

LIDAR SPRECTROSCOPY INSTRUMENT (LISSI): AN INFRASTRUCTURE FACILITY FOR CHEMICAL AEROSOL PROFILING AT THE UNIVERSITY OF HERTFORDSHIRE

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ABSTRACT

The lidar development at the University of Hertfordshire explores the feasibility of using Raman backscattering for chemical aerosol profiling. This paper provides an overview of the new facility. A high-power Nd:YAG/OPO setup is used to excite Raman backscattering at a wide range of wavelengths. The receiver combines a spectrometer with a 32-channel detector or an ICCD camera to resolve Raman signals of various chemical compounds.

The new facility will open new avenues for chemical profiling of aerosol pollution from measurements of Raman scattering by selected chemical compounds, provide data that allow to close the gap between optical and microphysical aerosol profiling with lidar and enables connecting lidar measurements to parameters used in atmospheric modelling.

1 INTRODUCTION

Conventional Raman or high spectral resolution lidar is used for deriving profiles of aerosol optical properties and, if proper input data are provided, retrieving aerosol microphysical properties through inversion techniques. The lidar development at the University of Hertfordshire (UH) is centred on exploring the feasibility of using Raman backscatter of various target species to obtain mass concentration profiles of selected chemical compounds. This marks the logical next step in aerosol profiling with lidar and will allow for defining aerosol types according to their chemical composition rather than optical properties. Ultimately, this will unify the treatment of aerosol types in lidar remote sensing with that of in-situ measurements and atmospheric modelling. The feasibility of the underlying methodology has been proven for measurements of Raman backscattering at silicone dioxide molecules within Asian dust layers [1, 2, 3].

2 OBJECTIVES

The lidar spectroscopy instrument (LiSsI) will enable profiling of trace gases, chemical components in particles, and bio-aerosols in atmospheric aerosol pollution through the combination of different non-linear spectroscopy techniques (photoluminescence, fluorescence, Raman and coherent anti-Stokes Raman spectroscopy) in a single measurement platform. The work at UH includes (1) the development of an end-to-end simulator that allows us to model the processes of inelastic photoluminescence and Raman scattering by aerosol particles and gases in the context of lidar remote sensing, (2) the identification of the luminescence and Raman scattering characteristics for a set of key aerosol types and gases (of natural and anthropogenic origin) by means of laboratory experiments, (3) test measurements and case studies as a proof of concept of the technique under laboratory conditions, and (4) the characterisation and development of a mobile prototype inelastic lidar spectrometer receiver for field deployment at established lidar sites.

3 LABORATORY SETUP

Figure 1 presents a sketch of the setup of the LiSsI facility. The main components are a high power Nd:YAG laser (Continuum Powerlite Furie LD), an Optical Parametric Oscillator (Horizon OPO), a motorised beam combiner, a multiwavelength depolarisation Raman lidar receiver, a Horiba 1250M spectrometer that can be used with an intensified CCD camera (ICCD, Princeton Instruments PI-MAX4 1024i-HBf) or a 32-channel Licel PMT (32PMT), an Olympus BX51TRF-6 Raman/fluorescence microscope, and several custom-made gas chambers.

An overview of the properties of the transmitter is provided in Table 1. The combination of a high-power Nd:YAG laser and an Optical Parametric Os-

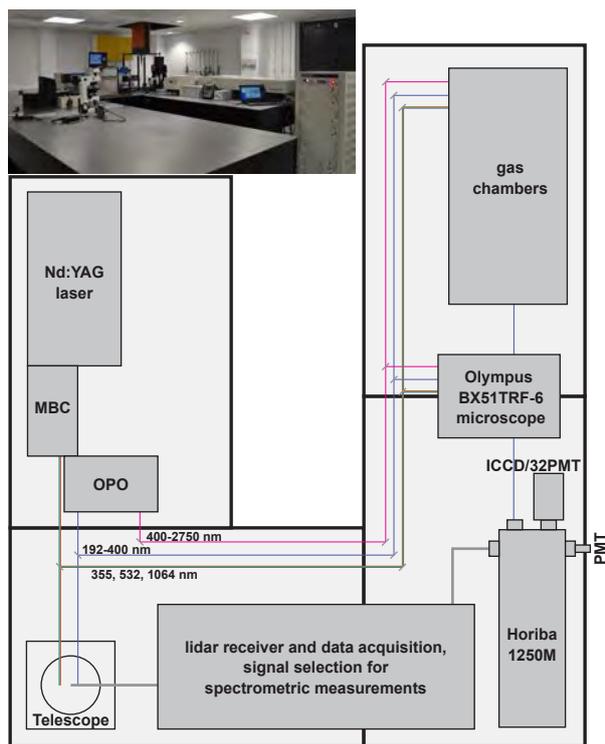


Figure 1: Sketch of the laboratory setup including a high-power Nd:YAG laser, a motorised beam combiner (MBC), an Optical Parametric Oscillator (OPO), an advanced lidar receiver, a spectrometer, an intensified CCD camera (ICCD), a 32-channel Licel PMT (32PMT), a Raman/fluorescence microscope, and several gas chambers.

illator allows for exciting Raman backscatter for a wide range of excitation wavelengths. The novel lidar receiver setup has been designed and simulated using ZEMAX and will combine the spectral resolution of spectrometers with a 32-channel detector to resolve Raman signals of a variety of chemical compounds. An overview of the configuration and properties of the receiver and data acquisition is provided in Table 2. Aerosol chambers will enable measurements of a range of chemical species to test the methodology for known compounds and to assess the respective detection limit with respect to applied laser power and resolvable particle concentrations. A Raman microscope will be used to measure the Raman scattering cross-sections of the selected target species—information that is needed to transform the detected signals into mass concentrations.

Table 1: Properties of the transmitter.

Continuum Powerlite Furie LD	
Laser type	Nd:YAG, injection seeded
Wavelength and pulse energy	7500 mJ @ 1064 nm 5000 mJ @ 532 nm 2500 mJ @ 355 nm
Beam divergence	0.5 mrad, 0.1 mrad after 5-fold beam expansion
Repetition rate	10 Hz
Linewidth	<0.003 cm^{-1}
Pulse duration	<15 ns
Horizon Optical Parametric Oscillator	
Pumping wavelength	355 nm
Wavelength and pulse energy	from 192 to 2750 nm scanning step 0.01 nm 120 mJ @ 400 nm 60 mJ @ 600 nm 25 mJ @ 300 nm
Beam divergence	<2 mrad (both axes)
Repetition rate	10 Hz
Linewidth	2-6 cm^{-1} , injection seeded
Pulse duration	<15 ns

The aim of the facility is to carry out precise inelastic spectroscopy experiments that target measurements of photoluminescence, fluorescence, and Raman spectra of aerosol and gas samples. These measurements will include the identification of spectra, absolute values of fluorescence, and Raman cross-sections that are currently poorly known or unknown.

4 APPLICATIONS IN LIDAR AND SPECTROSCOPY

The LiSsI facility has been designed to allow for comprehensive laboratory experiments as well as for atmospheric observations. The laser beam can be released into the atmosphere through a hatch in the roof of the laboratory. The backscattered light is collected with a 14-inch Schmidt-Cassegrain telescope and guided to the different components of the receiver (Table 2). Depending on the setup of the experiment, LiSsI can be used as multi-wavelength elastic backscatter lidar for measurements of aerosols and temperature from the troposphere to the mesosphere, as multi-channel spectroscopic Raman lidar (using Stokes and anti-Stokes, rotational, and rotational-vibrational Raman scattering), multi-channel spectroscopic photoluminescence/fluorescence lidar, high spectral resolution li-

Table 2: Properties of the receiver and data acquisition system.

Schmidt-Cassegrain telescope	
Focal length	3910 mm (14 inch)
Field of view	0.5-4.0 mrad (variable)
HORIBA 1250M Research Spectrometer	
Focal length	1.25 m
Aperture	F/9
Spectral range	0-1500 nm mechanical range (1200 g/mm grating)
Grating size	110 mm × 110 mm
Dispersion @500 nm	0.65 nm/mm
Accuracy	±0.15 nm
Repeatability	±0.005 nm
Gratings and resolutions @313.183 nm	2400 g/mm blaze 250 nm, max resolution 0.003 nm 1800 g/mm blaze 400 nm, max resolution 0.004 nm 1200 g/mm blaze 330 nm, max resolution 0.006 nm 600 g/mm blaze 500 nm, max resolution 0.012 nm
Detection	
Mie and Rayleigh scattering	355 nm, PMT HV-R9880U-20, bandwidth 1.3 nm 532 nm, PMT HV-R9880U-20, bandwidth 1.3 nm 1064 nm, APD InGaAs50, bandwidth 4 nm
Spectroscopic 1 Licel SP32-20	Hamamatsu H7260-20, 0.8 mm × 7 mm × 32 anodes spectral response 300-920 nm
Spectroscopic 2 Princeton Instruments PI-MAX4 ICCD camera	1024 × 1024 imaging pixels; 12.8 μm × 12.8 μm pixels Gen III filmless intensifier Sensitive range 290-710 nm QE>20% in range 355-700 nm; QE>40% in range 410-640 nm
Data acquisition system	
Mie and Rayleigh scattering	Licel transient recorders, 16 bit, 20 MHz A/D converters and photon-counters maximum count rate 250 MHz, variable range resolution
Multi-anode PMT	Single-photon counting system, maximum count rate 100 MHz, 50 ns resolution
ICCD	Digitization 16 bit, 32 MHz, minimum gate width 2 ns

dar (HSRL), polarization lidar, or infrared absorption and differential absorption lidar (DIAL).

In addition, LiSsI opens possibilities for a wide range of applications in spectroscopy: general spectroscopy (attenuation, transmission, and reflectance spectroscopy), high-spectral resolution spectroscopy, Stokes and anti-Stokes Raman spectroscopy of gas and solid material including applications involving a microscope, fluorescence spectroscopy of gas and solid material including microscope applications, laser-induced breakdown spectroscopy, coherent-anti-Stokes Raman spectroscopy, time-resolved spectroscopy, infrared gas analysis and materials processing.

5 FIRST RESULTS

The LiSsI laser has been installed in September 2017. Figure 2 shows one of the first measure-

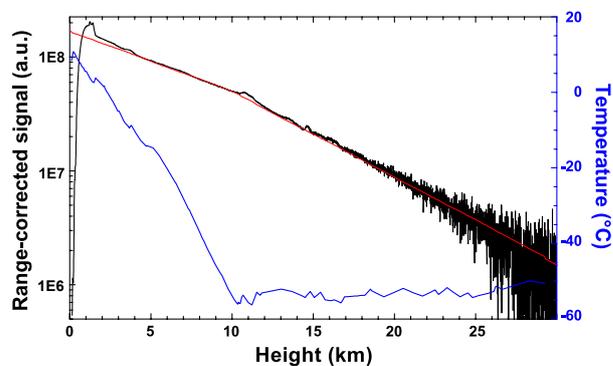


Figure 2: First measurement at 532 nm on 22 September 2017 (black) together with a molecular (red) and a temperature (blue) profile from a sounding launched at Nottingham at 0000 UTC on 22 September 2017.

ments performed between 1026 and 1330 UTC on 22 September 2017 using only a 532-nm elastic channel and a laser power of about 30 mJ. The analog counting signal is shown in the figure. Despite the low laser power, a rather small telescope and

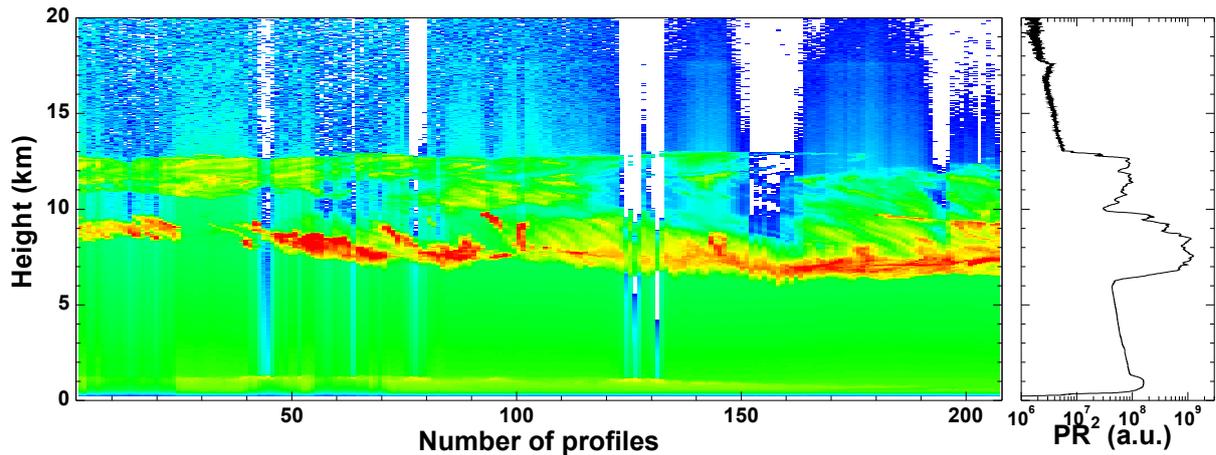


Figure 3: Time-height display (left) and average profile (right) of the range-corrected signed at 532 nm detected during a measurement between 1644 and 2010 UTC on 28 September 2017 with a temporal resolution of one minute.

observations being performed during daytime, reasonable signals can be obtained up to a height of 20 km. Aerosol can be seen in the lowermost 4 km as well as in layers at 7, 11, 14, 15, and 16 km height. The temperature profile of a radiosonde launched at Nottingham (about 150 km north of Hatfield) shows the tropopause at about 10 km height, putting the upper aerosol layers—likely originating from pyro-cumulonimbus events in Canada several weeks earlier—well within the stratosphere.

Figure 3 presents data from the first nighttime measurement performed with LiSsI. The configuration has been the same as during the measurements presented in Figure 2. Cirrus clouds are clearly visible between 6.5 and 13.0 km. The stratospheric aerosol layer from Figure 2 is still present between 17 and 18 km height.

6 NEXT MILESTONES

The coming milestones in developing the LiSsI facility are aligned with increasing the complexity of the optical setup. After the initial test measurements (Figure 2), we intend to perform atmospheric measurements of elastically and inelastically backscattered light at the different laser wavelength using full power. After that, the spectrometer will be integrated into the lidar receiver to allow for multispectral measurements in the atmosphere and using the gas and aerosol chambers in the laboratory.

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