

Identification of Atmospheric Mineral Dust Composition from Raman-Scattering Spectra

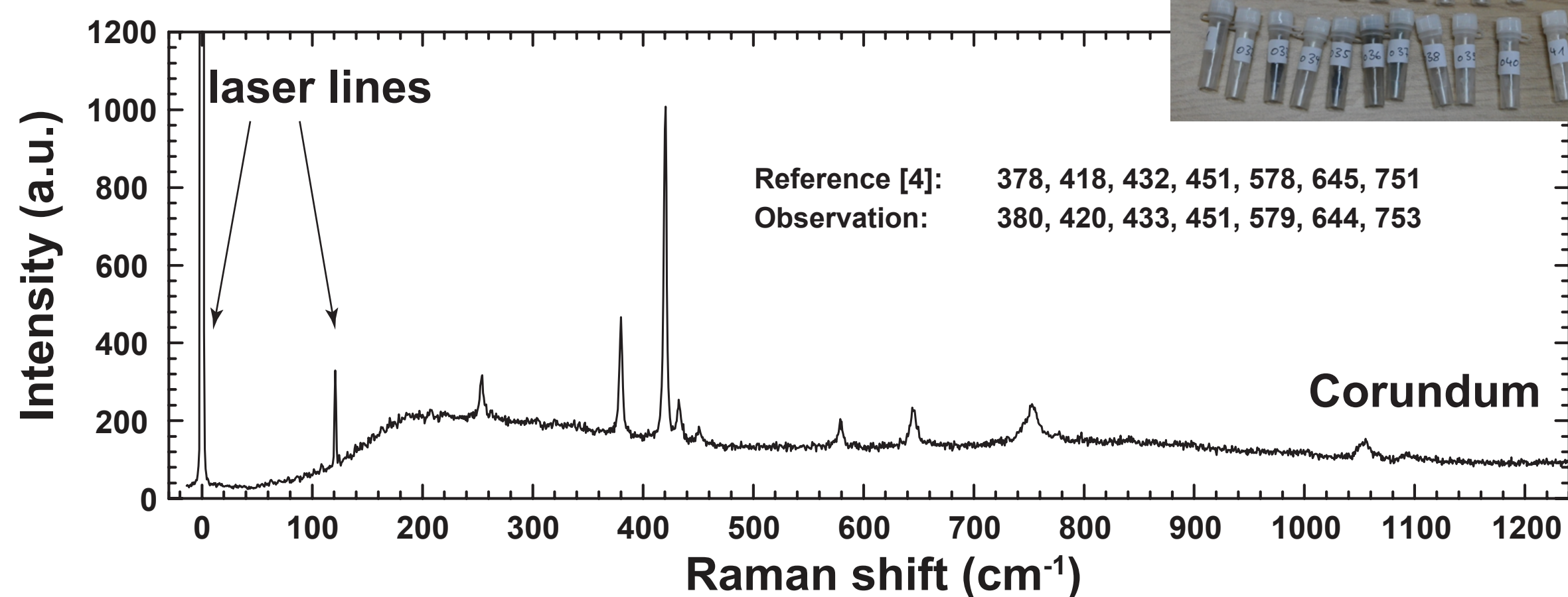
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Abstract

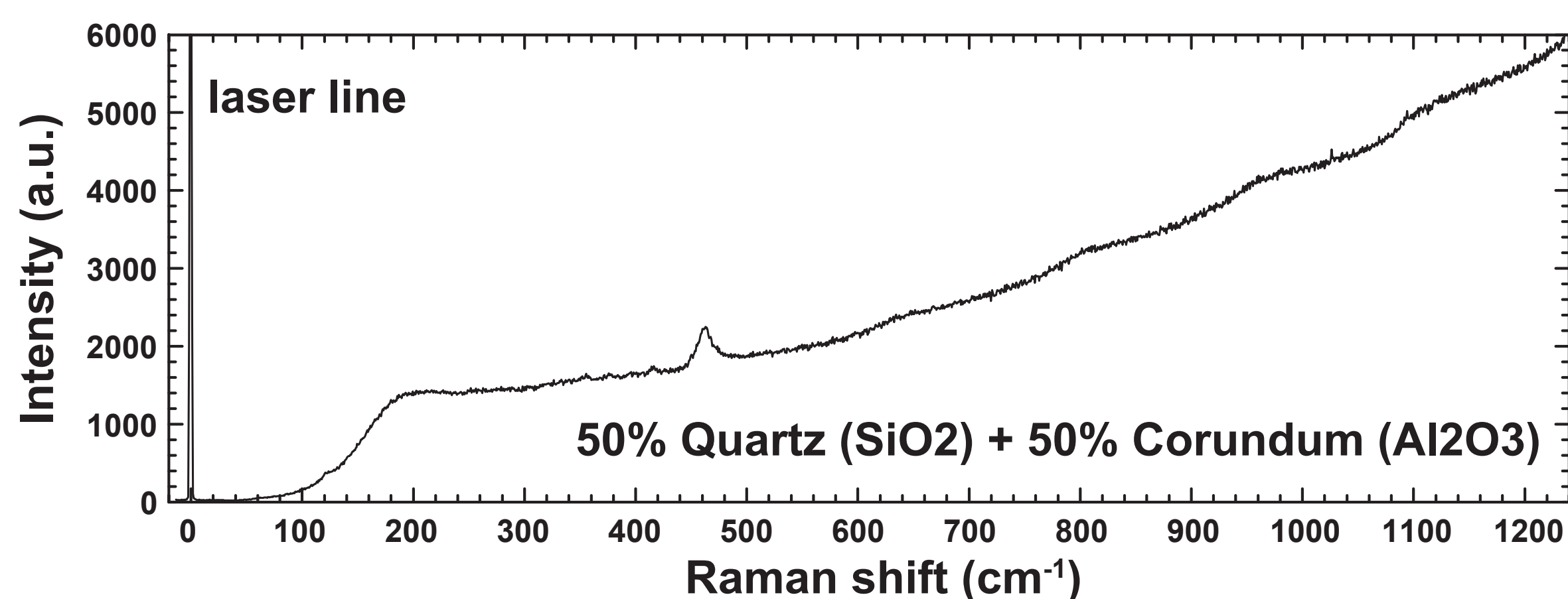
The aim of this work is to use a Raman microscope to collect “optical fingerprints” of mineral dust samples (i.e. pure materials and atmospheric samples collected from different source regions) in the form of Raman spectra. In particular, peaks in the Raman spectra will be investigated for their suitability for determining the composition of mineral dust in the atmosphere from light detection and ranging (lidar) measurements [1]. Raman spectroscopy represents a particularly powerful tool for laser remote sensing because it allows us to both identify and quantify the chemical constituents in a complex mixture - as is often the case for atmospheric aerosol pollution. Today, Raman lidar allows for an independent quantitative measurement of the aerosol backscatter and extinction coefficient profiles on the basis of Raman scattering from nitrogen or oxygen molecules. Previous work [2,3] has shown that the detection of Raman scattering by silicone dioxide in a lidar receiver (i.e. by introducing a dedicated measurement channel) can be used to infer the concentration of mineral dust in the atmosphere. We will develop a novel multi-channel spectrometric lidar system to shift the lidar paradigm towards measurements of Raman spectra that can give us information on chemical signatures characteristic of trace gases and the chemical components of aerosol particles. To achieve this vision, we need to obtain currently unavailable information on the Raman spectra of the aerosol species commonly found in the atmosphere.

Raman spectra of dust samples

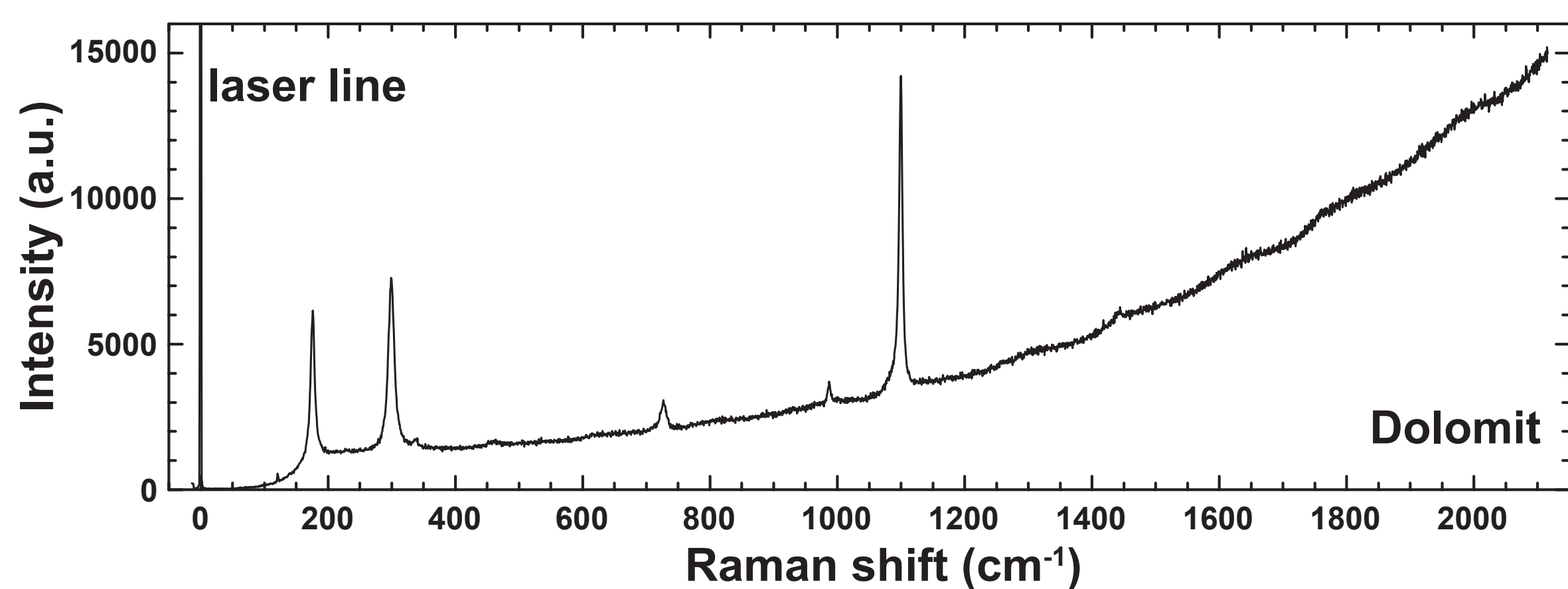
- ▶ Currently, we have a set of 40 dust samples from different desert regions
- ▶ Samples include Aluminium oxide, Quartz, Kaolinite, Dolomite, Goethite, Calcite, Corundum, Magnetite, Olivite, Apatite, etc
- ▶ Some samples are only designated by location of collection
- ▶ Results will be compared to reference data whenever available



Sample 01 / Corundum (Al_2O_3): measurement with a spectral resolution of 0.02 nm, 150 mW laser power, a grating with 2400 gr/mm, a slit width of 200 μm , acquisition of 1 s and 5 accumulations.



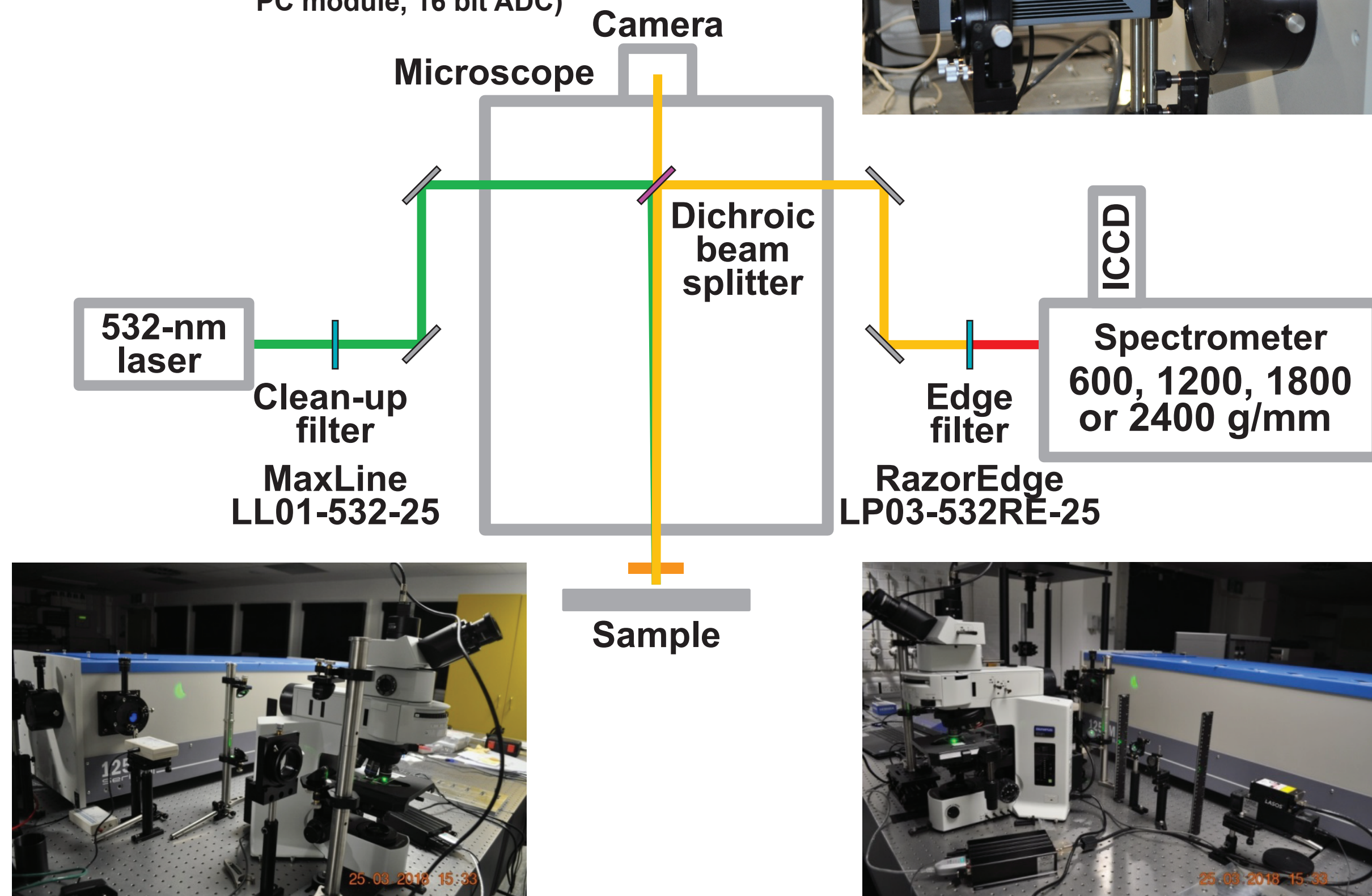
Sample 20 / 50% Quartz (SiO_2) + 50% Corundum (Al_2O_3): measurement with a spectral resolution of 0.02 nm, 150 mW laser power, a grating with 2400 gr/mm, a slit width of 200 μm , acquisition of 1 s and 5 accumulations.



Sample 28 / Dolomit ($\text{CaMg}(\text{CO}_3)_2$): measurement with a spectral resolution of 0.02 nm, 150 mW laser power, a grating with 2400 gr/mm, a slit width of 200 μm , acquisition of 0.1 s and 10 accumulations.

Raman microscope setup

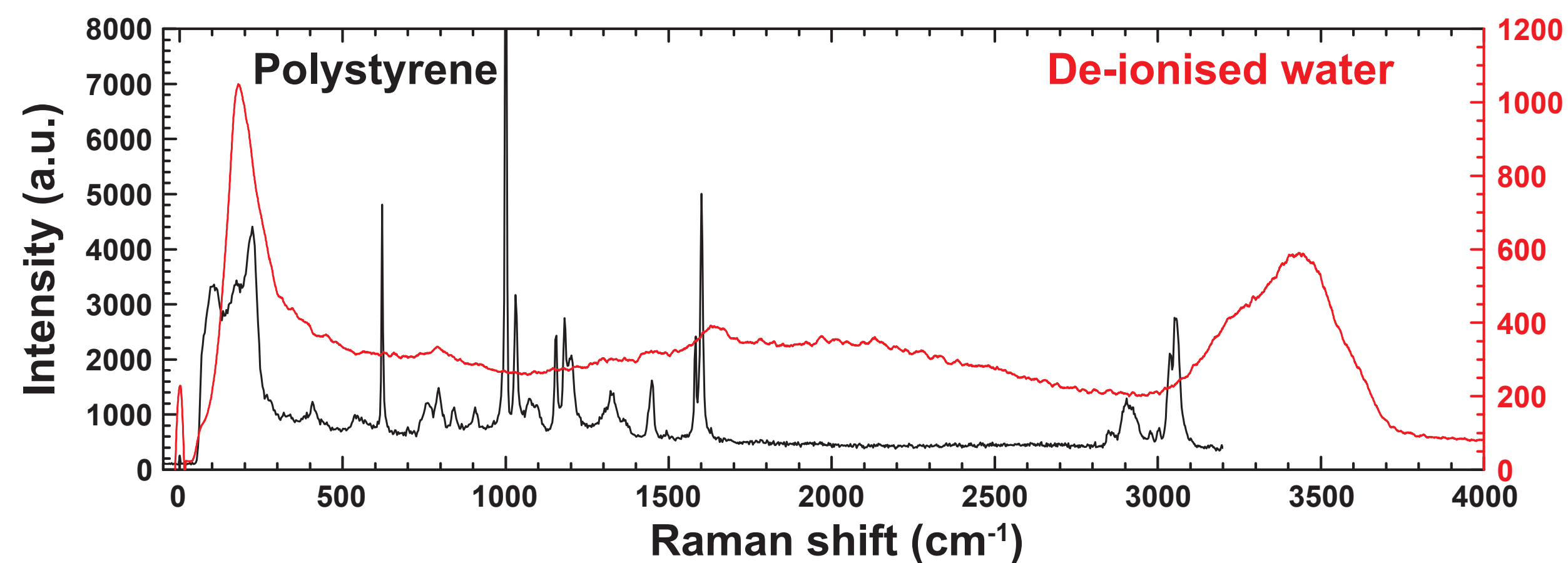
- Light source: ▶ LASOS DPSS GLK green diode laser, 150 mW at 532 nm
Microscope: ▶ OLYMPUS BX51
Spectrometer: ▶ HORIBA 1250M-SII, 2400 gr/mm grating
DAQ: ▶ Princeton Instruments PI-MAX4 ICCD
▶ Licel SP32-20 32-channel Multi-Anode PMT
▶ Single PMT with scanning software (R928P, 200-920 nm, SpectrAcq3 DAQ System, DM302 PC module, 16 bit ADC)



Calibration and measurement procedure

1. Characterisation of measurement system
 - ▶ Calibrate Spectrometer and ICCD: Raman standards (polystyrene), calibration source (Hg, Ar lamps)
 - ▶ Blank slide characterization
 - ▶ Laser emission line characterization
2. Measurement procedure
 - ▶ Prepare sample on microscope slide and mount on microscope
 - ▶ Focus image of sample and laser beam
 - ▶ Take image of sample through the microscope
 - ▶ Take coarse spectrum with ICCD or scanning PMT
 - ▶ Refine spectrometer setup and data acquisition setting
 - ▶ Take Raman spectrum
3. Data processing:
 - ▶ Noise reduction
 - ▶ Ambient light and cosmic rays correction
 - ▶ Account for contributions from slide and fluorescence

Polystyrene reference from www.chem.ualberta.ca/~mccreery/ramanmaterials.html, Raman shift (cm^{-1}):
McCreery Group, U Alberta: 620.9, 795.8, 1001.4, 1031.8, 1155.3, 1450.5, 1583.1, 1602.3, 2852.4, 2904.5, 3054.3
Our data, U Hertfordshire: 621.2, 794.4, 1002.3, 1030.7, 1155.9, 1449.2, 1583.4, 1601.1, 2849.5, 2905.2, 3052.5



Calibration measurement of Polystyrene (black) and de-ionised water (red) with a spectral resolution of 0.02 nm, 150 mW laser power, a grating with 2400 gr/mm, a slit width of 200 μm , acquisition of 0.1 s and 10 accumulations.

Summary and outlook

- ▶ The long-term objective of this work is to develop a complex lidar spectrometer that allows us to measure vertically resolved profiles of trace gases, chemical components in particles, and bio-aerosols in atmospheric aerosol pollution [5].
- ▶ Hypothesis: Combining non-linear spectroscopy methods (photoluminescence, fluorescence, Raman and CARS) into a single platform, i.e., lidar, will allow for vertically-resolved chemical characterisation of tropospheric aerosols and gases.
- ▶ We will measure the photoluminescence, fluorescence and Raman spectra of aerosol and gas samples in the lab to identify spectral features, absolute values of fluorescence and Raman cross-sections which are poorly known or unknown.
- ▶ The goal is a proof of concept under laboratory conditions that will allow us to define the hardware specifications and measurement sensitivity that is needed for a complex inelastic lidar spectrometer for atmospheric measurements.

References

- [1] Inaba and Kobayasi (1972), Laser-Raman radar -Laser-Raman scattering methods for remote detection and analysis of atmospheric pollution, Opto-electronics, 4, 101-123.
- [2] Müller et al. (2010), Mineral quartz concentration measurements of mixed mineral dust/urban haze pollution plumes over Korea with multiwavelength aerosol Raman-quartz lidar, Geophys. Res. Lett., 37, 2010GL044633.
- [3] Tatarov et al. (2012), Record heavy mineral dust outbreaks over Korea in 2010: Two cases observed with multiwavelength aerosol/depolarization/Raman-quartz lidar, Geophys. Res. Lett. 39, 2012GL051972.
- [4] Porto and Krishnan (1967), Raman Effect of Corundum, The Journal of Chemical Physics, 47(3), 1009-1012.
- [5] Tesche et al. (2018), Lidar spectroscopy instrument (LISSI): An infrastructure facility for chemical aerosol profiling at the University of Hertfordshire, EPJ Web of Conferences, 176, doi: 10.1051/epjconf/201817601008.