10.1117/2.1201605.006458

Detecting chemical threats at safe standoff distances

Damien Weidmann and Neil Macleod

An eye-safe, mid-IR hyperspectral active standoff detection system provides identification, detection, and confident quantification of chemical mixtures.

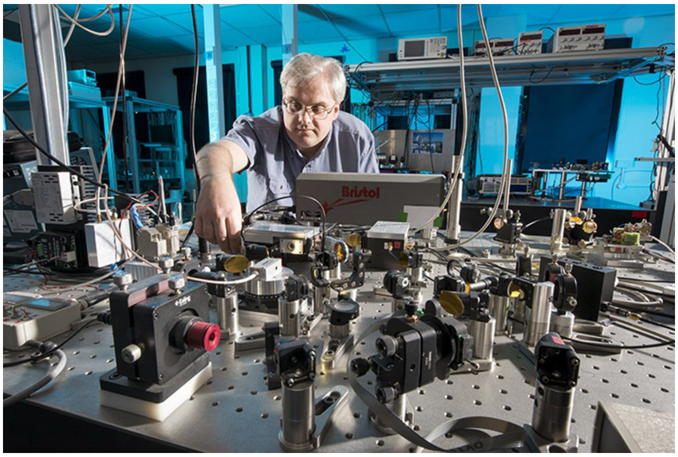

Figure 1. The very first benchtop active coherent laser spectrometer system developed for concept demonstration.

Standoff chemical detection is the ability to detect chemicals in an object that is some distance from the measurement instrument. This capability, along with chemical identification and quantification, is highly sought after in security and defense applications, which require remote analysis of explosives, toxic chemicals, and narcotics. However, the operational requirements of such systems for deployment in real-world environments are stringent. They include high chemical sensitivity and selectivity (even in cluttered chemical environments), simultaneous detection of both vapor phase chemicals and solid contaminants on surfaces, rapid response times, laser eye-safety, invisible illumination, and outdoor operation. Additionally, portable, power-efficient, and cost-effective devices are highly desirable.

The fulfillment of these combined requirements is challenging, and while optical standoff approaches are considered the most viable option, to date no satisfactory solution has been developed. Passive systems relying on background radiation are not sensitive enough. Active optical techniques, such as laser-induced breakdown spectroscopy, Raman spectroscopy, laser-induced fluorescence, and differential absorption lidar, do not always meet the eye-safety requirements, or cannot deliver both high sensitivity and high selectivity. For these reasons, we have investigated a solution that exploits recent progress in widely tunable mid-IR laser sources combined with the benefits of laser heterodyne detection. Heterodyne detection involves mixing light with that of a reference laser (the local oscillator) to enable spectral analysis and to operate frequency down-conversion. It enables spectrometry with ultra-high resolution and demonstrates high sensitivity by overcoming detector noise. The mid-IR region of the electromagnetic spectrum (2–20μm) is of particular interest for remote molecular detection since the atmosphere is transparent in the medium-wave IR (MWIR 3–5μm) and the long-wave IR (LWIR 8–12μm), which allows for long-range sensing. In the mid-IR region, molecules exhibit intense fundamental rotational-vibrational bands that create large molecular absorption cross sections, which enable enhanced detection sensitivity. Furthermore, the uniqueness of the spectral signatures, if properly resolved, provides information about functional groups and molecular fingerprints, which relates to chemical selectivity. To exploit these benefits, we developed the active coherent laser spectrometer (ACLaS), a mid-IR hyperspectral heterodyne lidar. We combined this approach with the development of spectral processing algorithms that are able to deconvolve the information contained in complex multi-chemical mixtures.

In our first ACLaS demonstrations, we used continuously tunable quantum cascade lasers (QCLs) to provide both the active illumination source and the local oscillator field (which was slightly shifted in frequency) of the laser heterodyne detection scheme. Optical heterodyne detection enables sensitive detection of the returned backscattered field, and ultra-high spectral resolution at the receiver, which is typically matched to the il-

Continued on next page

10.1117/2.1201605.006458 Page 2/2

luminating laser linewidth (of a few megahertz) and efficiently suppresses any background radiation. By continuously tuning the local oscillator/illuminator frequency, we obtained very high spectral resolution signatures of chemical absorption along the line of sight.

We tested two systems over standoff ranges covering 1–40m, using topographic targets such as brick, cardboard, painted metal, and aluminum foil. The first system was based on distributed feedback QCLs tunable over $\sim$5–10cm^{-1}, and was well-suited to study light molecules exhibiting narrow-band absorption features. A second system using an external cavity QCL (continuously tunable over $\sim$80cm^{-1}) demonstrated standoff detection of complex mixtures of vapors, including light and heavy molecules, which produced composite spectral signatures that mixed broadband and narrow-band structures.

High-resolution spectra are of no use to the end-user without their robust conversion into quantitative metrics. To that end, we developed a spectral processor based on the optimum estimation method. This approach relied on a forward model that describes the full physics known about the detection scenario and the instrument and its inversion, enabling us to recover molecular concentration parameters from spectral data. If necessary, we could add a priori parameters—and the level of confidence we had in them—to constrain the inversion further. This approach not only carries out the spectral inversion, it also provides a series of diagnostic tools to estimate error and bias propagation. As a result, we were able to quantify the level of cross-talk between retrieved parameters, and could provide a metric to estimate the level of confidence in the final output.[1]

We successfully tested the system (ACLaS and spectral processor) on nine different vapor phase molecules, and performed a very preliminary demonstration on surface deposits. The widely tunable mid-IR laser sources enabled good chemical versatility. The range of chemicals studied spanned atmospheric species (water, carbon dioxide, nitrous oxide, and methane), industrial gases (including the main components of natural gas), and explosives (ethylene glycol dinitrate and the precursor hydrogen peroxide). Using our processing algorithm, we confidently measured concentrations down to part-per-billion levels, even in spectrally cluttered environments.

In summary, combining the benefits of heterodyne detection (ideally offering shot-noise-limited detection together with high-spectral-resolution spectrometry) and the spectral processor, we have demonstrated a relatively simple, compact, and eye-safe system that achieves high detection sensitivity ($<$ 1ppm.m/$\sqrt{\text{Hz}}$ for the strongly absorbing molecules) and quantitative chemical selectivity. From these early investigations we have identified a number of limitations, and believe it may be possible to improve sensitivity by one or two orders of magnitude. To achieve this, in future work we will consider the use of laser sources covering the full MWIR and LWIR, such as optical parametric oscillators.

Author Information

Damien Weidmann and Neil Macleod
Rutherford Appleton Laboratory
Didcot, UK

Damien Weidmann leads the Spectroscopy Group in the Space Science and Technology Department. His work relates to research and development of novel laser-based instrumentation and applications for trace chemical analysis.

References

1. N. A. Macleod, F. Molero, and D. Weidmann, *Broadband standoff detection of large molecules by mid-infrared active coherent laser spectrometry*, **Opt. Express 23**, pp. 912–928, 2015.