roughness greater than the laser wavelength [4]. The interference pattern created by the supposition of a large number of independent oscillators is far from uniform. The high spatial resolution of coherent mixing typically images a single "speckle-lobe" which yields a spectral diversity parameter of  $\sim$  1 and a SNR of  $\sim$  1. Addition of independent and uncorrelated speckle patterns within the integration time of the instrument leads to significant improvements in performance. Assuming varying degrees of speckle noise suppression the model yields detection limits of 1-100 ppb.m for mid-infrared operation.

#### 3.2 Experimental results

The left hand panels of Figure 4 show images of the backscattered radiation recorded using an infrared camera (image size 11.6 by 15.5 mm) at a distance of 10 cm from a diffusely scattering solid target. The right hand panels show the corresponding mutual coherence function given by Equation 8. The upper panels show results without speckle suppression while the lower panels show data using a speckle reduction scheme involving a spinning target used to test the feasibility of speckle reduction.



Figure 4. Left, Images of the backscattered signal 10 cm from a roughened Aluminum target. Right, mutual coherence functions of the images. The intensity of the mutual coherence function is shown using a logarithmic scale. The top row shows results for a stationary target while the bottom row shows data for a rotating target (50 revolutions per second).

In the absence of any suppression mechanism, a clear speckle pattern (top left panel) is observed which disappears when active suppression is applied (bottom left panel). The speckle suppression is mirrored in the mutual coherence functions. The degree of speckle reduction can be quantified by a speckle diversity parameter (M) defined in Equation 7 where A is the receiver area,  $A_c$  is the coherence area of the speckle pattern and b = 0.866.

$$M = \left[1 + \left(\frac{A}{A_c}\right)^b\right]^{1/b} \tag{7}$$

The spatial size of the correlated area, or speckle lobe area ( $A_c$ ), can be calculated using the Van-Cittert theorem which expresses the mutual coherence function of the field in an observation plane as proportional to the Fourier transform of the source power profile. This in shown in Equation 8 where ( $\xi$ ,  $\eta$ ) refers to the target plane in which the backscattered power profile  $P_R(\xi, \eta)$  is considered, ( $\Delta x$ ,  $\Delta y$ ) refers to the observation plane located at distance *R* from the target and  $\lambda$  denotes the wavelength.

$$\mu_{12}(\Delta x, \Delta y) = \frac{\iint P_R(\xi, \eta) \cdot exp\left[j\frac{2\pi}{\lambda \cdot R}(\Delta x \cdot \xi + \Delta y \cdot \eta)\right] \cdot d\xi \cdot d\eta}{\iint P_R(\xi, \eta) \cdot d\xi \cdot d\eta}$$
(8)

Practically the mutual coherence function is calculated using the 2-dimensional Fourier Transform of the speckle images shown in Figure 4 with a unit conversion by multiplication by a factor of  $\lambda R$ . The coherence area (A<sub>c</sub>) is found by a Gaussian fit to a horizontal slice through the mutual coherence function. The speckle diversity parameter (M) is very small in the absence of any speckle suppression mechanism (M  $\sim$  3) but is greatly increased by active suppression through incorporation of a spinning, roughened Aluminium target (M > 7000). Accurate measurement of the M parameter under these conditions is limited by the finite size of the camera image which limits the resolution of the Fourier Transform. Clearly, speckle suppression via a spinning target is not feasible in any realistic detection scenario and has been used here for studies. Alternate means of increasing spectral diversity from within the instrument are still under investigation. At the time of writing, an M parameter of 1800 has been achieved with a stationary target.

Figure 5 shows absorption spectra measured by the ACLaS instrument using backscattered radiation from a solid target at a distance of 5.5 m from the instrument. The QCL was tuned over ~1.6 cm<sup>-1</sup> by application of a sawtooth modulation to the laser current. The total acquisition time was 100 seconds per scan. The target was a roughened Aluminium disc at a distance of 5.5 m from the instrument. The target was spun at 50 revolutions per second to suppress speckle noise. The left hand panel shows absorption spectra of the atmosphere between the instrument and the target. The principal feature is due to water. The right hand panel shows the absorption spectrum of a metal gas-cell containing ~ 1000 ppm of N<sub>2</sub>O. The gas cell was placed immediately in front of the target such that both the transmitted and backscattered beams pass through the cell to simulate a narrow plume of gas. The total path length was 0.38 m.



Figure 5. Absorption spectra of atmospheric water (left) and  $N_2O$  (right) in a gas cell placed in both transmitted and backscattered beams (~1000 ppm, 0.38 m path length). The target was a roughened Aluminium disc at a distance of 5.5 m from the instrument. The upper panels show the experimental data (black dots) and the fit (red trace) using an optimum estimation method. The lower panels show the residual of the fit. The heterodyne bandwidth was 1.5 MHz with an integration time of 100 ms. Total acquisition times were 100 s per scan.

The red traces show a fit to the measured data using an Optimum Estimation Method which has been developed as part of our previous work on passive remote sensing of the atmosphere [3]. The fitted variables are the concentrations of the species of interest (H<sub>2</sub>O and N<sub>2</sub>O) and a polynomial baseline term. The sloping baseline is a result of power modulation in the transmitted power during the wavelength scan. The LO power was kept constant during the scan by the active feedback loop. The fitted concentrations were 6782 ppm (H<sub>2</sub>O) and 538 ppm (N<sub>2</sub>O). In addition the OEM fit gives an estimate of the detection sensitivity for both species (91 ppm.m, H<sub>2</sub>O and 254 ppb.m, N<sub>2</sub>O). The detection limits are expressed as the minimum detectable concentration in a plume of 1 m width. The rather low sensitivity for water represents the small absorption cross-section for this particular absorption line which is ~ 200 times smaller than those of N<sub>2</sub>O. The wavelength of the QCL was chosen to lie within an atmospheric window where water absorption is minimised.

A rotating target provides efficient speckle reduction and has enabled us to characterize the ACLaS instrument. However, it is clearly not a practical option for any realistic detection scenario. Therefore, atmospheric absorption spectra of water have been obtained under more realistic conditions using a stationary target at a distance of 12.2 m from the ACLaS instrument. The target used was white cardboard which has a much lower diffuse reflectivity than roughened metallic targets and is a better approximation for the reflectivity of natural topographic targets. Speckle diversity was produced by an internal mechanism which mimicked a rotating target. Short acquisition time scans (4 s) recorded at intervals of 10 s show large changes in heterodyne signal (by a factor of ~ 3-6) along with alterations in the baseline due to residual spatial speckle. OEM fits of the experimental traces yield water concentrations that are within +/- 10 % of each other and of the value obtained with a spinning target. Factoring in the differences in acquisition time (a factor of 5 assuming Gaussian statistics) leads to degradation in detection limits by a factor of only 2.5 (at best) relative to the limits found for the spinning target at shorter range. We are currently working on improving the performance of the ACLaS instrument by optimising the internal speckle reduction mechanism and applying it to both the transmitted probe beam and the backscattered signal beam.

Analysis of the collected data indicates that the current ACLaS instrument is limited by speckle noise with diversity parameters that range from  $10-10^5$ . The sub ppm.m detection limits are still several orders of magnitude greater than the ultimate shot noise limit suggested by the forward model indicating that there is substantial room for improvement.

## 4. CONCLUSION

The concept of mid-infared Active Coherent Laser Spectrometer (ACLaS) for stand-off detection of chemicals has been demonstrated. The instrument is based on coherent (heterodyne) detection of backscattered radiation from a distant target, using active illumination from a quantum cascade laser. In combination with a comprehensive forward model, quantitative information on path integrated mixing ratios could be obtained down to ~100 ppb.m for chemicals with large absorption cross-sections such as N<sub>2</sub>O in the present demonstration. Coherent detection also provides a high spatial resolution of 2.1 cm<sup>2</sup> which is ideal for imaging localized sources of emission and a high spectral resolution which matches the illuminating laser bandwidth (~ 1 MHz). The performance of the demonstration instrument is far above the fundamental shot noise limit and is limited by speckle noise. Improvements in detection sensitivity by up to several orders of magnitude are expected through better speckle reduction methods. These improvements are currently being implemented and operation at longer, more relevant, ranges will be tested. The use of distributed feedback quantum cascade lasers limits the operating frequency range (to a few wavenumbers) and allows the study of narrow band absorbers only. In the near future, it is planned to operate the ACLaS instrument using widely tuneable external cavity QCL's and CW mid-infrared OPO's to extend the instrument capability to include broadband absorbers.

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