Fig. 1 The Schneefernerhaus research station on Mount Zugspitze. The Raman lidar telescope receiver is located in the small cylindrical structure topped with a white retractable dome, located on the uppermost roof of the facility.

Taking measurements to the top

A differential absorption lidar (DIAL) and a high-power Raman lidar have been developed and optimized on Mount Zugspitze.

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To improve the sensitivity and range of atmospheric water vapor measurements by ultraviolet Raman lidar, a powerful 308 nm industrial excimer laser was modified to deliver frequency-stabilized, narrowline output with low divergence and linear polarization.

The distribution of water vapor in the atmosphere plays a key role in climate and meteorological events. This distribution is highly inhomogeneous and dynamic. To enable improved understanding and modeling of both short-term weather events and long-term climate changes, scientists need improved measurements of water vapor at climate-relevant altitudes: the upper troposphere and the lower stratosphere. The troposphere is defined as the lowest layer of the atmosphere – up to an altitude of about 12 km. The stratosphere is the next layer, up to 50 km.

Several spectroscopic-based methods are in use to perform remote measurements using terrestrial systems at atmospheric observatories, as well as airborne instrumentation (planes, balloons). Two such systems have been developed and optimized on Mount Zugspitze, Germany's highest peak (2964 m, **Fig. 1**). Both a differential absorption lidar (DIAL) and a high-power Raman lidar were set up at the Schneefernerhaus research station, 300 meters below the summit.

DIAL is a laser-based method that compares backscatter intensity at two closely-spaced wavelengths which are either on- or off-resonance for a single near infrared water absorption line (817 nm) [1 – 3]. The system is based on a narrow-band Ti:sapphire laser system with pulse energy up to 250 mJ, and a 0.65 m diameter Newtonian telescope. The DIAL system has proved to be an excellent quantitative tool whose results are consistent with those of other methods. Also, unlike measurements based on solar absorption, it can be operated equally well during the day and at night. However, the practical altitude limit of this system is about 12 km.

Ultraviolet Raman lidar

To improve the range and sensitivity of water vapor measurements, other methods were considered. One laser-based method is Raman backscattering. This includes projecting a pulsed laser into the atmosphere and detecting Stokes-shifted Raman backscatter due to water rovibrational resonances. The signal is binned by time, which is then converted into distance (i. e., altitude). Raman lidar of water usually targets the Q branch of the O-H stretching vibration which is Stokes-shifted by 3654 cm^{-1} relative to the laser source.[1] Because the water concentration (mixing ratio) can only be a few parts per million above 12 km, signal-to-noise-ratio is a major challenge. Since the very weak Raman scattering intensity scales as $1/\lambda^4$, an ultraviolet wavelength is highly advantageous. Previous wide-range systems have all used the frequency-tripled output of Q-switched Nd:YAG lasers at 355 nm.

Various other factors influence the measured signal intensity including the efficiency of the signal collection optics and signal attenuation by ozone absorption. So in order to obtain quantitative measurements, it is typical to ratio the water signal to a simultaneous Raman signal for nitrogen, the major component of the atmosphere. This excludes the influence of instrumentation and provides, after calibration, the water mixing ratio. The strong nitrogen Q branch at a frequency shift of 2329.2 cm^{-1} is usually used for this purpose.

Commercial Nd:YAG lasers are limited to about 18 watts of power at 355 nm. Nonetheless, ground-based Raman lidar systems using these lasers have proved capable of quantitative water vapor measurements beyond 20 km altitude. However, this has required up to nine hours of signal averaging.

Modified industrial excimer laser

The most powerful laser source in the long wavelength ultraviolet spectral region is the XeCl excimer laser at 308 nm, so it was decided to develop a Raman lidar system based on this type of laser. While the shorter wavelength increases Raman signal-to-noise, this is unfortunately offset by increased atmospheric ozone absorption at 308 nm.

In the beginning of this project, the highest available power was from an excimer (Coherent LAMBDA SX[2]) delivering up to 1 Joule at repetition rates of up to 350 Hz. However, this laser was originally designed and optimized for industrial materials processing applications, such as annealing the silicon backplanes in flat-panel displays. So to perform quantitative Raman measurements over long distances, the beam and wavelength parameters required several modifications in order to obtain linearly polarized, narrow-line, frequency-stabilized output pulses with significantly reduced divergence.

The cavity was extended as shown in **Fig. 2**. A custom-made large intracavity etalon with 0.1 mm plate spacing and 70 mm diameter (SLS, specifications: $T = 0.9634$, $R = 0.55$) and a thin-film polarizer (Laseroptik, $T = 0.94$) were installed. In order to reduce the intensity incident on both the etalon and the polarizer, the narrow rectangular beam was expanded to a square cross-section shape (about 35×35 mm^2) with a cylindrical telescope. Apertures were also incorporated in order to avoid damage due to strong reflections or stray light. To allow automated tuning, and hence frequency stability, the etalon is mounted on a rotation stage that can be rotated manually or under computer control. However, this etalon is also the main source of loss that strongly grows at high repetition rates, limiting the output energy to about 180 watts (0.5 J at 350 Hz). This is a factor of about ten higher than powerful 355 nm Nd:YAG lasers.

The laser is operated on its high-frequency component (λ = 307.955 nm). All other contributions are very small. The measured spectral impurity can be as low as 0.5 %, but is mostly around 1 % at the highest repetition rates. The wavelength is controlled to within 0.025 nm by a calibrated grating spectrometer (Ocean Optics, model HR4000). Linear polarization of the radiation is important for achieving spectrally clean stimulated Raman shifting with high efficiency and for efficiently reducing the strong Cabannes components (purely rotational Raman) that are close to the Rayleigh backscattered signal. Insertion of the T = 94 % thin-film polarizer for this purpose delivers a degree of linear polarization of >99.5 % with an overall power loss of only 4 %.

The beam is then focused into a Raman shifter before being recollimated by a 10 m focal length concave

Raman Shifting

The illustration shows how the laser beam is focused into a Raman shifter that is 3.6 m in length, initially using an $f = 2$ m lens before final collimation. This Raman cell is filled with approximately 30 bar of hydrogen in order to generate a reference emission at 353.1 nm that is required for ozone data corrections. The long focal length was chosen to avoid radiation losses by optical breakdown as well as the generation of higher Stokes orders. At low repetition rates, a conversion up to 20 % was observed at 20 bar. For higher repetition rates, the conversion diminishes, since the pulse energy gradually decreases to about 0.5 J (which seems to be the threshold for Raman emission). Moreover, the generation of 353 nm radiation is extremely sensitive to the laser alignment, so the input lens has been reduced to a focal length of 1.75 m.

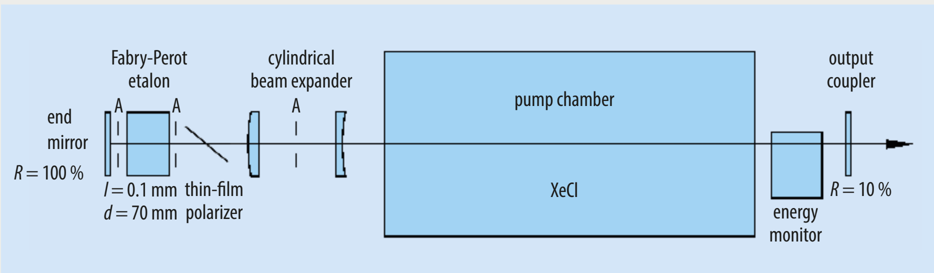

Fig. 2 Scheme of the modified laser system; A: sand-blasted aluminum apertures [4].

1) The Q branch is preferred because it consists of a narrow cluster of multiple overlapped rotational lines, providing a much stronger signal than a single, well-resolved rotational in the P or R branches.

2) Since this work was commenced, the 300-W LAMBDA SX model has been discontinued by Coherent and replaced by the more cost-effective 300-W LEAP laser.

mirror (**info box**). This results in a 5 times beam expansion (relative to the original laser cavity), which reduces beam divergence to less than 0.5 mrad. This is necessary for tight focusing of the backscattered light in the two receivers.

Receiver and signal processing

The signal intensity was predicted to span a nine-decade dynamic range. So, as in other Raman lidar systems, the decision was made to use two separate Newtonian receivers for near-field and far-field detection ($d = 0.38$ m, $f = 2$ m, and $d = 1.5$ m, $f = 5$ m, respectively). Because of the long focal length of the large mirror, the receivers are mounted in a tower on the terrace above the lidar laboratory, covered by a dome suitable for arctic conditions (Baader) – see **Fig. 1**. The large far field primary mirror (**Fig. 3**) has a 4 times larger area than an earlier 355 nm system. Together with the tenfold increase in laser power, this was expected to deliver a S/N increase by a factor of 40.

The radiation from both telescopes is focused into two six-channel polychromators created by a combination of polarization-sensitive optics and interference filters (Laseroptik and Materion Barr). The spectral widths are 0.25 nm (FWHM) for all channels except for the H_2O Q branch frequency, where a 0.75 nm filter with 70 % transmittance is used. The radiation is detected with Hamamatsu R7400 P-03 photomultiplier tubes (PMTs). To maximize the sensitivity of the far field signals, these are equipped with actively stabilized sockets enabling single-photon pulses without ringing (Romanski Sensors). The signal is processed by Licel 12-bit / 20 MHz transient digitizers together with a 5 GHz photon counting system (FAST ComTec).

Calibrated high quality H_2O data

As already mentioned, the water Raman signal is divided by the N_2 signal to obtain the water vapor mixing ratio. But this measured ratio also depends on different cross-sections for the two Raman signals, as well as different optical efficiencies and atmospheric attenuations at the two shifted wavelengths. In theory these could all be calculated. However, it is simpler and more reliable to calibrate the water Raman data using near-simultaneous data from the co-located DIAL system up to about 6 km.

Having both the Raman lidar and a DIAL system of proven calibrated accuracy [3] at the same location means this calibration can be achieved even under highly inhomogeneous conditions. **Fig. 4** shows a particularly demanding case involving three dry stratospheric layers where the agreement is close in spite of the over 15 minute delay between the two measurements.

Fig. 3 Far-field detection uses a 1.5 meter diameter primary mirror for efficient signal collection. Left to right: Thomas Trickl (KIT), Ulrich Emmerichs (Coherent), Petra Wallenta (Coherent), Hannes Vogelmann (KIT) – project meeting at mount Zugspitze (2.962 m). UV-Coherent-Laser successfully works there for climate research.

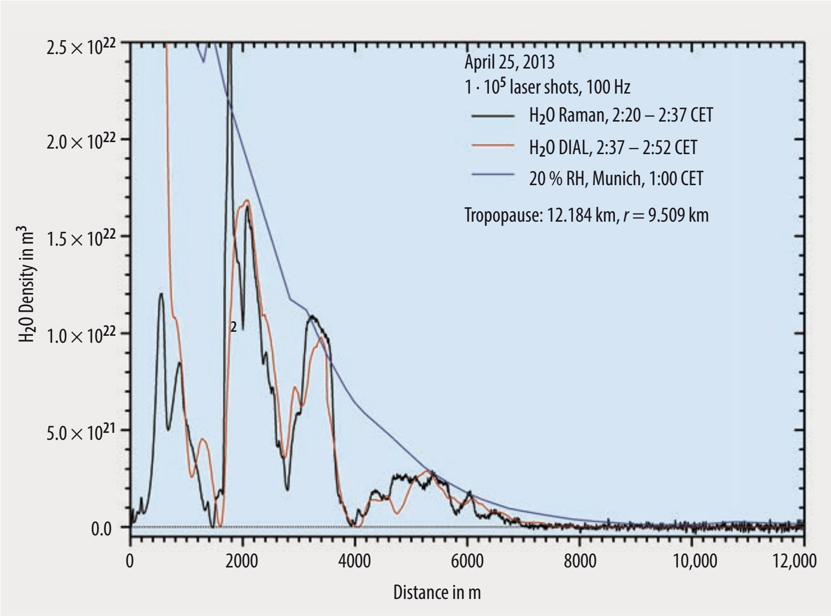

Fig. 4 Comparison of DIAL and Raman lidar measurements for a strongly layered H_2O distribution; "Distance" means vertical distance above the lab (2675 m a.s.l.); at two altitudes zero humidity was found by both systems [5]. The differences are explained mainly by atmospheric changes due to a time difference of roughly 15 min between the two measurements. For distances below 1 km the laser beam of the Raman lidar is not yet in the field of view of the telescope.

Summary

This Raman lidar system based on a modified industrial excimer laser can measure integrated water vapor to 18 km with a vertical resolution of 277 meters, and up to an incredible 22 km, with 1 km resolution. Up to 16 km, an uncertainty of the water vapor mixing ratio of 10 % and less with respect to the average stratospheric value of about 5 ppm has been demonstrated. And importantly, the integration times are typically only one hour instead of a full night for earlier 355-nm based systems. As a by-product, the system is expected to yield temperature measurements up to more than 80 km. The temperature will be derived in a standard procedure from the atmospheric density that governs the backscatter signals.

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