Review on time-resolved two-dimensional gas concentration mapping in turbulent flows using molecule-sensitive light scattering techniques*

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Abstract. The applicability of laser-induced molecular fluorescence, Rayleigh and Raman scattering for making quantitative, time-resolved two-dimensional gas concentration measurements in turbulent flows has been reviewed. Preliminary experimental results obtained as well in cold and chemically non-reacting as in hot and reactive gas flows by several working groups indicate, that all the three techniques can be used to monitor the concentration distribution in selected sections of turbulent flows using an optical multichannel analyzer for signal acquisition. The resulting data allow to quantitatively study the turbulent mixing mechanism. Time-resolved information about large-scale structures is obtained.

On the other hand huge efforts have been invested in the last years to develop spatially-precise, quantitative point measurement techniques of gas concentrations for flow and combustion applications, see e.g. [5–7]. The measurement location is translated to map out the field, point by point, which is, of course, time consuming. But it can reveal coherent structures of appropriate periodicity.

The development of optical multichannel analyzers like vidicons and diode array cameras has now introduced the possibility to obtain qualitative flow visualization using light scattering methods. First results have been obtained using the Lorentz/Mie scattering from small aerosol particles seeded to the gas component to be investigated [8]. This technique, however, is not applicable in highly accelerating flow regions where the large size of the particles relative to the gas molecules does not allow them to follow the flow. Seeding with particles could also lead to erroneous information about the flow structures in chemically reaction flows, e.g. flames, and in flow regions where molecular diffusion processes are important. For such experimental conditions a molecule-sensitive measurement mechanism would overcome these problems.

In this paper the molecule-sensitive techniques Raman and Rayleigh scattering and laser-induced fluorescence (LIF) will be discussed, and the two-dimensional extension of these methods obtained very recently by several working groups will be reviewed showing the great potential of these techniques for future flow investigations.

1 Introduction

Large scale structures (coherent structures) are widely recognized to play a dominant role in the formation of turbulence and mixing in reactive and non-reacting flows [1]. To gain information about these structures, current non-intrusive experiments are making greater use of flow visualization techniques. However, these are in general more qualitative in nature [2–4].

* Dedicated to Professor E. R. G. Eckert on the occasion of his 80th birthday

2 Molecule-sensitive scattering techniques

On irradiating a gas sample with laser of intensity \( I_0 \) an energetic interaction takes place between the laser photons and gas molecules, atoms and aerosols present in the sample, which is schematically shown in Fig. 1. This process is strongly dependent on both partners (photon and particle) and can be observed and distinguished by the scattered light. In general, the scattered light intensity \( I_s \) collected perpendicular to the laser propagation direc-
Fig. 1. Two representative molecular electronic states with vibrational splitting and the molecule-sensitive scattering processes discussed in this paper.

The fluorescence intensity may be expressed by

$$I_{sf} = K l_0 / \Omega n_i (d\sigma/d\Omega) f_i$$  \hspace{1cm} (1)

Here, $l$ indicates the length of the laser beam within the sample volume; $\Omega$, the solid angle of the collection optics; $K$, a setup calibration constant; $n_i$, the total number density of the scattering particles; and $f_i$, the Boltzmann fraction in the molecular state being investigated. $(d\sigma/d\Omega)$ is the differential scattering cross section of each process. For equal setup conditions, the signal strength of different scattering mechanisms relative to the others can easily be compared by comparing the cross section value, which is given in Table 1 for the techniques of interest here. As can be seen, the molecular-sensitive techniques are orders of magnitude weaker than the aerosol-sensitive Mie scattering process which has been used in Ref. 8 giving, even in situations with very low aerosol particle densities much higher scattering intensities than we can expect to obtain by the other processes. In the following parts of this chapter, the three different techniques, fluorescence, Rayleigh and Raman scattering will be treated separately somewhat more detailed explaining briefly the different characters of each single one.

2.1 Laser-induced fluorescence

Laser-induced fluorescence (LIF) occurs, see Fig. 1, when the irradiated laser frequency is tuned to coincide with a molecular absorption line or band causing the molecule to make a transition to an higher excited electronic level followed by a reemission of light at the same frequency (resonance fluorescence) or somewhat frequency shifted. Because of the requirement of a suitable absorption region in the visible or near ultraviolet, fluorescence has a less general applicability than Raman scattering. So all homonuclear diatomic molecules, e.g. nitrogen, oxygen and also air, cannot be probed directly by the laser fluorescence technique because their lowest electronic transition is in the VUV region [10].

The fluorescence intensity is several orders of magnitude higher than that observed in Raman or Rayleigh scattering (Table 1). The effective fluorescence scattering cross section can be expressed by

$$\frac{d\sigma}{d\Omega}_{lf} = \sigma_{abs} F \frac{A_{21}}{4\pi A_{21} + Q}$$  \hspace{1cm} (2)

where $F$ is the fraction of the monitored fluorescence dependent on the detector bandwidth. $\sigma_{abs}$ is the absorption cross section for the initial absorption process, which is only a function of molecular constants besides its dependence on the effective linewidth. $A_{21}/A_{21} + Q$ is the Stern-Volmer factor with Einstein coefficient $A_{21}$ for spontaneous emission and quenching rate $Q$, i.e. the rate for non-radiative electronic transitions. This factor describes the reduction of the fluorescence intensity by the competing quenching process. Because $Q$ is a function of the temperature, the individual quenching rate of each species and the concentration of each species, it is very difficult to obtain the exact value for the quenching correction for each different possible condition in a flow. But in several cases it is possible to select a molecular absorption frequency and to assume a fairly constant species distribution, so that the proportionality factor between the fluorescence signal and the total absorber number density does not vary significantly throughout the flowfield being studied [11, 12].

Another approach to overcome the quenching influence on LIF is the saturation of the absorbing transition by using pulsed lasers with high spectral intensity, see e.g. [13–16]. The fluorescence intensity at this point is then independent on laser irradiance and quenching rates. It requires, however, laser spectral intensities in the order of $10^6$–$10^8$ W/cm² cm⁻¹ [10] which is sometimes difficult to