# Remote detection of Raman scattering by use of a holographic optical element as a dispersive telescope

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We describe the retrieval of nighttime lidar profiles by use of a large holographic optical element to simultaneously collect and spectrally disperse Raman-shifted return signals. Results obtained with a 20-Hz, 6-mJ/pulse, frequency-tripled Nd:YAG source demonstrate profiles for atmospheric nitrogen with a range greater than 1 km for a time average of 26 s. 

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Lidar measurements based on backscattered radiation that is due to Raman scattering have routinely been used for environmental monitoring.[1,2] Although the cross sections are considerably smaller than in Rayleigh scattering, Raman signal detection offers a powerful multiplexing advantage in that a fixed, single-wavelength laser source can yield spectroscopic information on a wide variety of molecular species. Systems such as the field-deployable NASA Goddard Space Flight Center Scanning Raman Lidar[3] and the one permanently installed at the Southern Great Plains Cloud and Radiation Test-bed site[4] provide day and nighttime monitoring of water-vapor profiles of the lower troposphere, permitting study of the temporal and spatial evolution of atmospheric features. Recently, holographic optical elements (HOE's) have been used to reduce dramatically the size and weight of beam-scanning lidar systems by combination of the telescope and steering mirror that are normally required for collection of the backscattered signal.[5,6] To date, only elastic-scattering lidar systems have been demonstrated with this approach; however, HOE's inherently exhibit wavelength dispersion as a result of diffraction, thus spectral information from inelastic processes such as Raman scattering can in principle be retrieved. As part of the development of a compact Raman lidar for airborne measurements of water-vapor profiles, we investigated designs for combining the telescope, full aperture scanning, and a spectral disperser in a single HOE. In this Letter we describe a HOE that is designed and fabricated for detection of Raman lidar backscatter signals and demonstrate its operation in an experimental system.

The detection of Raman-shifted wavelengths in the backscattered return signal in this lidar application is based on using a single HOE to perform simultaneously the functions of a telescope and a spectrometer. This is accomplished with a HOE with focusing power generated by recording of the interference between a collimated reference beam at angle $\alpha$ and an on-axis spherical wave front (object beam), as shown in Fig. 1(a). A refractive-index modulation within the holographic film is obtained that corresponds to the recorded interference pattern. When it is illuminated with a conjugate reference beam from a collimated source as shown in Fig. 1(b), the reconstructed conjugate object beam on the opposite side of the HOE is brought to a focus point that is consistent with the curvature of the phase fringes within the hologram volume. The HOE's focal length can be approximated by the simple analytical expression that is commonly used for an interferometric zone plate, $z = \eta/\lambda$, where $\eta$ is a constant corresponding to the focusing power and $\lambda$ is the wavelength of the reconstruction light. The focus position is also displaced laterally according to the well-known grating formula $m\lambda = b(\sin\alpha \pm \sin\beta)$, where $m$ is the diffraction

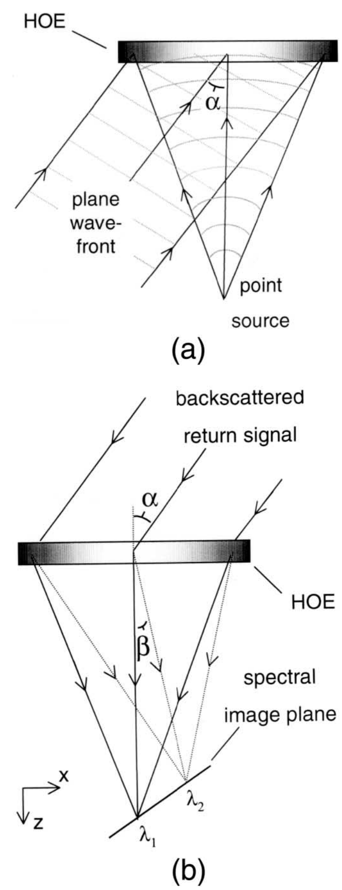


Fig. 1. (a) Holographic exposure setup used for recording the interference pattern for a transmission HOE with focusing power. (b) HOE used as a lidar receiver for monitoring wavelength-shifted light that is due to Raman scattering.

 

order, $b$ is the grating spacing in the plane of the HOE, $\alpha$ is the angle of incidence of the input beam, and $\beta$ is the diffracted angle. Combining the expressions for the simultaneous refraction and dispersion properties, and assuming a small angle on the converging side, we can represent the focus position coordinate $\mathbf{P}$ in the $z-x$ plane with the system of equations

$$\mathbf{P} = \begin{pmatrix} z \\ x \end{pmatrix} = \frac{\eta}{\lambda} \begin{pmatrix} 1 \\ \dfrac{\lambda}{b} - \sin \alpha \end{pmatrix}.$$

For a fixed angle $\alpha$, a change in $\lambda$ to longer reconstruction wavelengths results in a near-linear diagonal movement of the focus position closer to the HOE, as depicted by the tilted image plane in Fig. 1(b). When the HOE is used to collect the backscatter from a pulsed laser source propagating in the atmosphere, focused images of wavelength-shifted spectra that are due to Raman scattering appear along the corresponding position in the $z-x$ plane.

We fabricated large-diameter (40-cm) HOE plates for this effort, using dichromated gelatin (DCG) recording material and designed them to provide high coupling efficiency into the first diffraction order over a wavelength range from 355 to 408 nm. In addition to elastic scattering, this wavelength range covers Raman-shifted spectra from $N_2$, $O_2$, $H_2O$ (vapor and liquid phases), $NO_2$, $SO_2$, $CH_4$, and $CO_2$ when a frequency-tripled (355-nm) Nd:YAG laser source is used. First, a master hologram was fabricated by use of the geometry shown in Fig. 1(a). From geometrical ray-trace studies and numerical analysis of first-order diffraction efficiency, it was determined that an available argon-ion laser line at 457.9 nm could be used to obtain a HOE that exhibits high efficiency throughout the 355–408-nm range with an $\sim$2.5-m focal length and $<$0.6-mrad spot sizes. When aberration-correction lenses were included, ray-trace simulations indicated that focal spots $<$0.2 mrad could be obtained; however, these elements were not implemented for this initial experiment. By use of collimated light at a 45° angle and an on-axis point source at a distance of 1987 mm, a 4-$\mu$m-thick DCG film on a 40-cm-diameter, 6-mm-thick plate was exposed to 0.13-mW/cm$^2$, 457.9-nm radiation for 5 min. Development of the plate followed established procedures for DCG films,[7,8] including soaking the plate in a water bath to remove the photosensitive dichromate and immersing it in an alcohol bath to remove the water prior to drying. Through contact exposure, copies of the master were used to produce duplicate DCG plates that were processed following the same procedure as that for the master hologram. We then sealed the duplicate HOE's by applying a 6-mm cover glass with epoxy to form robust optical elements suitable for laboratory evaluation.

The spectral performance of two different HOE plates was evaluated over the 355–408-nm wavelength range by use of light generated by a high-pressure xenon arc lamp. The output of the lamp was first passed through a monochromator with a 0.2-nm spectral width and then coupled to the input of a multimode fiber. Aligning the output of the fiber with the focus point of a 40-cm-diameter, 2-m focal-length off-axis parabolic mirror yielded a collimated beam, permitting full-aperture illumination of the HOE. Measurements obtained at different HOE incident angles and wavelengths indicated that maximum diffraction efficiency occurred at $\sim$35° angle of incidence and varied from 60% to 65% from 355 to 408 nm. The position and orientation of the spectral line focus were consistent with numerical simulations, and analysis of the focus spots revealed $1/e^2$ diameters $\leq$1.5 mm, corresponding to a spectral resolution of 0.4 nm and a field of view of 0.6 mrad.

To demonstrate the measurement of atmospheric profiles of wavelength-shifted light that is due to Raman scattering, we assembled the system shown in Fig. 2. A frequency-tripled (355-nm) Nd:YAG flash-lamp-pumped laser was used to generate 6-ns, 6-mJ pulses of light at a repetition rate of 20 Hz. The output of the laser was directed vertically into the atmosphere through an opening in the laboratory ceiling. A large flat turn mirror adjacent to the outgoing laser beam reflected the backscattered radiation to the HOE, which was mounted at 35° angle of incidence. As shown in Fig. 2, this setup an also incorporated an optical path that permitted illumination of the HOE with reference light from the lamp–monochromator source and a parabolic collimator that was used in the diagnostic measurements described above. Removing the flat turn mirror allowed collimated light from the reference path to pass through the HOE, which facilitated alignment and established focus position prior to atmospheric measurements.

For this initial demonstration the Raman backscatter of atmospheric nitrogen at 387 nm was examined, since it has a well-known cross section, its signal is relatively strong, and its atmospheric density profile is stable. Figure 3 shows the lidar profile recorded for the Raman-scattered signal corresponding to atmospheric nitrogen for a time average of 26 s (512 laser pulses) at nighttime. We obtained this result using a photomultiplier tube (PMT) placed at the 387-nm focus position and filtering the light with a 0.3-nm-wide bandpass filter that provided $>10^{-10}$

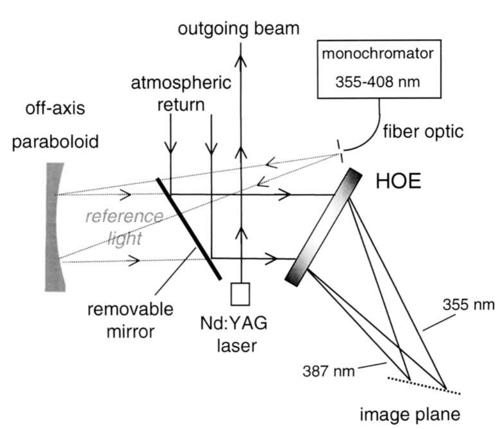


Fig. 2. Experimental setup used to recover Raman-shifted backscatter from atmospheric nitrogen.

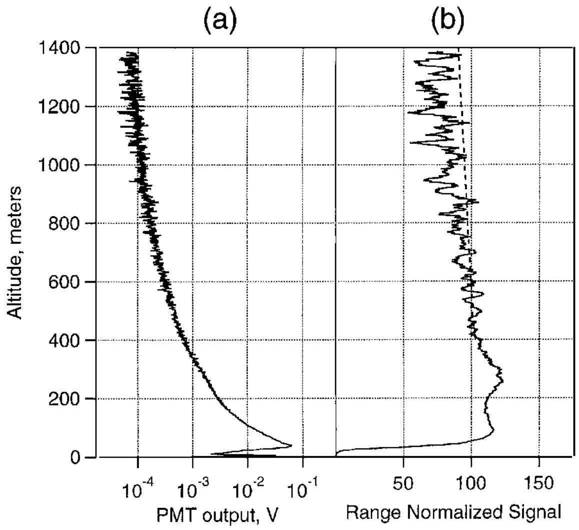


Fig. 3. (a) Raw PMT signal corresponding to Raman-shifted backscatter from atmospheric nitrogen and (b) profile corrected for atmospheric transmission and range-squared normalized.

out-of-band rejection to prevent stray scattered radiation (at 355 nm) from laboratory surfaces from saturating the PMT. The raw PMT signal, shown in Fig. 3(a), exhibits the characteristic decay shape associated with a lidar measurement of molecular nitrogen. The background-subtracted range-squared normalized result shown in Fig. 3(b) is smoothed to 16-m vertical resolution and corrected for standard atmospheric transmission. Although some anomalous features appear in the near-field region from 0 to 400 m, above 400 m the signal decreases at a stable rate that is slightly faster than expected for a pure molecular atmosphere, represented by the dashed line. This difference in decay corresponds to an optical depth of 0.15 from 0.4 to 1.4 km and is attributed to the presence of boundary-layer aerosols. Random noise in the 1–1.4-km range corresponds to ~10% rms of the signal for a 26-s time average at 16-m vertical resolution. These results indicate that this type of HOE receiver could be used for monitoring water vapor in the troposphere at nighttime when used with commercially available Nd:YAG lasers in the 100–300-mJ/pulse range. Performance during the daytime would be significantly reduced because of increased background light levels and would likely require improvements in the field of view and the spectral rejection.

A new method for retrieval of Raman lidar measurements by use of a single holographic optical plate to simultaneously perform the functions of a telescope and a spectrometer has been described. The HOE's fabricated for this research exhibited a spectral resolution of 0.4 nm in the wavelength range from 355 to 408 nm, which is suitable for retrieving Raman-shifted spectra from a frequency-tripled Nd:YAG laser source. Using this technique, we observed Raman scattering from atmospheric nitrogen from near the surface to ranges above 1 km at night, using a low-power laser source. This approach provides the unique advantage of enabling one to recover a wide range of Raman-shifted wavelengths, using a simple receiver geometry.

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### References

1. S. H. Melfi, D. N. Whiteman, and R. A. Ferrare, J. Appl. Meteor. **28,** 789 (1989).
2. R. A. Ferrare, S. H. Melfi, D. N. Whiteman, and K. D. Evans, Geophys. Res. Lett. **19,** 1599 (1992).
3. D. N. Whiteman and S. H. Melfi, J. Geophys. Res. **104,** 31,411 (1999).
4. J. E. M. Goldsmith, F. H. Blair, S. E. Bisson, and D. D. Turner, Appl. Opt. **37,** 4979 (1998).
5. G. K. Schwemmer, "Conically scanned holographic lidar telescope," U.S. patent 5,255,065 (October 19, 1993).
6. D. V. Guerra, G. K. Schwemmer, A. D. Wooten, S. S. Chaudhuri, and T. D. Wilkerson, J. Geophys. Res. **104,** 22,287 (1999).
7. R. D. Rallison and S. R. Schicker, Proc. SPIE **2404,** 217 (1995).
8. R. D. Rallison and S. Arnold, Proc. SPIE **2689,** 292 (1996).