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Laser remote sensing

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Laser remote sensing is a fast developing area of laser spectroscopy with numerous applications. Strengths and weaknesses of direct remote sensing, e.g. DIAL, LIDAR, and indirect sensing through fiber optics are discussed. Details of different configurations, instrumentation, and mobile platforms for remote sensing are also given. Examples from environmental analytical spectroscopy, respectively process analysis, are covered for both types of remote sensing. ©1998 Elsevier Science B.V. All rights reserved.

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1. Introduction

The term 'remote sensing' is prone to a number of different meanings depending on the discipline involved. At present the term is used most often for imaging objects near the Earth's surface by means of observation from airborne or satellite platforms. However, this article will concentrate (by a rather subjective

selection of examples) on remote 'chemical' sensing with lasers, omitting other techniques with a non-chemical objective.

In general, spectroscopic remote sensing can be performed either in a passive mode, employing natural radiation sources like the sun, or in an active mode utilizing a laser or lamp as a light source. In the following we will refer only to active laser remote sensing. The entire process of remote sensing can be divided in two fundamental steps, i.e. (1) excitation of the analyte within a matrix using a proper optical frequency, and (2) collection of a light signal carrying the analytical chemical information from this target by a suitable optical system (e.g. a lens, telescope, or fiber optic) and subsequent processing by a detector system not in physical contact with the object of analysis.

The spectral information emanating from the target can be utilized in a number of different ways, e.g. to identify single components or a class of analytes, to categorize by type (fingerprinting) and chemical composition (major, minor, and trace constituents) either on a threshold level or a continuous scale, to analyze dynamics and distribution as a function of space and time, or to evaluate related secondary target properties (e.g. thermal properties).

Following the history of remote sensing, which started as early as 1963 [1], we will distinguish in the following between direct remote sensing, i.e. both the laser and signal are used along an open (atmospheric) path, and indirect remote sensing, i.e. the laser or the signal is brought through fiber optics or some other means into contact or near the target.

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2. Direct sensing

Being coherent and basically only diffraction-limited in terms of propagation, the special features of laser light favor direct remote sensing, which at present is mainly related to environmentally relevant subjects as global warming, stratospheric ozone depletion, urban photochemical smog, forest decline, detection of oil spill on surface waters, and depth-resolved oceanographic measurements of organic materials and phytoplankton. An extensive review of laser techniques for direct remote sensing especially in the atmosphere is given in [2–4], respectively literature therein and in the classic textbook on remote sensing by Measures [1].

In principle, fluorescence, absorption, and Raman techniques are suitable for direct remote sensing (for more details concerning the underlying principles and typical instrumentation see [2,4,5]). Raman scattering, i.e. inelastically scattered radiation from molecules, is limited to moderate distances (< 1 km) and concentrations in the order of hundreds of ppm due to the low Raman cross-section; in some cases improvements are possible with a resonance Raman approach. The scattering cross-sections are much higher for elastically scattered radiation at particles (Mie scattering) or molecules and atoms (Rayleigh scattering).

As elastic scattering provides no analytical information per se, it has more importance for research on targets like clouds and aerosols. However, elastic scattering can be favorably used in combination with absorption spectroscopy (see below), the most popular technique for sensing of gaseous analytes. This stems not only from the higher cross-section of absorption and selection of wavelengths with the current laser technology (UV, VIS, NIR and IR regions), but also from the fact that fluorescence spectroscopy is hampered by severe collisional quenching in open atmospheric sensing. Only in the rare case of saturation of optical transitions over the entire distance of sensing is fluorescence spectroscopy employed for direct sensing. Otherwise, it is mostly used for targets in the upper atmosphere (mesospheric atoms), the hydrosphere, or terrestrial targets.

A straightforward approach to direct sensing is long-path absorption based on the same principles as a conventional spectrophotometer, using a well collimated monochromatic laser beam with a path length of several kilometres [6]. The analyte concentration can then be calculated as path-averaged concentration with the well known Beer-Lambert law. For calibration, the required differential absorption cross-section

of the molecule can be measured under laboratory conditions for a defined concentration, while the instrumental function is corrected mathematically. As in all open atmosphere sensing the light signal is affected by a priori unknown quantities and atmospheric backscattering, a differential approach (also known as DOAS, differential optical absorption spectroscopy) is often realized to cancel these effects, which are almost independent of wavelength.

In this differential mode (see also Fig. 2A), the laser is either tuned across the absorption line of interest or switched from a wavelength with peak absorption ('on' position) to a nearby wavelength with negligible absorption ('off' position). To compensate atmospheric turbulence, the measurement must be taken at a rate faster than 10 Hz. For long-path measurements either single or double-ended optical paths can be realized by way of retroreflectors or topographical targets (see Fig. 1). Range-resolved information can be acquired by using targets at different ranges or angles. However, for topographic targets problems arise from differential spectral reflectance of the target, which has to be accounted for.

Long-path absorption spectroscopy can be utilized in two major spectral regions, the middle or fundamental IR or the UV/VIS. In the former, the so called fingerprint region, the absorption is due to fundamental (as well as some overtone) bands and vibrational-rotational combination bands, whereas in the latter case electronic transitions with vibrational-rotational structure contribute to the absorption. For atmospheric long-path detection of small absorptions only spectral windows below $2.5 \mu\text{m}$, between $3 \mu\text{m}$ and $5 \mu\text{m}$ and from 8 to $14 \mu\text{m}$ can be used due to the atmospheric H_2O and CO_2 , which reduce the transmission beyond this range. The strength of open long-path measurements as compared to traditional sampling approaches using long-path multipass White cells is the absence of wall losses (important for unstable species like the OH radical), the potential for real time measurements, and the easy calibration. A serious disadvantage is atmospheric pressure broadening which not only decreases sensitivity (usually in the ppb range). Hence, long-path absorption works well for molecules with small pressure broadening coefficients and/or wide line spacing such as CH_4 and HCl , which limits the method often to molecules having less than 10 atoms.

Both sensitivity and selectivity are related to the spectral brightness and tunability of the source and to the spectral resolution and responsivity of the detector. In the IR mainly tunable diode lasers (TDL) based on lead salt semiconductors and molec-

ular gas lasers (tunable CO and CO₂ laser) are currently employed, in future also optical parametric oscillators and high-power quantum-well diode lasers will be seen more often. In the UV/VIS range excimer- or Nd:YAG-pumped dye lasers or Raman shifters are mostly employed. While the detectors available for the UV/VIS range (e.g. photomultipliers (PMT), photodiodes, photodiode arrays or CCDs) have sufficient quantum efficiency and low noise, the sensitivity of the photodetectors (mostly InSb and PbSe) in the MIR is often significantly improved by a first- or second-order derivative absorption measurement.

The light detection and ranging (LIDAR) technique using a pulsed laser as a transmitter and an optical telescope as a receiver in a RADAR-like fashion directly provides a range-resolved, three-dimensional mapping of pollution concentrations including meteorological parameters (temperature, wind speed and humidity). Tropospheric pollution monitoring is most frequently done using differential absorption LIDAR (DIAL) [7–10]. Range-resolved optical transients due to elastic aerosol backscattering are recorded for ‘on’ and ‘off’ laser wavelengths (see above) in alter-

nating laser pulses. The losses for the ‘off’ wavelength reflect only $1/r^2$ -losses of the divergent laser, respectively the signal, while the ‘on’ wavelength is also attenuated by absorption from gas (analyte) molecules. The spatial resolution is roughly twice the depth of the scatter layer. The ratio of the resulting curves yields the absorbance (i.e. concentration) as a function of distance (typical range for SO₂, NO_x, O₃, and Hg are 0.5–5 km with ppb detection limits), at the same time unknown atmospheric parameters are eliminated. Fig. 2A illustrates the DIAL principle, i.e. the backscatter intensity vs. distance, respectively the concentration vs. distance. As fluxes can be registered in real time, DIAL is particularly valuable for urban and industrial monitoring as well as analysis of gases from geogenic origin. Although DIAL is used mostly in a single-ended version, it can be set up also in a double-ended version with topographic targets or retroreflectors. As the signal strength received relies on effective backscattering (as ‘distributed mirror’), DIAL is most useful in the UV/VIS due to the strong Mie (λ^{-2} dependence) from particles and Rayleigh (λ^{-4} dependence) scattering from molecules. Mie

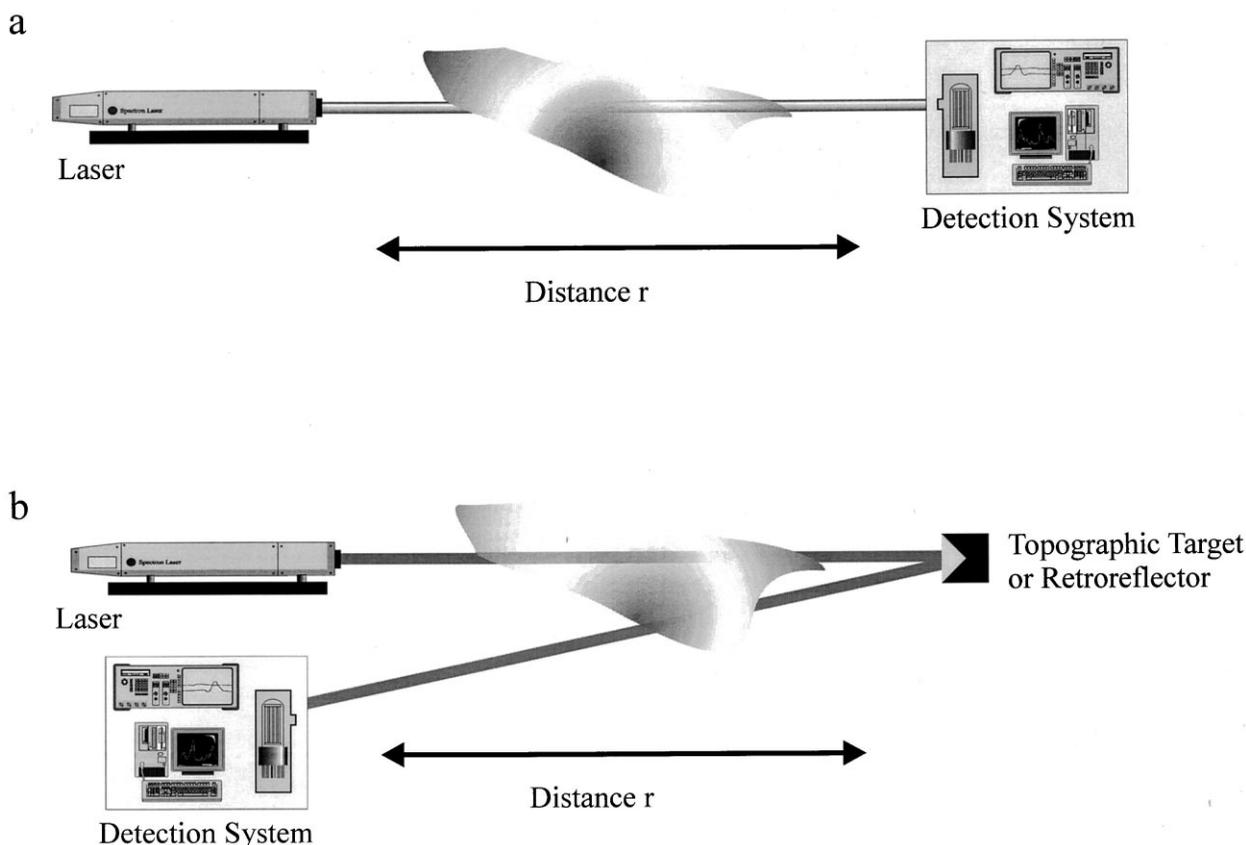


Fig. 1. a: Single- and b: double-ended configurations for long-path laser absorption spectroscopy.

A

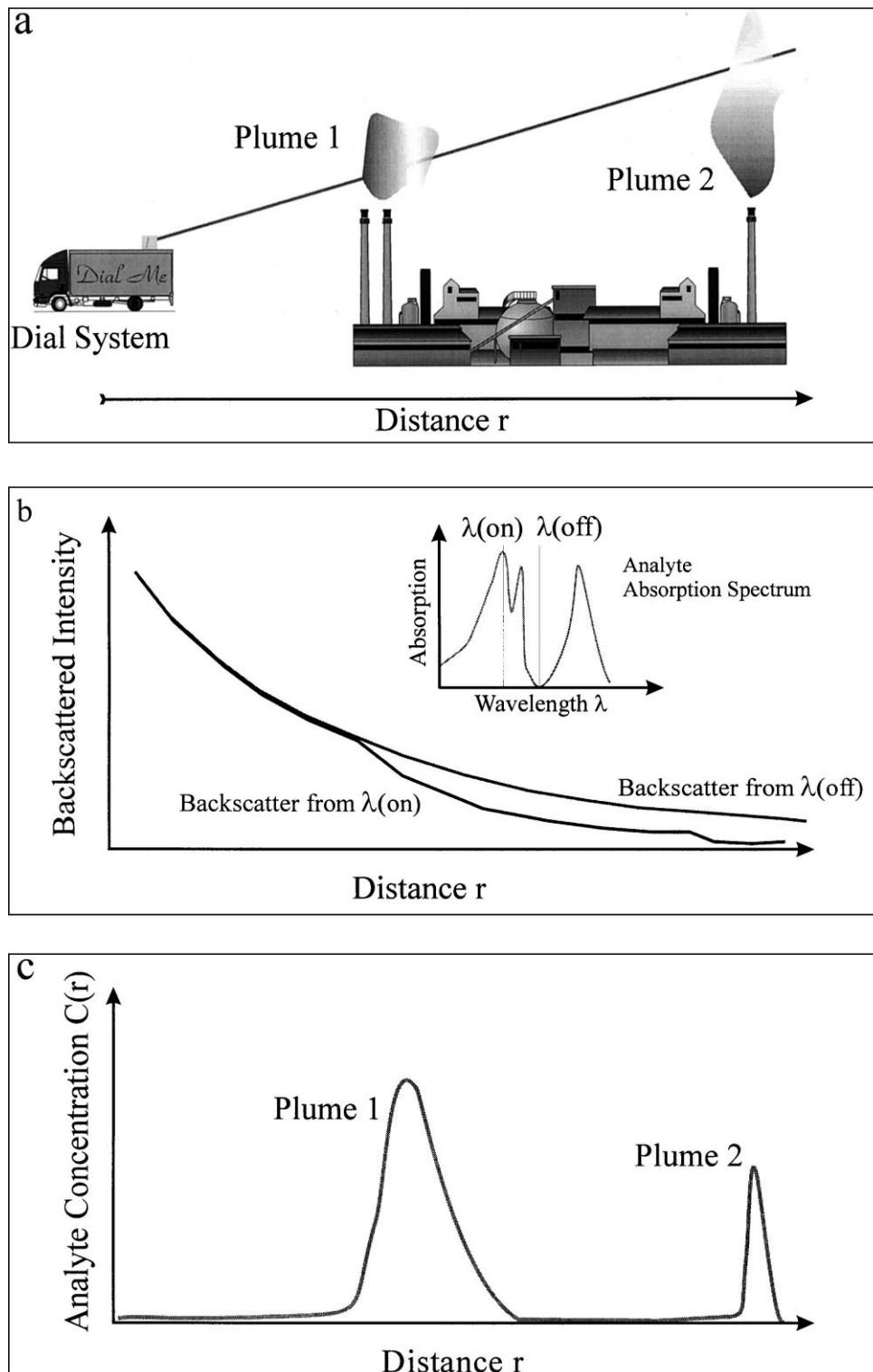


Fig. 2. A: The DIAL principle (adapted from [8]). a: Typical set-up for a DIAL measurement, b: range-resolved backscatter curves from the 'on' and 'off' laser wavelengths, c: analyte concentration as a function of distance (ratio of backscatter curves from b). B: Typical DIAL set-up based on a excimer-pumped dye laser.

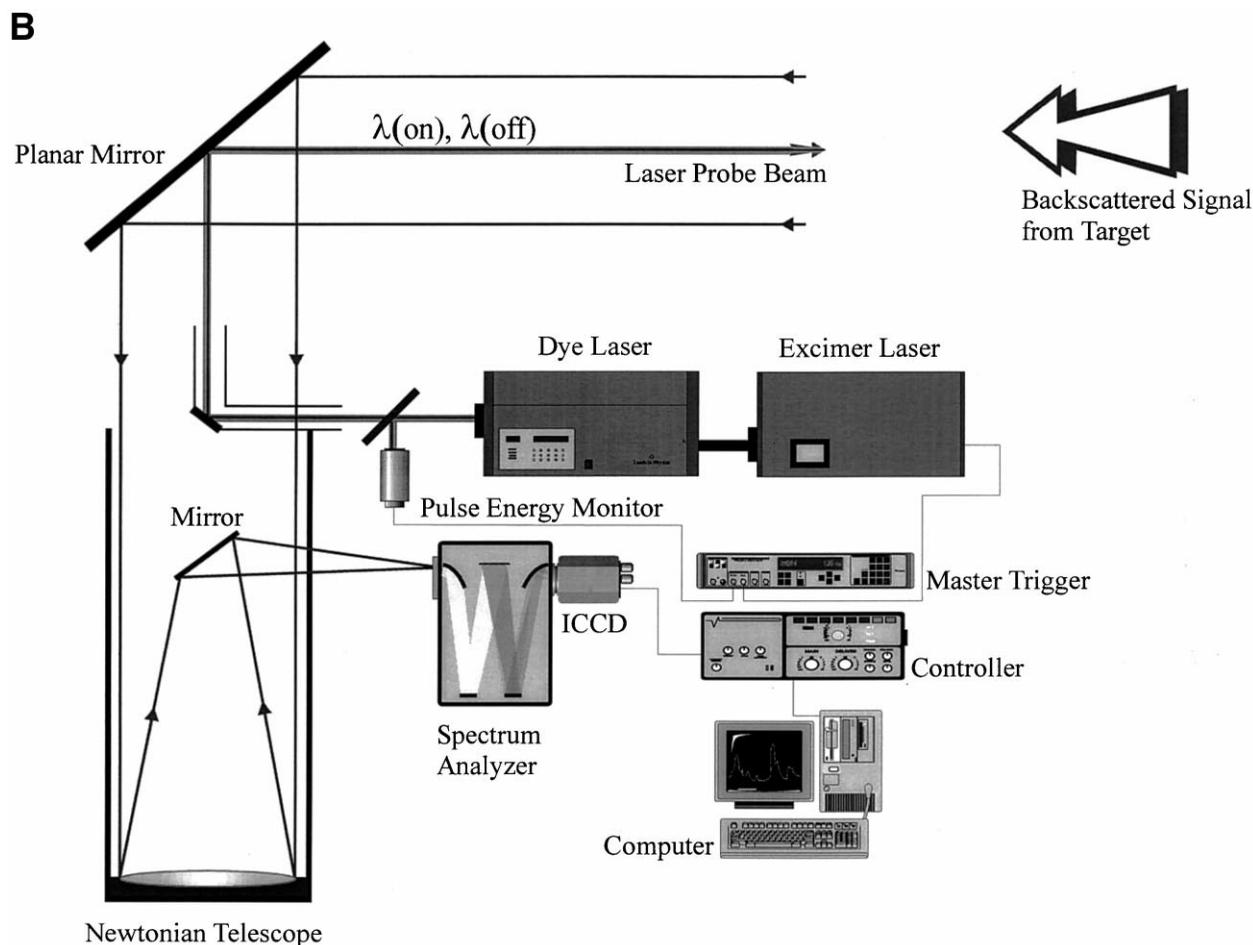


Fig. 2 (continued).

scattering provides additional information and range mapping for atmospheric particles (e.g. stratospheric dust from volcanic eruptions).

Since in this case Mie scattering involves inaccessible parameters about the particle distribution and morphology, a quantitative evaluation is often impossible. Instrumentation for DIAL is especially profiting from the developments of tunable solid state lasers, examples are diode laser-pumped Nd:YAGs for Ti:sapphire lasers or Nd:YAG-pumped Ce:LiSAF laser, a new UV vibronic laser (tuning range 280–320 nm) [11]. Other systems are based on excimer lasers (e.g. KrF laser in an oscillator-amplifier configuration) which can be favorably combined with Raman shifters [12]. Problems arise from the dynamics of the backscattered signal which often covers more than eight orders of magnitude and can be compensated by using two receiving telescopes, geometrical compression in the receiver optics, logarithmic

amplification, or in case of a PMT as detector dynode-chain ramping triggered by the laser pulse. Fig. 2B depicts a typical DIAL set-up with a Newtonian telescope as receiver optic, a small spectrograph with an ICCD as detector system, and an excimer-pumped dye laser, which can be equipped with two gratings to generate λ_{on} and λ_{off} .

For major atmospheric constituents like O_2 , N_2 , and H_2O , trace components over short ranges (some hundred meters), and vertical temperature profiles, Raman spectroscopy can be combined in a similar fashion with a LIDAR set-up [13,14].

Fluorescence LIDAR is quite limited for atmospheric research as pointed out before, but is employed successfully for solid targets in the biosphere. Pollution monitoring of oil spill, bathymetric measurements of sea depth, and algal bloom patches (see e.g. [15–17]) are probably the best known applications of fluorescence LIDAR in hydrospheric research. Airborne

measurements of maritime oil spill permit a sensitive classification and quantification of surface films in the order of $1\ \mu\text{m}$ [17] and are performed nowadays on a routine basis. The water Raman signal can be simultaneously used for normalization of the observed volume as well as the water temperature. A multidimensional approach including fluorescence lifetimes or excitation-emission spectra provides even more rapid diagnostics of water conditions (organic materials, eutrophication) and the capability to use soft modelling chemometrical methods to work on spectral signatures of analyte classes. Algae fluorescence monitoring becomes increasingly important for assessing the total marine activity, which is strongly connected to global climate and microscopic phytoplankton. It should be noted that due to the minimum absorbance of ocean water in the blue-green region of the spectrum, underwater LIDAR systems based on the second harmonic of Nd:YAG lasers [18] can additionally provide data on fish stock parameters (e.g. estimation of floating egg abundance, fish aggregation at thermal fronts).

Forrest decline studies and advanced vegetation remote sensing from LANDSAT satellites and similar spaceborne sensors based on multispectral reflectance also stimulate an increasing interest in complementary remote monitoring of plants by fluorescence LIDAR ('ground-truth control'). Plant activity and growth are affected in a complex manner by changes in environmental parameters (e.g. light, water, and CO_2). Hence, to understand these phenomena on a larger scale, remote sensing is needed to assess plant status, biomass productivity, and early detection of plant stress (e.g. due to herbicide inhibiting photosynthesis, and to biological stress, i.e. damage by mites) over medium size areas like crop fields. The observed fluorescence originates from the excitation of chlorophyll and other fluorescent leaf pigments [19,20]. It has been known for a long time that a correlation exists between the activity of the important photosystem II and the fluorescence quantum yield [21,22], i.e. the fluorescence of the antenna of the chlorophyll molecules is expected to have a longer decay time if the energy transfer to the reaction center is obstructed (Kautsky effect). For example, a five days water shortage reduces the lifetime of chlorophyll from 1 ns to 0.45 ns. However, measurement of quantum yields via the fluorescence lifetime is difficult due to variations in the total illuminated area of leaf, distance, and changes in the canopy. If the measurement is performed on an entire plant canopy, several leaves are excited by the laser spot. The total observed fluores-

cence decay is then the sum from individual leaves at different positions, i.e. the propagation time will introduce a variable delay among the decays. Canopy heterogeneity can be modelled by the reflectance signal, which can then be used to retrieve a mean lifetime. While the instrumentation for vegetation fluorescence LIDAR needs ps lasers (e.g. third or fourth harmonic of a mode-locked Nd:YAG) and fast detectors like transient analyzers, streak cameras, or ultrafast intensified CCD (ICCD) cameras for high time resolution, oceanographic applications can be performed on a nanosecond time scale with conventional Nd:YAG or excimer lasers, respectively PMTs or ICCDs.

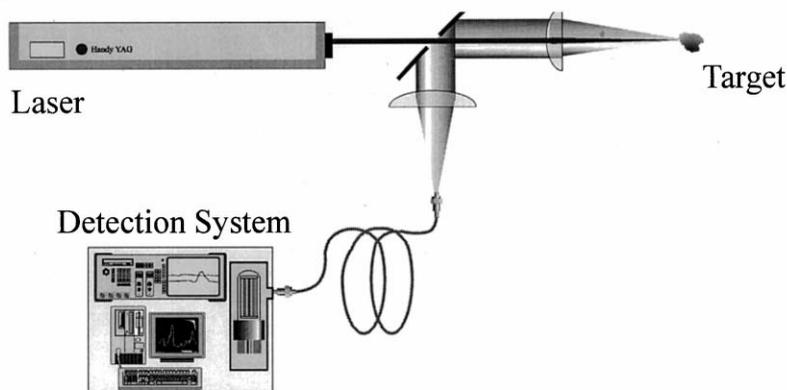
Another area of direct remote sensing which per se relies on a quantitative method working 'on-the-fly' is combustion and exhaust diagnostics [23,24]. During the last decade various remote laser methods were developed to study spatial and temporal composition, density, temperature, or flow velocities in processes like premixed gas flames, automobile engines, coal combustion, and waste incinerators. Application of techniques like laser-induced fluorescence, Rayleigh and Mie scattering, laser photofragmentation (LPF) [25], NIR emission and absorption spectroscopy can lead to considerable fuel optimization and pollutant emission minimization. Examples can vary from mapping of distributions of NO, OH, and NH_3 in a flame [24] to detection of the two-dimensional NH_3 distributions and spatial inhomogeneities in a flue gas channel [26].

3. Indirect remote sensing

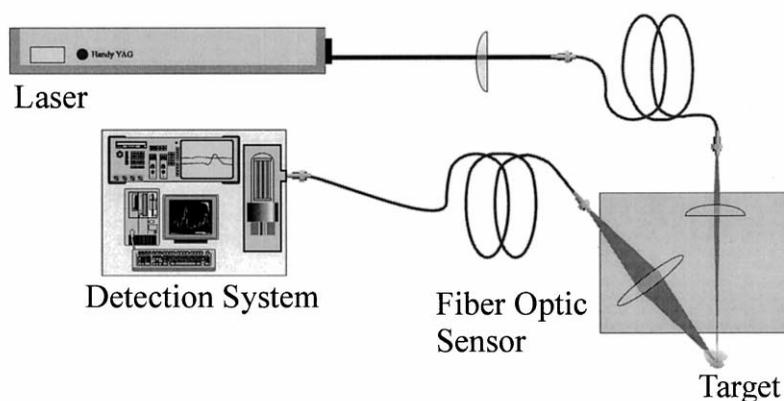
The development of fiber optical materials for long distance sensing provides an opportunity to avoid significant limitations in conventional direct laser remote sensing e.g. the $1/r^2$ reduction of radiation power. Fiber optics have revolutionized a number of fields including telecommunications, endoscopy and physical, respectively chemical sensing. The fiber's flexibility, long-range transmission, high bandwidth, small size, and imaging capability allow a huge variety of design options. Fiber optical transmission is possible for both steps of remote sensing (see above), i.e. either the laser and/or the signal from the target can be guided via fiber optics (see Fig. 3).

Nowadays, fiber optical sensor concepts exist for numerous chemical analytes (see [27,28] and references therein) and physical parameters of different matrices. Table 1 summarizes the major spectroscopic techniques suitable for fiber optical remote sensing.

Type I



Type II



Type III

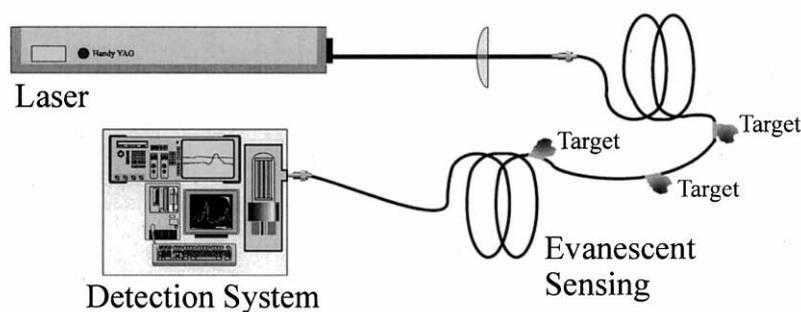


Fig. 3. Configurations for indirect fiber optical remote sensing (type I: direct laser excitation, fiber optical guided signal; type II: fiber optical guided signal and laser excitation; type III: sensing through evanescent field).

Laser-induced plasma spectroscopy (LIPS, or laser-induced breakdown spectroscopy, LIBS) is a popular approach to multielemental analysis for fast screening and process analysis. Usability of type I and II configurations for LIPS are strongly depending on the

matrix-laser interaction, i.e. the plasma threshold. Laser-induced fluorescence (LIF) is well established for environmental relevant analyte classes like polycyclic aromatic hydrocarbons (PAH) or oils and can be used also in a multidimensional mode, i.e. with

Table 1
Techniques suitable for fiber optical remote laser sensing

Technique	Analytical information	Type ^a		
		I	II	III
Laser-induced plasma spectroscopy (LIPS)	Elemental composition	++	+	
Laser-induced fluorescence spectroscopy (LIF)	Native fluorophores or fluorescent-labelled molecules	++	++	+
Raman spectroscopy	Inorganic or organic vibrational structure	++	++	
Laser photofragmentation (LPF)	Luminescence from fragments	+		
Photothermal spectroscopy (PTS)	Inorganic and organic electronic and vibrational structure	++	+	
UV/VIS/NIR absorption spectroscopy	Inorganic and organic electronic and vibrational structure	++	++	+

^aConfigurations for indirect fiber optical sensing (see Fig. 3): ++, good applicability; +, limited applicability.

time-resolved spectra and/or excitation-emission spectra. Raman spectroscopy, including surface-enhanced Raman spectroscopy (SERS) and resonance Raman techniques, is becoming attractive due to the simple and robust set-up and the wealth of chemical information. This renaissance is mainly technology driven, i.e. the commercial availability of holographic notch filters, NIR diode lasers, and sensitive CCDs has triggered new research, especially in the area of fiber optical sensors and imaging solutions. Laser photofragmentation (LPF) can be applied to small molecules, which are not easily detected by conventional spectroscopy. The method is based on a fragmentation step followed by a detection step, the later relies mainly on the luminescence of small excited fragments (e.g. SO, OH, CH, and CN) from the target molecule. Photothermal methods like photothermal deflection are especially interesting for non-destructive remote sensing, e.g. for the semiconductor industry or agricultural research (see e.g. [29]). With fiber optical microphones even photoacoustic spectroscopy can be performed remotely. Methods based on absorption, scattering and reflection either in a direct way or via the evanescent wave approach (see below) are probably the best established remote fiber optical sensing techniques these days [27,28,30], especially NIR spectroscopy became a routine tool in many industrial applications [31].

The first generation of fiber optical sensors is based on intrinsic sample/analyte properties, i.e. luminescence, Raman scattering, or absorption, where the fiber optics act simply as a light guide. Consequently, the second generation sensors ('optrodes') implement a sensor chemistry for enhancing selectivity and sensitivity to address all kinds of analytes. Early sensor

chemistry approaches were based on chelates (for example for metal ion sensing [32]), indicator dyes, and other complexing agents established in wet chemical analysis. These agents were mostly immobilized in polymeric supports or covalently bound to the fiber surface. Emerging sophisticated concepts are based on molecular imprints, tailored probes and switches (see e.g. [33]), or sol-gel chemistry.

Molecular imprinting produces stable synthetic polymers that possess selective recognition properties due to sites within the polymer matrix which are complementary to the analyte (shape, functional groups). Thus, these polymers can provide high affinity constants and selectivities comparable to natural systems leading to biomimetic sensor approaches [34]. Sol-gel materials provide a microporous support matrix in which analyte-sensitive species are entrapped and into which smaller analyte molecules may diffuse. Especially advantageous is this technique for sensitive biological sensing chemistry (antibodies, enzymes), which can be trapped without chemical modification. The gel can be tailored with considerable flexibility and ease of production allowing monoliths, thin films, as well as more elaborate structures [35].

In addition to being a simple light guide, fiber optical sensors open up also new sensing schemes not possible in direct remote sensing. Sensing is not limited to the tip of the fiber, but also possible along the fiber via the evanescent field around the fiber cladding [36]. With techniques like time domain reflectometry a high spatial resolution can be achieved for detection of changes in absorption, fluorescence, or refractive index, which makes this approach valuable for monitoring tasks at large sites, for example at waste disposals or refineries. The use of fiber optical imaging

bundles, which consist of 300–6000 independently fused fibers (typical diameter about 200 μm), permit a combined chemical sensing and imaging. The dimension of the bundle combined with a suitable sensing chemistry permits the utilization of μm -sized sensing spots, i.e. a chemical sensor array. Other possibilities arise from the shaping of the fibers, i.e. due to the different materials of the fiber core and cladding a single fiber reacts differently in a wet-etch process. This allows producing small wells for trapping analytes and reagents similar to a microtiter plate [37], or producing an imaging bundle for near field microscopy.

Fiber optical sensors can be produced in a wide variety of geometries like single, dual, or multiple fiber design, lens-coupled, or beveled tips suitable for different spectroscopic techniques as well as different matrices and analytes. In combination with two-dimensional detectors like CCD cameras and imaging spectrographs multiple fibers can be simultaneously used, so that fibers carrying different wavelengths or signals from spatially separated sensors (e.g. in a soil column or around a laser plasma) can be observed at the same time.

Strongly connected to remote sensing are mobile platforms for remote sensing, either direct or via fiber optical interfaces. After the success of the 'Sojourner' during the Mars Pathfinder mission, there is probably not much doubt left that robotic systems or rovers are a suitable platform for remote sensing. Future planetary missions will employ laser techniques (Raman spectroscopy seems to be the most promising candidate), but we will also see platforms for more earthly missions, e.g. robotic submarine systems with telepresence and micro robots in contaminated areas. Already at work are robots for taking subsurface samples below waste disposals or inspecting waste water tubes [38]. At present, most instruments, especially LIDAR and DIAL systems, are employed on airborne platforms (see e.g. [39] for a sophisticated OH radical measurement aboard a NASA ER-2 aircraft) or balloons. An interesting alternative platform for urban sensing are solar-driven zeppelins, as they can be operated securely in densely populated areas, have low costs and very long flight times, and provide a good spatial and temporal resolution due to their low air speeds. A popular approach to remote sensing of terrestrial subsurface environments, i.e. soil and aquifer systems, is a cone penetrometer [40]. Cone penetrometer testing has emerged as an effective means to introduce fiber optical probes for in situ analysis of contaminants in soil and ground-

water matrices. A variety of in situ chemical sensors based on Raman, LIF, LIPS or NIR spectroscopy has been developed that can be driven into the subsurface to explore the vertical and horizontal extent of contamination.

4. Conclusion and outlook

Driven by technological advances, increasing global environmental awareness, sustainable development, and improved quality control measures, laser remote sensing is a fast developing area of laser spectroscopy with numerous applications. While direct sensing will profit in future immediately from improvements in laser technology, indirect sensing is mostly application oriented, which involves mostly process analysis and other production related analytical tasks. This later development follows the general paradigm shift in analytical spectroscopy where instrumental sensitivity seems to be levelled off, and an orientation to production, process control, and monitoring seems to take place. To reduce the total cost of ownership for lasers and detectors the future will probably bring a combination of different techniques by way of fiber optical interfaces. For example a set-up based on a Nd:YAG laser with an intensified CCD could be employed via fiber optical sensors for a sensitive screening of large contaminated sites with both LIF and LIPS allowing to access analytes as different as oils and heavy metals. Although this approach will probably not result in a universal laser spectrometer, this 'hyphenated spectroscopy' could give new insights into complex matrices and analytes, including also a multiplexed and distributed sensing. Another inherent advantage of hyphenating spectroscopic techniques stems from the combination of approaches which provide independent ('orthogonal') dimensions of information. For non-destructive analysis or ultra-trace analysis where a wet chemical dissolution and separation are no longer possible, 'orthogonal' laser remote sensing is the only viable alternative.

Further advantages are foreseen for samples where analyte distribution (the inhomogeneity of the sample) corresponds to the dimensions of the fiber optical interface. This 'nano meets macro' development can be observed for optical near field spectroscopy [41], like near field fluorescence or near field surface-enhanced Raman spectroscopy (SERS) [42] and fiber optical confocal set-ups [43] for fluorescence and Raman spectroscopy.

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