

## Zeitverschiebungsverfahren zur Charakterisierung von transparenten Partikeln

### Time-Shift Technique for Characterization of Transparent Particles

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Droplets, Spray, Particle Characterization

#### Abstract

The goal of this work is the development of a measurement technique for the characterization of individual transparent particles and droplets in a flow. In the present study, the already known time-shift technique has been further developed; in particular new validation criteria have been introduced in order to use the time-shift technique for the reliable and simultaneous measurement of the size and the velocity, as well as the relative refractive index of individual particles or droplets in a flow. The example measurements demonstrate the reliability and the accuracy of the time-shift technique for characterizing transparent particles and droplets.

#### Introduction

Although the time-shift technique was already introduced by Semidetnov in 1985 [1], it has since not seen widespread use in either academic or industrial research, neither is the technique available as a commercial instrument. One reason for this reluctant employment of the technique is its inability to reliably distinguish individual droplets at even moderately high drop concentrations, an aspect which is directly addressed and partially remedied in the present study. Nevertheless, some attempts at further developing the technique have been made since 1985. Hess and Wood (1993) [2] 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 towards smaller droplets. In their instrument velocity was measured using the laser Doppler or the time-of-flight technique and they referred to the technique as. In their study they called this technique the pulse displacement technique. Lin et al. (2000) [3] also worked in forward scatter and employed three illuminating light sheets, extending the measurement capability to include relative refractive index. In Damaschke et al. (2002) [4] and Albrecht et al. (2003) [5] 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. (2002) [4] also examined the sensitivity of the time-shift technique to non-sphericity of the scattering particle as well as limitations for small particle sizing.

After first briefly reviewing the measurement principle, an implementation of the time-shift technique employing a single illuminating light sheet, two detectors positioned in backscatter and a new method of velocity measurements will be introduced and demonstrated.

### Measurement Principle

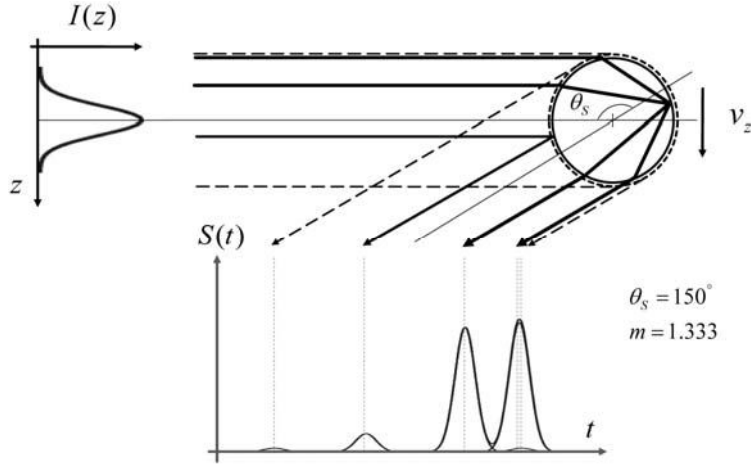


Figure 1: Basic principle of the time-shift technique.

The measurement principle of the time-shift technique is based on the light scattering of a single particle from a shaped light beam. The term '*shaped beam*' refers to the fact that the intensity of the illuminating beam varies significantly across the diameter of the measured droplet. Hence, a pre-requisite for the time-shift technique is that the illuminating beam must be smaller than the size of the smallest particle to be sized. In the configuration presented here the smallest sizeable particle was approximately three times larger than the beam width, other configurations have been presented where this factor is approximately one.

When a particle with a given velocity and size passes through a shaped beam, typically Gaussian in intensity distribution, it transforms the intensity of the light beam in space into a time dependent signal on a detector focused onto the scattering event, the signal being the sum of all scattering orders, for example reflection ( $p = 0$ ) and second-order refraction ( $p = 2$ ) with their respective modes ( $p = 2.1$  and  $p = 2.2$ ) for backscatter and for a relative refractive index of  $m=1.33$  (see Fig. 1). The time dependent signal can then be expressed in the general form:

$$I(z) = I_0 g(z, b) \xrightarrow{v_z = z/t} S(t) = \sum_p A_p g_p(v_z(t - t_0^{(p)}), b) \quad (1)$$

where  $b$  is the width of illumination beam with maximum intensity  $I_0$  and intensity distribution  $g$ ,  $A_p$  the amplitudes of the various scattering orders,  $v_z$  is the component of the particle velocity in the scattering plane and normal to the illuminating beam and  $t_0^{(p)}$  the relative time position of the scattering order  $p$  or of a mode of a particular scattering order. The quantities  $A_p$  and  $t_0^{(p)}$  depend on the scattering angle, relative refractive index and particle size.  $S(t)$  will be referred to as the time-shift signal. If the scattering angle, relative refractive index and velocity  $v_z$  are known, then the particle size can be extracted from the signal  $S(t)$ . The number of scattering orders seen by the detector will depend on the scattering angle and the relative refractive index. The signal shown in Figure 1 illustrates a water droplet with detection in

the near backscatter, resulting in the scattering orders: short-path surface wave, reflection, two modes of second-order refraction, and long-path surface wave.

Furthermore, the time width of the individual peaks in a time-shift signal each resemble the intensity profile of the illuminating laser beam and can be characterized by full width at half maximum (*FWHM*) given by

$$\Delta s = FWHM \left\{ A_p g_p \left( v_z (t - t_0^{(p)}), b \right) \right\} = \frac{b}{v} \quad (2)$$

which is related to the particle velocity  $v$  and to the width of the illumination beam  $b$ , also measured at *FWHM*. According to Eq. (2) the width of individual peaks is inversely proportional to the particle velocity, so that the particle velocity can be calculated by knowing the size of the illuminating beam and measuring the width of the peaks in the signal.

The time separation between peaks (scattering orders) can be expressed as:

$$\Delta t_{(p=a, p=b)} = \frac{d}{v} f(m, \theta_s) = t_0^{(p=a)} - t_0^{(p=b)} \quad (3)$$

and is related to the particle size and velocity. The times  $t_0^{(p)}$  can be taken as the apex of the respective peak in the time-shift signal. If the velocity is known, for instance using Eq. (2), then the particle size can be computed using Eq. (3). The function  $f(m, \theta_s)$  can be determined beforehand using geometric optics or Lorentz-Mie scattering codes.

### Measurement Setup and Signal Interpretation

The major advantage of the time-shift technique is that the detectors and the light source can be located on one side of the measurement point with a small angle between them. Consequently, only one optical access to the measurement position is required. In order to use this configuration for the characterization of transparent particles, and additionally for applying the signal validation discussed later, the scattering angle is best chosen such that reflection ( $p = 0$ ) and two modes of second-order refraction ( $p = 2.1$ ,  $p = 2.2$ ) are detected. The optical configuration in backscatter is illustrated schematically in Fig. 2, in which not one, but two detectors at different scattering angles have been employed.

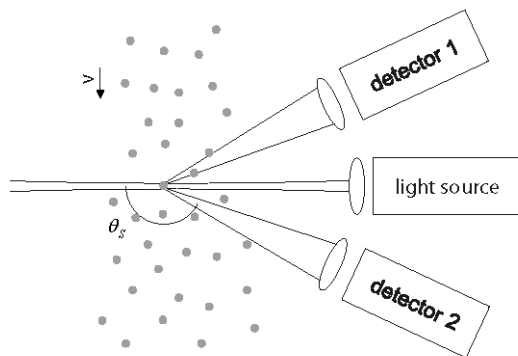


Figure 2: Schematic illustration of the time-shift technique in backscatter

For the experimental setup presented here the time-shift signals each consist of three main peaks: reflection ( $p=0$ ) and two modes of second-order refraction ( $p=2.1$  and  $p=2.2$ ). The contributions from the surface waves can be neglected. Accordingly, the associated time-shift signals can be written for a Gaussian beam profile in following form:

$$S(t) = A_{p=0} \exp\left(-\frac{2(t-t_0^{(p=0)})^2}{b^2}\right) + A_{p=2.1} \exp\left(-\frac{2(t-t_0^{(p=2.1)})^2}{b^2}\right) + A_{p=2.2} \exp\left(-\frac{2(t-t_0^{(p=2.2)})^2}{b^2}\right) \quad (4)$$

Consequently, the individual time-shift signal in backscatter includes three peaks, as illustrated in Fig. 3, so that a single signal includes two time independent separations and the two time-shift signals from the two detectors includes further information related to size and relative refractive index of the particle.

The time duration between peaks illustrated in Fig. 3 are given by simple relations:

$$\Delta t_{2222} = \frac{d}{v} \sin(\theta_i^{(p=2.2)}) \quad (5)$$

$$\Delta t_{2121} = \frac{d}{v} \sin(\theta_i^{(p=2.1)}) \quad (6)$$

$$\Delta t_{00} = \frac{d}{v} \cos\left(\frac{\theta_s}{2}\right) \quad (7)$$

$$\Delta t_{220} = \frac{d/2}{v} \left( \cos\left(\frac{\theta_s}{2}\right) + \sin(\theta_i^{(p=2.2)}) \right) \quad (8)$$

$$\Delta t_{210} = \frac{d/2}{v} \left( \cos\left(\frac{\theta_s}{2}\right) + \sin(\theta_i^{(p=2.1)}) \right) \quad (9)$$

$$\Delta t_{2221} = \frac{d/2}{v} \left( \sin(\theta_i^{(p=2.2)}) - \sin(\theta_i^{(p=2.1)}) \right) \quad (10)$$

where  $\theta_i^{(p)}$  refers to the angle between the illumination direction and the incident point of the respective scattering order on the surface of the particle. Clearly, these equations, together with Eq. (2) provide sufficient information to determine velocity, size and relative refractive index of the spherical, transparent particle.

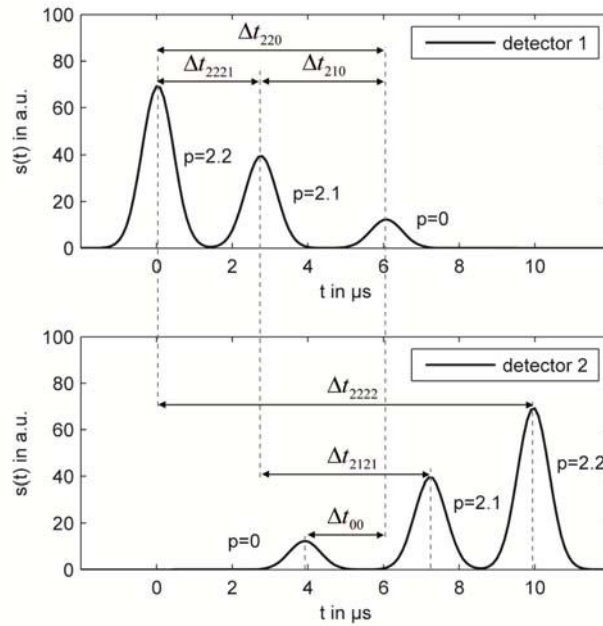


Figure 3: Time-shift signals in backscatter with  $p = 0$ ,  $p = 2.1$  and  $p=2.2$

## Signal Validation

The time-shift technique can only be applied if the detected time-shift signal originates from a single particle. However in some applications, for instance in high density sprays, more than one particle could be illuminated at the same time. This results in overlapping signals, which leads to errors in the analysis, as do signals coming from strongly non-spherical particles. Therefore, a validation at the signal processing stage is required before calculation of particle size, velocity and refractive index, in order to reject incorrect signals. In this study we present two such signal validations for the time-shift technique. They will be referred to as the *gamma* ( $\gamma$ ) and *beta* ( $\beta$ ) conditions; these criteria can be derived using geometric optics and are valid only for transparent particles

The characteristic signal of the time-shift technique includes the signals of reflection ( $p = 0$ ) and two modes of second-order refraction ( $p = 2.1$  and  $p = 2.2$ ). The time separations of the peaks in the time-shift signals are given by Eqs. (5-10). According to these equations, each time separation between peaks is a function of scattering angle, relative refractive index, particle size and velocity, whereby the ratio between time separations are independent of the particle size and velocity and must be constant for a given scattering angle and relative refractive index.

The ratios between different time separations are given by the *gamma* and *beta* functions defined by following relations:

$$\gamma(m, \theta_s) = \frac{\Delta t_{220}}{\Delta t_{210}} = \frac{\frac{d}{2} \left( \cos\left(\frac{\theta_s}{2}\right) + \sin(\theta_i^{(p=2.2)}) \right)}{\frac{d}{2} \left( \cos\left(\frac{\theta_s}{2}\right) + \sin(\theta_i^{(p=2.2)}) \right)} = \frac{\left( \cos\left(\frac{\theta_s}{2}\right) + \sin(\theta_i^{(p=2.2)}) \right)}{\left( \cos\left(\frac{\theta_s}{2}\right) + \sin(\theta_i^{(p=2.2)}) \right)} \quad (11)$$

$$\beta(m, \theta_s) = \frac{\Delta t_{222}}{\Delta t_{2121}} = \frac{\frac{d}{v} \sin(\theta_i^{(p=2.2)})}{\frac{d}{v} \sin(\theta_i^{(p=2.1)})} = \frac{\sin(\theta_i^{(p=2.2)})}{\sin(\theta_i^{(p=2.1)})} \quad (12)$$

These *gamma* and *beta* functions could be used to characterize all ratios between different time separations. For instance:

$$1 - \frac{1}{\gamma(m, \theta_s)} = \frac{\Delta t_{2221}}{\Delta t_{220}} \quad (13)$$

$$1 - \gamma(m, \theta_s) = \frac{\Delta t_{2221}}{\Delta t_{210}} \quad (14)$$

The numerical calculation of *beta* and *gamma* functions as a function of scattering angle for different relative refractive indexes are depicted in Figs 4 and 5. They are monotonic functions and consequently, for a given relative refractive index and scattering angle, only one value of *gamma* and *beta* exists. For that reason, the signal(s) originating from a single spherical particle must have approximately the expected value of *gamma* and *beta* for that particular refractive index. If it is not the case, these signals will be rejected.

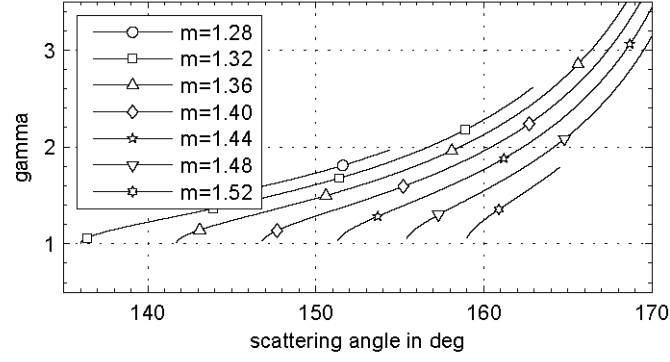


Figure 4: *Gamma* as a function of scattering angle, calculated for different refractive indexes.

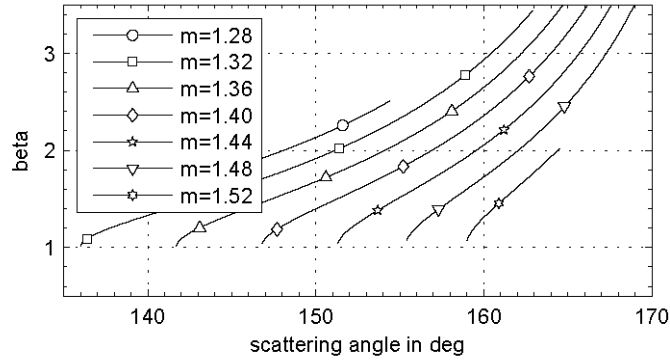


Figure 5: *Beta* as a function of scattering angle, calculated for different refractive indexes.

Additionally, there is an option to determine the incident angles, which are related to the relative refractive index and scattering angle. If the detectors are located symmetrically, the incident angles can be computed from the *beta* and *gamma* functions using the relation

$$\sin(\theta_i^{(p=2.2)}) = \cos\left(\frac{\theta_s}{2}\right) \left(\frac{\gamma-1}{\beta-\gamma}\right) = \frac{1}{\beta} \sin(\theta_i^{(p=2.1)}) \quad (15)$$

and the relative refractive index can be calculated from these incident angles by the relation:

$$m = \frac{\sin(\theta_i^{(p=2.1)})}{\sin\left(\frac{\pi}{4} - \frac{\theta_s}{4} + \frac{\theta_i^{(p=2.1)}}{2}\right)} = \frac{\sin(\theta_i^{(p=2.2)})}{\sin\left(\frac{\pi}{4} - \frac{\theta_s}{4} + \frac{\theta_i^{(p=2.2)}}{2}\right)} = f(\theta_s, \beta, \lambda) \quad (16)$$

Consequently the relative refractive index is a function of scattering angle, which is assumed to be known parameter.

### Measurement Results

The experimental setup consists of two photodetectors placed symmetrically around the light source at a scattering angle of about  $165deg$  (see Fig. 2). The laser beam is focused to a laser sheet with thickness of approx.  $38\mu m$  and approx. width of  $1000\mu m$ . This defines a measurement volume of approx.  $1mm^3$ . The minimal particle size, which can be measured by this experimental setup, is about  $100\mu m$ , because the thickness of the laser sheet is  $38\mu m$ .

First measurements were performed on glass beads with a narrow particle size distribution around  $250\mu m$ . For comparison the particle size distribution of the glass beads was first measured by direct imaging. These transparent glass particles have an approximate spheri-

cal shape and the refractive index is 1.51. About 20000 images were processed to calculate the reference size distribution using direct imaging.

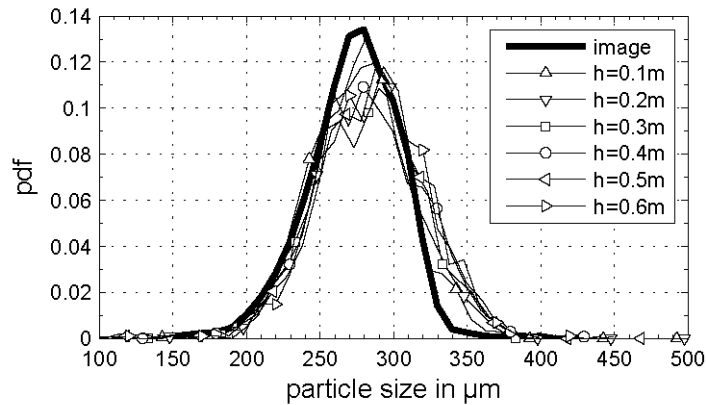


Figure 6: Particle size distribution of glass beads measured by direct imaging (black line) and by the time-shift technique (symbols).

The glass beads were allowed to flow out of a container through an opening of 0.7mm, accelerating due to gravity with distance from the exit orifice. The flow of beads from the orifice was such that often several beads passed through the measurement volume at once, similar to situations in sprays, in which multiple droplets are present in the measurement volume. The measurement position was varied at several distances from the exit orifice between 100mm and 600mm; resulting in increasing velocities with increasing distances.

The particle size distribution measured by the time-shift technique is depicted in Fig. 6, where measurements performed at different distances from the nozzle ( $h$ ) are shown. The measurements of falling glass beads from different heights results in approximately the same distribution, indicating a high repeatability of the measurements. Additionally the particle size distribution measured by the imaging method is compared with results performed by the time-shift technique. It can be seen that the results are in good agreement.

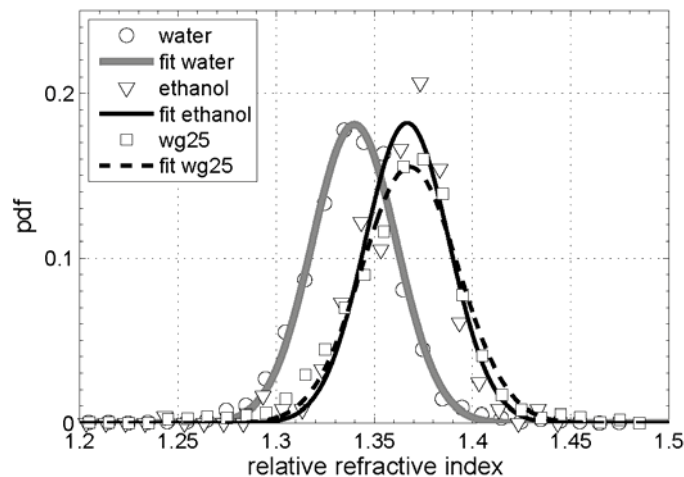


Figure 7: Relative refractive index distribution measured by the time-shift technique.

The same experimental setup (see Fig. 2) was used to measure the relative refractive indexes of different sprays: water, water/glycerin mixture with 0.25 concentration (wg25) and ethanol, using the  $\gamma$  and  $\beta$  functions. For these measurements the scattering angle was

changed to  $155\text{deg}$ . The entire particle size distribution in such a spray cannot be measured, because in the experimental setup the minimal particle size being detected is around  $100\mu\text{m}$ . The flat fan nozzle was operated by  $2\text{bar}$  relative pressure creates typically particles with a size distribution beginning from approximately  $10\mu\text{m}$ , so that there are also small particles below  $100\mu\text{m}$ . Consequently, only the signals originated from particles larger than  $100\mu\text{m}$  could be used for calculation of relative refractive index.

The experimental results are illustrated in Fig. 7, where the histogram of measured relative refractive index for water, water-glycerin mixture and ethanol are shown. These results indicate also the approximate resolution of the measured relative refractive index, being about  $0.01$ , which is caused by errors in calculating values of  $\beta$  and  $\gamma$ . Although not high, this resolution is sufficient for some applications in which different materials must be distinguished.

## Conclusion

The primary aim of the present work was the further development of the time-shift technique for droplet/particle characterization in spray applications. Although this measurement technique has been known for over 20 years, until now it has not been successfully applied to spray characterization because of several secondary problems which have been outlined and partially solved in this study. The analysis and processing of a time-shift signal has been described for computing the characteristic parameters of a single particle. The study concludes with example measurements, demonstrating the functionality of the measurement technique. It has been demonstrated, that the signal validation works successfully and the calculation of particle size is plausible. In addition, using the novel validation conditions presented in this study, the relative refractive index of transparent particles could be estimated from the measurement results performed by the time-shift technique.

## Acknowledgements

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