Evaluation of Light Scattering Data from Bimodal Turbid Suspensions: A Simple Fit Procedure

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SUMMARY: Whereas correlation spectroscopy gives reliable information on the size of immersed particles in those cases where the size distribution is narrow, large problems arise for more complex particle distributions. For instance, samples containing distinctly different particles of rather similar size lead to correlation functions which are very close to those of monodisperse samples. We present a measurement technique which is based on angle dependent measurements of 3D cross correlation functions and an evaluation scheme which uses the results of the Mie theory. The experimental technique warrants applications to strongly scattering samples. Having tested this procedure with mixtures containing standard latex particles we applied it to a sample of skimmed, homogenized milk.

Introduction

The evaluation of measured correlation functions in dynamic light scattering experiments on polymodal suspensions is based on the fact that these functions can be expressed in a simple way by the size distribution function of the particles and the size dependent correlation function of a uniform particle ensemble[1)]. Despite this simple connection the determination of the particle size distribution on the basis of the measured correlation functions, however, is a difficult task which has attracted a large amount of interest in the last two decades[2,3)]. In this context the signal to noise ratio plays an important role. Reliable application of the various inversion techniques employed so far is only possible for smooth correlation functions of relatively high quality. A small increase of noise often leads to significant variations of the particle size distributions calculated.

In this paper we are going to consider the special case of a bimodal distribution of particle

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sizes, i.e. the considered samples contain two types of spherical particles with a fairly well defined particle size each. Under the condition that the sizes of the two types of scatterers are very different the measured correlation functions show systematic and detectable deviations from a simple exponential behaviour. Then the inverse Laplace transformation for instance by CONTIN[4,5] yields the size and the concentration ratio of the scattering particles. This procedure becomes problematic when the particle sizes are rather similar. In such cases inverse Laplace procedures require correlation functions of high quality and of low noise levels. Otherwise deviations of the resulting correlation function from a simple exponential can hardly be recognized, not to mention be evaluated.

In order to enhance the resolution one has to perform measurements for different scattering angles. Furthermore the evaluation procedure of the correlation functions implies knowledge about the scattering form factors of the contributing particles. For homogeneous and spherical particles the form factors can be obtained by Mie[6] theory. In this way multiangle Laplace inversion techniques are capable of resolving bimodal samples with particle size ratios down to 1:2 [7–9]. However, the results of the Laplace inversion techniques are still sensitive to noise on the experimental data which could lead to broadened size distributions and unreproducible peak positions[10]. Additionally, such multiangle procedures are only useful when the particle form factors show a pronounced angle dependence, thus there is no improvement of resolution for scatterers with diameters $a < \lambda/10$ where λ is the wave length of the illuminating laser beam.

Unfortunately, in common light scattering experiments one is confronted with additional difficulties caused by multiple scattering. Even for samples which are usually considered as weakly scattering, multiple scattering leads to systematic errors[11,12]. In dynamic light scattering the influence of multiple scattering leads to a higher value for the initial slope of the auto correlation functions. This effect is especially pronounced for small particle diameters a and for small values of the scattering angle θ. Contrary to this behavior the influence of multiple scattering in static light scattering experiments increases with increasing θ at least for particle diameters $a > \lambda/10$ [11]. Clearly, this situation complicates the evaluation of data recorded in combined static and dynamic light scattering experiments.

In the last decade a number of different measurement schemes based on cross correlation techniques have demonstrated that these techniques are powerful tools for an effective suppression of the disturbing influence of multiply scattered light[12–26]. However, for weak scattering samples the theoretical limit of the signal to noise ratio for cross correlation experiments is significantly smaller than for auto correlation functions even for an ideal alignment of the set-up. Furthermore, the signal to noise ratio of cross correlation functions strongly depends on the alignment of the optical elements, which for such experiments is highly pretentious. These disadvantages are counterbalanced by the fact that no systematic errors due to multiple scattering occur. Moreover, cross correlation experiments allow the investigation of strongly scattering samples which again enhances the signal to noise ratio. However, for very strongly scattering samples where the fraction of singly scattered light becomes small, the statistical accuracy of cross correlation experiments decreases again.

In this paper we present a simple multiangle evaluation scheme for a set of angle dependent cross correlation functions and light scattering intensities. Our procedure makes use of the variation of the averaged particle size and of the scattering intensity with the scattering angle.

We investigated turbid suspensions containing latex particles with size ratios of 1:3.4 and 1:1.9. In these cases the deviations of the measured correlation functions from the mono exponential form are extremely small and the only reliable information that can be obtained from the experimental cross correlation functions is the averaged (or apparent) particle size. We show that the angle dependence of this quantity often yields first estimates of the contributing particle sizes. Combined with the measured intensities of the singly scattered light, this procedure yields the correct particle sizes and concentration ratios for bimodal suspensions, even for correlation functions which do not satisfy quality standards as high as necessary for performing inverse Laplace transformations. Furthermore we applied the procedure to skimmed, homogenized cow milk and compared the results with TEM (transmission electron microscopy) measurements.

Theoretical Background

We start with a definition of the normalized 3D cross correlation function:

$$C(\theta,\tau) = 1 + [R(\theta)\ G(\theta,\tau)]^2 \qquad \text{with} \qquad G(\theta,\tau) = \frac{\langle E_A(\theta,0)\ E_B^*(\theta,\tau)\rangle}{\sqrt{\langle |E_A(\theta,0)|^2\rangle\ \langle |E_B(\theta,0)|^2\rangle}}$$

In these equations $\langle \dots \rangle$ denotes averages, $E_{A,B}(\theta,\tau)$ are the electric field amplitudes of the singly scattered light at the detectors A and B, respectively, τ is the delay time, and θ

is the scattering angle. For monomodal suspensions containing spherical, non-interacting particles $G(\theta, \tau) = \exp(-\Gamma \cdot \tau)$. Here Γ equals $D \cdot q^2$, with the length of the scattering vector $q = 4\pi n \sin(\theta/2)/\lambda$, and the refractive index n. The diffusion constant D is related to the diameter of the suspended particles by the Stokes-Einstein equation $D = k_B T/(3\pi\eta a)$, where k_B is the Boltzmann constant, η the viscosity of the medium and T the absolute temperature. $R(\theta)$ is an amplitude factor which – especially in cross correlation experiments – depends extremely sensitively on deviations from the ideal alignment. Therefore the 3D cross correlation functions were normalized to the amplitudes $R_{c\to 0}(\theta)$ measured for nearly transparent samples[12,25]. The resulting amplitude $R_*(\theta) = R(\theta)/R_{c\to 0}(\theta)$ directly yields the intensity ratio of the singly scattered to the total scattered light.

For suspensions with scatterers of two different sizes the correlation function of the electric field amplitudes is given by[1]

$$G(\theta, \tau) = \frac{\langle I_1(\theta)\rangle}{\langle I_{tot}(\theta)\rangle} \exp(-\Gamma_1(\theta) \cdot \tau) + \frac{\langle I_2(\theta)\rangle}{\langle I_{tot}(\theta)\rangle} \exp(-\Gamma_2(\theta) \cdot \tau) \tag{1}$$

where the indices 1 and 2 refer to the two types of scatterers. In this equation $\langle I_{tot}(\theta)\rangle$ denotes the sum of the intensities $\langle I_1(\theta)\rangle$ and $\langle I_2(\theta)\rangle$ which result from single scattering processes at the particles. $\Gamma_{1,2}(\theta) = D_{1,2} \cdot q^2$ reflects the diffusive motion of the particles 1 and 2, respectively.

For scatterers of extremely different particle sizes eq. (1) leads to significant deviations of $G(\theta, \tau)$ from a single exponential. In contrast to that measurements with suspensions containing particles of rather similar size lead to correlation functions which usually look like single exponentials. The initial slope of such correlation functions is given by

$$\Gamma_{tot}(\theta) = \frac{\langle I_1(\theta)\rangle}{\langle I_{tot}(\theta)\rangle} \Gamma_1(\theta) + \frac{\langle I_2(\theta)\rangle}{\langle I_{tot}(\theta)\rangle} \Gamma_2(\theta) \tag{2}$$

with $\langle I_{1,2}(\theta)\rangle = N_{1,2} \cdot i_{1,2}(\theta)$. Here $N_{1,2}$ is the number of particles in the scattering volume and $i_{1,2}(\theta)$ is the intensity at the detector which results from scattering by a single particle. The particle concentration of spheres with density ϱ and diameter a is related to the mass concentration $c_{1,2}$ by $N_{1,2}/V = 6\, c_{1,2}/(\pi a^3 \varrho)$, where V is the volume considered.

The only quantities which can be determined experimentally are $\langle I_{tot}(\theta)\rangle$ and $\Gamma_{tot}(\theta)$. All other quantities appearing on the right hand side of eq. (2) are not directly accessible by experimental means. Of course, for given values of particle sizes and concentrations the quantities $\Gamma_{1,2}(\theta)$ and $i_{1,2}(\theta)$ can be obtained by the Stokes-Einstein relation and the Mie theory, respectively. Therefore, we start calculations for the fit procedure with initial values for particle sizes and concentrations which can often be estimated from the experimental data as will be shown in the next section. The next step is to improve the agreement between experimental and theoretical data of $\langle I_{tot}(\theta)\rangle$ *and* $\Gamma_{tot}(\theta)$ by varying the particle sizes $a_{1,2}$ and concentrations $c_{1,2}$ for the calculations. This is repeated until the estimated values for $a_{1,2}$ and $c_{1,2}$ yield angular dependencies for both, the decay constant $\Gamma_{tot}(\theta)$ *and* the total scattered light intensity $\langle I_{tot}(\theta)\rangle$ which agree with the results obtained experimentally.

Experiments

All measurements were performed with a 3D cross correlation instrument described in detail in [12]. Since this measurement scheme avoids any deterioration of the measured correlation function by multiply scattered light it allows the investigation of samples up to fairly high turbidity levels. We performed static (3D-SLS) and dynamic (3D-DLS) light scattering experiments with bimodal suspensions containing standard latex particles in de-ionized water. The evaluation of the experimental data is then performed by the fit procedure described in the previous section to the whole set of measured correlation functions and light intensities.

The diameters of the latex particles obtained by TEM measurements provided by the manufacturer (Dow) are 453.0 ± 9.0 nm, 236.0 ± 6.8 nm and 132 nm (no standard deviation given by the manufacturer). The refractive index of the latex spheres according to the manufacturer is $n = 1.59$ and the density is $\varrho = 1.05$ g/ml.

We checked the values for the particle size by light scattering experiments. We took advantage of the 3D-SLS method[11,25] which allows to select that part of the scattered light intensity which stems from single scattering processes. The angle dependence of the singly scattered light intensity obtained with monomodal samples was then evaluated by means of the Mie theory which yields values for the average particle size $a = 462 \pm 18$ nm, 238 ± 2 nm and 143 ± 3 nm. Furthermore, the evaluation of the time dependence of 3D cross correlation functions (3D-DLS) measured for the bigger particles yield $a = 445$ ± 5 nm and 246 ± 10 nm. All values are in rather good agreement with the TEM-data provided by the manufacturer.

The particle size ratios of the investigated bimodal suspensions were 1:3.4 and 1:1.9. For all measurements the temperature was kept at 20.6 ± 0.2°C. The turbidity of the samples was obtained using the relation $\ln(I_o/I_{trans})/l$, where I_{trans} is the intensity of the transmitted laser light in the presence of the sample, I_o is the corresponding value for the cell filled with pure water and $l = 10$ mm is the thickness of the sample. The turbidity level of the bimodal suspensions we used for our measurements ranged from 0.17 cm^{-1} to 2.79 cm^{-1}. As shown in [25] these values correspond to a concentration range where interactions of the 493 nm particles may cause a shift of the apparent particle size of ten percent maximum. This effect is negligible compared to the size differences in the bimodal solutions considered in this paper.

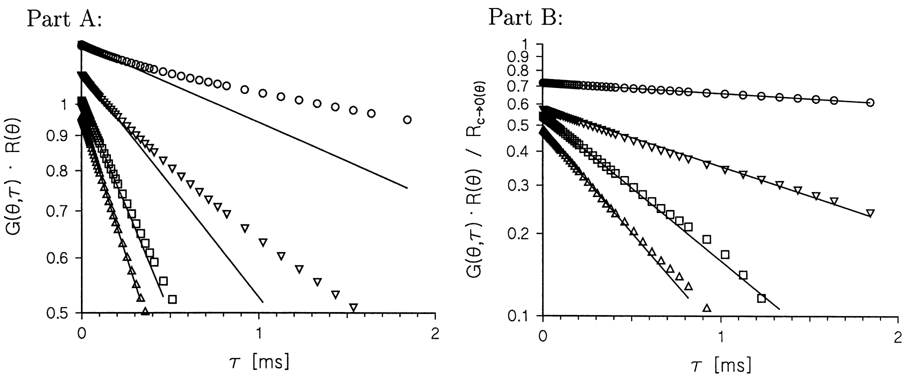

Fig. 1: Part A: Auto correlation functions measured at a bimodal suspension containing standard latex particles with TEM diameters 453 nm and 132 nm. The ratio of the mass concentration c_1/c_2 of the particles was 1:20.2. The scattering angles were $\theta = 30°$ (○), 60° (▽), 90° (□), 120° (△). The turbidity of the suspension was 2.60 cm^{-1}. For clarity of presentation the data of the auto correlation functions for $\theta = 30°, 60°$ and 90° have been given an offset.
Part B: 3D cross correlation functions measured with the same sample as in part A.

The first figure shows a comparison of auto and 3D cross correlation functions of the electric field amplitude for several scattering angles. The correlation functions were measured with a strongly scattering bimodal sample containing particles with diameters of 453 nm and 132 nm (fig. 1). The comparison demonstrates the strong influence of multiple scattering which – in case of the auto correlation technique – could lead to erroneous interpretations of the data. As can be clearly seen the measured auto correlation functions in part A of fig. 1 show significant deviations from a mono exponential decay, especially for small scattering angles. These deviations could be interpreted as if they were caused by a broad distribution of particle size. However, in this case the observed deviations do not result from different contributions of scattering particles but from the influence of multiple scattering.

This is confirmed by considering 3D cross correlation functions (part B of fig. 1) measured with the same sample. Here the normalized amplitudes $R_*(\theta)$ directly yield information about the influence of multiple scattering: For $\theta = 30°$ only 70% of the scattered light stems from single scattering processes. This is worse for $\theta = 120°$ where the intensity ratio of the singly to the total scattered light drops down to 50%. Moreover, the time dependence of the measured 3D cross correlation functions – which is caused by single scattering processes only – does not show such systematic deviations from a simple exponential as observed for the auto correlation functions. As mentioned above, this is due to the relatively small size ratio of 1:3.4 where the two correlation functions sum up to a cross correlation function which appears to be mono exponential. Therefore, it is difficult or even impossible to yield reliable information about the constituents from a single cross-correlation function.

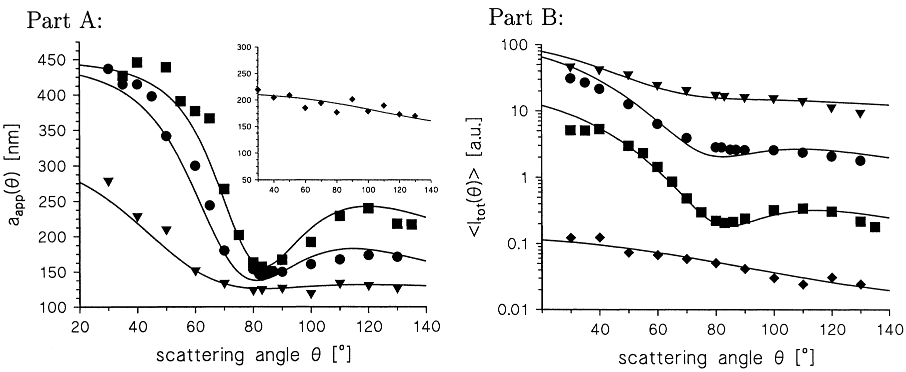

Fig. 2: Part A: Apparent particle size $a_{app}(\theta)$ of bimodal suspensions. $a_{app}(\theta)$ was calculated from the initial slope of 3D cross correlation functions using the Stokes-Einstein relation. The TEM diameters of the suspended particles were 453 nm and 132 nm, the concentration ratios were 1:1.1 (■), 1:2.0 (●) and 1:20.2 (▼), the turbidity levels were 1.08 cm^{-1}, 0.17 cm^{-1} and 2.60 cm^{-1} respectively. The inset shows $a_{app}(\theta)$ for a sample containing 236 nm and 132 nm particles with c_1/c_2 = 1:1.9.
Part B: Intensity of the singly scattered light $\langle I_{tot}(\theta)\rangle$ of the same samples as in part A. The solid lines represent the results (see tab. 1) of the fit procedure.

As mentioned in previous section we continue considering the initial slope $\Gamma_{tot}^{exp}(\theta)$ of the measured cross correlation functions which correspond to an apparent particle size $a_{app}(\theta)$. Part A of fig. 2 shows the results for $a_{app}(\theta)$ for several concentration ratios. Again, the diameters (TEM) of the suspended latex particles were 453 nm and 132 nm. The inset of fig. 2A shows results of a mixture containing 236 nm and 132 nm latex particles.

For all values of c_1/c_2 the apparent particle size $a_{app}(\theta)$ increases with decreasing scattering angle. This can easily be understood by considering the fact that the fraction of light scattered in forward direction increases with particle size. Therefore, the time dependence of the correlation functions measured at small θ essentially reflects the Brownian motion of the larger particles whereas for larger scattering angles the influence of the smaller particles increases. Based on these observations one can get first estimates for the sizes of the suspended particles.

Furthermore, the apparent particle size $a_{app}(\theta)$ depends differently on the scattering angle θ for the different concentration ratios. Conspicuous are the minima occurring around $\theta = 80°$ which are smeared out if the smaller particles contribute strongly to the total scattered light. Similar considerations hold for the angular dependence of the singly scattered light intensity $\langle I_{tot}(\theta)\rangle$ (Part B of fig. 2).

The solid lines in fig. 2 represent results obtained by the fit procedure described in the previous section with particle diameters and concentration ratios as free parameters. As can be seen, the angle dependence of the experimentally determined data for $a_{app}(\theta)$ and $\langle I_{tot}(\theta)\rangle$ are well described by the results obtained in this way.

Table 1: Particle sizes and concentration ratios of the samples investigated by light scattering and values obtained from the fit procedure outlined in the previous section.

samples		results from fit	
particle sizes [nm]	concentration ratio	particle sizes [nm]	concentration ratio
453/132	1 : 1.1	460 ± 10 / 140 ± 5	1 : 0.55 ± 0.1
453/132	1 : 2.0	470 ± 10 / 130 ± 5	1 : 1.6 ± 0.1
453/132	1 : 20.2	460 ± 5 / 125 ± 5	1 : 14.5 ± 0.1
236/132	1 : 1.9	240 ± 5 / 120 ± 5	1 : 1.3 ± 0.1

Furthermore, the values for the particle sizes obtained by the fit procedure are in good agreement with the values provided by the manufacturer and by our measurements on monomodal suspensions (see table 1). We conclude, that the presented simple model is suitable to get reliable information on the diameters of spherical, non-interacting particles in bimodal suspensions. Additionally, the results give a rough measure of the concentration ratio.

After having demonstrated the reliability of our measurement scheme using a rather artificial sample, we applied it to the more complex system of commercially available homogenized, skimmed milk. Our sample was supplied from Goldblume-O'LACY'S GmbH, Düsseldorf, Germany and contained 1 - 3 g/l milkfat and 3.4 g/l protein. For investigation by light scattering and by TEM the sample was strongly diluted with de-ionized water. TEM images (fig. 3) clearly show that the laser light is primarily scattered by two different types of particles. According to literature[27,28] the larger ones can be identified as fat globules and the smaller ones as casein micelles. Quantitative analysis of a set of TEM images yields a mean diameter for the fat globules of $(5.6 \pm 1.8)\cdot 10^2$ nm and for the casein micelles of $(1.5 \pm 0.4)\cdot 10^2$ nm.

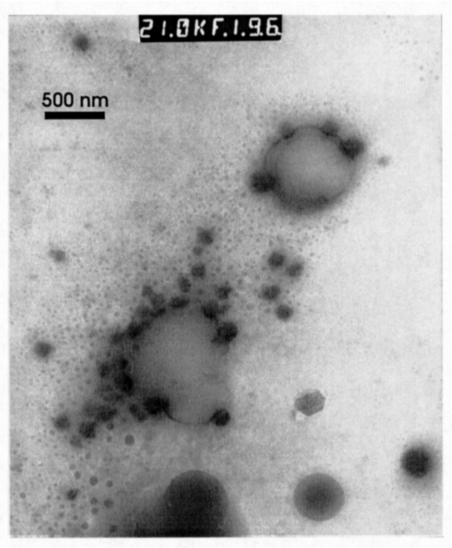

Fig. 3: TEM image of skimmed preserved milk. The milk fat forms large globules while the small dark droplets represent casein micelles. The light grey dots may be submicelles.

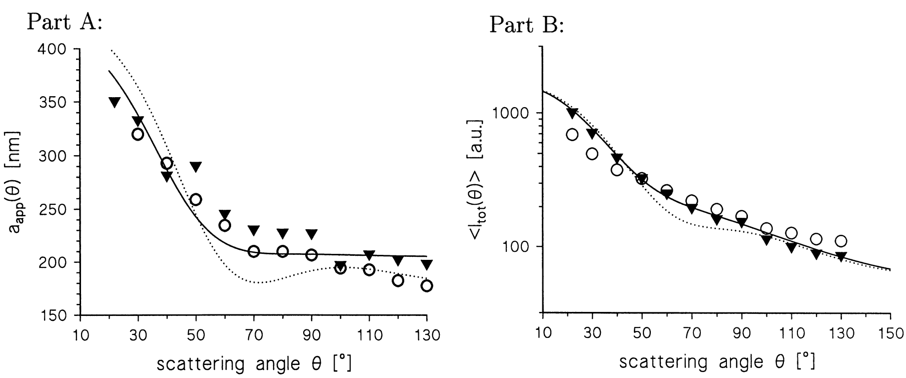

Fig. 4: Part A: Apparent particle size $a_{app}(\theta)$ for skimmed, homogenized milk containing 0.1 - 0.3 % fat and 3.4 g/l protein. The turbidity levels of the diluted samples were 0.83 cm^{-1} (○) and 1.94 cm^{-1} (▼).
Part B: Intensity of the singly scattered light $\langle I_{tot}(\theta)\rangle$ of the same samples as in part A. The lines in part A and B represent the results of the fit procedure. Dotted lines: results of the described fit procedure assuming narrow size distributions of casein micelles and fat globules.

Our TEM image also shows small particles with diameters of approximately 50 nm which may be interpreted as 'submicelles'. This would be in accordance with light scattering experiments of other workgroups which investigated milk for different dilution steps. According to [9] the casein micelles decompose into submicelles upon dilution. The appearance of such submicelles would also explain the finding of Sinn et al.[29] who report a shift towards smaller values and an increase of polydispersity of the particle size distribution for diluted samples. However, Fig. 3 gives clear evidence that even in strongly diluted milk samples large amounts of intact casein micelles are present which are the main interest of this paper.

The turbidity levels of the investigated samples were 0.83 cm^{-1} and 1.94 cm^{-1}. Fig. 4 part A and B shows the results for $a_{app}(\theta)$ and for the singly scattered light intensity $\langle I_{tot}(\theta)\rangle$. We think that the angle dependence of the apparent particle size results from two types of scatterers with distinct sizes, as is also suggested by TEM measurements. In this sense we suppose that the values for $a_{app}(\theta)$ at large scattering angles result from contributions of small particles with $a_{app}(\theta) \simeq 180$ nm. For smaller θ the light scattered by larger particles clearly dominates.

Fig. 4 shows the results obtained by the fit procedure for which we used the following values for the refractive index n and density ϱ: milk fat: $n_{fat} = 1.455$, $\varrho_{fat} = 0.931$ g/ml, casein: $n_{cas} = 1.55$, $\varrho_{cas} = 1.45$ g/ml [27,28]. Following the TEM measurements we assumed a relatively broad width of the particle size distributions and used 180 nm for the fat globules and 40 nm for the casein micelles. The solid lines in fig. 4 represent results of the fit procedure which yields $a_{fat} = 550$ nm as mean value for the diameter of the fat globules and $a_{cas} = 180$ nm for the casein micelles. For comparison, the dotted lines in fig. 4 represent results of calculations which do not take into account the rather large width of the size distribution of the milk constituents. The results obtained for the particle diameters are in good agreement with the values found by TEM. For the concentration ratio we obtained $c_1/c_2 = 1 : 2.3 \pm 0.1$ which also agrees with the specification given by the producer.

Summary and Discussion

In this paper we demonstrated a successful evaluation procedure for light scattering data obtained from measurements on bimodal suspensions. The aim was to determine the composition of suspensions containing scatterers with small size differences. As experimentally determined cross correlation functions can hardly be discriminated from single exponentials, an evaluation with inversion techniques like CONTIN does not yield detailed information about the composition of those samples. We presented a fit procedure which is based on the combination of angle dependent static and dynamic light scattering measurements. Under the condition that the scattered light stems from single scattering processes only, the procedure is applicable in the size regime $a > \lambda/10$. We investigated samples with size ratios 1:3.4 and 1:1.9, and could demonstrate that the presented simple model yields reliable results concerning the composition of the samples.

The advantage of the presented procedure is its easy handling: one only needs to determine the initial slope of the correlation functions, which in general can easily be done. This also means, that the correlation function does not need to be as smooth as for an evaluation using CONTIN. This point becomes especially important when the signal to noise ratio is low as e.g. for very weak or strongly scattering samples, where the contribution of singly scattered light is small. In such cases the CONTIN-procedure generally does not yield very reliable results, it may even happen that artificial peaks appear in the calculated size distribution functions.

Furthermore, the procedure appears to be quite stable: Even if the fit procedure starts with physically unreasonable values it ends up with correct values for the sizes of the contributing scatterers.

It is surprising to us that the presented model is also well applicable to a complex sample like milk. As known from TEM images as well as from literature the size of casein micelles and fat globules in milk have rather broad size distributions with relative widths of $\simeq 25\% - 32\%$. It can clearly be seen that the consideration of this size distributions mainly affects the results in the region of the scattering minimum of the larger fat globules. Nevertheless, our analysis shows that this does not have any considerable effect on the performance of the fit procedure. Only near the scattering minimum the agreement between experimentally obtained data and the results of the fit procedure is not satisfactory.

To summarize we can state that the presented fit procedure can be considered as a helpful complement to the well established CONTIN program in special cases.

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