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C. Russell Philbrick, "Raman lidar measurements of atmospheric properties," Proc. SPIE 2222, Atmospheric Propagation and Remote Sensing III, (29 June 1994); doi: 10.1117/12.177985



Event: SPIE's International Symposium on Optical Engineering and Photonics in Aerospace Sensing, 1994, Orlando, FL, United States

## Raman lidar measurements of atmospheric properties

C. R. Philbrick The Pennsylvania State University Department of Electrical Engineering and Applied Research Laboratory University Park, Pennsylvania 16802 (814)865-2975 Fax:: (814)863-8457 E-mail: crp@ecl.psu.edu

### ABSTRACT

The capability of Raman lidar techniques to make accurate measurements of the structure properties and species of the atmosphere has been investigated. The LAMP lidar which was developed at PSU during the past several years has focused on the application of Raman vibrational and rotational scattering results. Measurements have been carried out during several campaign periods which demonstrate the performance of Raman lidar techniques compared to standard rawinsonde balloon payload measurements. The investigation has included water vapor and molecular nitrogen profiles determined from the 1st Stokes vibrational Raman transitions from laser wavelengths of 532 nm, 355 nm and 266 nm. The profiles of the N<sub>2</sub> vibrational Raman scatter provide true extinction measurements in the lower atmosphere. Water vapor profiles are determined from the ratio of signals measured at the following wavelength pairs: 660/607, 407/387 and 294/283. The fact that the profile is determined from a signal ratio removes most of the factors which would result in errors in the profiles. The temperature structure has been measured using the rotational Raman scattering in the region between 526 and 532 nm. Measurements have been carried out to evaluate the performance and show the capability of Raman lidar to measure the profiles of atmospheric structure properties and water vapor in the lower atmosphere during night conditions and to determine the daytime measurement capability.

### **1. INTRODUCTION**

During the past twenty years, researchers at several laboratories have demonstrated that lidar has special capabilities for remote sensing of many different properties of the atmosphere. One of the techniques which shows a great deal of promise for several applications is Raman scattering. In this presentation, our application of the Raman scattering techniques to obtain profiles of water vapor and temperature in the lower atmosphere is described. The first Raman measurements of atmospheric properties with lidar were carried out in the late 1960's by Leonard<sup>1</sup> and Cooney<sup>2</sup>. Two years later, Melfi, et al.<sup>3</sup> and Cooney<sup>4,5</sup> showed that it was possible to measure water vapor using the Raman lidar technique. In 1972, a significant contribution was made by Inaba and Kobayasi<sup>6</sup> in suggesting several species that could potentially be measured using vibrational Raman techniques. While the early tests<sup>3,4,5,7</sup> showed that it was possible to measure the water vapor with limited range and accuracy, recent investigations have shown significant improvements. Particularly, the investigations of Vaughan, et al.<sup>8</sup>, Melfi, et al.<sup>9</sup>, and Whiteman, et al.<sup>10</sup> have demonstrated rather convincingly that the Raman technique has a high potential for making accurate water vapor measurements. A most useful review of the Raman and DIAL lidar techniques applied to the water vapor measurement has been given by Grant.<sup>11</sup> The measurements of water vapor during the daytime have been demonstrated by Renaut and Capitini<sup>12</sup> using the solar blind region of the ultraviolet spectrum. Their work showed that the optimum wavelength for the measurement was near the fourth harmonic for the Nd: YAG laser. At this wavelength, the measurements of  $N_2$  and  $H_2O$  are contaminated, at least to a small degree, by the absorption of ozone and  $SO_2$ in the lower troposphere, however it appears that an adequate correction can be obtained from the use of the measured Raman signals of the  $N_2$  and  $O_2$  compared to the known mixing ratio.

At tropospheric altitudes, the Raman  $N_2$  profile together with the multi-wavelength backscatter should allow the separation of the extinction, backscatter due to particles and the molecular backscatter signals. The advantage in using the Raman signals in the lower atmosphere is clear from the profiles shown above. Figure 1 shows a representation of the spectral signatures which would be expected from the back-scatter due to the 532 nm laser radiation in an atmospheric volume.<sup>6,13</sup> The laser is injection seeded to give a line-width of about 80 MHz and thus the particle back-scatter is of that spectral width, while the molecular peaks are broadened by the thermal Doppler velocity spreading. The vibrational Raman scattering peaks are shown for  $O_2$ ,  $N_2$  and  $H_2O$ , and around each of the peaks is also shown the broadening due to the rotational state distribution around each vibrational state. Only the first Stokes vibrational states are indicated, since the simple molecules have large vibrational energy state separation and the anti-Stokes lines are not normally populated. The figure shows the large cross-section differences between the processes involved. The Raman H<sub>2</sub>O signal measured as a ratio to the Raman N<sub>2</sub> signal provides a profile which is proportional to the water vapor concentration. The N<sub>2</sub> fraction of the atmospheric profile is known, and the atmospheric profile can be obtained from the temperature profile combined with a surface pressure value. The error caused by the extinction differences between the backscatter wavelengths is small (few percent) and can be corrected using the results from multiple wavelengths.



Figure 1. Descriptive representation of the vibrational and rotational Raman signals expected for radiation of an atmospheric volume with 532 nm laser.

While demonstrations by several researchers have shown the potential of lidar to measure many properties of the atmosphere, there have been few efforts to develop the lidar techniques sufficiently for them to be used for routine measurements. The research lidars generally require attentive interaction by highly specialized personnel to obtain useful measurements and thus the investigations have been generally limited to short and intensive measurement periods. The transition of the technical capability of lidar to operational applications in meteorological data collection, atmospheric physics investigations, studies of the environment, investigations of radiative transport and global climate analysis requires that instrumentation be improved and automated. All of the properties measured by current rawinsonde balloons and meteorological rockets can be measured by lidar remote sensing techniques which have been demonstrated to varying degrees in research laboratories. Lidar will probably not fully replace the need for conventional balloon and rocket techniques because there are optical extinction conditions which limit lidar, but there are also many factors which limit the use of current techniques, i.e. wind conditions that can prevent the release of balloons or the launch of meteorological rockets. The current established techniques require the use of expendable hardware which is manpower intensive and the spent package results in a degree of pollution that is not a problem for lidar applications. The initial expense of lidar sites is high compared to current technology, however the cost difference could be easily amortized within a few years. The lidar techniques have unique advantages in providing high resolution measurements in both time and space. One of our research goals is to make lidar a useful instrument for routine measurement applications within the next few years. Toward that goal, we have sought to perfect the techniques, and to automate the operation and data retrieval from lidar. The multi-wavelength Raman lidar techniques have been chosen to provide the profiles of density, temperature, water vapor and optical extinction in the troposphere because they appear to have the best capability for meeting the measurement requirements. The additional parameter required is wind velocity and several laboratories are currently working on both direct and coherent detection techniques for wind velocity profiles.

The LAMP lidar development was originally focused on the question of using two-wavelength measurements to define those regions where the backscatter profile represents the molecular profile. In those regions, the relative density profile can be used for measurements of the temperature structure in the stratosphere. With the LAMP instrument, the possibility of using the vibrational  $N_2$  Raman profile for temperature measurements in the regions contaminated by particle and aerosol scattering has been investigated. The investigation of Rau<sup>14</sup> has shown that the  $N_2$  Raman profile can be used for temperature determinations in some cases. However, in the presence of layers of particles and aerosols in the lower stratosphere and troposphere, its use is very limited. The necessary corrections for the extinction can only be applied when no significant cloud scattering layers are present and when simplifying assumptions about the particle scattering can be made. In several selected clear weather cases, he was able to calculate satisfactory temperature profiles in the lower stratosphere and troposphere. However, the severe restrictions imposed upon the technique by scattering layers or changing the type or size of the scattering particles make it impractical for use in typical atmospheric conditions.

During the past two years, we have focused the LAMP lidar efforts on demonstration the capability of vibrational Raman techniques for water vapor measurements and on the use of rotational Raman for temperature measurements, in the presence of a background of aerosols and clouds.

The rotational Raman technique for temperature measurements was reported by  $Cooney^{13}$  in 1972. A double grating monochrometer was used by Arshinov, et al.<sup>15</sup> to measure the rotational Raman spectrum in 1983. Hauchecorne, et al.<sup>16</sup> and Nedeljkovic, et al.<sup>17</sup> demonstrated the capability to measure the temperature using narrow-band filter technology in the upper troposphere and lower stratosphere. Figure 2 shows the way that we are using the rotational Raman signal for temperature profile determination. We are making use of the rotational Raman envelop of several lines which pass through a narrow band filter. The molecular species of the atmosphere, principally N<sub>2</sub>, O<sub>2</sub> and to some degree H<sub>2</sub>O, contribute to the envelop of lines on either side of fundamental laser wavelength. In our case, we measure the lines in the rotational states at the 530 and 528 nm filter bands. The envelop shape is determined by the population of the rotational states under the temperature distribution of the gas in the volume, which is illuminated by the doubled Nd: YAG laser at 532 nm. There are envelops on both the long and short wavelength sides of the fundamental exciting frequency, however we have chosen to work on the short wavelength, higher energy, side of the distribution in case there is any excitation from fluorescent transitions. In Figure 2, the ratio between the intensities of the two filter bands is indicated as a way of directly determining the temperature. While this curve is based upon a calculation, we use a method of comparison of a rawinsonde balloon measurements with the measured results to develop a calculation curve based upon that empherical fit. With the measured temperature profile and a ground based measurement of the surface pressure, the profiles of the ideal gas law. The calculated profile of density can be used to obtain the N<sub>2</sub> profile to place an absolute density on the water vapor from the ratio of vibrational Raman signals.



Figure 2. Representation of the use of the rotational Raman spectra for measurements of the temperature profiles in the troposphere.

The first major field experiment using the LAMP lidar was the LADIMAS (<u>LA</u>titudinal <u>DI</u>stribution of <u>Middle A</u>tmosphere <u>S</u>tructure) campaign<sup>18-19</sup>. The results from the LADIMAS experiment<sup>18-26</sup> have provided a unique data set to improve our understanding of the middle atmosphere properties. The project included coordinated ship-board measurements between 70°N to 65°S and measurements at the Andoya rocket range to study the structure, dynamics and chemistry of the atmosphere. Results on dynamical processes, such as gravity waves, as well as, the formation of the layers of meteoric ion and neutral species, have been obtained using lidars, digisonde, microwave radiometer, and spectrometers. The cooperative study of the atmosphere was undertaken by researchers from several laboratories, including Penn State University, University Bonn, University Wuppertal, Lowell University, and others. Instruments were assembled aboard the German research vessel RV *Polarstern* while this vessel was sailing from the Arctic to the Antarctic between October 8, 1991 and January 2, 1992. Some of the results on atmospheric properties which have resulted have been published in several reports and theses.<sup>18-26</sup>

The second major field investigation included a set of three measurement periods using the LAMP lidar at NAWC Point Mugu, CA, during the period July-November 1993. This location provided the opportunity to measure the properties of the marine costal environment at a time of concentrated atmospheric measurements associated with a program called VOCAR (Variability of Coastal Atmospheric Refractivity). In addition, the instrument has been used for extensive testing at Penn State University while not involved in these field experiments.

### 2. LAMP INSTRUMENT DESCRIPTION

The LAMP (Lidar Atmospheric Measurements Program) lidar profiler was placed in service at Penn State University during the summer of 1991. The LAMP lidar uses two wavelengths in the upward propagating beam and up to eight detectors in the receiver. The instrument is arranged in a coaxial configuration, which permits useful measurements in the near field, as well as in the far field, see Figure 3. Two detector systems have been prepared for the instrument, one for high altitude measurements and one which is specialized on the low altitude region. The high altitude detector system is used to obtain data between 1 km and 80 km. It uses a mechanical shutter to block the high intensity, low altitude signal, from the two high altitude detectors until the beam has reached an altitude of 18 km. The low altitude detector has the capability of measuring two N<sub>2</sub> and two H<sub>2</sub>O vibrational Raman channels simultaneously and measuring two wavelengths of the rotational Raman spectrum for temperature determination. The detector was originally designed to directly attach the telescope by an optical transfer relay, however, the current use of an optical fiber permits the use of either detector. The Nd: YAG laser has an output of 1.5 j at 20 Hz at the 1064 nm fundamental output. The beam is passed through a doubling crystal and a mixing crystal to produce the 532 and 355 nm, or 266 nm, beams which are used for the lidar measurements. The primary receiver is a 42 cm diameter Cassegrainian telescope. The low altitude backscatter signals of the visible and ultraviolet beams can be detected as analog signals and digitized at 10 MSps to provide 15 meter resolution from the surface to 25 km, or detected as photon count signals with 75 m altitude resolution. The high altitude signals, obtained by photon counting techniques, are accumulated into 500 nanosecond range bins to provide 75 meter resolution, from 20 to 80 km. The detector also contains low altitude photon counting channels which measure the first Stokes vibrational Raman signals of the N<sub>2</sub> and the H<sub>2</sub>O Raman scatter. The transmitter, receiver, detector, and data system combination have been integrated into a standard shipping container, which serves as a field laboratory. We have investigated the possibility of measuring the temperature profile in the turbid lower atmosphere using the  $N_2$  vibrational Raman signal. However, this approach has proven to be quite limited due to the accuracy with which the extinction can be determined when cloud layers are present.<sup>14</sup> However, measurements of the rotational Raman backscatter have proven to be the appropriate way to measure the temperature profiles in the presence of clouds, aerosols, and in the boundary layer.



Figure 3. A diagram of the LAMP lidar shows the laser on the bottom of the table, the detector on the middle shelf, and the receiving telescope at the top with the beam steering mirror.

### 3. MEASUREMENTS

Figure 4 shows an example of the raw lidar signals, corrected for  $1/R^2$  dependance, which are typical of the signals measured on several of the data channels. The profiles show the signals from boundary layer aerosols, tropospheric clouds, high cirrus clouds and stratospheric aerosol lavers. In this case, the low altitude channels for 532 and 355 nm receive about 5% of the collected signal intensity and the measurements are made in analog mode with an A/D converter at 10 MHz (15 meter altitude steps) with 12 bit resolution. The high altitude channels are mechanically shuttered below 15 km to prevent the PMT's from being saturated by the large signal received at low altitudes. The high altitude channels and the Raman channels for N<sub>2</sub>, at 607 nm, and for H<sub>2</sub>O, at 660 nm, use photon counting detectors, with range bins of 500 nanoseconds (75 meter altitude steps). The back-scatter and extinction associated with the stratospheric aerosols, clouds and the boundary layer can be readily observed in the profiles at these two wavelengths. Notice that the scattering ratio of the 532 nm compared to the 355 nm changes significantly with the changing size of the particle scatterers. When the stratospheric aerosol scattering intensities are compared to those for the tropospheric clouds, the change in extinction and back-scatter cross-section with particle size is obvious. One of the more important observations from the LADIMAS results is the significance of the two-wavelength lidar approach in defining the particle scattering layers of the atmosphere, particularly the lower stratospheric aerosol and particle layer. The boundary where the high altitude signal, above about 30 km, can be analyzed to provide density and temperature profiles can be identified using the two-wavelength approach. The LADIMAS measurements have allowed the preparation of a map<sup>20,22</sup> of the Pinatubo volcano aerosol distribution and an investigation of the latitudinal variation in the density and temperature structure<sup>21</sup> of the middle atmosphere. Studies of the distribution of tropospheric water vapor and tropospheric aerosols, and an investigation of the optical scattering properties of the atmosphere have also been carried out using the results.<sup>23-26</sup>



Figure 4. Examples of the raw signals, corrected for  $1/R^2$ , for the low and high altitude channels as a profile. The slope change below 1 km is due to telescope focus and geometrical factors. The profiles of the particle backscatter intensity at the two wavelengths are compared by using a reference model for the molecular scattering component tied to the profile at 40 km.

The two-wavelength lidar shows a large difference in relative backscatter intensity from the stratospheric aerosols, primarily due to the smaller molecular scatter component at 532 nm, but also due to the fact that the stratospheric aerosols are sufficiently small that the wavelength dependence of scattering is significant. The results indicate that the particle size is about 0.7  $\mu$ , comparable to the wavelengths of the lidar. When the molecular scattering component is removed and the particle scattering is examined, the size

distribution of stratospheric aerosols is found to be remarkably stable, exhibiting a uniform ratio in the scattering intensity for the two wavelengths, see Figure 4 between 15 and 22 km. The two-wavelength lidar backscatter is important for determining the minimum altitude where a molecular profile analysis is applicable, other authors have presented single wavelength cases with invalid results. The extinction profiles for the stratospheric aerosols were calculated from the backscatter intensities using standard inversion proceedures.<sup>14, 24</sup> However, the backscatter inversion techniques have not been found to be satisfactory for cloud layers in the troposphere. The results show that the particle scatterers in the stratosphere are of uniform size and optical character, which is not the case for tropospheric clouds. The backscatter inversion calculations require a uniform particle distribution, which does not generally apply. However, the N<sub>2</sub> Raman signal appears to provide an adequate way of measuring the optical extinction.

Examples of the results from the Raman measurements of water vapor during the August 1993 field program at Point Mugu, CA, are shown in Figure 5. The results of Figure 5 show the comparisons of near simultaneous lidar measurements and rawinsonde balloon measurements on 18 and 31 August 1993. The bar shows the one standard deviation range for the signal statistics. Figure 6 shows an example of the results from simultaneous lidar measurements at two wavelengths. The  $H_2O$  and  $N_2$  Raman signals, from scattering of the 532 and 355 nm laser radiation, at the 660/607 and the 407/387 have been used to examine the small scale structure of the water vapor profiles. Preliminary results from the 266 nm lidar measurements using the solar blind portion of the spectrum are shown in Figure 7. The ratio of the 295/284 can be used to determine the water vapor concentration without significant interference from the solar radiation scattered background during the daytime. One correction to be made to these results is the absorption of ozone and other minor species in the troposphere, which can require a 10-20% correction at 1 km altitude. The  $N_2$  (284 nm) and  $O_2$  (277 nm) Raman signals can be used to determine the correction since the proper ratio of these signals is known. One advantage of the lidar technique is the opportunity to follow the changing atmospheric properties with time resolution steps as short as one minute. Figure 8 shows the time variations with profiles separated by four minutes. The top of the boundary, near 2.2 km, and the dry and moist layers can be observed and followed. In this case, a moist layer is observed near 3.7 km and is moving to lower altitude during this period.

An example of the rotation Raman scattering signal used to measure the temperature is shown in Figure 9. The measured signals in filter bands near 530 and 528 nm can be formed into a ratio which is proportional to the temperature. The rotational Raman technique for temperature does not depend upon profile integration, as in the case of the vibrational Raman  $N_2$  approach. It appears to perform well in the presence of aerosols and clouds, as long as the signal strength after extinction permits an analysis to be carried out.



Figure 5. Examples of the water vapor concentration obtained from the Raman lidar signals, with one sigma error shown, compared with a rawinsonde balloon profiles.



Figure 6. The measurements obtained from two laser wavelengths at the same time have been investigated. The 660/607 signals are from the 532 nm laser radiation and the 407/387 signals are from the 355 nm laser radiation.



Figure 7. The water vapor profile determined from the 266 nm laser radiation requires a correction for the absorption of tropospheric ozone and minor species. The technique has the capability of measuring the water vapor during the daytime.



Figure 8. The time sequence of water vapor distribution is shown as a gray scale based upon profiles of water vapor mixing ratio analyzed each 4 minutes. Notice the boundary layer demarcation and the moist layer near 3.7 km.



Figure 9. The rotational Raman signals at 530 and 528 nm provide the opportunity to determine the atmospheric temperature profile. In this case, the standard deviation error and a balloon comparison are shown.

The studies have shown the importance of the two-wavelength approach in identifying the regions appropriate for analysis of the structure properties from the molecular profiles. The  $N_2$  Raman signal has proven to be the useful measurement for atmospheric extinction. The Raman lidar technique has been found to provide water vapor mixing ratio profiles which appear to be as good and any other technique available today, and with the added advantages of high time resolution, inexpensive profiles, and non-polluting operation. The rotational Raman technique is proving to be the best approach for measurements of temperature in the troposphere. Good comparisons of the lidar and standard techniques have been found. Improved measurements of the properties of the atmosphere are required for meteorological forecasting, operational test support, and for studies of the global environment. The long term goal of these efforts is to develop a lidar instrument which can provide routine measurements of the atmospheric properties.

### 4. ACKNOWLEDGEMENTS

The preparation of the LAMP instrument and these measurements have been supported by PSU/ARL project initiation funds, PSU College of Engineering, the Navy's Environmental Systems Program Office (SPAWAR PMW-165), the Naval Command Control and Ocean Surveillance Center (Code 54) and the National Science Foundation's CEDAR (Coupling Energetics and Dynamics of Atmospheric Regions) Program. The measurements on the RV *Polarstern* were made possible by invitation of the Alfred-Wegener-Institut which is gratefully acknowledged. The efforts of Professors O. Schrems and U. von Zahn in organizing the voyage are appreciated. The efforts of D. B. Lysak, Jr., R. E. Erdley, D. E. Upshaw, T. W. Collins, T. J. Kane, J. Jenness, T. Petach, T. D. Stevens, P. A. T. Haris, Y.-C. Rau, S. Rajan, B. Mathason, M. O'Brien, S. C. McKinley, S. Maruvada, G. Evanisko, G. Pancoast and E. Harpster have contributed much to the success of this project.

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