Meas. Sci. Technol. **9** (1998) 197–209. Printed in the UK PII: S0957-0233(98)86402-3

Standard PDA for measuring the size of inhomogeneous droplets

M Mitschke†, T Wriedt and K Bauckhage

University of Bremen FB4, Verfahrenstechnik, Badgasteiner Str. 3, D-28359 Bremen, Germany

Received 30 July 1997, in final form 15 October 1997, accepted for publication 22 October 1997

Abstract. A method to obtain the diameter of inhomogeneous droplets by using phase-Doppler anemometry (PDA) is described. Inhomogeneities included in a fluid droplet disturb light scattered by the droplet. Consequently the phase difference measured between two Doppler bursts deviates from the value calculated by geometrical optics theory for a droplet of diameter d. Because evaluation of the measured phase differences is based on a linear relationship between phase difference and particle diameter the accuracy of PDA is deteriorated. The difference between the real and the measured diameters of such droplets can be characterized by convolution with a Gaussian probability function. For monodisperse droplets and for a given type of inhomogeneity the standard deviation of the Gaussian function increases linearly with increasing mass concentration of these inhomogeneity. Applying infrared laser-PDA (IR-PDA) with a wavelength of 1312 nm, the measuring range to investigate droplets of, for instance, water–latex suspensions containing 450 nm polystyrene spheres can be extended to suspensions with six times the measurable concentration standard PDA using a He–Ne laser at $\lambda = 632.8$ nm. The relative improvement obtained by applying IR-PDA (in comparison with a standard PDA system equipped with a 632.8 nm laser) can be estimated by the decrease of the extinction efficiency Q_{ext} of the inclusions with laser wavelength. The linear relation between extinction E and standard deviation σ can be used by a blind deconvolution algorithm. Starting from the measured diameter distribution of the inhomogeneous droplets the real diameter distribution and the standard deviation of each size class of the particles can be calculated by the algorithm. From this standard deviation it is possible to determine the extinction E and the mass concentration of the inhomogeneities.

1. Introduction

With progressive process automization the demand for fast and high-precision measurement and automated control techniques increases rapidly. To avoid or minimize low-grade product charges, in-line measuring techniques which allow *in situ* process control are favoured. In different areas of industrial processes, atomization of liquids in connection with heat and mass transfer has become an important unit operation. For control and optimization of these unit operations, the diameter and the velocity of the atomized droplets has to be measured continuously. Optical measuring techniques are preferred because they work non-intrusively and with a short evaluation time. A well established optical technique is phase-Doppler anemometry (PDA), which allows simultaneous measurements of droplet diameter and velocity with a high temporal and spatial resolution. By applying Mie theory, the calibration curve of a specific PDA set-up can be evaluated. Mie theory is applicable to homogeneous spherical particles of constant refractive index. Therefore, PDA has been successfully applied to sprays of homogeneous media. Difficulties arise when the droplets to be measured contain optical inhomogeneities. The size of these inhomogeneities—in most cases superfine particles—is of the order of or larger than the wavelength of the laser light. Their refractive index is different from that of the liquid of the droplet. Examples of such processes are sprays of paints and process fluids which are spray dried in order to produce instants.

Mie theory and other light scattering theories have been expanded to simulate the scattering behaviour of non-homogeneous particles but their application range is limited either to a few larger particles in a droplet or to droplets with significantly smaller inclusions [1, 2]. Contrary to this theoretical progress, in most technical applications inhomogeneous droplets contain a large number of fine particles of size of the order of the laser wavelength. In the case of PDA the simple phase difference–diameter relation used to determine the particle diameter is no longer valid. The accuracy in determining particle velocity is also diminished, though the influence is not as significant as for the diameter determination.

† E-mail address: vt@iwt.uni-bremen.de

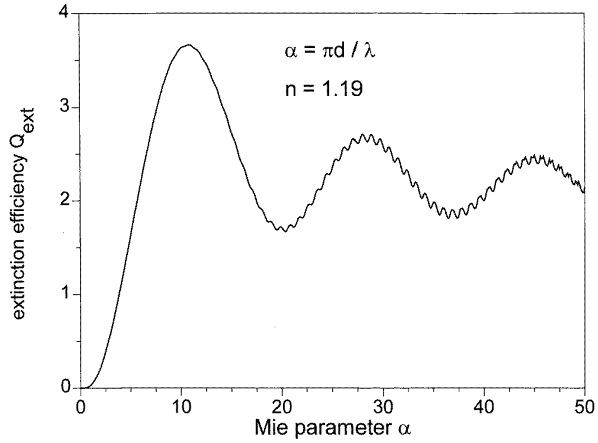

Figure 1. Extinction efficiency Q_{ext} for a relative refractive index n = 1.19 which is equivalent to polystyrene in water at 832 nm.

It has already been shown by Manasse *et al* [3] that the influence of the fine particles (inhomogeneities) can be interpreted as a Gaussian blur function. This function depends on the diameter of the inhomogeneities, their number concentration and the wavelength of the laser beam. For particles with an extinction efficiency proportional to the Mie parameter $\alpha = \pi d/\lambda$, the disturbing effect can be reduced by applying lasers with longer wavelengths [4]. This adjusts the measurement results closer to reality and improves the corrected results of the following inversion algorithm. This relationship is normally valid for Mie parameters of fine particles of up to $\alpha = 10$.

2. Background

The most important parameters influencing multiple scattering of inhomogeneous droplets are given by the equation for the extinction E within a droplet [5]:

$$E = N_V Q_{ext} A_P = \frac{c_V}{\rho V_P} Q_{ext} A_P. \qquad (1)$$

The number concentration N_V (or mass concentration c_V) as well as the particle projection area A_P and the volume V_P of an inhomogeneity are assumed here to be fixed values subject to the technical process considered. The only value that can be influenced is the extinction efficiency Q_{ext} [6] because it depends on the light wavelength (if one can vary the wavelength of the applied laser).

To demonstrate this effect, the extinction efficiency for polystyrene particles in water ($n = 1.19 + 0.0\text{i}$) was calculated and is plotted in figure 1. The extinction efficiency can be minimized if the Mie parameter can be reduced to less than ten by increasing the laser wavelength. By increasing the laser wavelength, the total scattering power of the carrier droplet, which is normally of the order of several 10 μm or 100 μm, decreases proportionally by the second power of the particle diameter. That means that if the noise level remains constant, the signal to noise ratio (SNR) of the measured Doppler burst decreases and signal evaluation becomes more difficult [7].

3. Experimental details

To verify the effect of the laser wavelength described above, three standard PDA set-ups were realized in order to measure the diameter and the velocity of passing particles simultaneously. In referring to standard PDA we mean the original PDA set-up proposed by Bauckhage in 1988 [8]. The complete experimental set-up is shown in figure 2.

The first PDA transmitting system consists of either a He–Ne laser (632.8 nm) or a diode laser (832 nm). Two avalanche photodiodes are integrated in the corresponding receiving optics. The second transmitter operates with a 1312 nm Nd-YAG laser and is shifted in the x–y plane by 35° against the first system. The signal is detected by two pin diodes. The set-up parameters are listed in table 1.

With these three PDA systems monodisperse droplets of 21 different latex–water suspensions at two off-axis angles ($\varphi = 30°$, $60°$) were examined. The diameters of the polystyrene particles (inhomogeneities) were determined by scanning electron microscopy (SEM) to be 0.45 μm, 0.72 μm and 3.5 μm. With these polystyrene particles monodispersed latex suspensions from 0.25% up to 3.0% mass concentration (0.25, 0.5, 1.0, 1.5, 2.0, 2.5, 3.0%) were produced. These suspensions were separated into (chains of) monodispersed droplets by a droplet generator having an orifice of 100 μm or 150 μm diameter. The diameter and the velocity of these droplets were measured by the three PDA systems. The diameter of the droplets was also observed by an Olympus microscope coupled to a SONY CCD camera. Photographs were analysed with regard to the mean droplet projection area by Global Lab image evaluation software. The accuracy of the image evaluation in diameter determination was expected to be ±5%. To avoid measuring inaccuracies due to variations in the latex concentration, the mass concentration of the suspensions was continuously monitored by spectroscopy. Variations in mass concentration were observed, caused by segregation in the feed tank of the suspension due to sedimentation or due to evaporation of water. A calibration curve was therefore determined by measuring the extinction of the suspension correlated to the mass concentrations at definite values. Samples of the examined suspensions were taken before and after each experiment. These samples were diluted (ratio 1:1000) with bi-distilled water. The extinction at 497 nm was measured and compared with the calibration curve. The results show that the change in mass concentration during one experiment was less than 0.1% in mass concentration for all suspensions.

The adjustment of the 1312 nm PDA transmitter was observed by an IR laser beam analyser, an IR power meter and an IR camera. Due to dispersion the real focus of the lenses in the receiving system was unknown. To verify the distance between the receiving system and the measuring volume, the PDA results were compared to results from image evaluation for water drops of two different diameters (70 μm and 200 μm).

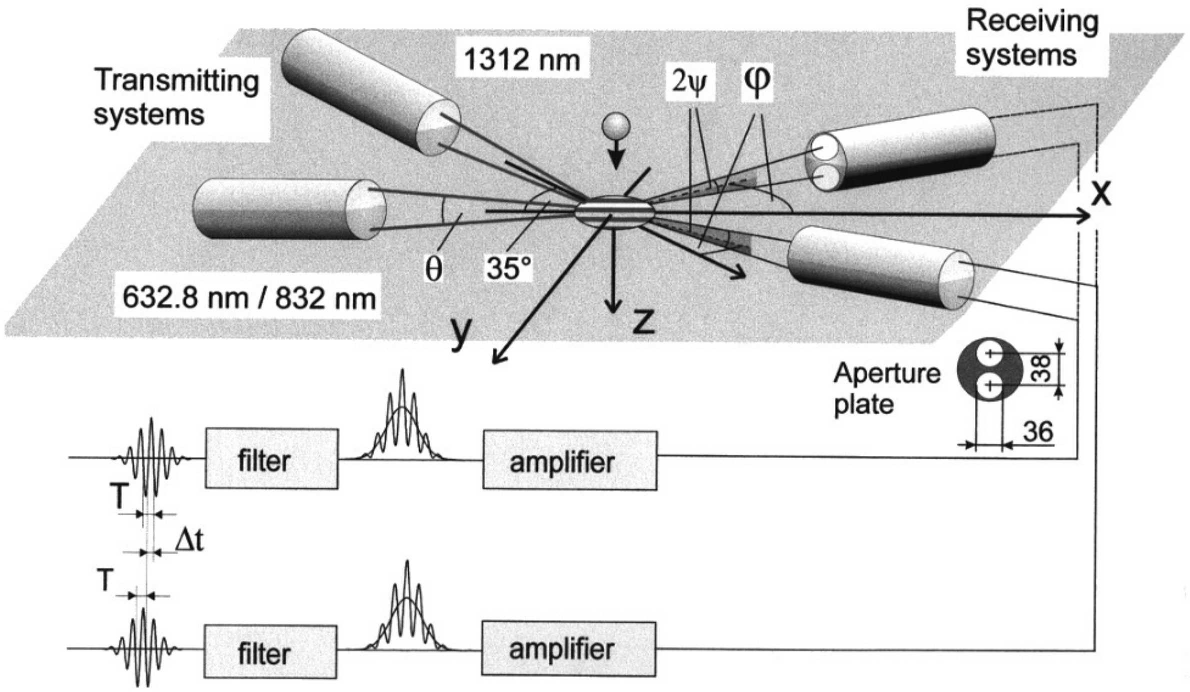

Figure 2. Standard PDA set-up. Two laser-beams intersect in the x–z plane under the crossing angle θ; the receiving systems are positioned in the x–y plane at the off-axis angle φ; the detectors are tilted against the x–y plane under the elevation angle ψ. The particle trajectory is parallel to the z direction. The time difference Δt between the measured Doppler bursts is proportional to the diameter and the frequency T is proportional to the velocity of the particle.

Table 1. Parameters of the three standard PDA set-ups.

Wavelength (nm)	Beam crossing angle (deg)	Power of laser beams (mW)	Diameter of measuring volume (y, z) (μm)	Elevation angle (deg)	Off-axis angle (deg)
632.8	2.37	30	650/570	±1.45	30/60
832.0	2.37	5	330/280	±1.45	30/60
1312.0	4.18	34	350/330	±1.45	30/60

4. Experimental results

All measured diameter density distributions were fitted by a Gaussian function characterized by the mean diameter d_m and standard deviation σ. The standard deviation is a direct quantity of the measuring accuracy of the PDA system. Figure 3 demonstrates the accuracy of the droplet generator as well as that of the PDA systems in determining the diameter of 5000 homogeneous water droplets. The standard deviation of the fitted Gaussian function is always better than 2.0 μm for a mean diameter of 182 μm with either the 632.8 nm or the 832 nm laser and for 181 μm with the 1312 nm laser. The difference between the mean diameter measured by PDA and that determined by image evaluation is of the order of 1 μm (equivalent to 0.6%).

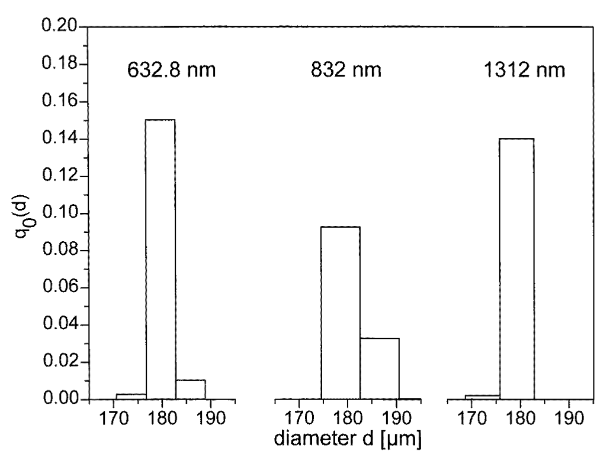

Figure 3. PDA measured number density distributions of water droplets, shifted for better differentiation. Each distribution is equivalent to 10 000 particles. Identical mean values were detected for each distribution.

Typical PDA results obtained for the same collective of monosized droplets of a latex–water suspension at three different wavelengths are plotted in figure 4. Here the measured diameter distributions have been shifted in order to distinguish between the different results in a more significant manner. The standard deviation is of the order of 42 μm (632.8 nm), 40 μm (832 nm) and 18 μm (1312 nm) for a mean diameter of about 170 μm (171 μm, 167 μm, 166 μm).

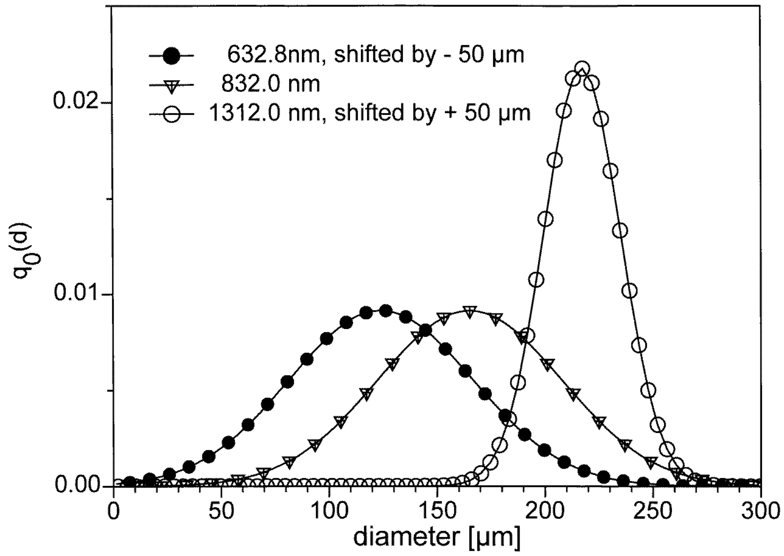

Figure 4. PDA measured number density distributions of droplets of latex–water suspensions (diameter of inhomogeneities = 720 nm), shifted for better differentiation. Each distribution is equivalent to 10 000 droplets with 0.5% mass concentration. The diameter, being equivalent to the mean particle projection area, was determined to 177 μm.

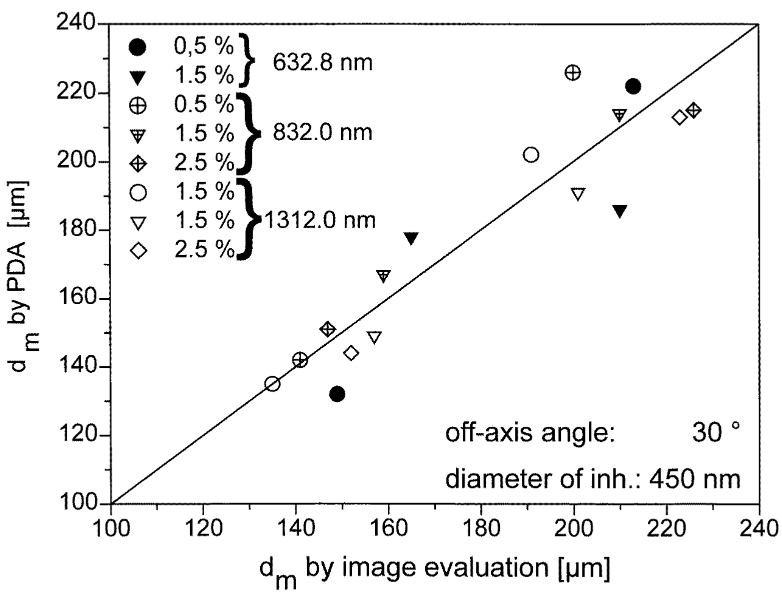

Figure 5. Mean diameter of the droplet collective measured by PDA and by image evaluation (Global lab).

The mean diameter of each droplet collective is given in figures 5, 6 and 7 for different values of diameter of inhomogeneity. For clarity figures 5, 6 and 7 show results only for suspensions of 0.5%, 1.5% and 2.5% mass concentration of inhomogeneities of 450 nm and 720 nm diameter. Results are shown for occasions when nearly identical droplet diameters were detected by both PDA and image evaluation.

Because no dependence of the standard deviation on the diameter of the droplets could be stated for the examined diameters between 120 μm and 240 μm, a mean standard deviation was calculated from measurements under identical conditions. Figures 8, 9 and 10 show the standard deviation as a function of the mass concentration and the laser wavelength. The series of measurements suggests a linear dependence of the standard deviation on the concentration of suspensions for constant wavelength.

In case of the 450 nm and the 720 nm latex–water suspensions (at 30° off-axis angle), it is obvious that the mean diameter of all suspensions can be detected correctly only with the 1312 nm system, (see figures 5, 6, 8 and 9). The standard deviation (at 1312 nm) of each distribution is less than 50 μm in the examined diameter range. The measuring accuracy of the PDA systems decreases with increasing mass concentration. This reduction is in good agreement with equation (1), where

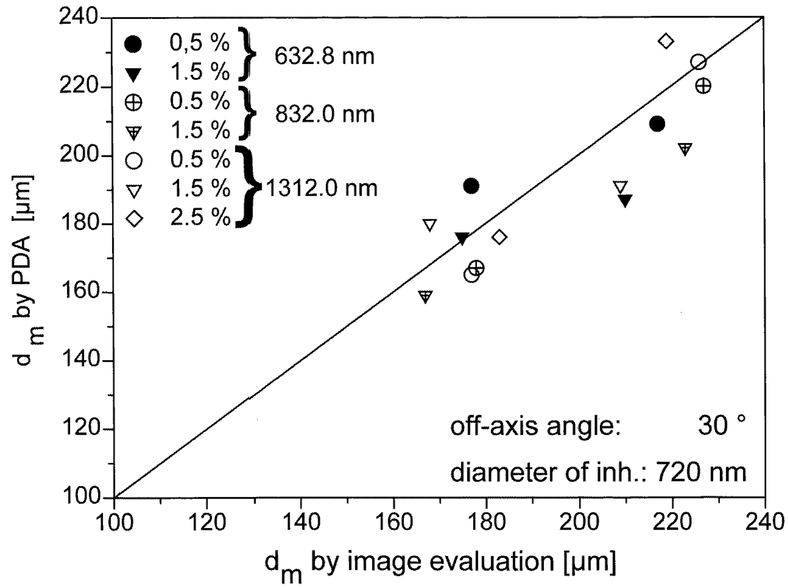

Figure 6. Mean diameter of the droplet collective measured by PDA and by image evaluation (Global lab).

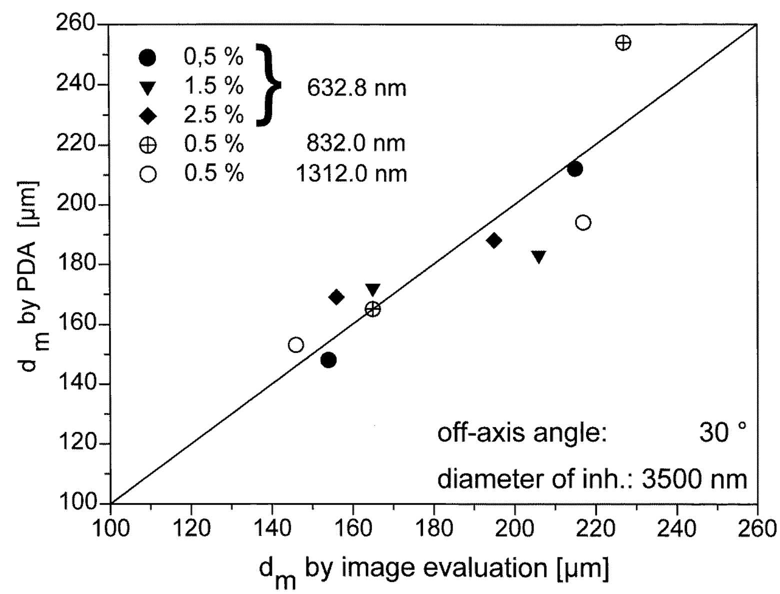

Figure 7. Mean diameter of the droplet collective measured by PDA and by image evaluation (Global lab).

the extinction E increases with increasing concentration. Suspensions with identical mass concentrations show an increase in standard deviation with decreasing wavelength. This effect results from a higher extinction efficiency Q_{ext} which is reciprocally proportional to the wavelength (for inhomogeneities with diameters lower than 2 μm, see figure 11). The gradient of the fitted straight line increases with decreasing wavelength as well. This cannot be explained by equation (1) and has to be attributed to another influencing parameter which is still under investigation.

A direct quantity for the quality of the signal is the signal to noise ratio (SNR) of the detected Doppler bursts [7]. It is calculated from the power spectrum of the Doppler bursts:

$$\begin{aligned} \mathrm{SNR} &= 10\log_{10}\frac{P_{signal}}{P_{noise}} \\ &= 10\log_{10}\frac{f_{i-1}+f_i+f_{i+1}}{\sum_{a=i-2}^{a=1} f_a + \sum_{a=i+2}^{a=n} f_a} \\ & \quad i = \text{bin of maximum power}. \end{aligned} \tag{2}$$

The SNR of the Doppler bursts decreases with increasing latex concentration and decreasing laser wavelength. Multiple scattering between the inhomogeneities within the carrier droplet leads to an isotropic distribution of the scattered light outside the droplet [9]. This effect blurs the Doppler bursts and reduces the quality of the

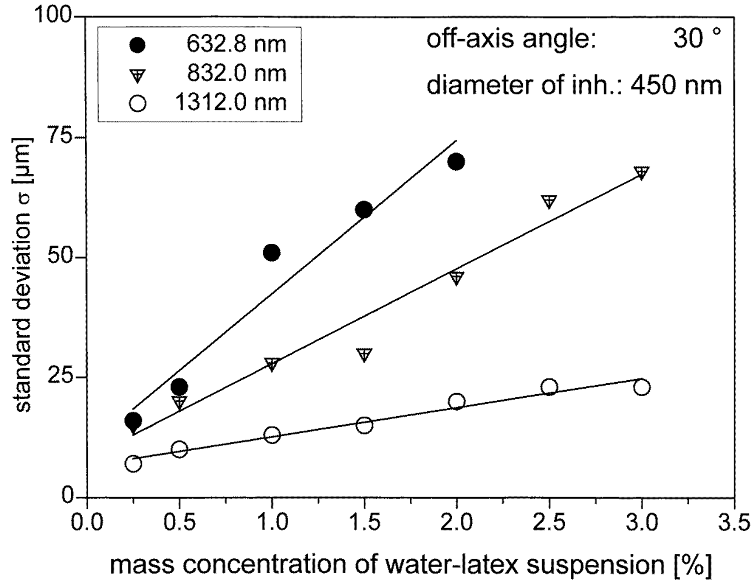

Figure 8. Standard deviation of the fitted Gaussian distributions.

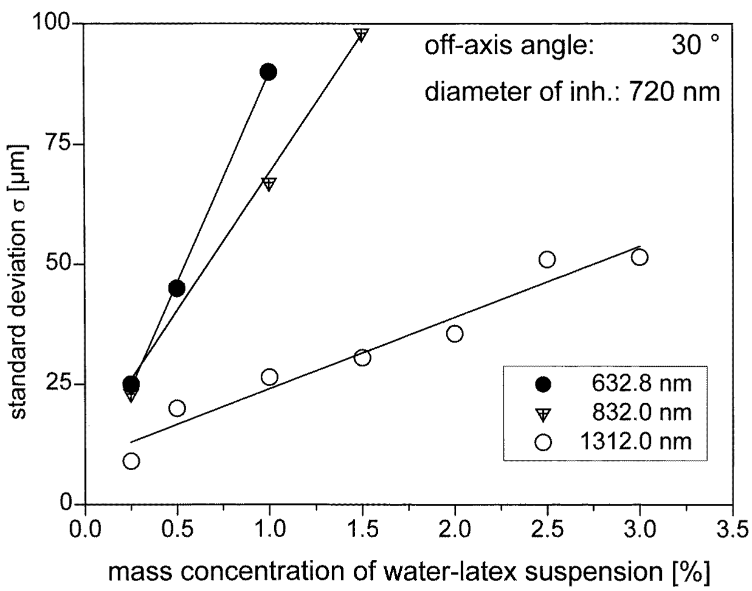

Figure 9. Standard deviation of the fitted Gaussian distributions.

signal. Because the disturbing effect of the inhomogeneities on the scattering process increases with decreasing laser wavelength ($Q_{ext} \sim 1/\alpha$) the quality of the bursts is improved by the longer wavelength (here 1312 nm). This means that the IR-PDA system is able to detect droplets of higher concentrated suspensions more effectively than the other two systems. Due to the reduced quality of the Doppler bursts, it was impossible to measure droplets of a 720 nm latex–water suspension with mass concentrations higher than 1.0% by the 832 nm and 0.5% by the 632.8 nm PDA system. The reduced accuracy of the PDA in measuring droplets of an inhomogeneous liquid is additionally deteriorated by a lower SNR, corresponding to the Cramer–Rao lower bound [7]. The reason for this is that with lower SNR values the calculation of the mean frequency of the Doppler bursts is less exact. Thus, determination of the velocity of the particles becomes more difficult with increasing latex concentration. Nevertheless, the velocity of the particles could be detected exactly in all cases where the mean diameter of the particle was determined correctly.

By changing the off-axis angle from 30° to 60°, the error in measuring the mean diameter of the suspension droplets increases for all wavelengths (compare figures 9 and 12). In the case of 632.8 nm (832 nm/1312 nm) the correct mean diameter can only be detected for mass

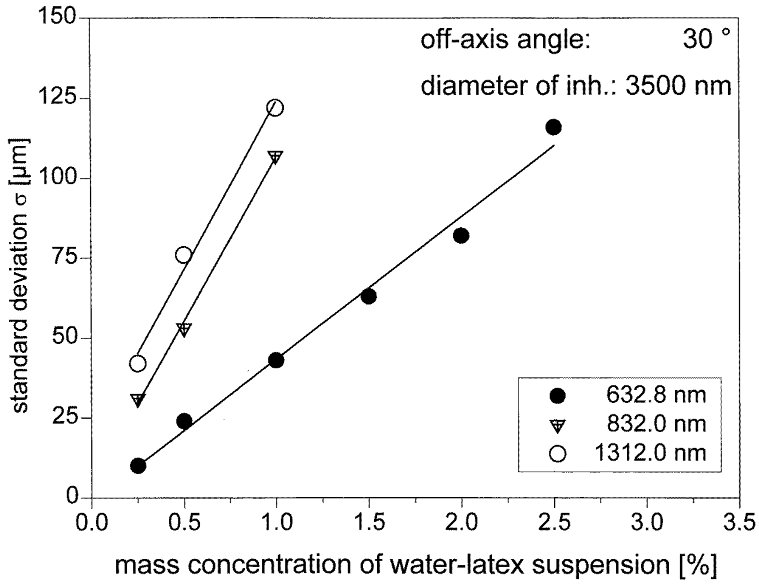

Figure 10. Standard deviation of the fitted Gaussian distributions.

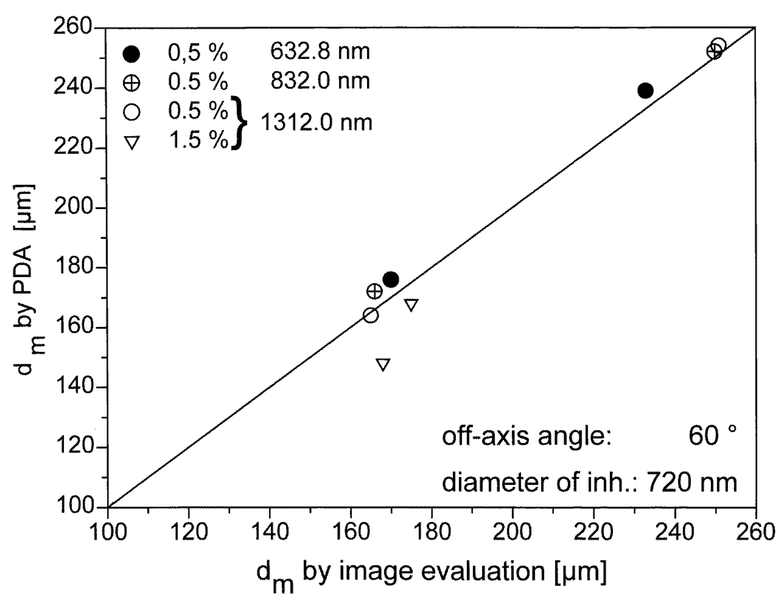

Figure 11. Mean diameter of the droplet collective measured by PDA and by image evaluation (Global lab).

concentrations of up to 0.5% (1.0%/2.0%). The higher the laser wavelength, the more concentrated the suspensions may be for accurate PDA measurements. The advantage of the higher ratio of refracted light to reflected light at $\varphi = 60^\circ$ (in the case of an optically homogeneous sphere) is more than compensated by multiple scattering within the carrier droplets (in the case of an optically inhomogeneous droplet). Monte Carlo simulations of the far field scattering of an inhomogeneous droplet show that in the case of a water droplet with inclusions of $n = 1.46$ and $d = 500$ nm the influence on the scattering field increases with increasing off-axis angle [9]. Therefore the measuring accuracy of PDA is worsened by changing the position of the detectors from 30° to 60° off-axis. This effect results in higher standard deviations of diameter determinations of the investigated suspensions.

Due to the reduced scattering intensity of the carrier droplets at 60°, the SNR of the Doppler bursts is worse than that at 30°. The SNR for the measurement of the 1.0% mass concentration suspension at 1312 nm decreases by 20% from about 13 dB (30° off-axis angle) to 9 dB (60°). This fact reduces the measuring range of the PDA system to lower concentrated suspensions. The maximum permitted mass concentrations are 0.5%, 0.5% and 1.5% for $\lambda = 632.8$ nm, 832 nm and 1312 nm respectively.

A totally different result was obtained for the

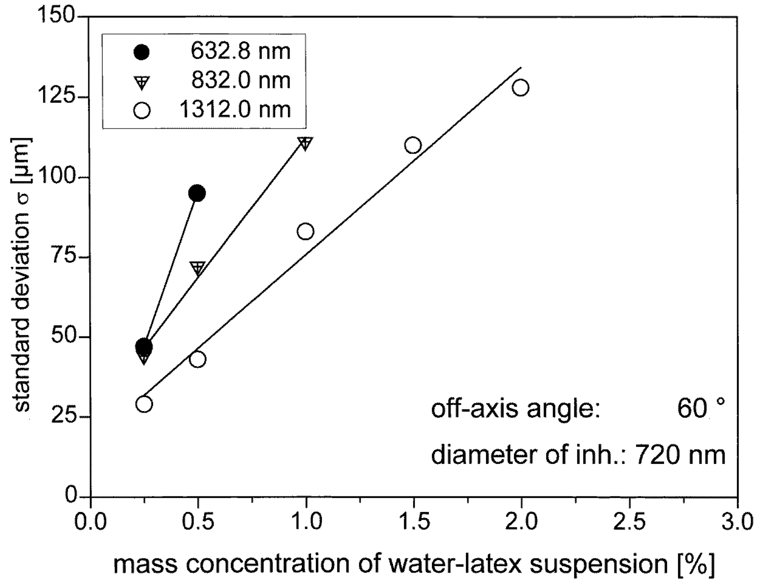

Figure 12. Standard deviation of the fitted Gaussian distributions.

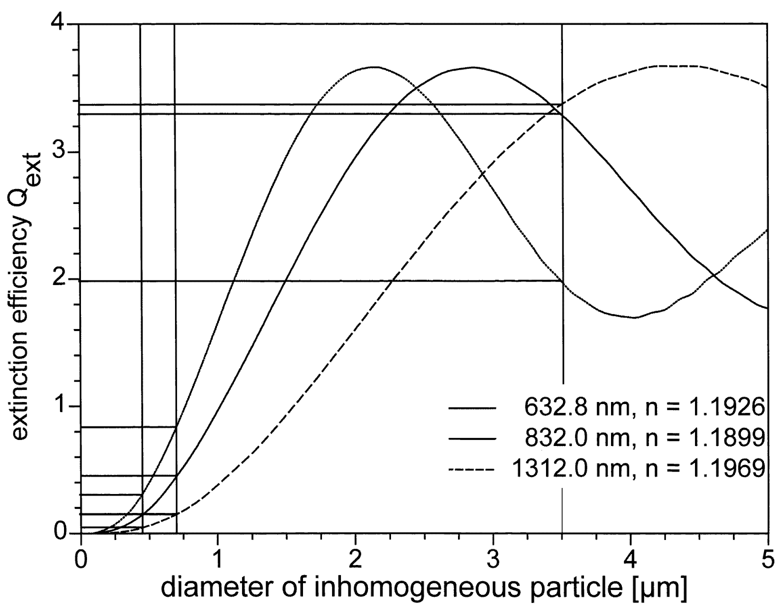

Figure 13. Calculated extinction efficiency Q_{ext} as a function of wavelength for polystyrene particles in water.

3.5 μm latex–water suspension (see figures 12 and 13). The standard deviation increases with increasing laser wavelength; the gradient of the fitted line is greater for higher wavelengths; and droplets of higher concentrated suspensions can be measured more accurately with lasers of lower wavelength (632.8 nm/2.5%; 832 nm/1.0%; 1312 nm/1.0%). At first glance, this result seems to be inconsistent with the previous explanations. However, it verifies the statement that the extinction efficiency Q_{ext} is the most important parameter in measuring characteristics of inhomogeneous particles. Figure 13 demonstrates that the change in correlation between laser wavelength and standard deviation is based on the oscillating structure of the extinction efficiency–diameter (inhomogeneities) relation. The extinction efficiency for a particle of 3.5 μm diameter is about two for 632.8 nm wavelength light instead of 3.4 for 1312 nm. Due to (almost) identical extinction efficiencies for wavelengths of 832 nm and 1312 nm, equal measuring accuracies for both PDA systems can be stated.

5. Theory and measuring results

Table 2 lists the extinction values ($E = N_V Q_{ext} A_P$) where N_V is the number of inhomogeneous particles in a droplet of a diameter of 100 μm. N_V is calculated from the mass concentration of the suspensions. A_P is given in

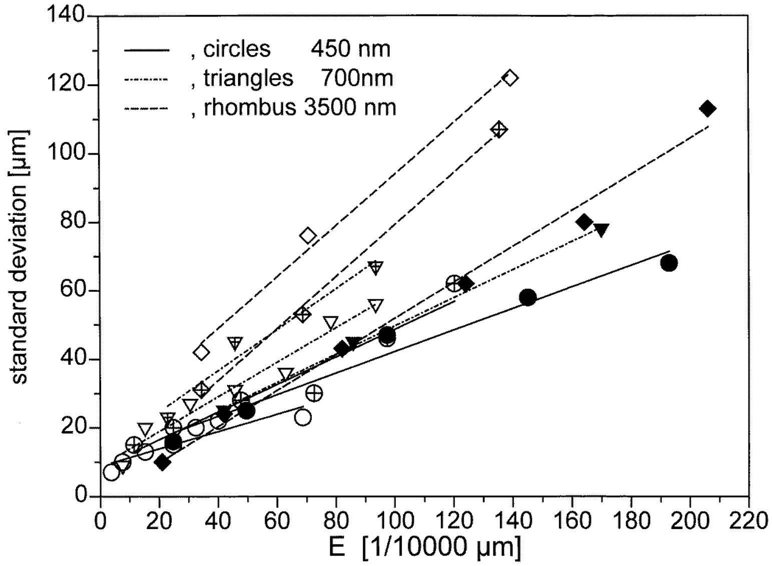

Figure 14. Standard deviation in micrometres of the fitted Gaussian distributions as a function of the extinction $E = N_V Q_{ext} A_P$ for the examined suspensions. (Closed symbols, 632.8 nm; crossed symbols, 832 nm; open symbols, 1312 nm).

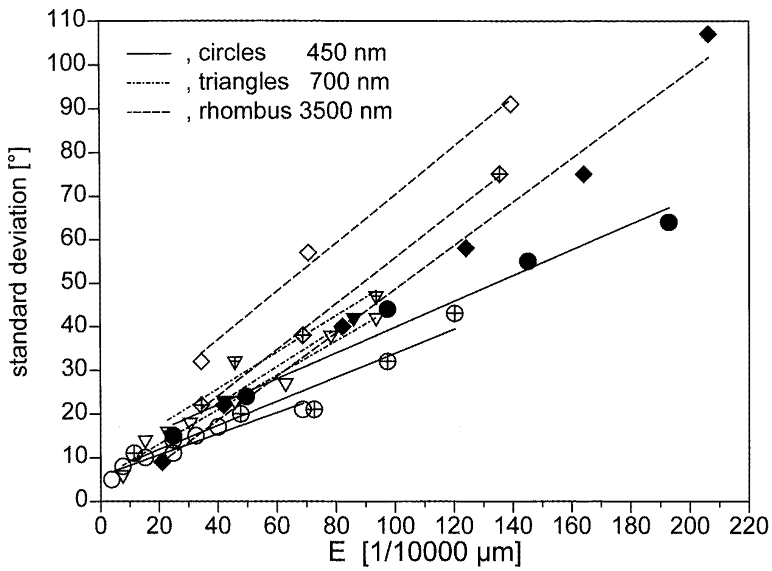

Figure 15. Standard deviation in degrees phase difference of the fitted Gaussian distributions as a function of the extinction $E = N_V Q_{ext} A_P$ for the examined suspensions. (Closed symbols, 632.8 nm; crossed symbols, 832 nm; open symbols, 1312 nm).

μm^2. The values of table 2 are plotted against the fitted standard deviations of the measured diameter distributions in figure 14. Figure 15 represents identical values and the corresponding standard deviations of the measured phase difference distribution, which is the original measured quantity. These standard deviations can be calculated by multiplying the standard deviations of the diameter distributions by the ratio of the 360° phase difference to the largest measurable diameter.

Figures 14 and 15 show a linear relation between the extinction E and the standard deviation. Furthermore, figure 15 illustrates that for suspensions with inhomogeneities of identical diameter the gradient of the fitted lines is nearly identical for all wavelengths. By changing the diameter of the inhomogeneities, the gradient of the curve varies: for suspensions of 450 nm latex spheres the gradient is about 0.3°/10 000 μm; for 720 nm latex spheres, about 0.4°/10 000 μm; and for 3500 nm latex spheres, about 0.5°/10 000 μm. It is also obvious (most significantly in case of the 3.5 μm suspension) that a systematic deviation exists between the curves for identical suspensions at different wavelengths. The graph shows that the mean diameter of droplets of suspensions with an extinction E higher than 0.02 could not be measured correctly. Thus, figure 15 can be used to estimate the accuracy of PDA measuring results beforehand by monitoring inhomogeneous droplets.

Table 2. Extinction $E = N_V Q_{ext} A_P$ for the examined suspensions.

Mass concentration (%)	0.45 μm polystyrene particles			0.7 μm polystyrene particles			3.5 μm polystyrene particles		
	632.8 nm	832 nm	1312 nm	632.8 nm	832 nm	1312 nm	632.8 nm	832 nm	1312 nm
0.25	25	11	4	42	23	8	21	34	34
0.5	50	25	8	86	46	15	42	69	71
1.0	97	48	15	168	94	31	82	136	139
1.5	145	73	25	252	138	46	124	208	207
2.0	193	97	32	336	183	63	164	277	277
2.5	241	120	40	420	229	78	206	346	346
3.0	412	196	69	505	275	94	254	415	415

6. Mathematical inversion algorithm

As discussed in section 5, the mean diameter of a monosize droplet collective can be determined correctly for an extinction E lower than 0.02 (under the presumption that the diameter of the inhomogeneities is less than 1 μm). In this case the standard deviation of the measured distribution is about 70 μm. In a real spraying process, where droplets of many different diameters exist, the profile of a measured diameter distribution is totally lost. In most technical processes (where the diameters of the droplets are of the order of several 10 μm), an accuracy of $\pm$10 μm seems to be sufficient so that suspensions up to an extinction E of 0.002 could be measured without problem. In contrast to standard (632.8 nm) PDA, suspensions of higher concentration could be investigated with an IR-PDA system due to the lower values of the extinction efficiency Q_{ext}.

The implementation of mathematical inversion algorithms is another possibility to evaluate the diameter distributions of higher concentration suspensions. Such algorithms can be applied if the diameter distribution results from at least 5000 measured droplets. The wider the distribution, the larger the minimum number of counted particles should be. The disadvantage of this method is that the size information of one single particle in conjunction with its velocity is lost. Only the common result during a certain time period and for a sufficient number of droplets can be interpreted.

To apply the inversion algorithm, the PDA measurement result has to be described mathematically. The relation between the measured distribution $w(d)$ and the real diameter distribution $q(x)$ [$q(d)$] is given by a Fredholm integral equation

$$w(d) = \int_{d_{min}}^{d_{max}} u(d, x) q(x)\, \mathrm{d}x \leftrightarrow \bar{\boldsymbol{w}} = \mathbf{U} \cdot \bar{\boldsymbol{q}}. \qquad (3)$$

The transfer matrix $\mathbf{U}$ describes the probability with which PDA interprets a droplet of real diameter x as a droplet of diameter d. Each column of this matrix corresponds to broadening of the according diameter class with mean diameter x. Fredholm integral equations of the first kind are generally ill posed due to the high sensitivity to measurement errors [10]. Thus, in the presence of noise, a simple inversion leads to highly oscillating curves. Therefore, special algorithms have to be applied to compute the unknown diameter distribution $q(d)$. The transfer function has to be known; it can be found by measuring a larger number of monodispersed droplets [11].

A more sophisticated way is the application of a blind deconvolution method. This method operates without detailed information on the standard deviation of the transfer function. Only two conditions have to be fulfilled: the shape of the transfer function has to follow a known parametric function and this function has to be independent of the diameter of the droplets. These two assumptions are fulfilled for latex–water suspensions or milk–water emulsions.

The structure of the algorithm is as follows.

First, the measured distribution is deconvoluted by an iterative inversion algorithm, where the number n of iteration steps is (here) fixed to 50, see equation (4). The number of iteration steps influences the quality of the restoration result. In general, this number has to be adapted to the unknown distribution $q(d)$: the smaller the distribution the higher the number of iteration steps required.

$$q^{(n+1)} = \mathbf{U}^T w + (1 - \tau \mathbf{U}^T \mathbf{U}) q^n \qquad n = 50. \qquad (4)$$

To calculate $q^{(n+1)}$ according to equation (4), the parameter of the transfer function u has to be chosen. The transfer function can be any cubic spline function but, with regard to the problem of optical inhomogeneous droplets, a Gaussian function is favoured. Thus, with the found ‘real distribution’, simulations were performed to optimize the number of iteration steps. Details about this optimization process can be found in [11]. For our application this optimization was further improved by distributing the random numbers according to the supposed diameter distribution. Thus with the computed amount of random numbers, the transfer matrix $\mathbf{U}$ was calculated. After optimization the actual blind deconvolution algorithm (BDA) starts.

In the first step of the BDA the unknown real diameter distribution is calculated by applying equation (4) with the optimized number of iteration steps n and a Gaussian transfer function with a standard deviation estimated by the user. Due to this presumed standard deviation, the determined distribution $q(d)$ represents only a first approximation to the real diameter distribution. Therefore, a further step is included to improve the first choice of standard deviation. This step profits from the assumption that the transfer function is independent of the total particle

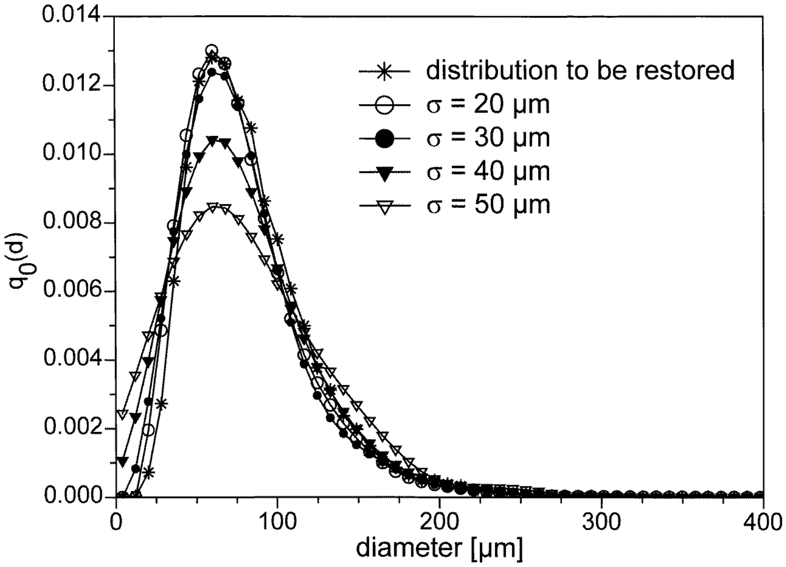

Figure 16. Real diameter distribution (basic log–normal distribution for the simulation) and corresponding diameter distributions computed by BDA.

diameter. In this case, the only difference between the columns of the transfer matrix **U** is that they are shifted by one row from one column to the next. On condition that the transfer matrix can be expressed by the values of the 'real distribution $q(d)$' (which was calculated before) and the transfer function u can be interpreted as a vector $\boldsymbol{u}$ equation (3) is transformed into

$$\boldsymbol{w} = \mathbf{Q} \cdot \boldsymbol{u}. \quad (5)$$

Equation 6 is again an ill-posed problem. This time it is solved by a least-squares method originally proposed by Tikhonov and Arsenin [12]

$$(\mathbf{Q}^T \cdot \mathbf{Q} + Y \cdot \mathbf{I}) \cdot \Delta \boldsymbol{u} = \mathbf{Q}^T \cdot (\boldsymbol{w} - \mathbf{Q} \cdot u_{alt}). \quad (6)$$

A minimum of the oscillation of the calculated distribution is reached by use of a smoothing operator Y. The value of the operator has to be chosen between 0 and 1 according to the distribution of interest $u_{new}(d) = u_{old}(d) + u(d)$. The χ^2 comparison of the presumed and the calculated standard deviation describes the quality of the presumed transfer function. If χ^2 is larger than a definite value, the process starts once again with the new calculated standard deviation and so on. At the end of the blind deconvolution both unknown functions—the real diameter distribution $q(d)$ and the transfer function $u(d)$—are determined. The information about the transfer function can be used to find the mean diameter of the inhomogeneities or their mass concentration, if the other parameter is known. Nevertheless the accuracy of the restoration result of the deconvolution procedure increases with decreasing width of the transfer function.

6.1. Simulations of restoration results

Four different diameter distributions were simulated to test the BDA. These distributions were calculated by convoluting a log–normal distribution with Gaussian functions of different width ($\sigma = 20$ μm, 30 μm, 40 μm, 50 μm). Each simulated distribution should be equivalent to a PDA measured diameter distribution of 10 000 droplets of an inhomogeneous medium.

First, the probability density function of the log–normal distribution, representing the PDA measuring result of a homogeneous medium, was calculated. Each distribution was simulated by 10 000 random numbers as discussed in [13].

These distributions were convoluted with the Gaussian functions mentioned above. Therefore, Gaussian functions of identical width were calculated by a random process [13] for each diameter class. These functions differed in the amount of random numbers by which they were built. The amount was determined with regard to the 10 000 random numbers and the shape of the log–normal distribution. Thus, computed distributions were inverted by applying the BDA. The result is plotted in figure 16 and the standard deviation is given in table 3.

Figure 16 and table 3 show that with the aid of the BDA the original distribution could be restored. Furthermore the standard deviation of the Gaussian function could be computed in most cases. The algorithm fails only for Gaussian functions with standard deviations of more than 50 μm. In our case, those standard deviations correspond to latex–water suspensions of 3% mass concentration (1312 nm, latex spheres of diameter 720 nm) or to suspensions of 0.5% mass concentration in case of the 632.8 nm PDA system (same latex spheres).

These computations make clear that, with the application of IR lasers in standard PDA set-ups and simultaneous use of the BDA, droplet collectives of inhomogeneous media can be measured and evaluated.

On the condition that the inhomogeneity diameter is known and that the standard deviation is independent of the diameter, the standard deviation of the Gaussian function

Table 3. Real standard deviation of the convolution procedure and standard deviation computed by the BDA.

Standard deviation of the given Gaussian function	20 μm	30 μm	40 μm	50 μm
Standard deviation of the BDA computed Gaussian function	23 μm	32 μm	39 μm	40 μm

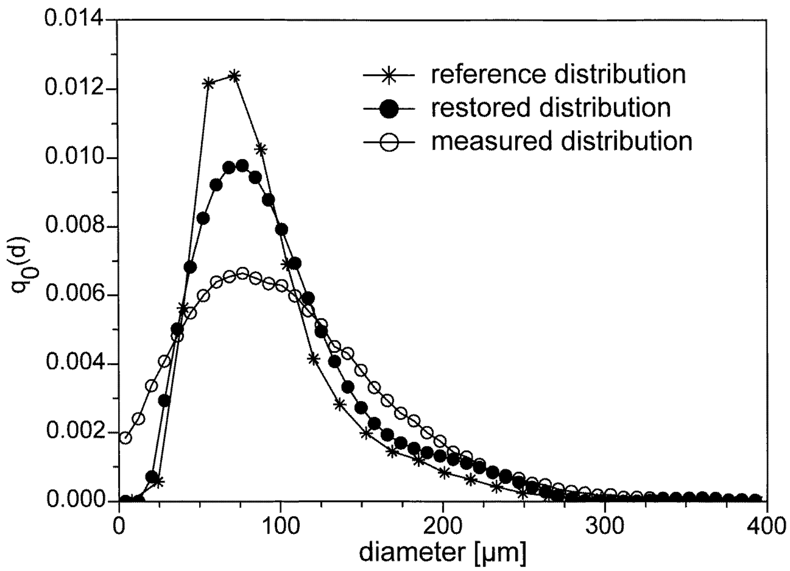

Figure 17. PDA measured diameter distribution of a real emulsion (16% condensed milk diluted with water) and corresponding diameter distribution restored by BDA.

can be well computed by the BDA. This information can be used to determine the mass concentration of the suspension. The accuracy in determining the mass concentration will be discussed elsewhere.

The diameter distribution of an emulsion of 16% condensed milk and water was measured during a spraying process. This original distribution was restored by the BDA algorithm. The measured, the restored and a reference diameter distribution are shown in figure 17.

It is obvious that the restored distribution is identical to the reference distribution. The standard deviation calculated by the BDA is about 42 μm which is of the order of that determined by a fitting algorithm from the measured diameter distributions of collectives of monosize droplets (40 μm). Deviation of the computed standard deviation from the fitted standard deviation can be attributed to different values of the *SNR*. In the case of a collective of monosize droplets, all parameters of the PDA set-up (trigger level, amplifier settings) are adapted to the defined diameter of the droplets. In a real spraying process these parameters have to be adapted to produce identical measuring conditions for a wide range of diameters (in this special case for droplets with diameters between 30 μm and 300 μm).

7. Conclusions

With the extinction E, the PDA user has a parameter to estimate beforehand the possibility of measuring inhomogeneous droplets of suspensions containing fine monosized spherical particles. If the extinction E is below 0.02 PDA detects the correct mean diameter of a monosized droplet collective. The standard deviation of the measured values from the mean diameter is of the order of 100 μm (100° phase difference) for droplets of 120 μm. Measuring droplets consisting of inhomogeneous media, standard PDA operating at 630 nm fails even at low concentrations. In the range where the extinction efficiency Q_{ext} is proportional to the Mie parameter PDA can be extended to suspensions with six times higher concentrations. The experimental results also illustrate that changing to higher wavelengths does not always lead to more realistic results. On the contrary, the proper selection of the laser wavelength has to be based on Mie calculations of the extinction efficiency. Higher wavelengths are the better choice only for a Mie parameter of less than five (dependent on the refractive index). For larger values of the Mie parameter the relation between the extinction efficiency and wavelength is ambiguous and because of this statements relating to improvements obtained by changing the laser wavelength cannot be made without Mie calculations.

With the assumption that the transfer function—which is equal to the PDA measurement result of a monosized droplet collective—is independent of the total droplet diameter a blind deconvolution algorithm (BDA) can be used to correct the influence of the included inhomogeneities. Applying this algorithm, the original real diameter distribution can be computed up to Gaussian transfer functions with standard deviations of the order of 50 μm. The BDA also determines this standard deviation which is coupled to the extinction E. If the diameter of the inhomogeneities is known, this information can be used to determine the mass concentration of the inhomogeneities within the droplet. Nevertheless, the user has to realize that with the application of BDA information about the single particle and the diameter–velocity relation is lost.

Acknowledgments

We acknowledge the support of research project BA 542/25-2 by the Deutsche Forschungsgemeinschaft (DFG).

References

[1] Fuller K A 1995 Scattering and absorption cross sections of compounded spheres. III. Spheres containing arbitrarily located spherical inhomogeneities *J. Opt. Soc. Am.* A **12** (5) 893–904

[2] Fikioris J G and Uzunoglu N K 1979 Scattering from an eccentrically stratified dielectric sphere *J. Opt. Soc. Am.* **69** (10) 1359–66

[3] Manasse U, Wriedt T and Bauckhage K 1994 Reconstruction of real size distributions hidden in phase-Doppler anemometry results obtained from droplets of inhomogenoues liquids *Part. Part. Syst. Charact.* **11** 84–90

[4] Manasse U, Wriedt T and Bauckhage K 1992 Phase-Doppler sizing of optically absorbing liquid droplets: comparison between Mie theory and experiment *Part. Part. Syst. Charact.* **9** 176–85

[5] Van de Hulst H C 1981 *Light Scattering by Small Particles* (New York: Dover)

[6] Bohren C F and Huffman D R 1983 *Absorption and Scattering of Light by Small Particles* (New York: Wiley)

[7] Wriedt T, Bauckhage K and Schöne A 1989 Application of Fourier analysis to phase-Doppler-signals generated by rough metal particles *IEEE Trans. Instrum. Meas.* **38** (5) 984–90

[8] Bauckhage K 1988 The phase-Doppler-difference-method, a new laser-Doppler technique for simultaneous size and velocity measurements *Part. Part. Syst. Charact.* **5** 16–22

[9] Göbel G, Lippek A, Wriedt T and Bauckhage K 1998 Monte Carlo simulation of light scattering by inhomogeneous spheres *Proc. Radiance Transfer Symposium (Kusasdasi, Turkey) 1997*

[10] Twomey S 1977 *Introduction to the Mathematics of Inversion in Remote Sensing and Indirect Measurements* (New York: Elsevier)

[11] Köser O and Wriedt T 1996 Iterative inversion of phase-Doppler-anemometry size distributions from sprays of optically inhomogeneous liquids *Appl. Opt.* **35** (15) 2537–43

[12] Tikhonov A N and Arsenin V Y 1977 *Solutions of Ill-Posed Problems* (New York: Winston-Wiley)

[13] Press W H, Flannery B P, Teukolsky S A and Vetterling W T 1987 *Numerical Recipes* (New York: Cambridge University Press)