

# Phase Doppler sizing of optically absorbent single- and multicomponent liquid droplets using semiconductor devices

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**Abstract.** After a successful application of the phase Doppler technique to homogeneous transparent or opaque dispersed media, which are mainly used for model experiments, it was the aim with this method simultaneous size and velocity measurements of real process fluid droplets that are optically absorbent and/or inhomogeneous, in order to open the way to in-line measurements for process control. Starting with an optically absorbent homogeneous liquid (black ink), phase Doppler measurements were taken at off-axis angles  $\varphi$ , where either refracted light or reflected light dominates. Varying both the sizes of the monodisperse droplets and their optical absorption showed the limits of the  $\Delta\Phi$ - $d$  relation for dominant refraction and a transition to dominant reflection at an off-axis angle  $\varphi$  of  $30^\circ$ . On the basis of previous experiments and the idea of using a larger wavelength of 830 nm (semiconductor laser) instead of 488 nm (Ar ion laser), ambiguous phase difference-particle diameter relationships due to absorption were avoided. In a second step the phase Doppler technique was applied to monodisperse droplets of inhomogeneous liquids. Using the larger wavelength the broad size distributions, experimentally achieved by employing the Ar ion laser, became much narrower. It was therefore concluded that the use of semiconductors for the phase Doppler measurements can be advantageous for dealing with absorption and inhomogeneities.

For optically absorbent homogeneous liquid droplets, the Mie theory alone proves to be an indispensable means for selecting the suitable PDA set-up parameters. For inhomogeneous liquid droplets, the Mie theory and Mie scattering computer programs can be used to describe and understand their scattering behaviour, if—as a precondition—it is valid to assign to them an effective complex refractive index. Thus, computer aided PDA (CAPDA) measurements open the chance for in-line process measurements in real process liquid spraying.

## 1. Introduction

Compared with other particle measurement techniques, phase Doppler anemometry (PDA) has the advantage of *simultaneously* measuring the size and velocity of spherical particles, and is therefore of interest in many industrial spray applications for quality assistance and/or process control. This method has so far been successful in the analysis of liquid atomization of water [1–3] and molten metals [4, 5] as well as in the diagnosis of model experiments with glass spheres or bubbles in water flows [6, 7]. While all these dispersed media are either transparent or opaque, most real process fluids—for instance in the chemical, pharmaceutical and food industries—are not only simply optically absorbent but also inhomogeneous.

This leads to difficulties in applying PDA to these real process fluids. As described in a previous publication [8], we concluded that the use of a larger laser wavelength should reduce the problems. This idea was taken up for further study and has been validated in this article.

## 2. Theory

The basic principles of phase Doppler anemometry have been described elsewhere [1, 7, 9, 10], with most of these publications based on the work of Durst and Zaré [11]. Since a detailed description exists in the literature [12],

we confine ourselves to a brief description of PDA and then direct our attention to scattering theory and geometrical optics.

**2.1. Phase Doppler anemometry**

Two laser beams focused at an angle  $\theta$  form the interference volume of a phase Doppler anemometer (figure 1). As an extension of the well known laser Doppler anemometer, the light scattered by a spherical particle passing through the interference volume is detected by two (instead of only one) photodetectors. These photodetectors are usually arranged at the same off-axis angle  $\varphi$  and symmetrically to the interference planes with equal elevation angles  $\psi$ .

The scattered light registered by both detectors has a temporal frequency due to the interference fringes  $f_D = 1/T$  (see figure 2) and is directly related to the particle velocity according to equation (1)

$$|v_z| = \frac{f_D \lambda}{2 \sin(\theta/2)} \tag{1}$$

Only when either refraction or reflection dominates, the time difference  $\Delta t$  measurable as a phase difference  $\Delta\Phi = 2\pi \Delta t/T$  (see figure 2) between a pair of signals (Doppler bursts) can simply be measured and is linearly related

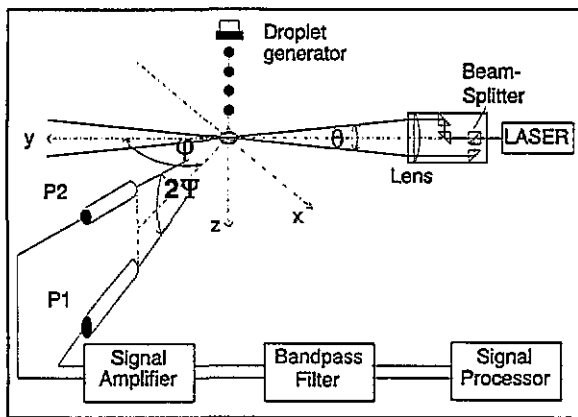


Figure 1. Phase Doppler anemometer (schematic).  $\theta$  beam crossing angle;  $\varphi$  off-axis angle;  $\psi$  elevation angle.

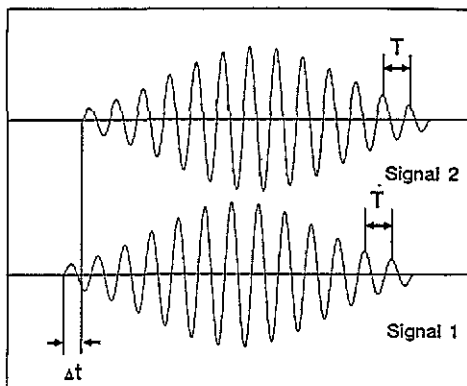


Figure 2. Phase difference  $\Delta\Phi = 2\pi (\Delta t/T)$  between two Doppler bursts.

to the diameter  $d$  of the spherical particle as expressed by

$$d = \frac{1}{2b} \left( \frac{\lambda}{\pi n_c} \right) \Delta\Phi \tag{2}$$

where  $n_c$  is the refractive index of the continuous phase and  $b$  a function of the geometrical arrangement. The term  $b$  for both cases is listed in table 1 as equations (3) and (4). The scattering behaviour of a liquid droplet determines the off-axis angles  $\varphi$  where reflected or refracted light is dominant and where the positions of the photomultipliers have to be arranged. For a homogeneous liquid, Mie theory [13] usually provides a sufficient description of the light scattering but is not exact, since it assumes plane incident wavefronts.

Equations (2)–(4) can be used without any difficulty for the analysis of PDA signals resulting from dispersed materials with simple optical properties. But for more complicated optical properties of the dispersed phase the validity of these quantities may become doubtful.

**2.2. Electromagnetic scattering theory**

**2.2.1. Light scattering by optically absorbent single-component liquid droplets.** Already by 1908 Mie had obtained, on the basis of the electromagnetic theory, a rigorous solution for the light scattering of a plane monochromatic wave by a homogeneous sphere of any diameter situated in a homogeneous medium. For ‘Mie’ parameters  $\alpha = (\pi d/\lambda) > 10$ , light scattering can be well approximated by geometrical optics. This allows the separation into diffraction, reflection and refraction as well as into higher orders of scattering [14]. Quantities such as refraction and absorption, necessary to describe the optical properties of the dispersed and continuous medium, are used in the Mie calculations in the form of a real and imaginary part of a complex refractive index given by

$$n = n' - i \kappa \tag{5}$$

where  $\kappa = K\lambda/4\pi$  is the absorption coefficient,  $K$  the absorption constant and  $n'$  the refractive index.

The refractive index  $n'$  is determined usually by means of an Abbé refractometer using the phenomenon of total reflection. Optical absorption in a refractometric homogeneous sample can give rise to errors much larger than the precision typical of critical angle refractometry [15]. This error (about  $1 \times 10^{-3}$  in  $n'$  at  $\kappa = 1 \times 10^{-2}$ ) can, however, be ignored when using Mie calculations for phase Doppler anemometry.

The absorption constant  $K$  of a homogeneous liquid is obtained with a spectrophotometer (accuracy  $\pm 1\%$  [16]) measuring the attenuation of light traversing a distance  $x$  through a particulate medium [17]:

$$\frac{I_{\text{transmitted}}}{I_{\text{incident}}} = \exp(-Kx). \tag{6}$$

Usually the attenuation, that is to say the extinction,

**Table 1.** Term  $b$  for reflection and refraction.

$$b_{\text{reflection}} = \sqrt{2} \left[ \left( 1 + \sin \frac{\theta}{2} \sin \Psi - \cos \frac{\theta}{2} \cos \Psi \cos \varphi \right)^{1/2} - \left( 1 - \sin \frac{\theta}{2} \sin \Psi - \cos \frac{\theta}{2} \cos \Psi \cos \varphi \right)^{1/2} \right] \quad (3)$$

$$b_{\text{refraction}} = 2 \left\{ \left[ 1 + n'^2 - \sqrt{2} n' \left( 1 + \sin \frac{\theta}{2} \sin \Psi + \cos \frac{\theta}{2} \cos \Psi \cos \varphi \right)^{1/2} \right]^{1/2} - \left[ 1 + n'^2 - \sqrt{2} n' \left( 1 - \sin \frac{\theta}{2} \sin \Psi + \cos \frac{\theta}{2} \cos \Psi \cos \varphi \right)^{1/2} \right]^{1/2} \right\} \quad (4)$$

$$n' = \frac{n_d}{n_c}$$

is caused by absorption and scattering [15]

$$K_{\text{ext}} = N (C_{\text{abs}} + C_{\text{sca}}) = K + K_{\text{sca}} \quad (7)$$

where  $N$  is the number of particles per unit volume,  $C_{\text{abs}}$  the absorption cross section and  $C_{\text{sca}}$  the scattering cross section.

It should be noted that in media classified as homogeneous, the dominant attenuation mechanism is absorption.

**2.2.2. Light scattering by optically absorbent multicomponent liquid droplets.** Many process fluids—for instance in the chemical, pharmaceutical and food industries—are not only optically absorbent but also inhomogeneous. Naturally, homogeneity is relative, but usually a medium can be treated as homogeneous when the difference between the refractive indices of the constituents is small or when the constituents are small compared with the wavelength [17].

At present, light-scattering theories concerning inhomogeneous liquid droplets still have to be seen being at the beginning stages of development [18–22] because they are capable only of calculating the scattering behaviour of drop sizes of the order of the light wavelength  $\lambda$  or smaller. This is mainly due to the validity range of the approximations or the necessary computer storage capacity or the computation time that is required. However, to overcome this problem, effective medium theories were put forward using an effective complex refractive index of a composite medium for calculating absorption and the scattering behaviour of this medium [23–25]. Several different mixing rules exist for calculating the effective refractive index which are well summarized and compared by Srivastava [26].

The 'effective medium' theories may serve also for calculating the fundamental scattering behaviour of microscopically inhomogeneous liquid droplets by means of the well established Mie theory. By using an effective complex refractive index for calculating the scattering behaviour, the liquid is treated as macroscopically homogeneous. While the calculation of the complex refractive index—especially in the case of multicomponent liquids—is extremely complicated and in most cases impossible, the mean or effective refractive index is measured quite simply: it is done experimentally in the same way as described above.

If the liquid is non-homogeneous, the precision in determining the refractive index  $n'$  is reduced, not only

by optical absorption originating from conversion of light into heat, but also from scattering inside the medium or even from both. Nevertheless, the critical angle refractometric technique gives fairly precise ( $\Delta n' \approx \pm 0.003$ ) refractive index data [27].

The extinction determined by means of a photometer is a result of both absorption and scattering. For a better understanding of the influence of the inhomogeneities on the PDA signals it is necessary to know the relative contributions of absorption and scattering to extinction, which is also a function of the wavelength. However, a first overview of the scattering angle intervals with dominant reflection, refraction, etc. can be calculated using the measured extinction coefficient instead of the absorption coefficient for Mie calculations. Using a too high absorption coefficient enables one at least to check deviations from linearity of the  $\Delta\Phi$ - $d$  relation for normally dominant refraction. So far we have not considered multiple scattering, i.e. when the light is scattered several times in succession before it leaves the medium. It occurs when the number of inhomogeneities is very high and their separation very small. Equations (6) and (7) are only valid when multiple scattering is negligible [17].

### 3. Experimental set-up

A phase Doppler anemometer similar to that shown in figure 1 is realized as follows. The blue beam ( $\lambda = 488$  nm) of a 4 W Ar ion laser is divided into two parallel beams of the same intensity with adjustable polarization and then focused to yield the interference volume of the anemometer. Two photomultipliers symmetrically positioned at an elevation angle  $\psi$  relative to the horizontal scattering plane and at an off-axis angle  $\varphi$  are used to detect the scattered light. In the semiconductor device the Ar ion laser is replaced by a laser diode emitting at  $\lambda = 830$  nm and two avalanche photodiodes take the place of the photomultipliers.

The use of semiconductor components has some principal, i.e. independent of the liquid under investigation, advantages: it allows the construction of miniaturized instruments of higher robustness, better signal-to-noise ratios, and higher quantum efficiencies when compared with conventional equipment based on gas lasers and photomultipliers [28].

In tables 2(a) and 2(b) the exact geometrical parameters are summarized. The phase difference and the frequency information of the bandpass-filtered Doppler bursts were obtained by FFT processing [29].

For testing the applicability of the phase Doppler technique to optically absorbent liquids, droplets of known sizes are necessary. Therefore a stream of mono-disperse droplets passing through the measuring volume was produced by an impulse jet technique, which is comprehensively discussed by Heinzl and Hertz [30]. A pressurized liquid jet is broken into a stream of uniformly sized droplets by a mechanical disturbance caused by a piezoelectric tube [31]. Photos of the droplets, illuminated by a xenon flashlamp stroboscope (illumination time=150 ns) and enlarged 63 times by means of a microscope, are taken to achieve reference values.

#### 4. PDA experiments

##### 4.1. Phase Doppler technique applied to an optically absorbent single-component liquid

Black ink (Pelikan 4001, Brillant Schwarz) seemed to be a promising sample for investigating the influence of

absorption on the  $\Delta\Phi-d$  relation. Due to its high absorption ( $\kappa \approx 1 \times 10^{-2}$ ) it should be possible to adjust the imaginary part of the refractive index over a wide range by diluting it with distilled water. Furthermore, it was treated as a homogeneous liquid, as analysis of the particles inside the black ink by laser diffraction led to the result that their size is smaller than  $0.5 \mu\text{m}$ . According to the specifications of the manufacturer most of the pigments should be dissolved. Even if there are still (very small) particles inside, absorption dominates over scattering and contributes primarily to extinction.

All refractive index  $n'$  measurements were made with a high-precision ocular Abbé instrument (ZEISS, Model B) at wavelengths of 488 and 632.8 nm. The absorption was obtained with a spectrophotometer (Hamamatsu, UV-2102 PC). Table 3 contains the complex refractive indices of the chosen ink solutions for  $\lambda=488 \text{ nm}$ .

At an off-axis angle  $\phi$  of  $30^\circ$  for water, refraction dominates (see figure 3) and the high scattered light intensities in the forward scattering (see figure 4) lead to good signal-to-noise ratios. Therefore, as in many industrial applications, it was at this angle that we carried out an investigation of the influence of absorption on the  $\Delta\Phi-d$  dependency.

Mie calculations showed [8] that it should be possible to apply the phase Doppler technique for particle

Table 2(a). PDA optical parameters, Ar ion laser device.

Transmission optics		
Laser wavelength $\lambda$ (nm)	488	
Lens focal length $f$ (mm)	1200	
Beam crossing angle $\theta$ (deg)	1.88	
Measuring volume ( $\mu\text{m}$ )	$d_x$	564
	$d_y$	34394
	$d_z$	565
Receiving optics		
Off-axis angle $\phi$ (deg)	30	90
Collection aperture diameter (mm)	52	52
Distance from measuring volume (mm)	1000	800
Elevation angle $\psi$ (deg)	1.85	4.00
Sizing range: ( $\mu\text{m}$ )	refraction	300 214
	reflection	239 302

Table 2(b). PDA optical parameters, semiconductor device.

Transmission optics		
Laser wavelength $\lambda$ (nm)	830	
Lens focal length $f$ (mm)	1000	1185
Beam crossing angle $\theta$ (deg)	2.86	2.42
Measuring volume ( $\mu\text{m}$ )	$d_x$	264 190
	$d_y$	10586 8985
	$d_z$	264 190
Receiving optics		
Off-axis angle $\phi$ (deg)	30	90
Collection aperture dimensions (mm)	$33.4 \times 40.5$	$33.4 \times 40.5$
Distance from measuring volume (mm)	500	160
Elevation angle $\psi$ (deg)	1.91	5.90
Sizing range: ( $\mu\text{m}$ )	refraction	324 192
	reflection	259 271

Table 3. Refractive indices of the ink solutions.

Concentration (ink: water) weight per cent	Complex refractive index $\lambda=488 \text{ nm}$ , temperature = $20^\circ\text{C}$
0	$1.33 - i$
2	$1.33 - i 0.78 \times 10^{-4}$
4	$1.33 - i 6.43 \times 10^{-4}$
7	$1.34 - i 9.03 \times 10^{-4}$
12	$1.34 - i 1.28 \times 10^{-3}$
25	$1.34 - i 2.49 \times 10^{-3}$
50	$1.34 - i 3.96 \times 10^{-3}$
75	$1.34 - i 6.37 \times 10^{-3}$
100	$1.35 - i 0.89 \times 10^{-2}$

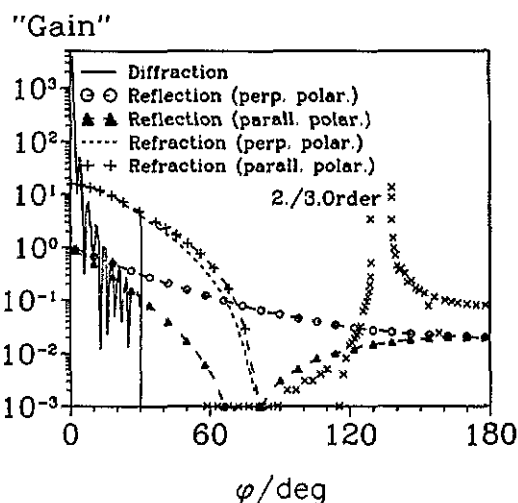
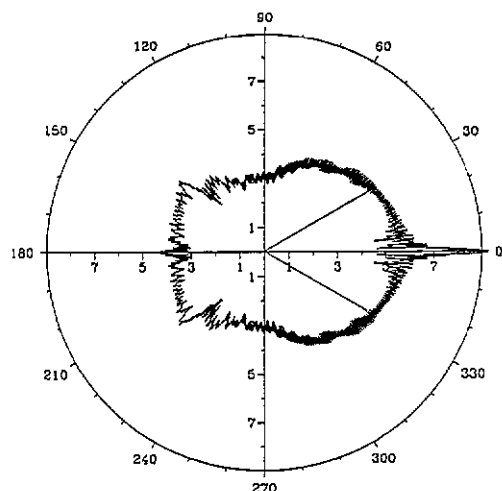


Figure 3. Geometrical optics for water ( $n=1.333$ ).



**Figure 4.** Logarithm of the scattered light intensity as a function of the scattering angle for a water droplet ( $d = 50 \mu\text{m}$ ,  $n = 1.33$ ,  $\lambda = 632.8 \text{ nm}$ ).

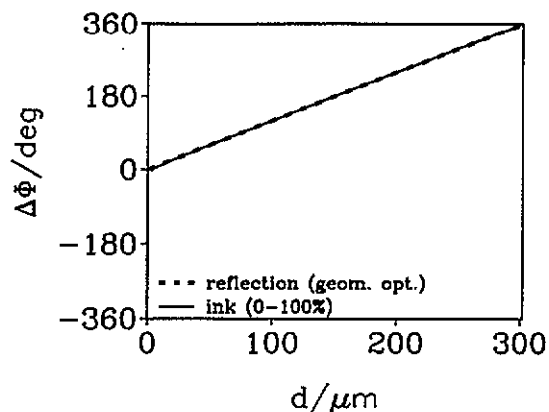
sizing by using parallel polarized laser light and the  $\Delta\Phi$ - $d$  relation for dominant refraction up to an ink concentration of 12%. This, however, reduces the sizing range to  $150 \mu\text{m}$ . However, for ink concentrations equal to or larger than 25% the  $\Delta\Phi$ - $d$  relation for dominant reflection in combination with perpendicular polarization has to be used.

This could be demonstrated by applying the PDA to monodisperse droplets of each ink concentration over a range of three different diameters [8]. The generated droplet diameters were roughly twice the orifice diameter (25, 50 and  $100 \mu\text{m}$ ). The maximum error in determining the droplet diameter by phase Doppler technique was of about 8% [32]. The vagueness of photography was equivalent to 3–12% ( $\Delta d = \pm 6 \mu\text{m}$ ).

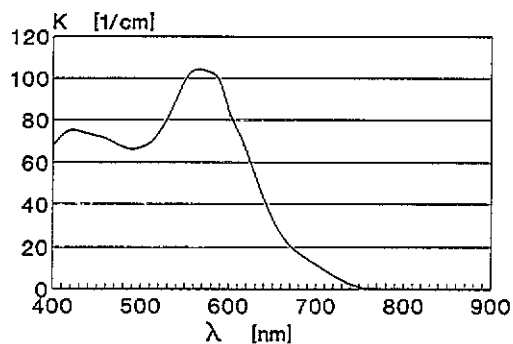
Due to a possible mass and heat transfer in spraying processes a change of concentration of an optically absorbent droplet constituent can happen, for instance by evaporation of solvent [33]. This means that the absorption of the liquid droplets varies and that for each concentration a  $\Delta\Phi$ - $d$  relation different from the assumed linear  $\Delta\Phi$ - $d$  dependency is valid. To avoid this ambiguity the photodetector should be set at an off-axis angle  $\varphi$  where reflected light dominates, because it is uninfluenced by absorption.

Figure 5 shows the  $\Delta\Phi$ - $d$  relations predicted by Mie theory (chosen off-axis angle  $\varphi = 90^\circ$ ) for water and pure ink. The results were identical to the simplified relation of the geometrical optics. This was verified by an excellent agreement between the droplet sizing results of PDA and photography [8].

However, it was also desirable to have a linear  $\Delta\Phi$ - $d$  relation and high scattered light intensities. Therefore we referred to the absorption constant of black ink as a function of the wavelength (see figure 6). In contrast to the imaginary part of the refractive index it is the absolute measure for absorption or scattering. Unlike the strong absorption in the visible region, the absorption was very weak in the near infrared, and therefore a

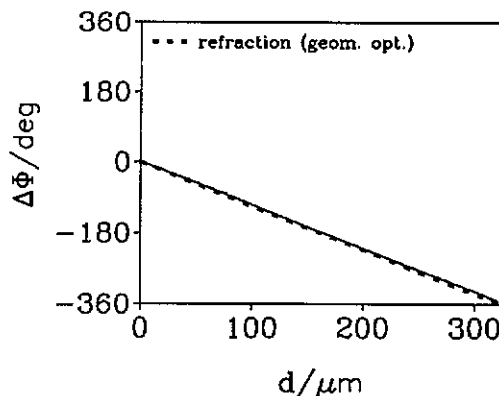


**Figure 5.** Mie calculations for water and pure ink,  $\varphi = 90^\circ$ , perpendicular polarized light,  $\lambda = 488 \text{ nm}$ .



**Figure 6.** Absorption constant of black ink solution,  $c = 4\%$ , as a function of the wavelength  $\lambda$ .

laser emitting in this region was needed. This was realizable by means of a semiconductor laser emitting at  $\lambda = 830 \text{ nm}$  where the imaginary part of the refractive index of pure black ink was only  $1.14 \times 10^{-5}$ . The calculated  $\Delta\Phi$ - $d$  relation (see figure 7, pure ink,  $\varphi = 30^\circ$ ), as well as the good agreement between the droplet sizing results of PDA and photography (see table 4), shows that it is possible to use the simplified  $\Delta\Phi$ - $d$  relation for dominant refraction and for all ink concentrations. Non-linear  $\Delta\Phi$ - $d$  relations or a shift to the  $\Delta\Phi$ - $d$  relation for



**Figure 7.** Mie calculations for pure ink,  $\varphi = 30^\circ$ , parallel polarized light,  $\lambda = 830 \text{ nm}$ .

**Table 4.** Sizing results,  $\varphi = 30^\circ$ ,  $\lambda = 830$  nm, 2000 counts.

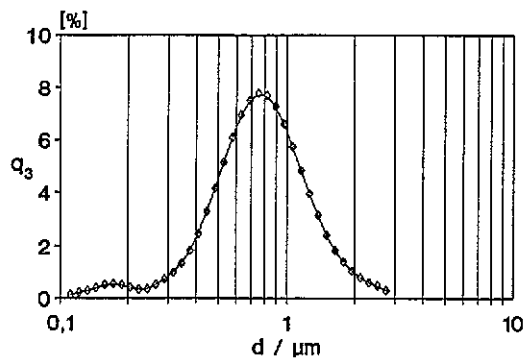
Ink concentration (%)	Result by photography		Result by PDA		by equation (2) and
	$d$ ( $\mu\text{m}$ )	$ \Delta d $ ( $\mu\text{m}$ )	$d_{10}$ ( $\mu\text{m}$ )	$ \Delta d $ ( $\mu\text{m}$ )	
[0]	57	6	53	4.2	(4)
	101		105	8.4	
	170		185	14.8	
[12]	57	6	51	4.1	(4)
	95		88	7.0	
	152		157	12.6	
[100]	48	6	56	4.5	(4)
	97		86	6.9	
	158		159	12.7	

dominant reflected light are absent. Consequently, the use of a semiconductor laser emitting in the near infrared can improve the applicability of PDA to optically absorbent liquids. However, due to its dependence on the absorption of the liquid under investigation in the near infrared, it is not a general solution.

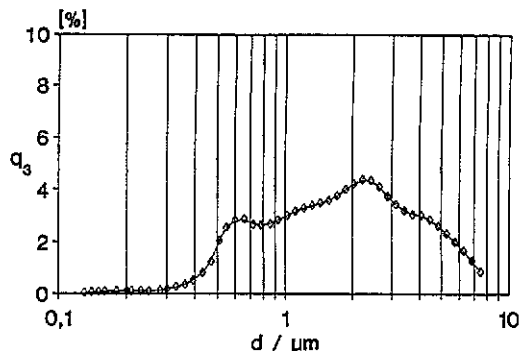
**4.2. Phase Doppler technique applied to an optically absorbent multicomponent liquid**

Solutions of instant coffee (Nescafé KSX) and condensed milk (Glücksklee, 7.5%) represent typical process fluids since in the food industry the types of fluid are processed by spray drying to yield powders. The lipids and proteins in condensed milk were dispersed as globules of sizes up to 3  $\mu\text{m}$  (see figure 8). The refractive index of the milk fat globules is real and varies between 1.4527 and 1.4566 [34]. This means that condensed milk is an emulsion of very weakly absorbing particles which therefore attenuate light primarily by scattering. Solutions of instant coffee contain dispersions with dimensions in the order of up to 7  $\mu\text{m}$  (see figure 9). Therefore both solutions have to be treated as inhomogeneous liquids.

Their complex refractive indices were determined experimentally in the same way as described above and are summarized in table 5. In the case of condensed milk the imaginary part of the refractive index is chiefly a result of scattering, whereas in the case of solutions of



**Figure 8.** Volume density distribution of particles in condensed milk.



**Figure 9.** Volume density distribution of particles in instant coffee.

**Table 5(a).** Refractive indices of milk and coffee solution,  $\lambda = 488$  nm.

Liquid	Complex refractive index $n = n'_{\lambda=488\text{nm}} - i\kappa$ $\lambda = 488$ nm Temperature = 20 °C	Extinction constant $K_{\text{ext}}$ [ $\text{cm}^{-1}$ ]
Condensed milk solution (8%)	1.3398 - $i 2.58 \times 10^{-4}$	66.4
(48%)	1.3544 - $i 3.04 \times 10^{-4}$	78.3
Instant coffee solution (8%)	1.3522 - $i 4.47 \times 10^{-4}$	115

**Table 5(b).** Refractive indices of milk and coffee solution,  $\lambda = 830$  nm.

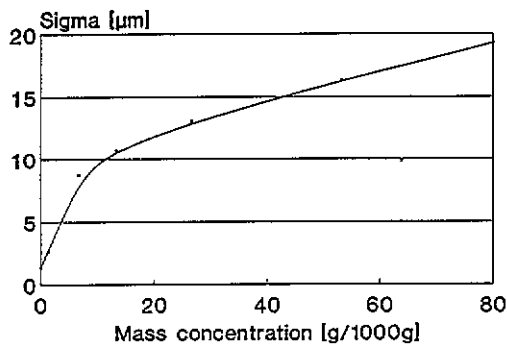
Liquid	Complex refractive index $n = n'_{\lambda=830\text{nm}} - i\kappa$ $\lambda = 830$ nm Temperature = 20 °C	Extinction constant $K_{\text{ext}}$ ( $\text{cm}^{-1}$ )
Condensed milk solution (8%)	1.3348 - $i 5.86 \times 10^{-5}$	8.9
Instant coffee solution (8%)	1.3468 - $i 8.50 \times 10^{-5}$	12.9

instant coffee it is a combination of strong scattering and weak absorption. It was technically impossible to determine the refractive index  $n'$  at a light wavelength  $\lambda = 830$  nm with a commercial refractometer. (Normally the deviation of the used refractive index  $n'_{\lambda=632.8\text{nm}}$  from the real refractive index  $n'_{\lambda=830\text{nm}}$  is small, but due to the different dispersive behaviours of the liquids this cannot be generalized.)

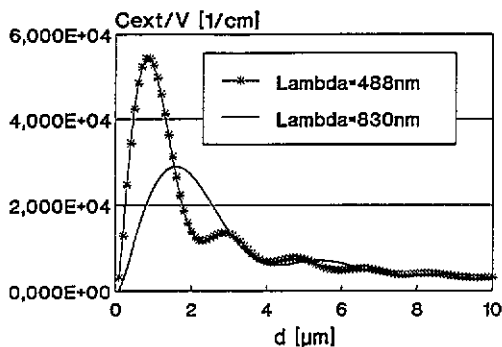
Treating the measured quantities as the effective refractive indices, it should be possible to apply PDA for droplet sizing at an off-axis angle  $\varphi$  of  $30^\circ$ —as predicted by Mie theory. Nevertheless it has to be taken into account that the real mechanism of light attenuation is scattering by inhomogeneities in the droplet. Accordingly the detected refracted light can be disturbed by scattering inside the droplets in such a way that the phase difference  $\Delta\Phi$  deviates from that for a straight optical path. In fact

it turned out that the experimentally verified size distributions of the monodisperse droplets were extremely broad, although the mean diameters  $d_{10}$  were in good agreement with the results by photography [8]. The latter seems to prove that the scattering behaviour of such liquid droplets is basically similar to that predicted by the Mie theory using an effective refractive index, but that the refracted light is disturbed by inhomogeneities inside the droplet. Admittedly changes of concentration can occur, but they affect the real and the imaginary parts of the refractive index very little (see table 5).

The scattering of the refracted light by inhomogeneities inside the droplets is subject to their (mass or number) concentration. Figure 10 exhibits this when one treats the standard deviation of the number distribution obtained by PDA applied to monodisperse condensed milk droplets (diameter about  $100 \mu\text{m}$ ) as a rate for scattering. Furthermore the scattering of the inhomogeneities and therefore its negative influence on the Doppler bursts depends on the size of the inhomogeneities. Figure 11 shows the extinction cross section per unit particle volume of Latex particles in water, which serves as a model for condensed milk (fat globules in water), as a function of their size. For  $\lambda=488 \text{ nm}$  Mie theory shows a strong peak at  $d \approx 1 \mu\text{m}$ , whereas at  $\lambda=830 \text{ nm}$  the peak is shifted to about  $1.7 \mu\text{m}$ , less pronounced and



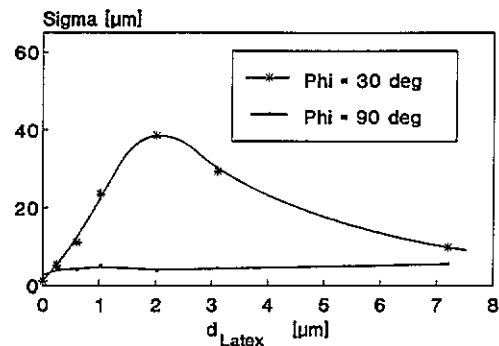
**Figure 10.** Standard deviation of PDA sizing results applied to monodisperse droplets of condensed milk,  $d \approx 100 \mu\text{m}$ , 2000 counts,  $\varphi = 30^\circ$ ,  $\lambda = 830 \text{ nm}$ .



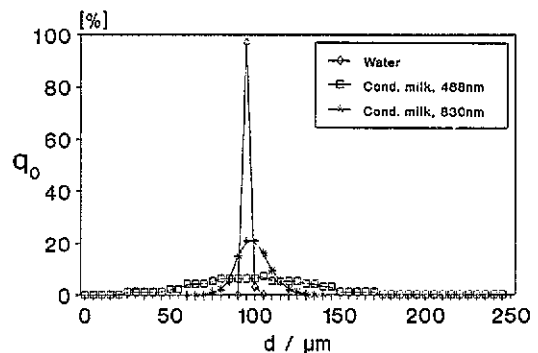
**Figure 11.** Extinction cross section per unit particle volume according to Mie theory for latex particles in water.  $n_{\text{water}, \lambda=488 \text{ nm}} = 1.337$ ,  $n_{\text{latex}, \lambda=488 \text{ nm}} = 1.603$ ,  $n_{\text{water}, \lambda=830 \text{ nm}} = 1.328$ ,  $n_{\text{latex}, \lambda=830 \text{ nm}} = 1.579$ .

on the whole the extinction is smaller. In figure 12 the standard deviations of the number distributions achieved by PDA applied to monodisperse droplets (diameter about  $150 \mu\text{m}$ ) of monodisperse Latex suspensions (Latex particles in water) are shown as a function of the Latex particle diameter. For an off-axis angle of  $30^\circ$ , there is a peak at  $2 \mu\text{m}$  and in general the curve is similar to that predicted by Mie theory. Detecting the reflected light by measuring at an off-axis angle of  $90^\circ$  indicates that in reality the generated droplets are nearly monodispersed Latex and that the broad size distributions at an off-axis angle of  $30^\circ$  are only the result of scattering by inhomogeneities. Due to the lower scattering that is to be expected, the idea of repeating the PDA measurements with a laser emitting in the near infrared to size condensed milk droplets seemed to suggest itself. As predicted by the model calculations, experiments using the semiconductor PDA device provided narrow sized diameter distributions (as can be seen in figure 13) with a peak more closely packed than that obtained by the Ar ion laser at the same off-axis angle  $\varphi$  of  $30^\circ$ .

Although the scattered light intensities at an off-axis angle  $\varphi$  of  $90^\circ$ —where normally reflection is the only light scattering component—are low, additional experiments were executed using the Ar ion PDA device as well as the semiconductor device. For this angle, light passing through the droplet and therefore affected by the



**Figure 12.** Standard deviation of size distributions obtained by PDA applied to monodisperse water droplets ( $d \approx 150 \mu\text{m}$ ) containing monodisperse latex particles, 2000 counts,  $\lambda = 830 \text{ nm}$ .



**Figure 13.** PDA results belonging to droplets of  $99 \mu\text{m}$  in diameter, 2000 counts, detection of refracted light ( $\varphi = 30^\circ$ ).

inhomogeneities inside the liquid should not contribute to the detected light. Indeed these experiments provided size distributions which can be described as much more narrow, but not as monosized (see figure 14). Once again there is fairly good agreement in the mean diameter values between PDA and photography, as summarized in table 6. The width of the size distributions may be influenced by the surface roughness caused by the inhomogeneities peeping partly out of the droplet.

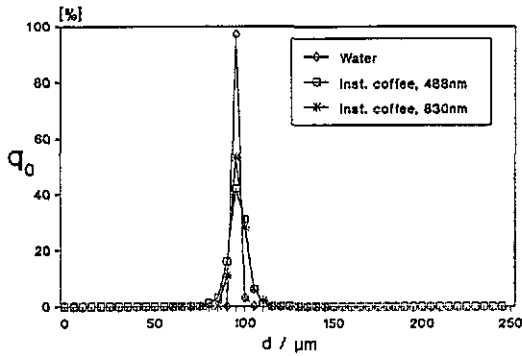


Figure 14. PDA results belonging to droplets of 101 μm in diameter, 2000 counts, detection of reflected light ( $\varphi = 90^\circ$ ).

Table 6. Sizing results for condensed milk and instant coffee solutions,  $\varphi = 90^\circ$ ,  $\lambda = 488$  nm, 2000 counts.

Liquid	Result by photography		Result by PDA		By equation (2) and
	$d$ (μm)	$ \Delta d $ (μm)	$d_{10}$ (μm)	$ \Delta d $ (μm)	
Water	59	6	58	4.6	(3)
	86		85	6.8	
	137		137	11.0	
Condensed milk solution (8%)	63	6	59	4.7	(3)
	89		87	7.0	
	178		167	13.4	
Instant coffee solution (8%)	58	6	51	4.1	(3)
	101		99	7.9	
	148		142	11.4	

Using the semiconductor device the width of the size distributions is once again reduced (see table 7). In this table the standard deviations of some PDA results are summarized.

It is obvious that, especially at off-axis angles where refracted light dominates but also at off-axis angles where reflected light dominates, the use of a semiconductor laser (i.e. the use of a larger wavelength) reduces the influence of the inhomogeneities and therefore allows or improves the application of PDA to optically absorbent, inhomogeneous liquids.

### 5. Conclusions

It has been shown that the use of semiconductor devices can facilitate or improve the application of PDA to optically absorbent homogeneous and inhomogeneous liquids. This offers a wider range of applications in the diagnosis of real process liquids spraying and its in-line process control. If one is forced to detect the scattered light in the forward direction, i.e. the refracted light, the use of a larger wavelength  $\lambda$  can remove the problem of absorption of light in the visible wavelength depending on the liquid's absorption properties and provide a linear  $\Delta\Phi-d$  relationship. In the case of optically absorbent inhomogeneous liquids, the use of a larger wavelength is universally advantageous because it reduces the scattering by these inhomogeneities inside the droplets.

As long as the inhomogeneities remain larger than the light wavelength  $\lambda$  of the semiconductor laser, it is recommended to employ PDA only at off-axis angles  $\varphi$  where the detected light has not passed through the particle but has been reflected.

The Mie theory is a suitable aid to describing the scattering behaviour of optically absorbent homogeneous liquids as well as to describing approximately the scattering behaviour of optically absorbent inhomogeneous liquids and can be used to find the appropriate parameters of the optical set-up in combination with a correct  $\Delta\Phi-d$  relation.

Table 7. Standard deviation  $\sigma$  for characterizing the sizing results by PDA applied to optically absorbing inhomogeneous liquids, 2000 counts.

Liquid	Type of laser	Off-axis angle $\varphi$ [deg]	$d_{10}$ (μm)	Standard deviation $\sigma$ (μm)		
Water	Ar ion	30	74.6	3.1		
		90	104.6	3.3		
	Semiconductor	30	106.7	2.0		
		90	97.9	1.5		
Condensed milk solution (8%)	Ar ion	30	100.4	34.8		
		90	84.4	11.0		
	Semiconductor	30	101.4	11.9		
		90	94.1	7.0		
		Instant coffee solution (8%)	Ar ion	30	99.2	41.0
				90	98.5	7.8
Semiconductor	30		100.3	18.7		
		90	103.1	5.1		

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