Barium Nitrate Raman Laser Development for Remote Sensing of Ozone

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Introduction

In order to understand the impact of anthropogenic emissions upon the earth’s environment, scientists require remote sensing techniques which are capable of providing range-resolved measurements of clouds, aerosols, and the concentrations of several chemical constituents of the atmosphere. The differential absorption lidar (DIAL) technique is a very promising method to measure concentration profiles of chemical species such as ozone and water vapor as well as detect the presence of aerosols and clouds. If a suitable DIAL system could be deployed in space, it would provide a global data set of tremendous value. Such systems, however, need to be compact, reliable, and very efficient.

In order to measure atmospheric gases with the DIAL technique, the laser transmitter must generate suitable on-line and off-line wavelength pulse pairs. The on-line pulse is resonant with an absorption feature of the species of interest. The off-line pulse is tuned so that it encounters significantly less absorption. The relative backscattered power for the two pulses enables the range-resolved concentration to be computed.

Preliminary experiments at NASA LaRC suggested that the solid state Raman shifting material, Ba(NO₃)₂, could be utilized to produce these pulse pairs. A Raman oscillator pumped at 532 nm by a frequency-doubled Nd:YAG laser can create first Stokes laser output at 563 nm and second Stokes output at 599 nm. With frequency doublers, UV output at 281 nm and 299 nm can be subsequently obtained. This all-solid state system has the potential to be very efficient, compact, and reliable.

Raman shifting in Ba(NO₃)₂ has previously been performed in both the visible and the infrared. The first Raman oscillator in the visible region was investigated in 1986 with the configurations of plane-plane and unstable telescopic resonators. However, most of the recent research has focused on the development of infrared sources for eye-safe lidar applications.

Current Experiments

We have recently completed an initial set of measurements with the Raman oscillator in the visible region. The experimental apparatus for these tests is shown in figure 1. A compact, Q-switched frequency-doubled Nd:YAG laser serves as a pump laser for the Raman oscillator. Its PRF can be set at 30/N Hz by triggering
the Q-switch on every \textbf{Nth flashlamp} pulse. A half-wave plate (HWP) and polarizer cube (P) combination provides variable attenuation for the pump beam. The two lens-telescope provides control over the pump beam collimation and diameter. The \textbf{Raman} oscillator consists of a high reflector (HR), an output coupler (OC) and the \textbf{Raman} medium. The Ba(NO$_3$)$_2$ crystal (1x1x5 cm) is mounted in a sealed housing filled with index-matching fluid. The exit windows are A-R coated. The HR is a mirror which is 99.5\% reflective at the Stokes wavelengths, but 90\% \textbf{transmissive} for the 532 nm pump beam. Three sets of output couplers were designed to optimize the generation of first Stokes and second Stokes wavelengths separately and both Stokes wavelengths simultaneously. Doubling crystals convert the visible wavelengths into the \textbf{UV}.

Figure 2 is a plot of output energy from the \textbf{Raman} oscillator at the second Stokes wavelength vs. 532-nm pump energy for two pulse repetition rates. Pump energy is measured inside the resonator just before the barium nitrate crystal. There is virtually no first Stokes output for this cavity mirror configuration. The beam diameter at the crystal is 2.5 mm. Pump \textbf{fluence} must be maintained below the damage threshold for this material, which is approximately 8J/cm$^2$.

![Diagram of Raman oscillator configuration](image)

Figure 1. The barium nitrate \textbf{Raman} oscillator, configured for \textbf{UV} generation.
Figure 2. Output energy at the second Stokes wavelength vs. pump laser energy for two pulse repetition frequencies.

Conclusion
We are developing a compact, all-solid state UV source for DIAL measurements of ozone. We have Raman-shifted 532 nm pulses with up to 40% efficiency for the first Stokes and 48% efficiency for the second Stokes with our initial oscillator configuration. Beam divergence in the visible was measured to be 1.5 to 3.5 mrad with this particular pumping geometry.

At the conference, we will present results from our improved oscillator configurations for the visible and the UV.

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References