The Time-shift technique for characterization of non-transparent, spherical particles

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Abstract

The time-shift technique, also known as the pulsed-displacement technique, is re-visited as a means of measuring size, velocity and relative refractive index of spherical, non-transparent particles. Building on the basic measurement principle, several new innovations are introduced, making the technique significantly more attractive for use outside of the laboratory. These include validation criteria for two-detector arrangements, and approaches for achieving higher size bandwidths, in particular lower measurable sizes. However, the main novelty introduced in this contribution is the ability to measure non-transparent droplets. Such droplets are quite common, for example in spray drying processing or in paint sprays.

In this contribution the basic working principle of the time-shift techniques will be reviewed, followed by guidelines for the optical layout. Example measurements are presented.

Introduction

The characterization of spherical, transparent particles in terms of size, velocity and possibly relative refractive index is of major interest in a variety of applications, but especially when investigating sprays of pure liquids. Counting techniques, i.e. techniques which measure and count individual droplets, are often desirable over integrating techniques such as the laser diffraction technique (LDT), since the velocity information, together with the counts can yield flux densities and concentration estimates. Several techniques fulfill these expectations, in particular direct imaging and interferometric techniques, such as the phase Doppler technique, holography, rainbow refractometry or interferometric particle imaging. Available techniques for this purpose have been recently summarized in a review article (1).

However also the time-shift (TS) technique is an interesting candidate in this respect, and for several reasons there are very good grounds to re-visit this technique with the intention of making it more suitable for practical applications. One motivation is that the TS technique does not require coherent light and can therefore take advantage of low-cost, high power light sources which have recently become available. Furthermore, the TS technique can be optically configured to work in the near backscatter, allowing transmitting and receiving optics to be aligned through a single optical access to the measurement position. This immediately opens the possibility of a probe construction, i.e. all optical components in a single housing; hence achieving a higher degree of robustness.

The time shift technique itself is not new, its origins go back to the Ph.D. thesis of Semidetnov in 1985 (2). Hess and Wood (3) presented several different optical configurations of the time-shift technique, all operating in forward scatter, i.e. employing scattered light from reflection and first-order refraction. One focus of their development was to enlarge the measurable size range, especially for smaller droplets. In their instrument velocity was measured using the laser Doppler or the time-of-flight technique. In their study they called this technique the pulse displacement technique. Lin et al. (4) also worked in forward scatter and employed three illuminating light sheets, extending the measurement capability to include relative refractive index. In Damaschke et al. (5) and Albrecht et al (6) configurations suitable for backscatter detection were introduced, enabling more compact optical arrangements and easier optical access to the measurement position, while at the same time enabling size and refractive index to be obtained using only one illuminating beam. The laser Doppler technique was used for velocity measurement. Damaschke et al. also examined the sensitivity of the time-shift technique to non-sphericity of the scattering particle as well as limitations for small particle sizing.

It can be noted that the distinguishing feature of the time-shift technique compared to other techniques for size and velocity measurements of particles is the fact that the time-shift technique uses a shaped beam, meaning that the intensity variation of the beam varies considerably over the diameter of the particle to be measured. Hence, the time-shift technique utilizes a fundamentally different measurement principle then for instance the grating anemometer introduced by Semiat and Dukler (7) or Cartellier (8). The condition of a shaped beam can naturally arise, especially with larger particles and/or bubbles, and a technique closely related to the time-shift technique has been introduced by Brankovic et al. (9), known also as the triple-peak technique (Yu and Varty (10)). These realizations have also relied on the laser Doppler technique for velocity measurement. Indeed, these and several other variations of this technique (11) can be considered special cases of the time-shift technique. All of these techniques assume sphericity of the particle.

However, general principles for optically configuring the time-shift instrument for a given application remain lacking as do validation strategies for enabling the technique to be used in situations with higher particle densities. Furthermore, the use of the laser Doppler technique or the time-of-flight approach for the particle velocity measurements add complexity and cost to the time-shift technique. The present work attempts to alleviate some of these shortcomings.

Measurement Principle

The measurement principle of the time-shift technique is only briefly summarized here, since adequate descriptions can be found elsewhere (Albrecht et al. (2003)). The measurement principle of the time-shift technique is based on the light scattering of a single particle from a shaped light beam. Whether the intensity variation is Gaussian, as would be the case for a laser beam in TEM_{00} mode, or any other form, is not important in principle. However some advantages can be recognized for the subsequent signal processing if a Gaussian beam is used.

The light scattered from a single particle can be interpreted as the sum of all scattering orders present at the detector location. This decomposition of scattered light into various scattering orders is well described by the Debye series (12) expansion of the Mie (13) scattering functions, or by using a geometric optics (14) approach to compute the scattered field. Considering the propagation of individual rays of light through the particle, as depicted in Figure 1, the point at which the incident ray intersects the particle surface is known as the incident point. The angle of an incident point is called the incident angle θ_i. The point where the light ray exits a particle after reflection or a number of refractions is called a glare point (15).

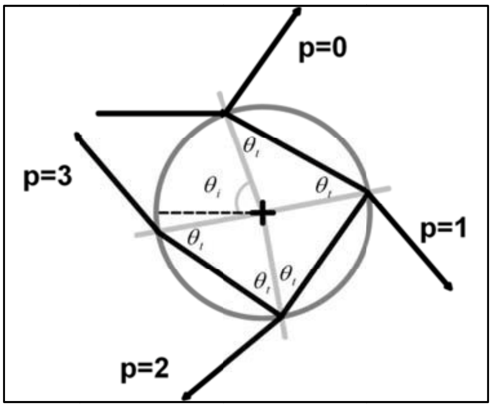

Figure 1: Schematic illustration of the scattering orders according to geometrical optics. Abbreviations: GP - glare point,; p=0, reflection; p=1, first-order refraction; p=2, second-order refraction; p=3, third-order refraction; θ_i, incident angle

If the illuminating beam is 'shaped', effectively a different measurement volume is realized for each received scattering order, and each measurement volume is displaced in space from one another. This is in contrast to illumination by a homogeneous plane wave, for which all scattering orders are received with the same incident intensity, regardless of the particle position. The magnitude of the displacement between the illuminated volume and the measurement volume depends on the particle size, refractive index, scattering order and the scattering angle. The displacement in space translates through the particle velocity component in the direction of measurement volume separation into a temporal separation of the received signals; hence the two different designations of this technique: the pulse displacement technique or the time-shift technique.

When a particle passes through these different measurement volumes, the measurement volume displacement appears as a time shift of the signal on any one single detector. The number and relative position of the measurement volumes in space are related to the relative position of the respective incident points of the scattering orders and their modes of the received light. The position of these incident points depend in turn on scattering angle and relative refractive index, and can be calculated by ray tracing methods. In order to achieve adequate separation of the different measurement volumes in space, the particle size must be larger than the size of the shaped light beam. The size ratio which is adequate for reliable measurements will depend to some extent on the exact optical configuration; in general forward scattering arrangements can size smaller particles.

An example time-shift signal originating from a water droplet passing through a Gaussian shaped beam and collected at a scattering angle of 150deg from forward scatter is illustrated in Figure 2, in which signal components are observable from the following scattering orders: surface wave long path, reflection (p=0), second-order refraction mode 1 (p =2.1), second-order refraction mode 2 (p=2.2) and surface wave short path. The time between individual peaks in the time-shift signal $S(t)$ will depend on the detection angle, the relative refractive index, the particle velocity normal to the illuminating beam, and the particle size. If the first two parameters are known and the particle velocity is measured, e.g. using a laser Doppler or time-of-flight technique, then the particle size can be deduced from the time shift between two of the signal peaks. In the example shown in Figure 2 three main signal peaks are obtained; hence two independent measures of particle size would be possible, e.g. $t_{p=0} - t_{p=2.1}$ or $t_{p=0} - t_{p=2.2}$.

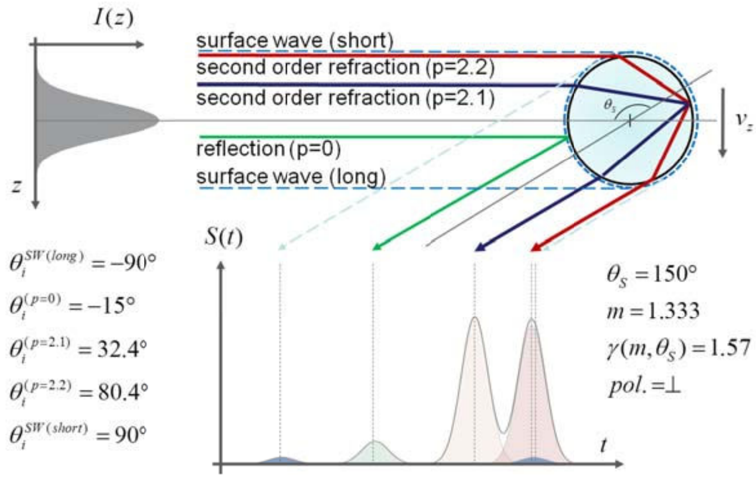

Figure 2: Time-shift signal originating from a water droplet passing through a Gaussian beam and with a detector placed at a scattering angle of 150deg from forward scatter.

Measurement System for Non-Transparent Particles

For non-transparent particles such as emulsions, suspensions or dispersions, as one would expect in a spray dryer or with painting applications, the light scattering properties of single droplets are expected to change significantly. In particular, the transmission of light through the particle is expected to be greatly reduced and scattering from internal scattering centers can be expected. This changed situation is depicted graphically in Figure 3. Therefore, the signal obtained at a given detector will comprise reflective scattering and scattering from within the droplet. Direct detection of higher order refractive components (p>0) can only be expected for very dilute droplet mixtures.

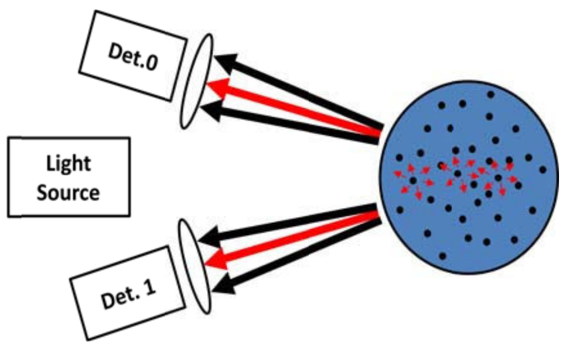

Figure 3: Schematic illustration of light scattering from a non-transparent droplet.

A suitable optical configuration for the size measurement of such droplets is shown in Figure 4, in which two detectors are used in a near backscatter arrangement. This configuration can be extended by employing a Doppler or time-of-flight measurement of droplet velocity. Two detectors are used at different scattering angles, since the arrival time of the reflective signal obtained from the droplet will depend now on scattering angle, droplet velocity and droplet size (no longer relative refractive index.) Knowing the scattering angle of each detector (shown symmetric in Figure 4) and the droplet velocity from the time-of-flight measurement, allows the drop size to be computed from the time shift between the reflected peaks of the time-shift signal.

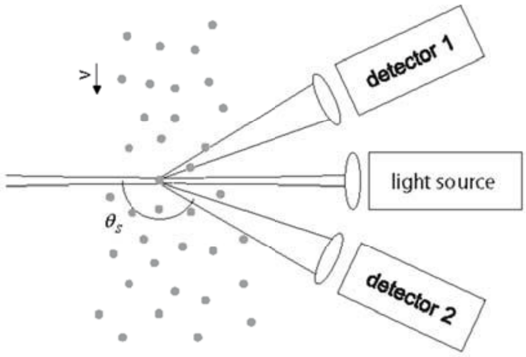

Figure 4: Optical configuration for a two-detector time-shift device operated in backscatter.

The signals expected from each detector arising from a single illuminated volume are depicted in

Figure 5. This signal exhibits two main features: the first is a baseline signal arising from the internal scattering in the particle and extending over a residence time *d/v*, i.e. the particle diameter divided by the velocity of the particle normal to the illuminating beam. The second feature is the reflective peak, appearing at different times for each detector, the arrival time being dependent on the respective detection angle, the particle velocity and the droplet diameter. Knowing the geometric parameters of the optical configuration, the time shift between the two reflective peaks is linearly related to the particle diameter, if the droplet velocity is known.

Any one signal from each detector can be expressed analytically as:

$$S(t) = \underbrace{C_0 w_0 \left(d^2 - 4v^2 (t-t_0)^2\right)}_{baseline} + \underbrace{\sum \frac{A_p}{C_p} \exp\left\{-\frac{2\left((t-t_0)-t_p\right)^2}{\sigma^2}\right\}}_{refraction} - \underbrace{A_0 \exp\left\{-\frac{2\left((t-t_0)-t_{p=0}\right)^2}{\sigma^2}\right\}}_{reflection} \quad (1)$$

which exhibits three components. The first component corresponds to the baseline signal, whose amplitude will somehow be related to the scattering center concentration in the droplet *C*. This component has been approximated by a width of (d/v), where *d* is the particle diameter and *v* the particle velocity. The second component is related to higher-order refractive components, but will be neglected for all but the most dilute particles (non-transparency). The last component corresponds to the reflective scattering and is represented here by a Gaussian shape of width σ, assuming that the illuminating beam also exhibits a Gaussian intensity profile.

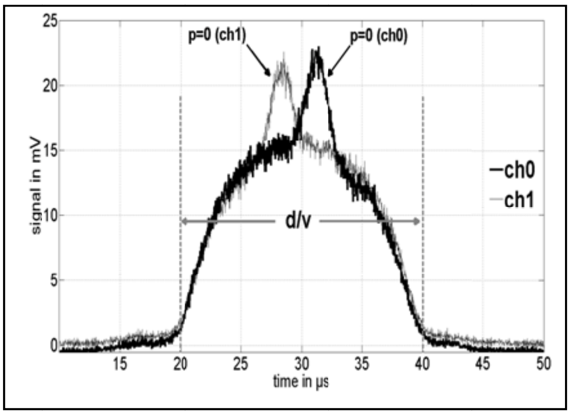

Figure 5: Typical signals expected from a two-detector time-shift measurement system.

Note that the velocity of the particle can also be deduced from the width of this reflective peak, if the width of the illuminating beam (*b*) has been measured beforehand using, for instance, a beam scan device. Then the velocity is given as: $v = b / \sigma$. To make use of Eq. (1), the acquired signal is fit to the analytic expression (1) by adjusting the parameters *C*, A_0, t_0, and $t_{p=0}$. The quantities t_0, and $t_{p=0}$ refer to the arrival time of the particle in the illuminating volume and the time shift of the reflective peak for the particular detector. Such a fitting procedure (model parameter estimation) can be accomplished using different methods; in the present study a least squares regression has been used.

Signal Validation and Example Measurements

In practice the signals shown in the previous section contain noise and can also be distorted when multiple particles traverse the measurement volumes simultaneously. This experience is illustrated in Figure 6a, where signals arising from other particles being in the measurement volume are shown, as opposed to signals suitable for further processing, shown in Figure 6b. To distinguish between suitable signals and non-suitable signals, the baseline portion of the signal is examined in more detail. Specifically, the amplitude of each baseline signal from detectors 1 and 2 must coincide within pre-determined bounds. Otherwise, the signals are rejected from further statistics.

Further signal validation is conceivable. From the previous section it is clear that the particle velocity can be estimated from the width of the reflective peaks, knowing the width of the illuminating Gaussian beam beforehand. However the velocity can also be estimated from the time-of-flight between signals on a single detector, knowing the separation of the illuminating volumes. This validation is only applicable if the time-of-flight technique is used for the velocity measurement. Each of these estimates can be performed with each detector; hence redundancy is available, which can be used for validation purposes.

Also the particle size can be estimated twofold. The main approach is to use the time-shift between reflective peaks of different detectors. However the particle size will also directly influence the width of the baseline signal. This is indicated in

Figure 5, with the quotient *d/v*. Hence, if the velocity is known, for example from the Doppler or time-of-flight measurement, then a further estimate of particle size can be performed. This may be of special interest for non-spherical particles, e.g. for ice crystals.

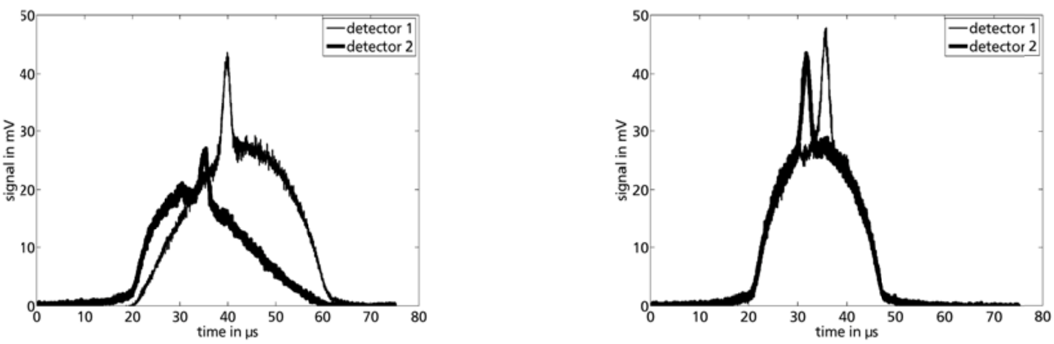

Figure 6: Example time-shift signals. a) Signals with multiple particle in measurement volume simultaneously; b) signals suitable for further processing

The measurement system and signal processing procedure described above has been used to perform measurements in a milk spray. A simple pressure atomizer was used (Lechler 632.363) at various pressures. The time-shift signal used a laser source at wavelength 405nm. For velocity measurement the time-of-flight technique has been used. The scattering angles of detectors were 147deg for detector 1 and 160deg for detector 2 and a detection volume has been used of 1mm, determined by the aperture used in front of the detector.

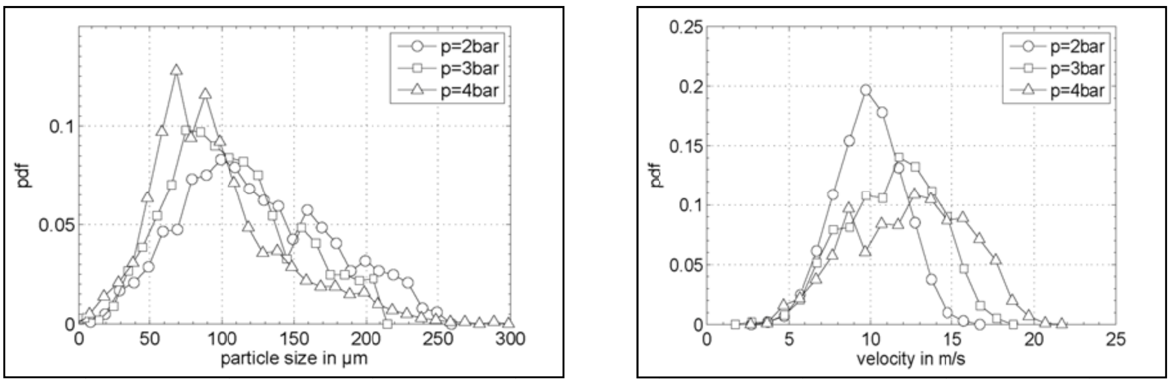

Figure 7: Measured milk droplet size and velocity at different atomizer pressures

The results of these measurements are presented in Figure 7. Figure 7a shows the probability density function of the droplet sizes for three different nozzle pressures. The higher pressure resulted in a smaller mean droplet diameter. Figure 7b shows the corresponding velocities of the particles. With increasing nozzle pressure the mean velocity increases.

Conclusions

The suitability of the time-shift technique for the size measurement of non-transparent droplets has been discussed. For such droplets, reflected scattered light is used. Scattered light arising from internal scattering within the droplet contribute to the obtained signals in the form of a baseline signal, which also exhibits dependencies on particle size and velocity. A measurement system has been introduced, operating in backscatter and using two detectors and two illuminating beams. The velocity measurement is performed using the time-of-flight principle; the size measurement is derived from the time shift between reflective components of the signal on different detectors. Overall considerable redundancy is available in both the size and velocity measurement, offering numerous possibilities for signal validation.

The feasibility of this technique for measuring non-transparent particles has been demonstrated with the measurement of a milk spray from a simple pressure driven atomizer.

Acknowledgements

This work was financially supported by the Deutsche Forschungsgemeinschaft through the Research Training Group GRK 1114 “Optical measurement techniques for characterizing interfacial transport phenomena”.

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