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# Capabilities of a Multipurpose Raman Lidar

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**Abstract.** The present paper reviews different applications of the Raman lidar technique and describes its capabilities by citing examples of measurements carried out with the GKSS Raman lidar in its latest state of development.

## 1. Introduction

When the lidar idea had come into existence, prompt inelastic scattering was among the first candidates for the basic effect to be used as the interaction of light with matter on which the extraction of information about remote parts of the atmosphere could be based. Well known as the Raman effect and well understood, it promised to lend itself to the measurement of concentration profiles of a number of gases because of several desirable characteristics, viz.,

- the gas-specific (Raman) shift of the return signal,
- the prompt appearance of the Raman radiation, and
- the low requirements as to laser wavelength,spectral width, and spectral stability.

There was a price to pay, though, because of

- the small magnitude of Raman as compared to elastic scattering cross sections: the Rayleigh backscatter cross section of $N_2$ alone (no aerosols present) is roughly 140 times larger than the $N_2$ vibrational-rotational Raman cross section; and
- the small differences of Raman cross sections of different gases, in view of the large difference in concentration between nitrogen and oxygen and most of the interesting gaseous species in the atmosphere: hardly ever are Raman cross sections of atmospheric gases more than a factor of 10 larger than that of the $N_2$ Q branch (Inaba 1976, Cherlow and Porto 1976, Schrötter and Klöckner 1979, Eliseev et al. 1982).

In spite of these problems, profiles of atmospheric gases were the first target of Raman lidar measurements. In the pioneering work by Cooney (1968) and Melfi et al. (1969) and by Inaba and Kobayasi (1969, 1972) it was demonstrated that water vapor and pollutant gases can in principle be determined with this method. Although at the time the available laser sources were too low-power ($N_2$) or too long-wavelength (Ruby) and certainly inadequate for routine work, moisture profiling has in the meantime developed into an important application of Raman lidar systems. To the authors' knowledge, pollution measurements with Raman lidars have, on the other hand, not gained widespead application partly because of the usually low concentrations of these gases even relative to water vapor, partly because of the need for multiwavelength discrimination of the return signals which are very much weaker than the Raman signals from $N_2$ and $O_2$, these being in turn several orders of magnitude smaller than the elastically scattered laser-wavelength return from air molecules and atmospheric aerosols.

Instead, it turned out that other features of the Raman effect could also very well be put to work in lidar systems. One is the fact that narrow-bandwidth Raman radiation is emitted by a compound only when in gaseous form, aerosols do not scatter the primary radiation into the wavelength window of one of the atmospheric gases. Another is the different Raman shifts from molecules in different rotational molecular states which, because of different occupation numbers at different temperatures, allows to infer the atmospheric temperature from differences in rotational Raman line intensities.

The Raman lidar designed and built at GKSS more than ten years ago and modified several times since followed the classical Raman lidar scheme only for two gases, carbon dioxide and water vapor, but allows to determine a number of other atmospheric parameters some of which cannot be profiled to date with any other means. In the present contribution this system is described in a state in which it allows to determine tropospheric water vapor, tropospheric ozone, stratospheric ozone, tropospheric temperature, stratospheric temperature, the coefficients of aerosol backscatter and aerosol extinction and the lidar ratio, and the thermodynamic phase of clouds. These parameters and their spatial and temporal variation often characterize cloud structures, especially those in the stratosphere, more completely than do in-situ measurements. They thus contribute substantially to our understanding of the underlying atmospheric processes.

In this paper the effects relevant for the determination of the different quantities are reviewed. The experimental setup is briefly sketched. System capabilities are demonstrated by desribing recent results, some of them unexpected or not observed previously. Results of model calculations necessary for the interpretation

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of measured data are presented, and improved Raman lidar schemes suited to further improve the performance of the system are discussed.

## 2. Measurement principle

### Carbon dioxide, water vapor

The original system had been designed to determine the lower concentration limit of a gas for practical, routine lidar work, and carbon dioxide appeared to be a substance suited for the purpose. $CO_2$ and $H_2O$ are both determined by dividing the respective Raman return signal by the Raman return from a reference gas. To minimize differences in atmospheric attenuation for the return from the gas to be measured and the reference, $O_2$ was chosen as a reference for $CO_2$, $N_2$ for $H_2O$. For calibration it turned out that a precise in-situ measurement with a received instrument provides better results than a derivation of the calibration factor from a determination of the properties of the different components of the lidar system.

### Ozone

Because of its low abundance and small Raman shift, ozone is not amenable to the same measurement scheme as the two other gases. $O_3$ is instead determined using the differential absorption lidar (or DIAL) technique (Schotland 1966); two lasers provide the necessary primary radiation. Gradients in atmospheric backscatter due to aerosols, however, cause large measurement uncertainties. Therefore nitrogen Raman returns are utilized instead of the elastic return signals from regions for which aerosol backscatter is not negligible. This Raman DIAL scheme (McGee et al. 1993) is often used in the troposphere and tropopause, the conventional DIAL technique with its larger signal intensities in the stratosphere up to a height of 50 km.

### Aerosol extinction and backscatter coefficients, lidar ratio

The coefficients of aerosol extinction and backscatter are obtained independently of each other and without model assumptions from the elastic and the $N_2$ or $O_2$ Raman backscatter signals. Whereas the elastic signal is determined by both backscatter and extinction properties of the aerosol, the Raman signal senses only the aerosol extinction (Ansmann et al. 1990, Ansmann et al. 1992b). The molecular contribution to extinction and elastic (Rayleigh) and inelastic (Raman) scattering must be known, and is known with sufficient accuracy to implement this scheme. The ratio of the extinction and backscatter coefficients, generally known as the lidar ratio, is indicative of aerosol microphysical parameters and allows, e.g., within certain limits the determination of the droplet mean square diameter if the optical constants of a liquid aerosol are known (Wandinger et al. 1995).

### Stratospheric temperature

Stratospheric temperature is deduced from the stratospheric density obtained from the elastic lidar return signal under the assumption that the atmosphere is in hydrostatic equilibrium. The resulting expression for the temperature requires integration from a reference altitude to the desired altitude; the temperature at the reference height must be known. The system works well if integration is done from a reference point at the remote end of the lidar range "downward" to the height of interest, the reference temperature taken from a model atmosphere is so much the less critical the lower the height of interest. Clearly, this is pure Rayleigh lidar and has nothing to do with Raman backscattering.

### Tropospheric temperature

The Rayleigh lidar scheme fails in the presence of aerosols or clouds. So even in thin clouds like subvisible cirrus and polar stratospheric clouds, another measurement principle is required. In these instances nitrogen Raman integration helps considerably. Below about 25 km, integration can be carried out down to the ground in very clear weather. Generally, however, Raman integration is limited to regions above the tropopause and with low aerosol load.

In the tropopause and below, purely rotational Raman scattering is used to advantage. Rotational Raman lines with low quantum number $J$ get less intense, high-$J$ lines get more intense as the temperature increases. So the ratio of the two groups of lines provides a direct measure of the temperature which, except for the small difference in extinction of the slighly different wavelengths, is independent of the presence of aerosol (Cooney 1972). If the anti-Stokes (short-wavelength) groups of rotational Raman lines are used, no perturbation by fluorescence must be faced, and excellent temperature profiles are obtained (Behrendt et al. 1998b, Behrendt and Reichardt 1999).

### Cloud particle phase

Liquid droplets are spherical, and spherical particles do not depolarize polarized laser light. Solid particles generally do. So from the depolarization of a polarized laser beam information about the thermodynamic phase or, in mixed-phase clouds, the mixing ratio can be drawn (Schotland et al. 1971).

## 3. System description

The GKSS Raman lidar in its present state of development is a two-laser, three-primary-wavelength, 13-receiver-channel system. The laser pulse energy and pulse repetition frequency data as well as the wavelengths and polarization data of the receiver channels are shown in Table 1.

The sensitive channels for the far-field signal are protected from the intense short-distance elastic returns by a mechanical chopper. The two-blade chopper rotates at 6 000 rpm, generating trigger signals for the lasers at a frequency of 200 Hz. The excimer laser is trigge-

Table 1. Receiver channels of GKSS Raman lidar

| Channel No. | 1 | 2 | 3 | 4 | 5 | 6 | 7 | 8 | 9 | 10 | 11 | 12 | 13 |
|---|---|---|---|---|---|---|---|---|---|---|---|---|---|
| Transmitter | XeCl excimer | | | Nd:YAG, 3rd harmonic | | | | | | Nd:YAG, 2nd harmonic | | | |
| Energy, PRF* | 170 mJ, 200 Hz | | | 350 mJ, 50 Hz | | | | | | 200 mJ, 50 Hz | | | |
| Wavelength, nm | 308 | 308 | 332 | 355 | 355 | 355 | 387 | 387 | 408 | 532.25 | 530.90 | 529.35 | 608 |
| Source† | EL | EL | $N_2$ R | EL | EL | EL | $N_2$ R | $N_2$ R | $H_2O$ R | EL | lo-$J$ RR | hi-$J$ RR | $N_2$ R |
| Near/Far | N | F | N+F | N | N | F | N+F | N+F | N+F | N+F | N+F | N+F | N+F |
| Polarization | laser not polarized | | | $\parallel$ | $\perp$ | $\parallel$ | $\parallel$ | $\perp$ | $\parallel$ | $\parallel + \perp$ | $\parallel + \perp$ | $\parallel + \perp$ | $\parallel + \perp$ |

* PRF, pulse repetition frequency

† EL elastic, R (Vibration-Rotation) Raman, RR rotational Raman

red by each one, the Nd:YAG laser by every fourth of these pulses.

The receiver optics consists of a 900-mm-diameter, f/2 parabolic mirror and an optical preselection unit near the primary focus which divides the atmospheric backscatter signal according to primary wavelength and polarization. Four fibers transmit the separated light to the receiver channels.

In the early days of the system a polychromator with an echelle grating and a holographic grating for order sorting was used. Today the polychromators work exclusively with filters. Clearly, the transmittance and blocking data of the filters are crucial for the proper functioning of the whole system. Details have been published along with its metamorphosis from a simple to an increasingly complex apparatus (Riebesell 1990, Ansmann et al. 1992a, Reichardt et al. 1994, Wandinger et al. 1994c, Reichardt et al. 1996b, Reichardt 1997).

To clarify the physical significance of the individual channels of the system, Table 2 gives a survey of the channels used for the determination of the different atmospheric quantities.

## 4. Performance and selected measurements

Instead of presenting a complicated list of performance parameters, the authors decided to demonstrate the capabilities of the system by presenting a number of measurements and examples.

### Cirrus geometrical and optical properties

Among the first applications of the system was the investigation of cirrus clouds. Cirrus enjoy particular interest among atmospheric scientists for a number of reasons. First, their contribution to the greenhouse effect can be either positive or negative, depending on the properties of the individual cloud. Second, cirrus occurrence depends to a certain extent on human activities. Numerous investigations have therefore been carried out with the Raman lidar in cirrus clouds (Ansmann et al. 1993a, Reichardt 1998a, Reichardt 1999).

Table 2. Survey of the quantities that can be measured with the GKSS Raman lidar, receiver channel numbers used, and measurement scheme applied

| Quantity of interest | Height range | Channel numbers used | Measurement scheme |
|---|---|---|---|
| Ozone molecule number density | 20 - 45 km | 2 and 6 | DIAL |
| | 5 - 27 km | 3 and 7 | Raman DIAL |
| | 5 - 20 km | 1, 4, and 5 | DIAL |
| Water vapor mixing ratio | 1 km - tropopause | 7 and 9 | VR* Raman lidar |
| Aerosol extinction coefficient | 4 km - UBSAL† | 7 | VR Raman lidar |
| Aerosol backscatter coefficient | 4 km - UBSAL | 4, 5, and 7 | VR Raman lidar |
| Lidar ratio | 4 km - UBSAL | 4, 5, and 7 | VR Raman lidar |
| Depolarization ratio | 4 km - UBSAL | 4 and 5 | polarization lidar |
| Temperature | UBSAL - 70 km | 6 | Rayleigh integration lidar |
| | tropopause - 25 km | 7 | Raman integration lidar |
| | 5 - 40 km with aerosols | 11 and 12 | rotational Raman lidar |

* VR vibration-rotation

† UBSAL upper boundary of stratospheric aerosol layer

### Cirrus and ozone

Third, but not least, cirrus, with their conspicuous anticorrelation of ozone within their geometric boundaries, may have a much more important effect on the ozone budget than is presently recognized. In fact, lidar measurements provided one evidence for this important phenomenon. Although the fraction of the ozone column that is at any moment affected by the presence of cirrus is only 3 %, the effect on the Earth's radiation budget may be quite important (Reichardt et al. 1996a, Reichardt 1997, Reichardt and Weitkamp 1997).

### Volcanic stratospheric aerosol

With the eruption of the Philippine volcano Mount Pinatubo in June 1991 large amounts of sulfur dioxide were projected into the stratosphere which in a period of weeks formed large stratospheric clouds that consisted of droplets of sulfuric acid. The stratospheric aerosol appeared over Central Europe as soon as August 1991 and was then measured in regular time intervals. From its height disitribution the temporal behavior of the boundaries of the voclanic cloud, of its center of gravity, of the radiative characteristics and of such important microphysical properties as the mean square particle diameter and specific particle surface were deduced which directly determine the chemical interaction potential with many gases including ozone (Ansmann et al. 1993b, Wandinger et al. 1995, Ansmann et al. 1996, Ansmann et al. 1997).

### Polar stratospheric clouds

The GKSS Raman lidar was the first instrument to detect polar stratospheric clouds (PSCs) from the North polar vortex as far south as 53.5°. The appearance of the vortex over Geesthacht (53° 24' N, 10° 26' E) on 4 and 5 March 1996 was accompanied by a drastic reduction of stratospheric ozone which, in the height interval between 15 and 30 km, went from 280 Dobson units (DU) before to 140 DU during and 260 DU after the PSC event (cf. Serwazi et al. 1996a for a preliminary evaluation).

PSCs, in particular those generated by leewave effects behind the Scandinavian Mountain Range, were also the subject of more recent investigations in the Arctic (Fierli et al. 1998, Fricke et al. 1998, Mehrtens and Reichardt 1998). The characterization of PSCs according to type and composition (Behrendt et al. 1998a) and the observation of dynamical processes such as the change of PSC characteristics on sunrise (Reichardt et al. 1998d) render Raman lidars that offer temperature profiling capabilities highly desirable.

### Carbon dioxide

One of the purposes for which the original system was built was the determination of the concentration limit down to which the Raman lidar scheme could be used on a routine basis. $CO_2$, with a natural abundance of 300 ppm by volume, appeared to be a suitable, though not an easy, candidate. The $\nu_1$ vibrational band is totally buried by the molecular oxygen O branch. The $2\nu_2$ band is still superimposed by the high-$J$ wing of $O_2$ O. With its cross section even smaller than that of the oxygen Q branch and the small Raman shift of 1285 $cm^{-1}$, it additionally suffers from the incomplete suppression of the elastic return signal. Nevertheless, these problems were all solved. The Rayleigh extinction correction for the differences in primary and Raman wavelength is almost linear in altitude and amounts to 1.5 % at a height of 5 km. The aerosol extinction correction varies, but can also be corrected for. What remains is that part of the background under the $CO_2$ $2\nu_2$ band that shows strong day-to-day variations and which we attribute to fluorescence from organic molecules. If $CO_2$ were an ordinary pollutant for which an accurtacy of 10 % is adequate, its profiling with Raman could be carried out without major problems. Because of its small variation, however, a precision 100 times better is required. It must thus be concluded that a the present time the measuremnent of $CO_2$ is not possible with the Raman lidar technique (Riebesell 1990).

### Water vapor

Water vapor can well be measured up to the top of the troposphere, even in the presence of cirrus clouds (Serwazi et al. 1994, Reichardt et al. 1996b). Simultaneously taken lidar temperature profiles are particularly useful for the presentation of the water vapor profiles in terms of relative humidity because they eliminate the need for data from a radiosonde ascent.

### Investigations into the Klett inversion method

With a means to determine the aerosol extinction and aerosol backscatter coefficient profiles separately and with no other a-priori assumptions than the validity of actual atmosphere models, it is possible to assess the quality of current algorithms for the inversion of signals from simple backscatter lidars. These investigations have shown that the current assumption that extinction-to-backscatter ratios of naturally occurring aerosols and clouds are constant at least within one cloud and that certain values taken from experience can be used for the inversion in a resonably large number of cases is totally inadequate (Ansmann et al. 1991a). It could be shown, however, that an iterative method of a modified Klett algorithm involving alternating forward and backward integration yields, after relatively few iteration steps, the correct *average* lidar ratio; the distribution of lidar ratios, may, however, come out totally wrong (Ansmann et al. 1991b).

### Participation in international measurement campaigns

More information about the possibilities the GKSS Raman lidar has proven to offer is available from the topical literature and final reports on the international programs in which it has participated. The programs include the International Cirrus Experiment 1989 (ICE'89), the Experimental Cloud Lidar Pilot Study (ECLIPS, Platt et al. 1994), the Lidar-In-Space-Technology Experiment (LITE, Serwazi et al. 1996b), the Lindenberg Complex Terrain - Fluxes between Atmosphere and Surface - a Long-Term Study (LITFASS-98), the European Leewave Project 1996-1998 and the Project POLECAT (**Po**lar stratospheric clouds, **Le**ewaves, **C**hemistry, **A**erosols and **T**ransport) 1996-1998.

## 5. Other Raman lidars at GKSS or with GKSS participation

Although the GKSS Raman lidar absorbed most of the Raman lidar activity at GKSS, it is not the only device that helped GKSS staff gain experience with Raman lidar systems.

### ATLAS

Another system is the **A**dvanced **T**emperature-Humidity **L**idar for **A**tmospheric **S**tudies (ATLAS). ATLAS was designed to measure moisture using the vibration-rotation Raman returns from water vapor and nitrogen and temperature with the rotational-Raman technique. Whereas the GKSS Raman lidar performs best at night when the background from the sky is greatly reduced, ATLAS works equally well in bright sunlight. On the other hand, ATLAS is a short-range system designed essentially for the lower troposphere (Lahmann et al. 1996, Zeyn et al. 1996).

ATLAS transmits radiation of a krypton fluoride excimer laser shifted in a hydrogen Raman cell from 248 to 277 nm. A special optical configuration was developed for the laser to reduce the fraction of amplified spontaneous emission from 8.1 % to 0.005 % (Luckow et al. 1994). The laser is fine-tuned to transmit, after frequency shifting, radiation of a wavelength of 276.787 nm. A thallium atomic vapor filter that absorbs exaxtly at this wavelength is used to keep the intense elastic backscatter signal off the receiver polychromator (Heimrath 1992). The important data of the system are given in Table 3.

### AWI lidar

GKSS also participated in the design, construction and test measurements of a mobile modular Raman lidar system for the Alfred-Wegener-Institut für Polar- und Meeresforschung (AWI) Bremerhaven. The system transmits frequency-doubled and frequency-tripled radiation from a Nd:YAG laser. Of its ten receiver channels four register the perpendicular-polarized and the short and long-distance parallel-polarized elastic as well as the $N_2$ Raman return for the longer of the primary wavelengths, five more channels the same plus the $H_2O$ return for the shorter wavelength, and the tenth channel provides an option for the 1064 nm elastic return signal. The system which was designed chiefly for aerosol work took part in a great number of campaigns and performed to full satisfaction (Schäfer et al. 1996).

Table 3. ATLAS technical data

| | |
|---|---|
| Laser: | |
| Type and wavelength | KrF excimer, 248-249 nm |
| Energy, PRF | 300 mJ, 250 Hz |
| Divergence | 0.3 mrad x 1.6 mrad |
| Raman shifter: | |
| Medium | hydrogen @ 1.5 bar |
| Optical length | 1 m |
| Lenses focal length | 1 m |
| Power efficiency | 25 % |
| Beam expansion | 8x |
| Receiver optics | Cassegrain 600 mm, f/3.3 |
| Dispersion system for rotational Raman subsystem: | |
| Type | echelle grating in Czerny-Turner mount |
| Number of channels | 4 (Stokes and anti-Stokes high-*J* and low-*J*) |
| Linear dispersion | 9 mm/nm |
| Free spectral range | 14 nm |
| Dispersion system for vibration-rotation subsystem: | |
| Type | filter |
| Number of channels | 4 (elastic, Raman $N_2$, $O_2$, $H_2O$) |
| Photomultipliers | Thorn EMI 9893 QB 350 |
| Data acquisition system: | |
| Number of channels | 12 |
| Maximum frequency | 300 MHz |
| Minimum dwell time | 100 ns |

### Rotation - Vibration-Rotation (RVR) Raman lidar

The Raman DIAL technique, although far superior to conventional DIAL in the presence even of small amounts of aerosol, suffers from residual uncertainties caused by differences in atmospheric attenuation and multiple-scattering effects on the two wavelengths. This uncertainty can be substantial in thick water and ice clouds.

To overcome this problem a novel technique has been developed that reduces these uncertainties and eliminates the need for a second laser. The approach is based on differential absorption by the gas of interest of the purely rotational Raman return signals from nitrogen and oxygen as the on-resonance and the vibrational-rotational Raman return from nitrogen or oxygen as the off-resonance wavelength. Because of this combination, rotational - vibrational-rotational, or RVR, Raman DIAL might be an appropriate expression for the new method. If the target is ozone, then the method is superior to the more obvious use of the $O_2$ vibration-rotation Raman return for the on and the $N_2$ vibration-rotation Raman return for the off-line signal because the primary wavelength need not be so much shorter than both signal wavelengths; this results in a considerably longer range.

The viability of the approach has been proven with a xenon chloride excimer laser of only 5 W average power and a 760-mm-diameter, f/4.5 Cassegrain telescope in the trailer of the Sandia Livermore water-vapor Raman lidar. Excellent ozone profiles were obtained between 3 and 21 km height. The measurement time was 220 minutes; with a 100-W average power laser ozone profiles from the ground to the middle stratosphere can thus be obtained in less than half an hour (Reichardt et al. 1997, Reichardt et al. 1998b, Reichardt et al. 1998c).

## 6. Simulations

For the understanding of the results of existing and the planning of improved Raman lidar systems theoretical and experimental work must proceed simultaneously. Scattering phase functions, polarization effects and multiple-scattering processes are the fields in which theoretical studies are most urgently needed. Simulations have therefore been carried out in both these fields and contributed significantly to the interpretation of measured phenomena (Wandinger 1994, Wandinger et al. 1994a, Wandinger et al. 1994b, Wandinger et al. 1994c, Reichardt 1997, Reichardt et al. 1999).

## 7. Conclusions and further development

Raman lidar is one of the powerful variants of the lidar principle. Raman lidar systems offer the possibility to measure several quantities simultaneously that together provide a much more comprehensive description of the state of the atmosphere than each of them alone.

The inherent drawbacks, all associated with the small Raman cross sections, are made up for by a multitude of desirable properties the most important of which is probably the missing backscatter contribution of aerosols to the Raman return signal. Raman lidars require careful design and a certain financial effort, but are easy to operate because of the relatively uncritical requirements as to wavelength stability and spectral width.

Future developments will probably put more emphasis on new concepts such as Raman DIAL and RVR Raman DIAL. The addition of a liquid-water channel is an important goal for applications in cloud physics. The simplification of experimental setups for increased mobility and eventual use from airborne and spaceborne platforms, though highly desirable, will, however, not easily be realized in the near future.

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