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Particle diagnosis in multiphase media

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Abstract - We review laser based optical techniques in use for the optical characterization of discrete particles contained in flows at rest or transported by moving flows. For simultaneous measurements of velocities and sizes, we first discuss the visibility, pedestal, phase-Doppler and top-hat beam techniques in which velocimetry is carried out by means of laser Doppler Anemometry. Next, we consider the Visible Infra-red Double Extinction method for simultaneous measurements of supermicronic sizes and particle concentrations in densely laden media, and the Quasi-Elastic Light Scattering Spectroscopy devoted to the investigation of submicronic particles. Most of the techniques are based on the interaction between laser beams and particles whose diameters are not much smaller than the beam diameters. Under such circumstances, the classical Lorenz-Mie theory which may be used to design instruments and interpret data leads to misleading results. We therefore introduce the Generalized Lorenz-Mie Theory which correctly describes the interactions between particles and arbitrary incident illuminating beams. The differences between plane wave theory and Gaussian beam theory are exemplified by discussing a case study taken from the phase-Doppler technique.

I. INTRODUCTION

When dealing with optical characterization of discrete particles in flows, we may be faced to a very large range of situations. First, particles may be perfect spheres but they also may be other regular particles like fibers or ellipsoids, irregular particles and even fractal particles. Very often, it is necessary to assume that particles are spherical even when they are not, an unsafe attitude which can lead to the so-called spherical chicken syndrome (ref.1). Next, particle concentrations may range from 0 (LDV tracers, dilute media) to the case of packed media like in fluidized beds below incipient fluidization. Also, difficulties may arise due to non homogeneity of the material and measurement techniques may have to cope with the value of the complex refractive index of the material which may be difficult to know, like in the case of soot in flames.

There is therefore a need of several kinds of instruments to cover such a broad range. This fact, the big number of potential applications, and the flexibility of optical techniques have produced a prolific literature which may in part be appreciated by having a look at some review papers (refs 2-4). To organize the development of the topic, a series of international conferences started in 1987 (ref. 5) in Rouen. The second conference was held in 1990, in Phoenix, Arizona and was organized by D. Hirleman. The third conference will be held in 1993, in Yokohama, Japan, and will be organized by M. Maeda. The newcomer in the field will find these conferences very useful to gain an up-to-date knowledge. The present paper also aims at helping this newcomer by providing a guideline through the labyrinth of optical particle sizing.

II. SIMULTANEOUS MEASUREMENTS OF SIZES AND VELOCITIES

We start by discussing simultaneous measurements of sizes and velocities by using optical techniques based on laser Doppler Velocimetry (LDV) (ref. 6). These techniques are essentially devoted to the study of supermicronic particles in low concentration media. A typical signal obtained from a LDV-probe is shown in Fig 1. Velocities are evaluated from the high-frequency modulation.

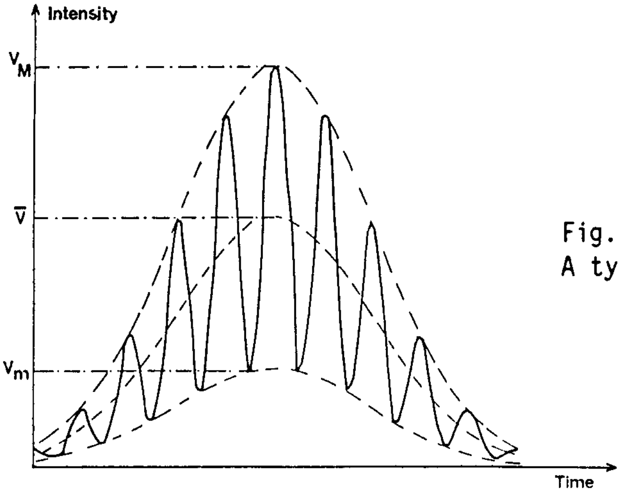

Fig. 1.
A typical Doppler signal

The first concept for simultaneous particle sizing has been introduced by Farmer (ref. 7) who defined the visibility :

$$\mathcal{V} = \frac{V_M - V_m}{V_M + V_m} \tag{1}$$

and showed that $\mathcal{V}$ is correlated with the diameter d of the particle by :

$$\mathcal{V} \simeq \frac{|J_1(\pi d/i)|}{\pi d/i} \tag{2}$$

in which J_1 is the first-order Bessel function and i the fringe spacing in the LDV-control volume.

In the pedestal technique, diameter is correlated with the pedestal level $\bar{V}$ (refs 8,9). There is clearly some trajectory ambiguity in this technique because big particles crossing the edge of the control volume made from a Gaussian beam will give the same pedestals as small particles having a median trajectory. The problem may be partly solved by using an inversion technique assuming equiprobability of trajectories but, consequently, only simultaneous measurements of size and velocity probability density functions are feasible.

Visibility and pedestal techniques are now of little use. The most popula method is the phase-Doppler technique introduced by Durst and Zaré (ref. 10). Commercial systems are available (Aerometrics, Dantec and Invent companies for instance). In this technique, Doppler signals are recorded by two detectors (actually more than two are often used) and the phase difference between signals from the two detectors is linked to the particle diameter. This technique is however not free of problems as we shall discuss later.

Visibility, pedestal and phase-Doppler techniques are one-colour technique : both velocity and size are extracted from the same signal produced by one laser wave-length. We now come to a two-colour technique, the so-called Top-Hat Beam Technique (THBT) (refs 11-13). In THBT, velocities are measured from a LDV-probe and sizes are measured from a second spatially superimposed probe, using another wave-length of the laser source. To avoid trajectory ambiguity effects, the sizing probe which is originally a Gaussian beam is corrected to a top-hat illumination profile. The height of the top-hat signal is correlated to the particle diameter. This technique may handle non-spherical particles. Although no commercial devices are available, THBT is likely to become more popular in the future.

III. SIMULTANEOUS MEASUREMENTS OF SIZES AND CONCENTRATIONS IN OPTICALLY THICK MEDIA

When media exhibit large optical thicknesses, multiple or even dependent scattering effects may arise, and the previously described techniques which rely on single scattering assumption do not work any more. We may then use the Visible Infra-Red Double Extinction (VIDE) technique. This technique theoretically relies on the fact that Beer-Lambert law remains valid under multiple scattering conditions when only collimated radiation is considered (refs 14,15). The Beer-Lambert law is expressed in terms of two indeterminates : particle diameter d and number-density N (assuming that the complex refractive index is known). If extinction by the medium under study is measured at two-well chosen wave-lengths, one then shows that d and N may be simultaneously measured. For particles in the range 10-100 μm, wave-lengths, may be λ_1 = 632.8 nm (He-Ne laser) and λ_2 = 337 μm (HCN laser), hence the name of VIDE. Validations of the technique have been carried out on a falling cloud of coal particles (ref. 16) and, later on, on spherical glass particles (ref. 17) by using specially designed standard media (ref. 18). According to Refs 14-15, reflectance measurements would also be possible but the theoretical concept has not yet been experimentally checked.

IV. QUASI-ELASTIC LIGHT SCATTERING SPECTROSCOPY (QELSS)

This technique is devoted to the characterization of submicronic particles, allowing for local simultaneous measurements of sizes and number-densities. It is now of wide use to study polymers, for example, and commercial devices are available. We however used it to study sooting laminar premixed flames (ref. 19).

The principle is as follows. A coherent beam (laser radiation) is directed on to the particle medium under study. The scattered light is collected by a photodetector. The scattering process induces a loss of correlation between photons due to the random character of Brownian motion (surrounding fluid is here assumed to be at rest). Since the efficiency of Brownian motion depends on particle diameters, a measure of the loss of correlation allows for the particle diameter measurements. A more refined analysis shows that number-densities may also be evaluated.

The loss of correlation information is obtained by measuring the photodetector photocurrent autocorrelation function, by using a photocorrelator leading to a technique called photon correlation spectroscopy. By using a spectrum analyzer, we obtain a power spectrum which is the Fourier Transform of the photocurrent autocorrelation function, leading to a technique called Diffusion Broadening Spectroscopy.

Any process leading to a loss of correlation adds information in the signal. Therefore, QELSS may also be used to measure velocities when the flow velocities through the control volume dominate Brownian motion. The technique then acts as a transit velocimeter. Very often, both velocity and Brownian motion information are contained simultaneously in the signal. It is then possible to retrieve each information by a numerical deconvolution procedure. Polydispersity information is also available. We finally comment that the study of turbulent flows is in principle possible althouth this issue has not been systematically investigated from an experimental point of view.

V. GENERALIZED LORENZ-MIE THEORY

The very classical Lorenz-Mie Theory (LMT) which is about one century old describes the interaction between a plane wave and a perfect sphere. It has been widely used to design instruments and analyze data in optical sizing. Unfortunately, most techniques use laser sources. Therefore, when the particle diameters are not small enough with respect to the beam width, LMT is invevitably misleading. We therefore needed a Generalized Lorenz-Mie Theory (GLMT) describing the interaction between a laser beam (Gaussian beam and more generally any kind of incident beam) and a perfect sphere. GLMT has been developed by steps in our team. See Refs 20-23 from which earlier references may be found.

The physics behind GLMT is very simple. It consists in solving Maxwell equations with boundary conditions at the surface of the sphere and limit conditions at the infinite of the sphere. The rest is only mathematical literature. Besides classical mathematical functions, GLMT introduces two new sets of coefficients, the so-called beam shape coefficients $g^m_{n,Tm}$ and $g^m_{n,TE}$ describing the incident beam. For practical computations, special algorithms and formulae must be derived for efficient numerical evaluations of these coefficients. The most efficient methods are the use of finite series (ref.24) and of the so-called localized approximation (refs 25-26). These numerical methods enabled us to design efficient computer programs for GLMT and to consider GLMT-applications to optical sizing.

A discussion of GLMT-applications to the top-hat beam technique is available from Ref. 27. However, our main effort is devoted to GLMT-applications to phase-Doppler technique in so far as this technique is certainly the most widely used nowadays. GLMT unambiguously reveals the existence of trajectory ambiguity effects in Phase-Doppler technique but, also, suggests remedies to cure this problem (refs 28-29). Obviously, GLMT is a quite new tool and applications to optical sizing only begin. It is likely that it will be a wave of the future and that many crops will have to be harvested. Concerning phase-Doppler techniques, our (provisional) opinion is that it is to be recommended, and that, although new problems are evidenced, most of them may be cured. However, the practice of phase-Doppler techniques might become more difficult as refinements will become required. May be the time is approaching where people will be pleased to have at their disposal another complementary opportunity. We suggest that THBT may provide such an opportunity.

VI. CONCLUSION

This paper provided a concise guideline on optical sizing techniques. This guideline is however biased by the author expertise and some readers may be disappointed of the lack of discussion of other techniques like Gabor microholography or diffractometry (but see Ref. 30). In any case, it is not possible in a concise paper to review all kinds of techniques available. We however hope that we may have helped the newcomer to easily gain an overview of the topic.

REFERENCES

1. B.H. Kaye, a random walk through fractal dimensions, VCH, (1989).
2. G. Gouesbet, G. Gréhan, Proceedings of the International Symposium on plasma chemistry, Zürich, 27th Aug-1st Sep, pp 603-616, (1979).
3. G. Gouesbet, Measurement techniques in heat and mass transfer, R.I. Soloukhin, N.H. Afgan, editors, Springer-Verlag, pp 27-50, (1985).
4. G. Gouesbet, Plasma chemistry and plasma processing, 5, n° 2, pp 91-117, (1985).

5. G. Gouesbet, G. Gréhan, edts, Optical particle sizing : theory and practice, Plenum Press, (1988).
6. F. Durst, A. Melling and J.H. Whitelaw, Principles and practice of laser-Doppler anemometry, 2 nd edn., Academic Press, New York, (1981).
7. W.M. Farmer, Applied Optics, 11, 11, 2603, (1972).
8. Don Holve and S.A. Self, Applied Optics, 18, 10, Part I, 1632 and Part II, 1646, (1979).
9. A.J. Yule, N.A. Chigier, S. Atakan, A. Vingut, Journal of Energy, 1, 4, (1977).
10. F. Durst and M. Zaré, Proc. LDA-Symposium, Copenhagen, pp 403-429, (1975).
11. G. Gréhan, G. Gouesbet, Applied Optics, 25, 19, 3527-3538, (1986).
12. F. Corbin, G. Gréhan, G. Gouesbet, Particle and Particle Systems Characterization, 8, 222-228, (1991).
13. M. Maeda and K. Hishida, in Ref. 5.
14. B. Maheu, J.N. Le Toulouzan, G. Gouesbet, Applied Optics, 23, 19, 3353-3362, (1984).
15. B. Maheu, G. Gouesbet, Applied Optics, 25, 7, 1122-1128, (1986).
16. G. Gouesbet, P. Gougeon, J.N. Le Toulouzan, M. Thioye, J.B. Guidt, Particle and Particle Systems Characterization, 5, 2, 51-56, (1988).
17. J.B. Guidt, G. Gouesbet, J.N. Le Toulouzan, Applied Optics, 29, 7, 1011, (1990).
18. J.B. Guidt, J.N. Le Toulouzan, M. Thioye, G. Gouesbet, Particle and Particle Systems Characterization, 7, 36, (1990).
19. N. Lhuissier, G. Gouesbet, M.E. Weill, Combustion Science and Technology, 67, 17-36, (1989).
20. G. Gouesbet, G. Gréhan, B. Maheu, J. Optics, 16, 2, 83-93, (1985).
21. G. Gouesbet, B. Maheu, G. Gréhan, J. Opt. Soc. Am. A, 5, 1427-1443, (1988).
22. B. Maheu, G. Gouesbet, G. Gréhan, J. Optics, 19, 2, 59-67, (1988).
23. G. Gouesbet, G. Gréhan, B. Maheu, in Combustion measurements, N. Chigier, ed., Hemisphere Publishing , 339-384, (1991).
24. G. Gouesbet, G. Gréhan, B. Maheu, J. Optics, 19, 1, 35-48, (1988).
25. G. Gréhan, B. Maheu, G. Gouesbet, Applied Optics, 25, 19, 3539-3548, (1986).
26. G. Gouesbet, G. Gréhan, B. Maheu, J. Opt. Soc. Am. A, 7, 6, 998-1007, (1990).
27. F. Corbin, G. Gréhan, G. Gouesbet, B. Maheu, Particle and Particle Systems Characterization, 5, 3, 103-108, (1988).
28. G. Gréhan, G. Gouesbet, F. Vannobel, J.B. Dementon, Fifth International Symposium on Applications of Laser Anemometry to Fluid Mechanics, July 9-12, Lisbon, (1990).
29. G. Gréhan, G. Gouesbet, A. Naqwi, F. Durst, International Conference on Multiphase Flows, University of Tsukuba, Japan, Sept 24-27, (1991).
30. G. Gouesbet, M. Ledoux, Optical Engineering, 23, 5, 631-640, (1984).