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Gas-correlation lidar

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Basic principles for the extension of gas-correlation techniques to the lidar situation are discussed. Favorable signal-to-noise ratios and relaxed laser requirements characterize the technique. Preliminary experiments on atomic mercury are reported.

The differential absorption lidar (DIAL) technique¹⁻³ is a powerful and widely used remote-sensing method for monitoring of atmospheric gases, e.g., air pollutants. In the present Letter the combination of lidar and gas-filter correlation techniques^{4,5} is proposed for achieving improved laser monitoring of the atmosphere. Basic operational considerations are given, and preliminary remote-sensing experiments on mercury are described.

In normal DIAL experiments, pulsed-laser radiation is transmitted into the atmosphere at two alternate wavelengths, one on an absorption line of the species of interest and one off the absorption line but still close in wavelength (reference wavelength). The range-dependent backscattering, which is mainly due to Mie scattering from particles, is recorded with an optical telescope equipped by a detector and time-resolving electronics. Atmospheric turbulence, which has a correlation time of less than 10 msec (Ref. 6) will largely determine DIAL performance. By using a pulsed laser of a repetition rate of 10 Hz and switching between the two wavelengths between all pulses (see, e.g., Ref. 7), large-scale inhomogeneities are canceled out in normal ground-based applications; but the signal-to-noise ratio is clearly impaired by turbulence, requiring some additional signal averaging for such a relatively simple but still useful system. By using two separate, individually tuned laser systems fired at an interval of some tens of microseconds, frozen atmospheric conditions are achieved and high-quality data are obtained for this rather complex system, which can still operate with a single detection system. (Both wavelengths pass the same narrow-band filter; the two lidar returns are captured on a single transient digitizer sweep. See, e.g., Refs. 6 and 8.) For a monitoring system on a fast-moving platform a dual-laser approach has been necessary.

In nonlaser (passive) long-path optical absorption monitoring the effects of atmospheric turbulence can be eliminated by fast scanning, such as in differential optical absorption spectroscopy,⁹ dispersive correlation spectroscopy,⁵ and gas-filter correlation spectroscopy.^{4,5} Simultaneous on-off monitoring can also be achieved by using optical multichannel (array) techniques or systems with beam splitters. Gas-correlation spectroscopy is a particularly simple and powerful technique,

in which the incoming light is either passed directly to a detector or first passed through a cell containing an optically thick sample of the gas to be studied. For the case of an atmospheric path free from the gas, the light intensities in a selected wavelength region are balanced out by using lock-in or electrical bridge techniques. With the gas present in the atmosphere the light passing through the gas cell is still the same, whereas the additional absorption in the direct beam results in an imbalance in the electronics, which after calibration can be directly expressed as a parts-per-10⁶ times meter (ppm · m) atmospheric-gas burden.

The gas-correlation concept can readily be applied to the lidar configuration, leading to important system simplifications and improvements in signal-to-noise ratio. In particular, only one fairly broadband laser is needed, and no laser tuning is necessary between pulses. On- and off-resonance wavelengths are transmitted and detected simultaneously. In order to describe the gas-correlation lidar technique we chose a simple model example. We consider the case of atmospheric (atomic) Hg monitoring, for which we recently reported ordinary DIAL measurements.¹⁰ The description follows with reference to Fig. 1.

The laser is tuned to the 253.7-nm Hg resonance line. (A pulsed frequency-doubled dye laser could be used.) The region of Hg absorption (considering isotope shifts, hyperfine structure, and Doppler and pressure broadening) is about 0.005 nm. The laser bandwidth is chosen to be about three times this value. If a short pulse (a few nanoseconds) is used, no pronounced mode structure will be obtained, and a smooth spectral distribution for the pulse is assumed for simplicity, as indicated in the figure. The laser pulse is transmitted into the atmosphere through a Hg cloud at some distance from the lidar system and finally hits a topographic target or a retroreflector. Backscattered light is received by an optical telescope, and overlap between the transmission and detection lobes is obtained some distance from the system. For a homogeneous atmosphere, a $1/R^2$ falloff of the recorded intensity is then received. With an interference filter the spectral region of interest is isolated for background-light suppression. In contrast to the normal DIAL system, a beam splitter and two detectors are now used instead of one. One of the beams passes a gas-filter correlation cell—in the

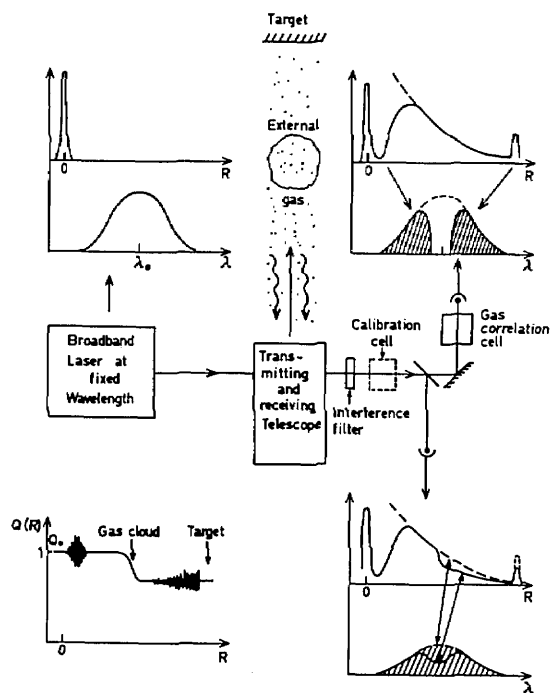


Fig. 1. Conceptual diagram of gas-correlation lidar.

example chosen, containing Hg of sufficient vapor pressure—to block out the central part of the resonance line. In DIAL language, in this detection arm the off-resonance signal is recorded (actually, two close-lying reference wavelengths are used simultaneously). In the other detection arm the whole spectral distribution is measured, which for the case of no atmospheric Hg is the same as the transmitted spectral distribution. For this case the detected signals in the two arms can be made equal (balanced out as in passive gas correlation) by beam attenuation or gain adjustments. If external Hg is present, less signal is detected in this arm, whereas the signal in the gas cell arm is unaffected. The imbalance between the two arms indicates the presence of the external gas. In Fig. 1 spectral and temporal curves at different points in the system are shown, illustrating the measurement process. In particular, spectral distributions could be considered for the final target echoes. By dividing the signals as illustrated in the figure—a procedure that is also common in DIAL⁷—a deviation from 1 is obtained in the presence of external Hg. Note that the ratio $Q(R)$ is independent of the laser-pulse energy, turbulence effects, etc., since the measurements are performed simultaneously on the same pulse. This is true for the signals' recorded range resolved at any one delay. For fast-moving platforms this is a great advantage. Note that the percentage deviation from 1 in the divided signal is the same as the one that would have been obtained in a DIAL measurement, in which the laser would be used once tuned on the absorption line and once tuned completely off the line. Since a linewidth larger than the absorption linewidth is used, the relevant absorption cross sections are dependent on the actual laser linewidth, and an optical depth dependence (deviation from the Beer-

Lambert law) also persists. Thus a gas-correlation lidar system is best calibrated by inserting cells with known ppm · m numbers in the light path between the telescope and the detector arrangement in direct connection with the actual measurements.

For a practical laser the spectral distribution within the laser bandwidth will vary from pulse to pulse, and this will result in a strongly increased noise level, since the two detection arms can no longer be balanced out. However, it is possible to monitor the relevant spectral fluctuations of the laser by detecting the ratio Q_0 of the intensity of the laser beam for a direct path to a detector and when passing an identical gas-correlation cell. No special arrangement is needed for this. The prompt signals that are due to light scattering in the telescope can be adjusted to the proper level and can be isolated from an atmospheric-backscattering background by an initial separation of the transmitted laser beam from the telescope's optical axis. The signals are recorded together with the atmospheric returns, as indicated in Fig. 1. If a low external gas concentration can be assumed close to the telescope and a laser power yielding a sufficient atmospheric backscatter as in the figure is used, the Q_0 value can also be obtained from the close-range backscattering. It can easily be shown that

$$\exp \left[-2 \int_0^R \alpha n(r) dr \right] \approx [kQ(R) - 1] / (kQ_0 - 1), \quad (1)$$

where α is the effective absorption coefficient in the bandwidth of the studied species of concentration $n(r)$, and k is the ratio of the signals for wavelengths not absorbed by the gas-correlation cell at the gas-cell and direct-pass detectors, respectively. If $Q(R)$ and Q_0 are recorded for every pulse, the integrated concentration value is not affected by turbulence, laser spectral fluctuations, etc., and a noise-free measurement situation has been achieved. Practically, and from the point of view of the approximate nature of Eq. (1), the system is most conveniently calibrated by inserting cells of known absorption in front of the beam splitter. Problems with possible Fabry-Perot fringes are then also largely eliminated.

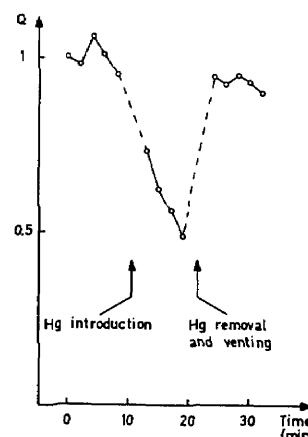


Fig. 2. Demonstration of remote Hg detection using the gas-correlation lidar technique. Each point corresponds to 100 laser pulses.

To demonstrate the gas-correlation lidar concept, some preliminary experiments on Hg with an experimental setup similar to the one in Fig. 1 were performed. An excimer-pumped dye laser, frequency doubled to the 254-nm region, was used. The lidar setup was similar to the one described in our previous paper on Hg.¹⁰ The laser beam was directed through a remote 2-m-long open-ended chamber (70-m distance), where a Hg-containing atmosphere could be obtained by introducing Hg droplets. After passing the chamber, the beam was retroreflected back to the lidar telescope, which had a diameter of 25 cm. The signals from the two detectors were recorded by a dual-channel boxcar integrator that was gated to the reflector echo and interfaced to a minicomputer. In Fig. 2 the ratio Q is plotted, illustrating the remote detection of the introduction and removal of the Hg droplets. No attempt to calibrate the system was made in the present demonstration, nor were the spectral fluctuations compensated for.

Although the system concept description and the experiments were related to Hg, it is evident that the same principle works for any gas in which close-lying wavelength regions with strong differential absorption exist. NO, with a sharp bandhead at about 226 nm, is such a case for which normal DIAL measurements have been performed.¹¹ The laser is tuned right to the bandhead, generating on- and off-resonance wavelength components simultaneously. Also, in the near-IR region, which is accessible, e.g., by rather broadband Raman-shifted dye lasers (see, e.g., Ref. 12), or through optical difference frequency generation, measurements on CH₄, CO, HCl, etc. should be feasible. For achieving an even more dial-like measurement situation and for better laser control, a laser simultaneously emitting two close-lying wavelengths (see, e.g., Ref. 13 and references therein) could be used together with gas-filter techniques as described above.

The sharp absorption features of gases permit a stable separation of signals at close-lying wavelengths without using sharp interference optics. It was recently suggested that these features be used to detect Mie and Rayleigh scattering separately in lidar systems for assessing atmospheric temperature.¹⁴ In this Letter the spectral correlation between an atmospheric-gas constituent and the gas contained in a cell is used instead for pollution monitoring. It should be noted that, whereas a perfect spectral match is achieved, an accidental coincidence by an interfering molecular absorption line would cause an error. By using a broad-

band laser (several nanometers), such as the ones used in broadband coherent anti-Stokes Raman spectroscopy (see, e.g., Ref. 15), true gas correlation⁴ with automatic rejection of interfering species through Zeeman or Stark scanning of the cell every second shot should be achievable, e.g., in a NO₂ lidar system. Further, the same concept should apply for properly selected wavelength regions of multiline HF/DF lasers, for continuously tunable high-pressure CO₂ TEA lasers, and for diode lasers.

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