DIAL technique for pollution monitoring: improvements and complementary systems

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Improvements to and complementary systems for a differential absorption lidar system for field work are discussed. A dual-wavelength laser and a dual-wavelength detection system, which make DIAL with simultaneous lidar measurements at two wavelengths possible, are described. A multiwavelength measurement routine, which will improve accuracy and detection limits, is proposed. A dual-beam lidar system for measurements of plume velocity is also described.

I. Introduction

The differential absorption lidar (DIAL) technique has been applied in remote sensing of atmospheric gases and has become an efficient tool for measurements on air pollutants. The method utilizes the differential absorption properties of molecular species as observed in backscattered lidar signals from the atmosphere and topographic targets. High performance systems employing dye lasers have been developed for measurements on SO₂, NO₂, and O₃.¹⁻⁹ The DIAL technique has been discussed by numerous authors, and in a recent book by Measures¹⁰ the theory and applications are discussed in detail. In a forthcoming book, the use of the DIAL technique for pollution mapping will be reviewed¹¹; thus only a brief review and discussion of a few details will be presented here.

A mobile DIAL system was developed in 1979,¹² and it was tested and evaluated in numerous field tests.^{4,5} Through these investigations information was obtained on the requirements and limitations of the technique in field applications. The design of a new mobile laser laboratory has been outlined, and it is now under construction. Some improvements to and complementary systems for a DIAL system for field work are proposed in this work.

Lidar measurements at two wavelengths, corresponding to an absorption wavelength of the gas mea-

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sured and a neighboring reference wavelength, are performed in a DIAL measurement. The two laser beams are generated with two different lasers or by one laser, which is then alternatively tuned to the absorption and reference wavelengths. For several reasons it would be advantagous if the two laser beams could be generated simultaneously with one laser. Several dual-wavelength lasers have been described.^{13–27} In this work a device for fast precision tuning of the wavelength of a dye laser is presented, which can also be employed to run the laser at two wavelengths. This device is easily applicable in a standard dye laser, and the power ratio of the two wavelengths is continuously variable.

With a dual-wavelength laser it is also necessary to separate the lidar signals in the receiver of the DIAL system. A prism separator is designed to accomplish this. It is mounted at the exit of a spectrometer and separates neighboring wavelength regions into two channels.

So far only two wavelengths have been employed in most DIAL measurements. However, it would be possible to increase the accuracy and detection limits if several wavelengths were employed. With current 16-bit minicomputers and dye lasers with intermediate repetition rates, it is possible to improve the measurement routines and carry out evaluations during the measurement cycle and also determine the precision in the measurement.

In many pollution studies, where the DIAL method is applicable, knowledge of the wind parameters is required as well as the concentration of the pollutant. For example, when the pollutant flow from an industrial area is measured the accuracy is limited by the difficulty in estimating the wind velocity at different altitudes. Several lidar methods for measuring wind parameters have been developed.^{28–42} In this work a simple device

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for a cross-correlation technique, which is intended to be complementary to a DIAL system, is reported.

II. DIAL with Simultaneous Lidar Measurements at Two Wavelengths

Average concentrations over selected range intervals are measured with the DIAL technique. The lidar signals at two wavelengths with different absorption cross sections for a certain molecule are measured and compared. The basic equation for the average molecular number density N_m between the range R_1 and R_2 applicable in the case where there is only one differentially absorbing species can be expressed as

$$\overline{N}_{m} = \frac{1}{2(R_{2} - R_{1})[\sigma_{A_{m}}(\lambda_{abs}) - \sigma_{A_{m}}(\lambda_{ref})]} \times \left\{ \ln \left[\frac{P(\lambda_{ref}, R_{2})P(\lambda_{abs}, R_{1})}{P(\lambda_{abs}, R_{2})P(\lambda_{ref}, R_{1})} \right] \right\},$$
(1)

where $\sigma_{A_m}(\lambda_{abs}) - \sigma_{A_m}(\lambda_{ref})$ is the difference in the absorption cross section between the absorption and reference wavelengths, and $P(\lambda_{abs},R)$ and $P(\lambda_{ref},R)$ are the backscattered lidar signal powers from the range R at the absorption and reference wavelengths, respectively.

There are several assumptions made in the reasoning leading to this equation. The absorption by other species must be identical for the two laser beams, and the scattering properties of the atmosphere must have the same relationship between the two wavelengths for all ranges. The detection efficiency of the receiver must also have a constant relationship for the two beams; i.e., the overlap functions of the laser beam and the field of view of the receiver must be identical. Furthermore, the spatial intensity distribution of the two laser beams and their temporal power profiles should be identical. In realistic cases these requirements are not completely fulfilled, and, in addition, the signal SNR of single lidar signals is generally too low to make the equation above meaningful. Thus only averaged lidar signals can normally be evaluated.

With current standard equipment the above requirements, that the laser beams should be identical except for the wavelength and that the atmospheric conditions should be the same for corresponding lidar measurements, lead to a trade-off between using one or two lasers. In a one-laser DIAL system the first requirement is more easily fulfilled, and in a two-laser system the beams can be transmitted with a short time delay during which the atmosphere can be considered to be frozen.

A technique where one laser is modified with a precision tuning device to produce a dual-wavelength beam and a corresponding dual-wavelength detector will be presented.

A. Dual-Wavelength Laser

Several techniques for generating two wavelengths simultaneously with a dye laser have been reported. The basic idea is to split the beam in the laser cavity into two beams, which can be individually tuned to optical resonance. This can be achieved with a glan prism or



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Fig. 1. Condensed schematic diagram of the Nd:YAG pumped dye laser operating at two wavelengths simultaneously. A precision tuning device covers half of the expanded beam in the dye laser cavity. The output wavelengths correspond to an absorption and a reference wavelength in an SO₂ DIAL measurement.

a dielectrically coated beam splitter and two separate gratings or a mirror arrangement.^{13–17} An alternative way to split the laser-cavity beam into two beams with different optical resonances is to employ a wedge plate.^{18–20} It has also been proposed to split the beam with a mirror and use two gratings²¹ to use two dielectric optical filters and a mirror arrangement²² or to split the pump beam and employ two regions of the dye cell.²³ Furthermore, dye lasers with gratings at grazing incidence as tuning elements can be operated at two wavelengths with proper mirror arrangements.²⁴⁻²⁷ In DIAL applications it is advantageous if the relative powers of the two wavelengths can be easily changed. An accessory to an ordinary dye laser is reported here, which has this property. This device can also be used for fast precision tuning of the laser wavelength and has some advantages regarding wavelength calibration in field applications.

The precision tuning device is made up of two circular wedge plates, which are rotated by means of stepping motors and gear belts. The principle of operation is shown in Fig. 1. The two wedge plates cover half of the expanded beam in the cavity and change its angle of incidence and reflection at the grating. The plates are fused silica and have a diameter of 50 mm. The wedge angle is 1.15°, which gives a total maximum refraction in the plane of incidence of about ± 20 mrad in the visible region. The refraction is changed by rotating the two wedge plates clockwise and counterclockwise, respectively. The device has been used in a Quanta Ray PDL-1 dye laser pumped by a Nd:YAG laser source. This laser has an echelle grating in Littrow mounting for wavelength tuning similar to the configuration shown in Fig. 1. The amplifier stage of the dye laser shown in Fig. 1 is simplied while the real laser has an additional beam expander and a different optical arrangement for the pumping beam. When the dye laser grating was operated in the fifth order and the nondeflected beam was tuned to 600.1 nm, the deflected part

of the beam could be tuned to any wavelength between 597.5 and 602.7 nm. The precision of the wavelength shift is basically determined by the wedge angles and the precision of the angular rotation and was in this case ~ 0.01 nm, which was less than the linewidth of the laser.

The output of the laser will be a two-wavelength laser beam, where the two wavelength parts will have the same direction but a slightly different spatial intensity distribution. The linewidth of each of the output beams will be somewhat increased compared with the one-wavelength output, since fewer lines of the grating are illuminated by each beam. The relative powers of the two beams are easily changed by lowering or raising the two wedges in the expanded laser beam.

The fine-tuning device can also be employed to change the laser wavelength between each laser pulse as is normally done in most current DIAL applications. The wedges are then lowered to cover the whole beam in the cavity and continuously rotated. The exact wavelengths are selected by triggering the laser at preset positions of the stepping motors.

The wedge arrangement needs no extra calibration since the wavelength shift is exactly determined by the position of the stepping motors, and in practice only the dye laser has to be calibrated at the nonrefracting wedge position. This is accomplished with a glow discharge lamp employing the optogalvanic effect. In the case of SO₂ DIAL measurements the calibration is made against the spectrum of neon.⁴³

Frequency doubling of the visible laser radiation to the UV region in, e.g., DIAL measurements on SO₂ and O₃ is accomplished with two frequency-doubling crystals mounted in series, which are tuned to the absorption and reference wavelengths. The serial arrangement attenuates the laser radiation somewhat but is advantageous compared with other arrangements regarding complexity and does not introduce any appreciable differential misalignment of the two beams.

B. Dual-Wavelength Detection System

A schematic diagram of the transmitting and receiving optics of a typical DIAL system is shown in Fig. 2 together with a new dual-wavelength detection system. The telescope and directing mirror arrangement are similar to the system described in Ref. 4 and to a new system under construction shown in Fig. 9. The telescope has a Newtonian configuration, and the telescope mirror has a focal length of 1 m and a diameter of 30 cm (new system: 40 cm). The original detection system is equipped with narrowband pass filters to suppress background light, and two neighboring wavelengths can be measured simultaneously only by employing a beam splitter and extremely narrow filters, which will substantially attenuate the signals.

The dual-wavelength detection system shown in Fig. 2 is a spectrometer based on a diffraction grating. The optical configuration is basically similar to an ordinary monochromator of the Czerny-Turner type, and it has an adjustable entrance slit, but instead of an exit slit it has a prism separator with two exits. This prism sep-



Fig. 2. Transmitting and receiving optics of a DIAL system with dual-wavelength detection.



Fig. 3. Detail of spectrometer with prism separator and two exit slits. The shaded areas correspond to two wavelengths dispersed by the grating and separated by the prism separator. The disperson is exaggerated for the sake of clarity.

arator, shown in detail in Fig. 3, consists of an equilateral prism of quartz. The dispersed wavelength spectrum enters the prism at normal incidence at one prism surface and is totally reflected at one or other of the two far prism surfaces. The wavelength corresponding to the position of the far edge of the prism will determine the dividing line of two spectral regions. There is one exit slit for each of the two regions, and these can be translated, and their widths can be independently adjusted. The exit focal plane of the spectrometer coincides with the position of the slits. The separation of the two regions will be independent of the width of the exit slits, and it is exclusively determined by the width of the entrance slit and the angular dispersion of the grating. This is an advantage since any leakage of light from one optical channel to the other would cause a systematic error in the DIAL measurement. Narrowband pass filters and absorption filters can be added in the optical path of the receiver or in the photomultiplier tube housings to improve the spectral rejection.

The receiver and detection systems should be adapted to each other so as to optimize the efficiency, and the diffraction grating of the spectrometer must be optimized for this application. In some cases, where the measurement wavelengths are very close, it may be advantageous to decrease the laser beam divergence with a beam expander to be able to decrease the telescope field of view and the entrance slit of the monochromator.

III. Multiwavelength DIAL

Only two wavelengths have been employed in most visible and UV DIAL measurements, and the evaluations have been made according to Eq. (1). However, it would be possible to improve the accuracy and detection limits a great deal if several lidar measurements at different wavelengths were performed and the results were fitted to an absorption profile of the studied gas. In conventional laboratory spectrometers this fitting procedure is a standard technique. A similar technique was recently employed in long-path absorption measurements on NO.44 Six wavelengths around the absorption line at 226.8 nm were then generated with a Raman-shifted frequency-doubled dye laser, and the backscattered light from a retroreflector was monitored. Long-path absorption measurements are performed with conventional light sources that have a continuous emission spectrum in the differential optical absorption spectroscopy (DOAS) technique.⁴⁵ Very low detection limits are then accessible as a large number of wavelengths are fitted to an absorption profile.

DIAL measurements using several wavelengths have been restrained by technical limitations. Dye lasers pumped by Nd:YAG lasers or fast flashlamps have had a maximum pulse repetition rate of ~10 Hz at maximum output power, and the minicomputers normally employed have had limited receiving and storage capacity. However, current laser systems can well have a repetition rate of 30 Hz, and new lasers with considerably higher repetition rates can be expected in the next few years. Furthermore, the current 16-bit minicomputers have a much greater receiving and storage capacity than prior 8-bit minicomputers. Thus it is possible to apply measurement routines where lidar signals at several wavelengths are performed.

The dual-wavelength laser and the dual-wavelength detection system can be useful in multiwavelength DIAL measurements. One fraction of the laser radiation can then be fixed at a reference wavelength which is detected in one of the detection channels, while the other one is changed between each laser pulse. An example of such a routine for DIAL measurements on SO₂ is shown in Figs. 4 and 5. One part of a measurement cycle for a laser with a repetition rate of 30 Hz is shown on a millisecond time scale in Fig. 4. Here the fixed laser wavelength is 299.3 nm, and the other laser wavelength is subsequently tuned to seven different









Fig. 5. Program for multiwavelength DIAL measurements with on-line evaluations.

neighboring wavelengths close to the SO_2 absorption line at 300.03 nm. The laser is physically blocked by a chopper every eighth laser pulse, while the background noise signal is recorded in both detection channels. The recorded lidar and background signals for a preset number of cycles are averaged in 16 files by the computer before subtraction of the background and calculation of the logarithm. Several logarithmic lidar signals are averaged before the final evaluation of the concentration profile. As indicated in Fig. 5 simple evaluations are performed on-line, and the precision in the measurement is determined. This proposed measurement routine will be easy to control with a currently available 16-bit computer. The potential systematic error due to the averaging procedure,⁴ which is required to improve the SNR before calculating the logarithm according to Eq. (1), will be easier to minimize with the proposed routine, and it is also possible to let the eval-



Fig. 6. Principle of dual-beam lidar system for measurements of plume velocity.

uated data and precision steer the continuing measurement.

The measurement routine outlined above is largely also applicable in a one-wavelength DIAL system. The dye laser is then subsequently tuned to a number of wavelengths of the absorption and reference spectrum, and the logarithms of the lidar recordings are fitted to the spectrum. However, the measurements will be more sensitive to varying atmospheric conditions in this case.

IV. Dual-Beam Lidar for Measurements of Wind Velocity

The DIAL method can be employed in measurements on pollutant flows. This is certainly one of the most interesting applications of the technique, since pollutant flows are extremely difficult to measure with conventional point monitoring instrumentation. The measurement or estimation of the wind parameters must then be as precise as the measurement of the pollutant concentration. An estimation is often difficult to make, and it is not unusual that plumes at different altitudes in an industrial area spread in completely different directions.

Several research groups have demonstrated methods whereby atmospheric wind velocities and 3-D wind fields can be measured. Laser Doppler velocimetry has been applied in heterodyne lidar systems employing coherent CO_2 laser techniques.^{28–33} Measurements on global winds, clear-air turbulence, gust fronts, and aircraft wake vortices are some of the possible fields of application of such Doppler anemometers. Wind measurements using the Doppler shift approach have also been demonstrated with lidar systems employing other laser sources.^{34–35} The Doppler anemometers measure frequency shifts of the scattered return compared to the transmitted laser beam. Another possible way of measuring wind data with the lidar technique is to study the spatial and temporal variations of the lidar signal in different directions. The motion of aerosol inhomogeneities, and consequently the wind, is then determined with correlation techniques.^{36–42} In general the correlation analysis requires large computers and a lot of computing time.

In this work the configuration of a simple dual-beam lidar system for plume velocity measurements is presented. The technical devices are intended to be complementary to a lidar system. The system is primarily meant to be employed for measurements of wind direction and wind velocity orthogonal to a DIAL charting of a pollutant flow. An existing plume is employed as the scattering medium.

The principle of the dual-beam lidar system is shown in Fig. 6. Two simultaneous laser beams are transmitted in slightly different directions, thus probing different scattering volumes. In general the two beams have the same elevation angle, but this is not a restriction. The backscattered lidar signals of the two beams are captured individually. The lidar setup is basically identical to the system shown in Fig. 2.

The transmitted laser beam is split into two beams with a small angular separation by means of a quartz wedge plate with a 3-mm² center hole. The wedge angle is 1.15° , which gives an angle of refraction of ~10 mrad in the visible region. The arrangement is shown schematically in Fig. 7, where the refraction is exaggerated for the sake of clarity. One part of the laser beam is transmitted through the hole and is unaffected by the wedge while the rest of the beam is refracted. By rotating the wedge the noncentral part of the beam is refracted in all directions around the unaffected beam fraction. This possibility of choosing the direction means that the two beams can be adjusted to the same elevation angle when rotating the directing mirror of the system shown in Fig. 6.

The backscattered lidar signals collected with the receiver telescope will have two separate images in the focal plane. For a telescope with a focal distance of 1 m, a divergence of 10 mrad will correspond to a separation of 10 mm in the focal plane. The lidar signals are picked up by two optical fibers with a diameter of 1 mm and are transferred to photomultiplier tubes. The positions of the two fibers determine the fields of view of the detection. Thus the same precision can be obtained regardless of laser beam size. One of the fibers is fixed to the optical axis, while the other is adjusted to the desired position in the focal plane. The adjustable fiber is mounted on a rotatable mounting which is turned to the correct elevation angle with the aid of a video system shown in Fig. 2. Thus, in practice, the

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Fig. 7. Wedge unit for separation and refraction of the laser beam in the dual-beam lidar system. The refraction is exaggerated for the sake of clarity.

laser beams and wedge position are coordinated with the field of view of the receiver and not the reverse.

The signals of the photomultiplier tubes are fed to a 100-MHz transient digitizer. A dual-channel instrument is desirable, but a digitizer with a single channel can also be employed. In the latter case a number of recordings of the lidar signal A corresponding to the upwind laser beam position are recorded, and then the digitizer is switched to the other lidar signal B. A flow chart diagram for such a wind measurement is shown in Fig. 8. The pulse repetition rate of the laser must be 10 Hz or more for measurements of wind velocities of typically 10 m/sec at a distance of 1 km. A wedge with a larger wedge angle is an alternative to higher repetition rates. In many cases the wind direction and velocity can be deduced directly by comparing the sequential series of lidar signals of a plume while a simple correlation program is required in other cases. A minicomputer is sufficient for the correlation calculation.



tion monitoring. The accuracy and detection limits must then be optimized for field applications. In this work a few improvements to the technique are presented. The principal ideas of these improvements have been tested in field applications with our old mobile DIAL system.^{4,5,12} A new mobile laser laboratory, which is intended to be an improved version of the old system, has been outlined and is now being constructed here in Lund. A schematic diagram of this new mobile system is shown in Fig. 9. The platform is a covered truck, and the laboratory space is $6.0 \times 2.3 \times 2.1 \text{ m}^3$. The dome construction with the directing mirror can be lowered, and the system will be applicable not only to DIAL measurements but also to differential optical absorption spectroscopy (DOAS) employing light sources other than lasers. The system is also intended to be a mobile laboratory useful for other laser techniques based on Raman and fluorescence scattering with several industrial applications.

The fine-tuning device presented here can be employed for fast precision steering of any light beam. For example, it can be used as a precision tuning device in



Fig. 8. Program for measurements of plume velocity with the dual-beam system and a single-channel transient digitizer.

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Fig. 9. Schematic diagram of the mobile laser laboratory now being constructed.

a monochromator. The dual-wavelength detection system can be employed in other applications, where two optical regions are studied simultaneously. In differential optical absorption spectroscopy, where one small wavelength region is repetitively scanned to measure the absorption spectrum of a molecule, the device can be employed to make measurements of two spectral regions simultaneously.

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research was produced by two professors, a botanist and an immunologist, whose collaboration came about by coincidence. "Their offices were next door to each other," said David W. Fraser, president of Swarthmore. "At a large university they would have been in separate buildings and it would not have been likely to have happened."

Stuart B. Crampton, a physics professor at Williams, said scientists are not automatically handicapped by doing their research at the small liberal arts colleges. "In physics, there are whole fields dominated by small groups and even by single investigators. Yet scientists at small liberal arts colleges do suffer in some ways. "We do our work isolated from peer colleagues," said Jeanne A. Powell, a biologist at Smith College in Northampton, Mass.

Presidents of the leading liberal arts colleges talk of another sort of pressure. They often find that those they would like to hire as new faculty members in the basic sciences want reduced teaching loads. Until now, most of these colleges have resisted such requests as inimical to their institutional character. "We could pull ourselves apart if we give in and decrease teaching loads and increase research in order to attract some of these scientists," said Robert H. Edwards, president of Carleton College in Northfield, Minn. President Fraser of Swarthmore adds that a move in this direction could undermine general faculty morale.

The other issue confronting such institutions is the changing demands of science and escalating costs of equipment. "What we have done so well may not be possible unless we focus on more limited aspects," said Mary P. McPherson, president of Bryn Mawr College in Pennsylvania, [who] anticipates having to spend \$27 million on its science facilities. "Science is now being done primarily with large groups of people working with very expensive equipment and it will mean a whole different model for us."

The first report was published in the Sunday edition of 16 June 1985, the second in the Education column of the Science Times section of 18 June 1985. ۲

ERRATUM

Contrast-invariant pattern recognition using circular harmonic components

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One author's name was incorrectly spelled on the title page of this paper published in the 15 July issue. The names should have read:

Henri H. Arsenault and Claude Belisle.

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