

***AN OVERVIEW OF SOME
METHODS FOR LASER
DIAGNOSTICS OF FLAMES***

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1. INTRODUCTION

This write up presents the basic concepts and some current techniques used in the study of combustion processes.

The aim is to stimulate international co-operation of scientists and technicians in this field with two purposes: to improve the efficiency of devices and systems where burning of any type of fuels is used, and to reduce the polluting effects of this. Therefore the scientific and technical approach is mainly addressed at investigating those processes that characterise flames.

For those who may wish to expand their information on particular issues that are herewith overviewed, a list of references is attached.

2. PREFACE

2.1. Thermal Energy - The burning question

The world is using an increasing amount of energy. Both the developed and less developed countries are demanding larger shares of world energy resources. At the same time, it is fully recognised that such resources are limited. Most are fossil fuels, formed underneath the surface of the Earth millions of years ago. They will not easily be replenished!

The burning of fossil fuels is one of the most important ways in which the world's energy needs will continue to be met. For example, fuels are burned in engines and in industrial processes (furnaces, boilers).

Sometimes fuels are wastefully and, possibly dangerously combusted in accidental fires and explosions. However, there are worries, too, about the emission of pollutants, both gaseous and particulate, from the deliberate burning of fuels for legitimate industrial purposes.

Probably the most plentiful source of thermal energy is *coal*. This is essentially carbon but contains a number of impurities such as nitrogen, sulphur, and chlorine. Another group of fuels is the *hydrocarbons*. These may be liquid or gaseous and are compounds of hydrogen and carbon in different proportions.

Coal is well suited to industrial processes such as power generation, but it is difficult to store and transport. Hydrocarbons provide convenient automotive power, whether on the road or in the air.

2.2. The combustion process

The key features of the combustion process are that a fuel and an oxidiser mix in suitable proportions and are ignited. The oxidiser is usually (but not always) oxygen in the air. In the process of combustion, oxides of carbon (CO or CO₂) and hydrogen (H₂O) will be formed.

The basic reaction during combustion is:

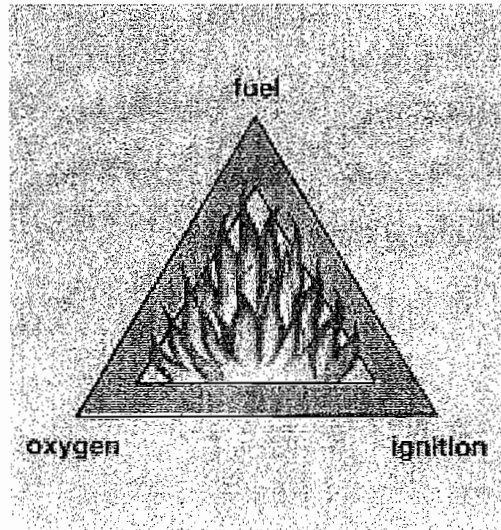
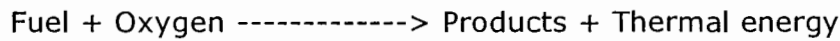


Figure A: The fire triangle

The flammable mixture of fuel and oxygen may be prepared in advance of the ignition process. An example of this is the carburettor and inlet system of a spark ignition engine. Alternatively, mixing may occur almost simultaneously with ignition, as in an aircraft jet engine.

2.3. Flames

Flames look quite different depending on how well mixed are the fuel and oxygen. Figure B shows, on the right, a bunsen burner with the vent open. The flame is well aerated, it is blue and luminous. The bunsen burner on the left has the vent closed. Oxygen supplied to the fuel only at the top of the tube (from the atmosphere). The air supply is restricted, combustion of the fuel is incomplete and a white luminous flame results.

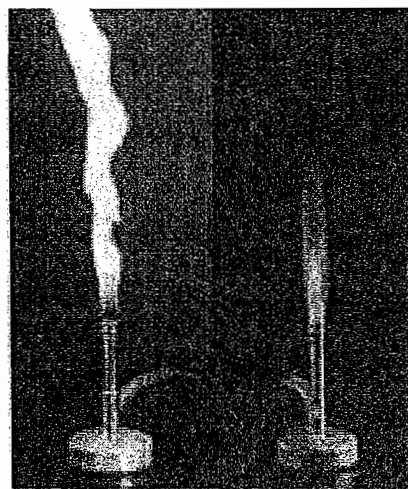


Figure B: Bunsen burner with vent closed (left), vent open (right)

Poorly aerated flames produce a lot of soot and smoke. They are inefficient because unburnt carbon will be deposited on surfaces above the flame and will restrict the transfer of heat. Another consequence of a poorly-aerated flame is the emission of carbon monoxide (CO), which is a highly poisonous gas.

Sulphur present in the fuel can result in the production of sulphur dioxide (SO₂). However, it is not always the fuel that is the major source of pollution. The nitrogen which comprises roughly four-fifths of the air commonly used as the oxidiser can itself be oxidised in the flame to produce oxides of nitrogen (NO_x).

If engineers are to design efficient, clean combustion systems to provide all the thermal energy we need, they must have a good understanding of the physics and chemistry of combustion.

2.4. Combustion research

In order to promote the efficient use of fuels, a lot of research has been carried out into combustion processes. This has required the use of a large number of analytical and research techniques. Many of these depend, for their operation, on fundamental principles of physics and mathematics. Investigations include the use of:

- lasers;
- high-speed photography;
- calorimetry and pyrometry;
- mathematical modelling.

2.5. Laser techniques

High temperatures and pressures in the combustion chamber make it a most hostile environment. Laser and other optical probes which cannot be destroyed in the combustion flow and do not disturb the flow are particularly important. Laser techniques have been used in a number of measurements, such as:

- air/fuel flows and velocities;
- combustion processes;
- identification of molecular components, etc.
- pollutant formation;
- droplet and particle sizes;

2.6. Mathematical modelling

Mathematical models, aided by computer systems, have been developed in a number of areas to test ideas. It would often be too expensive, or dangerous, to carry out a test at first hand – just to confirm a prediction! Here are two extreme examples. It would be much too costly to build a full-size furnace to determine how it would function in practice. It would be far too dangerous to have an explosion on an oil rig – just to see what happens! In both cases, scientists can devise mathematical models to simulate the real event.

In recent years, mathematical models have been developed for industrial burners and combustion chambers. Computers have been used to solve the basic equations for turbulent flow, heat transfer, combustion, and pollutant formation.

3. DIAGNOSTICS OF COMBUSTION PROCESSES

3.1. Background

Research in the field of combustion has been intensified recently because of the appreciation of the need for efficient combustion combined with low pollution. In order to obtain a deeper understanding of combustion processes it is necessary to perform the study on a molecular level. Laser spectroscopic techniques provide unique possibilities for non-intrusive measurements on the extremely aggressive media that burning or exploding gases constitute. Because of the unique properties of laser beams, both high spatial and temporal resolution can be achieved. Before we describe some measurement techniques we will give an elementary background to combustion processes.

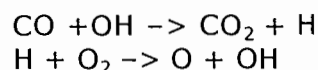
A detailed understanding of combustion must start with simple processes such as hydrogen, methane or acetylene combustion in oxygen or air. Normal liquid hydrocarbons are considerably more complex and wood or coal combustion can hardly be attacked on a molecular level. Below we give some "effective chemical reactions leading to a transformation of fuel and oxidant into carbon dioxide and water. The processes are strongly exothermic, which is, of course, a common feature for combustion processes (Table 1).

Flame release	Effective reactions	Temperature (K)	Energy (J/g)
H ₂ /O ₂	2H ₂ +O ₂ ->2H ₂ O	3,100	24,000
CH ₄ /O ₂	CH ₄ +2O ₂ ->CO ₂ +2H ₂ O	3,000	10,000
C ₂ H ₂ /O ₂	2C ₂ H ₂ +5O ₂ ->4CO ₂ +2H ₂ O	3,300	12,000

Table 1: Some effective chemical reactions in fuel combustion

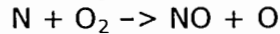
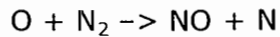
Combustion occurs with a large number of intermediate steps and even simple processes. With computer simulations it is possible to describe the interaction between the reactions, and concentration profiles can be calculated. In order to perform the computer calculations it is necessary to know the rate constants for the individual elementary reactions. Comparisons between theory and experiments are best made for a flat, premixed flame, which in its central part can be considered to have only one-dimensional (vertical) variation, allowing computer calculations to be performed comparatively easily.

As examples of important elementary processes we give the reactions

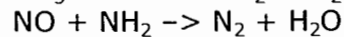
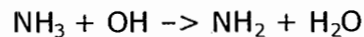


Reactive molecular fragments or radicals, such as OH, H and O are very important in combustion. The combustion zone of a stoichiometric CH₄/O₂ flame contains about 10% OH and 5% each of H and O. In the second of the two reactions given above the number of radicals is doubled. A fast increase in radical formation frequently leads to explosive combustion. Because of the high reactivity of radicals they cannot be measured by probe (extraction tube) techniques, since wall reactions immediately eliminate them. Thus, laser techniques are particularly valuable for radical monitoring. Pollution formation in

flames should also be considered. Nitrogen and sulphur oxides, incompletely burnt hydrocarbons and soot particles form important pollutants. It is of the utmost importance to understand which elementary reactions form and eliminate pollutants. The formation of nitric oxide is reasonably well understood. At temperatures above 2000 K the nitrogen in the air is attacked:



The two reactions constitute the so-called Zeldovich mechanism. NO is then oxidized to the toxic NO₂ by the oxygen in the air. NO can be eliminated from the post flame gases by the addition of NH₃. The following reactions occur



In the case of sulphur oxide SO it is immediately further oxidized to SO₂ in the flame. It seems that a reduction of SO₂ can only be obtained by using low-sulphur fuels. SO₂ and NO₂ are further oxidized to H₂SO₄ and HNO₃ in the atmosphere with subsequent acid rain formation.

Soot formation is the subject of many studies. Soot formation is enhanced at high fuel/air mixing ratios (rich flames) when using hydrocarbons with comparatively little hydrogen, and for bad mixing conditions. Since available fuels will become successively poorer in hydrogen, soot formation will become an increasing problem. The chemistry of soot formation is not well understood. Many processes, including polymerization of simple hydrocarbons to heavier ones and reactions with polyaromatic hydrocarbons, may be important.

Laser techniques have a great potential for studies of microscopic as well as macroscopic combustion in flames and engines. Combustion diagnostics with lasers was discussed in several reviews. We will here give examples of measurements of concentrations and temperature (flame kinetics) using fluorescence, Raman and coherent Raman techniques. In practical combustion systems turbulence is extremely important and we will also briefly discuss laser techniques for flow and turbulence measurements.

3.2. Laser-induced fluorescence and related techniques

In laser-induced fluorescence (LIF) experiments a laser is normally tuned to an allowed dipole transition from a lower to an upper state of the species under consideration, and the fluorescence light that is released upon the subsequent decay is observed. We will start this section by considering the corresponding spontaneous emission process. At the high temperatures in a flame, upper levels become thermally populated and a natural emission giving the flame its colour occurs. In Fig. 1 part of the emission spectrum from a C₃H₈/air Bunsen burner flame is shown, featuring strong bands due to the radical C₂. This emission was described by Swan as early as 1857 in one of the earliest molecular spectroscopy experiments. C₂ is responsible for the blue-green light from the lower parts of hydrocarbon flames. Schematic energy-level diagrams for C₂ and OH with the wavelengths of the individual bandheads are depicted in Fig. 2.

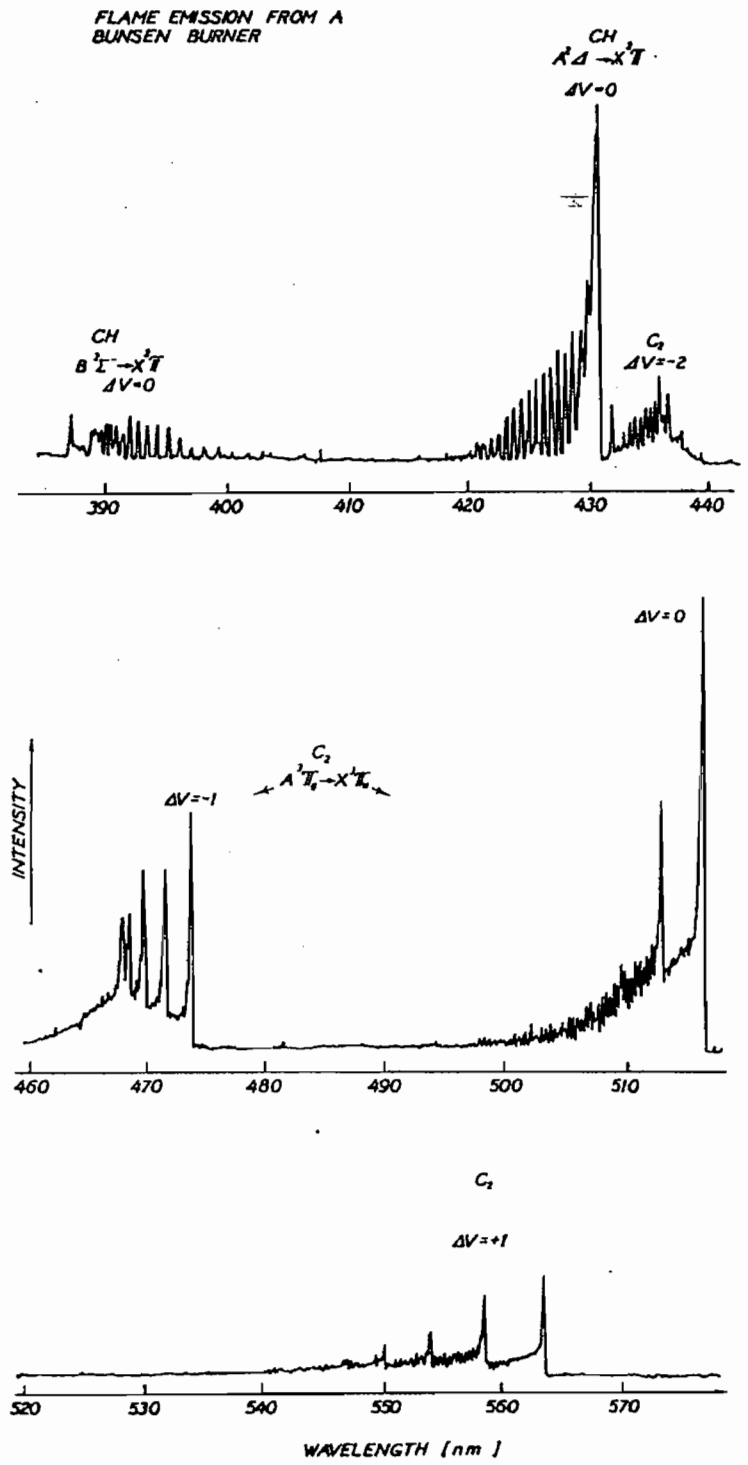


Figure 1: Emission from a propane/air Bunsen burner flame

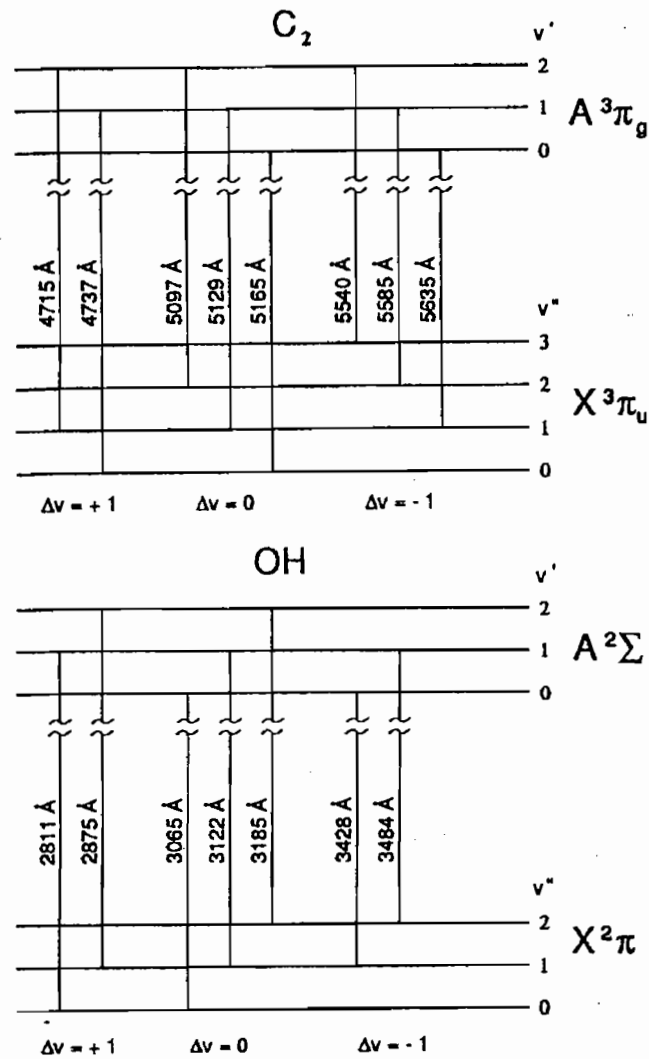


Figure 2: Energy level schemes for the C_2 and OH molecules

Hydrocarbon flames also exhibit strong bands due to the CH (~ 390 and 430 nm; especially in the flame front) and the OH radicals (~ 300 nm). In sooty flames the strong yellow light is due to incandescence of soot particles.

An experimental set-up for studying LIF in flames is shown in Fig. 3. The output of a Nd: YAG pumped dye laser can be frequency doubled and, if needed, the doubled output can be mixed with residual $1.06 \mu\text{m}$ radiation to achieve still shorter wavelengths. The beam is directed through the flame and the fluorescence can be spectrally analysed with the spectrometer shown in the upper part of the figure. The lower part of the figure shows how a diode-array detector can be used to obtain a one-dimensional image of the distribution of a radical across a flame. The streak of LIF is imaged onto the detector, which is gated by the laser firing. Figure 4 shows distributions of OH fluorescence at various heights in a CH_4/O_2 flame. A single 10 ns pulse is used for each recording.

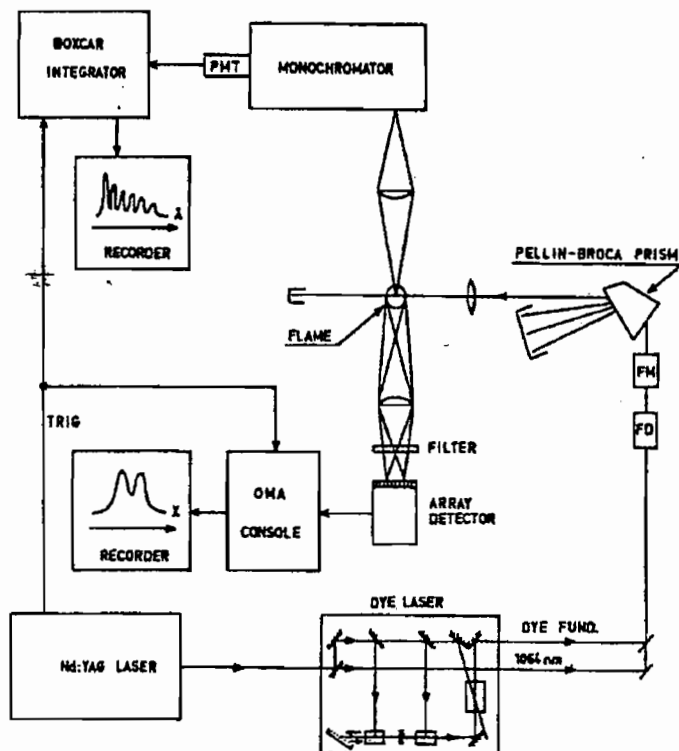


Figure 3: Set-up for spectrally resolved and imaging laser-induced fluorescence studies

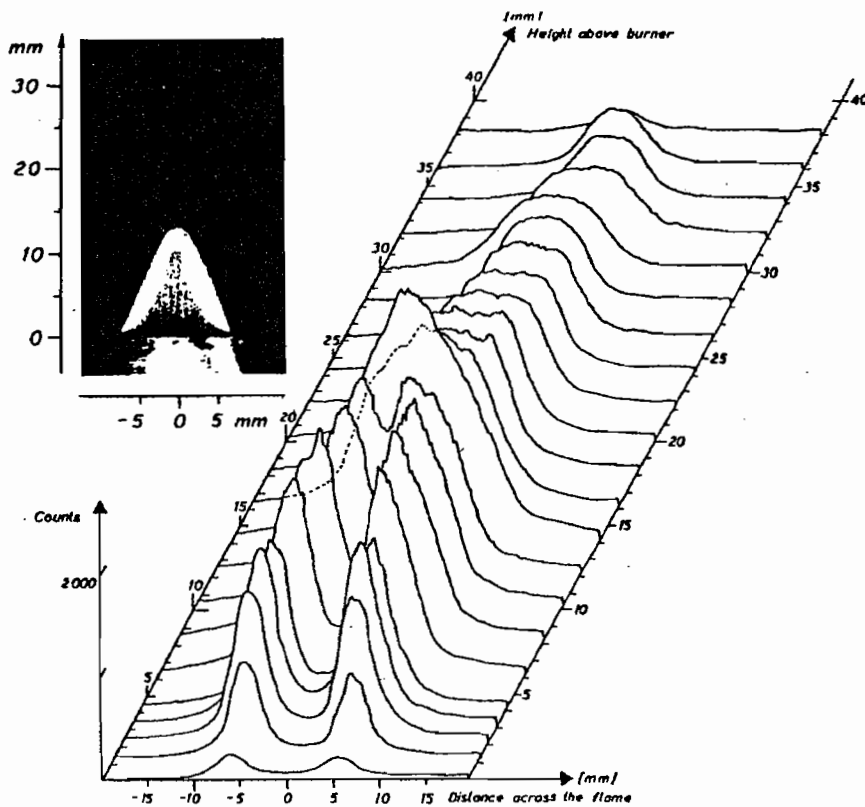


Figure 4: Spatial distributions of the OH radicals in a CH_4/O_2 flame. The excitation wavelength was 281 nm and the detection wavelength 308 nm

Using matrix detectors or Vidicon tubes, LIF imaging techniques can be extended to two-dimensional imaging. Many flame species, including OH, C₂, CN, and CH and NO, have been studied using single-photon LIF. Several important flame species have their single-photon excitation wavelengths in the VUV region where the flame gases absorb and tunable lasers are not readily available. It is then possible to use two-photon or even three-photon excitation.

Hydrogen atoms can be detected observing H α , or H β emission from the $n = 3$ or 4 levels following two- or three-photon excitation or step-wise excitations. CO molecules are also best-detected using two-photon excitation. The excitation of flame species to an upper level can also be detected by other means. Optogalvanic spectroscopy and photoacoustic spectroscopy employing pulsed lasers have been used. The former method is not non-intrusive in nature because of the need for electrodes. The latter technique utilizes the local pressure increase following excitation, and a microphone close to the flame is used for detection. Spatial resolution is limited since the signal is collected along the laser beam. Ordinary absorption measurements clearly constitute the best examples of such line-of-sight measurements. Diode lasers, as well as dye lasers, have been used successfully in flame absorption measurements. With tomographic techniques, similar to those used in medical X-ray imaging, spatially resolved information can be obtained from integrated absorption measurements in different directions through the flame. By using tomography three-dimensional information can also be obtained utilizing thermal flame emission and interferometry.

Laser beam deflection can also be used to detect optical resonance. In the region of optical excitation the index of refraction of the gas changes and a probing laser beam (frequently a He-Ne laser beam) crossing the excited region will be deflected.

3.3. Raman spectroscopy

Because of its insensitivity to quenching (the lifetime of the virtual state is $\sim 10^{-14}$ s), Raman spectroscopy is of considerable interest for quantitative measurements on combustion processes. Further, important flame species such as O₂, N₂ and H₂ that do not exhibit IR transitions can be readily studied with the Raman technique. However, because of the inherent weakness of the Raman scattering process only non-luminous (non-sooting) flames can be studied.

Extractive Raman measurements on stable flame species can readily be performed. Here gases are transferred from the flame through a thin tube to the scattering cell of a laser Raman gas analysing system. In Fig. 5 Stokes Raman spectra, obtained using an Ar⁺ laser operating on the 488 nm line are shown for the lower and upper part of a C₃H₈/air Bunsen-burner flame are shown. The conversion of fuel and O₂ into CO₂ and H₂O, constituting the over-all combustion process, is clearly demonstrated. The H₂O signal is prevented from increasing by a water vapour condenser in the gas feed-line. Soot particles, which give rise to a broadband LIF background, are also filtered away. The lower trace also displays signals due to CO and H₂, gases that will mostly burn up higher up in the flame. The weak signals in the shoulder of the strong Rayleigh line in the lower trace are due to pure rotational Raman transitions in the H₂ molecule, which, because of its small mass, has an exceptionally large rotation constant B. The main hydrocarbon signal at about 570 nm has many components corresponding to

slightly different C-H stretch vibrational frequencies. Overtone and combination bands are also observed at smaller Raman shifts.

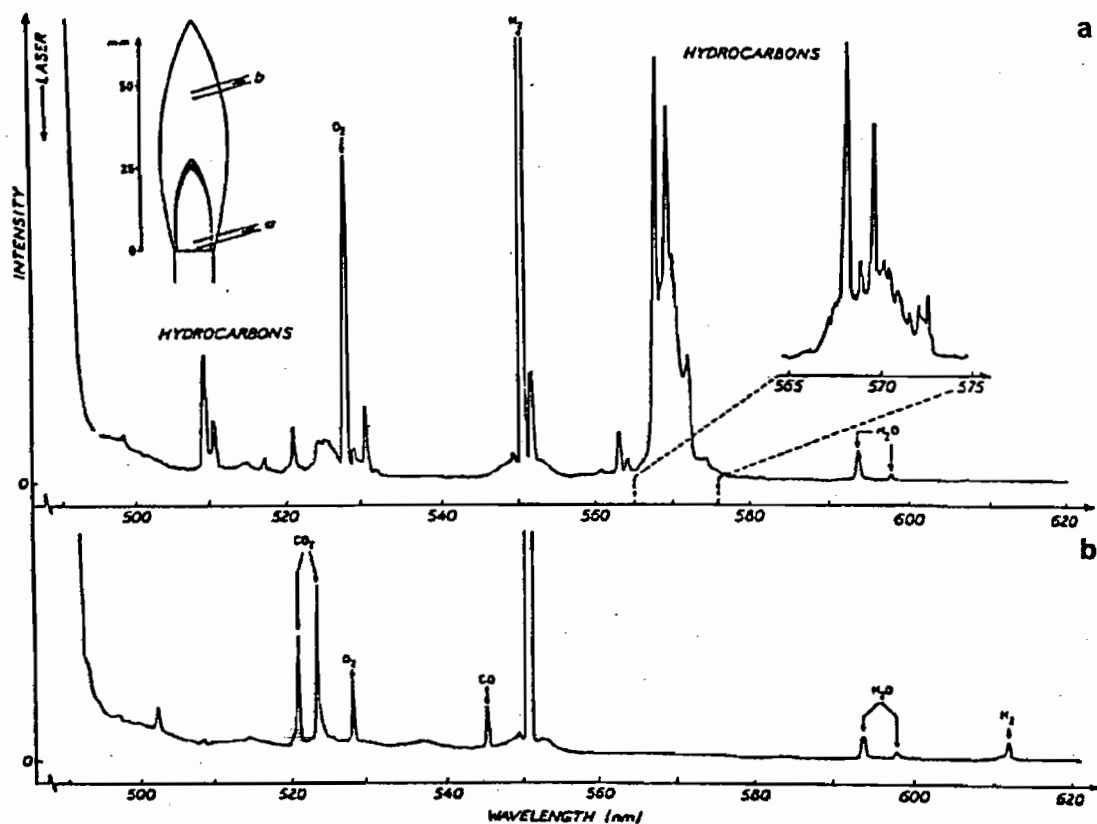


Figure 5: Raman spectra of extracted flame gases at two different heights in a Bunsen burner flame

Using pulsed lasers and gated detection electronics, Raman measurements can also be performed for major species in flames that do not contain too many particles. Temperature measurements can then also be made using the Stokes/anti-Stokes signal asymmetry.

3.4. Coherent anti-stokes Raman scattering

CARS spectroscopy is of particular interest for combustion diagnostics because of the strong signal available as a new laser beam emerging from the irradiated gas sample. Thus CARS is largely insensitive to the strong background light that characterizes practical combustion systems such as industrial flame and internal combustion engines. We recall that the spectroscopic information is contained in the third-order susceptibility term $\chi^{(3)}$. This term given by the sum of a complex resonant term, (proportional to the concentration of the studied molecule) and a non-resonant background term. Thus, when the expression $|\chi^{(3)}|^2$ governing the signal strength is formed interference between the resonant signal and the background occurs, resulting in asymmetric signals, very much like the case of polarization spectroscopy. For a molecule with vibrational and rotation levels, whose populations are temperature dependent, there are many closely lying resonances and the signal shape has to be calculated with a computer programme.

Recently, degenerate four-wave mixing has also been applied to combustion studies. In this technique the sensitivity of LIF is combined with the advantages of a coherent signal beam characteristic to CARS.

3.5. Velocity measurements

Laser Doppler Velocimetry (LDV) is an important non-spectroscopic laser technique for intrusion-free measurements of velocities in liquid or gaseous flows, including combustion flows. In this technique two laser beams (frequently from an Ar^+ laser) are crossed at a small angle in the medium to be studied, as shown in Fig. 6. A standing interference pattern with bright and dark fringes is then formed. If a small particle being carried by the flow passes the interference pattern it will produce periodic glimpses of light that can be detected by a photomultiplier tube. The frequency f_D of the periodic signal (the Doppler burst) can be analysed by Fourier transformation and the velocity v can then be determined since the fringe separation d is given by the laser wavelength λ and the beam crossing angle θ . We have

$$v = \frac{\lambda f_D}{2 \sin(\theta/2)}$$

$$f_D = \frac{2v}{\lambda} \sin \frac{\theta}{2}$$

Figure 6: Principle of Laser Doppler Velocimetry (LDV)

Clearly, if the particle is larger than the fringe separation, the contrast is reduced and thus LDV measurements also provide information on the particle sizes.

With one pair of crossing laser beams only one velocity component in the flow field can be determined. Furthermore, it is not possible to determine the sign of that velocity component since either flow direction produces the same Doppler burst. However, by frequency shifting one of the crossing beams with a Bragg cell (acousto-optic modulator), moving interference fringes are produced. It is then possible to decide from which direction a particle passed the interference field by noting if the burst frequency was shifted upwards or downwards from the frequency from a fixed object. A second velocity component can be measured by using two additional laser beams propagating in a plane perpendicular to the first laser beam plane and crossing at the same point. In order to be able to distinguish the Doppler burst from this interference pattern, different wavelengths are used for the laser beams. Thus, it is customary to use the 5145 \AA line of an Ar^+ laser for one velocity component and the 4880 \AA line for the other. The elastically scattered light is then detected through sharp interference filters in front of individual photomultiplier tubes. The third velocity component (in the

direction of the bisectrix of the crossing laser beams) is harder to measure. However, by using three crossing beams in the same plane and extracting the information pair-wise from the central beam and one or other of the external beams, the third velocity component can be projected out, although at lower accuracy. When the scattered intensity is low, e.g. because of small particles, it is still possible to extract velocity information from the time correlation of the recorded individual photons. Special autocorrelation techniques have been developed for optimum information extraction. It is frequently necessary to seed the flow with small particles. These particles do not necessarily truly follow the gas flow, which constitutes a complication with this technique.

We should conclude this description of LDV techniques with a comment on its common name. In our description of the technique we have not used "Doppler language" at all but rather expressed the observed phenomena in terms of a spatial interference pattern. However, we can alternatively consider the Doppler shift in the scattered light frequency caused by the motion of the particle. This shift is detected as a beat frequency against the light scattered from the other crossing beam. The beat frequency corresponds exactly to the frequency produced by a particle crossing the fringe pattern, so the two pictures are actually equivalent.

We will also consider a technique for velocity measurements, which is based on Doppler shifting of absorption lines of atoms or molecules in the flow. A narrow-band cw laser is tuned to a frequency ν in the shoulder of a Doppler-broadened absorption line, where fluorescence light of half the maximum intensity can be induced. If the gas molecules are displaced by a flow the whole Doppler-broadened profile will shift in frequency. Depending on the direction of this flow the fluorescence light intensity will increase or decrease. In order to measure the presence of a net flow, two alternate laser beams are sent through the medium in opposite directions. The fluorescence intensities are balanced out for a static gas, by adjusting the relative powers in the two beams. If the gas is moving, a modulation will be observed at the frequency of the beam direction shifting. By normalizing the signal it becomes independent of the number density of the molecules. We have

$$v = \frac{c I_{\text{left}} - I_{\text{right}} g(\nu)}{\nu I_{\text{left}} + I_{\text{right}} dg/d\nu}$$

where I_{left} and I_{right} are the two detected fluorescence intensities and $g(\nu)$ is the value of the line shape function at the chosen frequency ν .

4. A LABORATORY ON LASER DIAGNOSTICS IN DISCHARGES AND FLAMES THROUGH DETECTION OF CHARGES

An important application of laser spectroscopy is the determination of impurities present in several gaseous systems, such as for instance furnaces, combustion engines, burners and every other system, important in the determination of pollution.

Among the different laser methods utilized for this purpose a special role is taken by the methods based upon the detection of ions, namely through the current produced through the selective laser excitation of definite atomic or molecular species. The multiphoton excitation represents one method of selective excitation. Other important methods of selective ionization are based upon the energy exchange between excited atoms during the collisions. This particular method for the production of electric charges through laser excitation constitutes the basis of the optogalvanic detection of atoms and molecules.

The principle of the method is very simple: through a tunable laser a discharge is illuminated, using a resonance radiation with an optical transition between levels belonging to the species under investigation, and which are populated by the discharge, even at low concentration. One then observes, using an appropriate coupling electric circuit, the electric current passing through the discharge, or between two-detection electrode. The presence of a resonant radiation, resonating with a particular atomic or molecular species present in the system under investigation produces, through several different processes such as collision transfer, Penning ionization, associative ionization, etc, the electric current signal to be detected. The method shows a great sensitivity as compared with other diagnostic methods, more conventional, based upon the detection of photons, as a result of the experimental advantage of the detection of electric charges.

There are several advantages of the optogalvanic detection, and it is impossible to list them in a complete way; however, the optogalvanic techniques have allowed to obtain several spectroscopic information, such as level positions (atomic and molecular), both for states close to the ground state and for very high states, close to the ionization level. Moreover, the observation of the ionization produced by the resonant laser radiation has led to the understanding of different collision processes and to the measure of energy transfer cross sections. These characteristics of the application of the method to both measurements of fundamental physics as well as the utilization to the control of industrial processes make a very attractive feature the development of a new laboratory on this theme.

Moreover the study of optogalvanic phenomena is based upon a very simple instrumentation, which is therefore possible also for optics laboratories which possess very limited equipment.

One can use even lasers of a very low power to produce a measurable ionization in discharges. The use of a laser source of medium power allows the investigation of a large variety of systems. The observation of optogalvanic processes produced by an impulsed laser gives the possibility of applying techniques of temporal detection to the phenomenon of ionization, and therefore to obtain additional information on the ionization processes. Moreover, the impulsed lasers pumped by nitrogen lasers are very cheap, easy to operate and simple to maintain.

Utilizing discharges in commercial lamps it is possible to start to analyse the phenomena of impulsed ionization on some simple atomic and molecular systems, such as rare gases, where the optogalvanic observation has already produced positive results, and where some features of the temporal evolution of the phenomenon have not been fully investigated yet. In order to study the ionization in more complex systems, such as heavy metals or other elements

relevant in the pollution, it is required to build up discharge systems, which are not commercially available. It is therefore necessary to have the presence of mechanical technicians and glass blowers.

The list shown below covers all the necessary expenses, required to fully equip a laser spectroscopy laboratory, assuming that no previous instrumentation is available.

For the operation of an optics laboratory one needs to use appropriate optical tables; it is not needed to acquire expensive optical tables, such as those produced by the firms NRC in the USA or CINEL in Italy; good quality optical tables can be built easily by small mechanical workshops. Moreover, since in the experiments of optogalvanic spectroscopy it is possible the use of materials strongly reactive, it is necessary to have a chemical cowl over the optical table.

- Nitrogen pulsed laser (type LN 1000 by PAR)
- Dye laser (type LNA 107 by PAR, 4 Angstrom band width)
- Box car detection system
- Oscilloscope 500 Hz band-pass
- Fast amplifiers
- Vacuum system (rotary + diffusion pumps)
- Monochromator
- He-Ne laser
- He-Ne tube
- Hollow cathode lamps
- Regulated power supplies:
 - For high voltage
 - For hollow cathode
 - For integrated circuits
- Optical mounts
- Lenses
- Spare parts

The acquisition of the dye laser pumped by the nitrogen laser represents the most expensive part of the equipment for the experiments, which have been described. If one intends to set up three different optics laboratories, it is possible no proceed to the acquisition of a very high quality laser source, to be utilized part-time in the various experiments.

References

1. S. Svanberg: *Atomic and Molecular Spectroscopy* 2nd edn. (Springer, Berlin, Heidelberg 1992)
2. L.J. Radziemski, R.W. Solarz, J.A. Paisner (eds.): *Laser Spectroscopy and its Applications* (Dekker, New York 1987)
3. H. Medin, S. Svanberg (eds.): *Laser Technology in Chemistry*, Special issue. *Appl. Phys.* B46, No.3 (1988)
4. W.C. Gardiner, Jr.: The chemistry of flames. *Sci. Am.* 246/2, 86 (1982)
W.C. Gardiner, Jr. (ed.): *Combustion Chemistry* (Springer, Berlin, Heidelberg 1984)
J. Walker: The physics and chemistry underlying the infinite charm of a candle flame. *Sci. Am.* 238/4, 154 (1978)
5. M. Gehring, K. Hoyeremann, H. Schacke, J. Wolfrum: Direct studies of some elementary steps for the formation and destruction of nitric oxide in the H-N-O system. 4th Symp. on Combustion (Combustion Institute, Pittsburgh, PA 1973)
6. A. G. Gaydon, H. G. Wolfhard: *Flames, their Structure, Radiation and Temperature* (Chapman and Hall, New York 1979)
7. J. Wolfrum: Chemical kinetics in combustion systems: The specific effect of energy, collisions, and transport processes. 20th Symp. on Combustion (Combustion Institute, Pittsburgh, PA 1985)
8. A. C. Gaydon: *The Spectroscopy of Flames* (Chapman and Hall, New York 1974)
9. D.R. Crosley (ed.): *Laser Probes for Combustion Chemistry*, ACS Symp. Ser. Vol. 134 (Am. Chem. Soc., Washington 1980)
10. A.C. Eckbreth, P.A. Bonczyk, J.F. Verdick: Combustion Diagnostics by Laser Raman and Fluorescence Techniques, *Progr. Energy Comb. Sci.* 5, 253 (1979).
11. J.H. Bechtel, C.J. Dasch, R.E. Teets: Combustion research with lasers, in *Laser Applications*, ed. by R.K. Erf, J.F. Ready (Academic, New York 1984)
J.H. Bechtel, A.R. Chraplyvy: *Proc. IEEE* 70,-658 (1982)
12. TD. McCay, LA. Roux (eds.): Combustion diagnostics by nonintrusive methods. *Progr. Astronautics and Aeronautics*, Vol. 92 (1983)
13. A.C. Eckbreth: *Laser Diagnostics for Combustion Temperature and Species* (Abacus Press, Turnbridge Wells 1987)
14. K. Iinuma, T. Asanuma, T. Ohsawa, J. Doi (eds.): *Laser Diagnostics and Modelling of Combustion* (Springer, Berlin, Heidelberg 1987)

15. M. Aldén, H. Edner, S. Svanberg, T. Högberg: Combustion studies with laser techniques, Göteborg Institute of Physics Reports GIPR-206 (Chalmers University of Technology, Göteborg 1980)
16. M. Aldén, H. Edner, G. Holmstedt, T. Högberg, H. Lundberg, S. Svanberg: Relative distribution of radicals and temperature in flat flames, studied by laser-induced fluorescence and BOXCARS spectroscopy. Lund Reports on Atomic Physics LRAP-1 (Lund Institute of Technology, Lund 1981)
17. D.R. Crosley, G.P. Smith: Laser-induced fluorescence spectroscopy for combustion diagnostics. *Opt. Eng.* 22, 545 (1983)
K. Schofield, M. Steinberg: Quantitative atomic and molecular fluorescence in the study of detailed combustion processes. *Opt. Eng.* 20, 501 (1981)
18. R. Lucht Applications of laser-induced fluorescence spectroscopy for combustion and plasma diagnostics
19. N.S. Bergano, P.A. Janimaagi, M.M. Salour, J.H. Bechtel: Picosecond laser-spectroscopy measurement of hydroxyl fluorescence lifetime in flames. *Opt. Lett.* 8, 443(1983)
20. M. Aldén, H. Edner, P. Grafström, H.M. Hertz, G. Holmstedt, T. Högberg, H. Lundberg, S. Svanberg, S. Wallin, W. Wendt, U. Westblom: Imaging measurements of species concentrations, temperatures and velocities in reactive flows using laser-induced fluorescence, in *Lasers 86*, ed. by K.M. Corcoran, D.M. Sullivan, W.C. Stwalley (STS Press, McLean, VA. 1985) p.209
21. M. Aldén, H. Edner, G. Holmstedt, S. Svanberg, T. Högberg: Single-pulse laser induced OH fluorescence in an atmospheric flame, spatially resolved with a diode array detector. *Appl. Opt.* 21, 1236 (1982)
22. M.J. Dyer, D.R. Crosley: Two-dimensional imaging of OH laser-induced fluorescence in a flame. *Opt. Lett.* 7, 382 (1982)
G. Kychakoff, R.D. Howe, R.K. Hanson, J.C. McDaniel: Quantitative visualization of combustion species in a plane. *Appl. Opt.* 21, 3225 (1982)
G. Kychakoff, R.D. Howe, R.K. Hanson: Quantitative flow visualization technique for measurements in combustion gases. *Appl. Opt.* 23, 704 (1984)
G. Kychakoff, K. Knapp, R.D. Howe, R.K. Hanson: Flow visualization in combustion gases using nitric oxide fluorescence. *AIAA J.* 22, 153 (1984)
G. Kychakoff, R.D. Howe, R.K. Hanson, M.C. Drake, R.W. Pitz, M. Lapp, C.M. Penney: Visualization of turbulent flame fronts with planar laser-induced fluorescence. *Science* 224, 382 (1984)
R.K. Hanson: Combustion diagnostics: Planar imaging techniques, in *Proc. 21st Symp. on Combustion*, Munich 1986 (The Combustion Institute Pittsburgh, PA 1986)
B. Hiller, R.K. Hanson: Simultaneous planar measurements of velocity and pressure fields in gas flows using laser-induced fluorescence. *Appl. Opt.* 27, 33 (1988)
C. Véret (ed.): *Flow Visualizations IV* (Springer, Berlin, Heidelberg 1987)
23. M. Aldén, H. Edner, P. Grafström, S. Svanberg: Two-photon excitation of atomic oxygen in a flame, *Opt. Commun.* 42, 244 (1982)

- M. Aldén, H.M. Hertz, S. Svanberg, S. Wallin: Imaging laser-induced fluorescence of oxygen atoms in a flame. *Appl. Opt.* 23, 3255 (1984)
24. R.P. Lucht, J.P. Salmon, G.B. King, D.W. Sweeney, N.M. Laurendeau: Two photon-excited fluorescence measurement of hydrogen atoms in flames. *Opt. Lett.* 8, 365 (1983)
 25. M. Aldén, A.L. Schawlow, S. Svanberg, W. Wendt, P.-L. Zhang: Three-photon excited fluorescence detection of atomic hydrogen in an atmospheric pressure flame. *Opt. Lett.* 9, 211 (1984)
 26. J.E.M. Goldsmith: Two-step saturated fluorescence detection of atomic hydrogen in flames. *Opt. Lett.* 10, 116 (1985)
J.E.M. Goldsmith, R.J.M. Anderson: Imaging of atomic hydrogen in flames with two-step saturated fluorescence detection. *Opt. Lett.* 11, 67 (1985)
 27. M. Aldén, S. Wallin, W. Wendt: Applications of two-photon absorption for detection of CO in combustion gases. *Appl. Phys.* B33, 205 (1984)
 28. J.E.M. Goldsmith: Resonant multiphoton optogalvanic detection of atomic hydrogen in flames. *Opt. Lett.* 7, 437 (1982)
J.E.M. Goldsmith: Recent advances in flame diagnostics using fluorescence and ionization techniques
P.J.H. Tjossem, T.A. Cool: *Chem. Phys. Lett.* 100, 479 (1983)
 29. W. Persson, S. Svanberg (eds.): *Laser Spectroscopy VIII*, Springer Ser. Opt. Sci., Vol.55 (Springer, Berlin, Heidelberg 1987)
 30. K. Tennal, G.J. Salomo, R. Gupta: Minority species concentration measurements in flames by the photoacoustic technique. *Appl. Opt.* 21, 2133 (1982)
A.C. Tam: Applications of photoacoustic sensing techniques. *Revs. Mod. Phys.* 58, 381 (1986)
 31. R.K. Hanson, P.A. Kuntz, CH. Kruger: High-resolution spectroscopy of combustion gases using a tunable IR diode laser. *Appl. Opt.* 16, 2045 (1975)
K. Knapp, R.K. Hanson: Spatially resolved tunable diode-laser absorption measurements of CO using optical Stark shifting. *Appl. Opt.* 22, 1980 (1983)
 32. Special Issue on Computerized Tomography. *Proc. IEEE* 71, 291-435 (March 1983)
Special Issue on Industrial Applications of Computed Tomography and NMR Imaging. *Appl. Opt.* 24, 23 (1985)
 33. K.E. Bennett, G.W. Faris, R.L. Byer: Experimental optical fan beam tomography. *Appl. Opt.* 23, 2678 (1984)
 34. H.M. Hertz, G.W. Faris: Emission tomography of flame radicals. *Opt. Lett.* 13, 351 (1988)
 35. H.M. Hertz: Experimental determination of 2-D flame temperature fields by interferometric tomography. *Opt. Commun.* 54, 131 (1985)

36. A. Rose, G.J. Salamo, R. Gupta: Photoacoustic deflection spectroscopy. A new specie-specific method for combustion diagnostics. *Appl. Opt.* 23, 781 (1984)
 H. Sonntag, A.C. Tam: Time-resolved flow-velocity and concentration measurements using a travelling thermal lens. *Opt. Lett.* 10, 436 (1985)
 G.W. Faris, R.L. Byer: Beam-deflection optical tomography. *Opt. Lett.* 12, 72 (1987)
 G.W. Faris, R.L. Byer: Beam-deflection optical tomography of a flame. *Opt. Lett.* 12, 155 (1987)
37. M. Lapp, C.M. Penney: Raman measurements on flames, in *Advances in Infrared and Raman Spectroscopy*, ed. by R.J.H. Clark, R.E. Hester (Heyden, London 1977)
38. R.W. Dibble, A.R. Masri, R.W. Bilger *Combust. Flame* 67, 189 (1987)
 J.J. Valentini: Laser Raman techniques
39. M. Aldén, H. Edner, S. Svanberg: Coherent anti-Stokes Raman spectroscopy (CARS) applied in combustion probing. *Phys. Scripta* 27, 29 (1983)
40. D. Klick, K.A. Marko, L. Rimai: Broadband single-shot CARS spectra in a fired internal combustion engine. *Appl. Opt.* 20, 1178 (1981)
 G.C. Alessandretti, P. Violino: Thermometry by CARS in an automobile engine. *J. Phys. D* 16, 1583 (1983)
41. L.A. Rahn, S.S. Johnston, R.L. Farrow, P.L. Mattern: CARS thermometry in an internal combustion engine, in *Temperature*, Vol.5, ed. by J.F. Schooley (AIP, New York 1982)
42. M. Aldén, S. Wallin: CARS Experiment in a full-scale (10x10m²) industrial coal furnace. *Appl. Opt.* 24, 3434 (1985)
43. B. Attal, M. Pealat, J.P. Taran: *J. Energy* 4, 135 (1980)
44. A.C. Eckbreth: CARS thermometry in practical combustors. *Combust. Flame* 39, 133 (1980)
45. A.C. Eckbreth, P.W. Schreiber: Coherent anti-Stokes Raman spectroscopy (CARS): Applications to combustion and gas-phase diagnostics, in *Chemical Applications of Non-Linear Raman Spectroscopy*, ed. by A.B. Harvey (Academic, New York 1981)
 R.J. Hall, A.C. Eckbreth: Coherent anti-Stokes Raman spectroscopy (CARS): Application to combustion diagnostics, in *Laser Applications*, ed. by J.F. Ready, R.K. Erf (Academic, New York 1984) Vol.5
46. H. Haragushi, B. Smith, S. Weeks, D.J. Johnson, J.D. Wineforder: Measurement of small volume flame temperature by the two-line atomic fluorescence method. *Appl. Spectr.* 31, 156 (1977)
 R.G. Jolik, J.W. Daily: Two-line atomic fluorescence temperature measurements in flames: An experimental study. *Appl. Opt.* 21, 4158 (1982)
 M. Aldén, P. Grafström, H. Lundberg, S. Svanberg: Spatially resolved temperature measurements in a flame using laser-excited two-line atomic fluorescence and diode-array detection. *Opt. Lett.* 8, 241 (1983)

47. J. Pender, L. Hesselink: Phase conjugation in a flame. *Opt. Lett.* 10, 264 (1985)
P. Ewart, S.V. O'Leary. Detection of OH in a flame by degenerate four-wave mixing. *Opt. Lett.* 11, 279 (1986)
48. R.M. Osgood, S.R.J. Brueck, H.R. Schlossberg (eds.): *Laser Diagnostics and Photochemical Processing for Semiconductor Devices* (North Holland, Amsterdam 1983)
D. Bäuerle (ed.): *Laser Processing and Diagnostics*, Springer Ser. Chem. Phys., Vol.39 (Springer, Berlin, Heidelberg 1984)
D. Bäuerle, K.L. Kompa, L.D. Laudé (eds): *Laser Processing and Diagnostics II* (Physique, Les Ulis 1986)
D. Bäuerle: *Chemical Processing with Lasers*, Springer Ser. Mat. Sci., Vol.1 (Springer, Berlin, Heidelberg 1986)
L.D. Laudé, D. Bäuerle, M. Wautelet (eds): *Interfaces under Laser Irradiation*, NATO ASI Series (Nijhoff, Dordrecht 1987)
W.G. Breiland, M.E. Coltrin. P. Ho (eds): *Laser-Based Studies of Chemical Vapor Deposition*, Proc. Soc. Photo-opt. Instrum. Eng. 385, 146 (1983)
49. K.L. Kompa, J. Wanner: *Laser Applications in Chemistry* (Plenum, New York 1984)
V.S. Letokhov (ed.): *Laser Analytical Spectrochemistry* (Hilger, Bristol 1986)
T.R. Evans (ed.): *Applications of Lasers to Chemical Problems* (Wiley, New York 1982)
S. Svanberg: Laser spectroscopy applied to energy, environmental and medical research. *Phys. Scr. T* 23, 281 (1988); *Appl. Phys. B* 46, 271 (1988)
50. E.R. Pike, H.Z. Cummins (eds.): *Photon Correlation and Light Beating Spectroscopy* (Plenum, New York 1974)
51. L.E. Drain: *The Laser Doppler Technique* (Wiley, Chichester 1980)
52. E. Durst, A. Melling, J.H. Whitelaw: *Principles and Practice of Laser-Doppler Anemometry*, 2nd edn. (Academic, London 1981)
53. C.J. Dasch, J.A. Sell: Velocimetry in laminar and turbulent flows using the photothermal deflection effect with a transient grating. *Opt. Lett.* 11, 603 (1986), and references therein
R. Miles, C. Cohen, J. Connors, P. Howard, S. Huang, E. Markovitz, G. Russel: Velocity measurements by vibrational tagging and fluorescent probing of oxygen. *Opt. Lett.* 12, 861 (1987)
54. B. Hiller, J.C. McDaniel, E.C. Rea, Jr., R.K. Hanson: Laser-induced fluorescence technique for velocity field measurements in subsonic gas flows. *Opt. Lett.* 8, 474 (1983)
55. U. Westblom, S. Svanberg: Imaging measurements of flow velocities using laser-induced fluorescence. *Phys. Scripta* 31, 402 (1985)
U. Westblom, A. Aldén: Spatially resolved flow velocity measurement using laser-induced fluorescence from a pulsed laser. *Opt. Lett.* 14, 9 (1989)