Analysis of the light scattering of a colloid droplet on a Gaussian beam to determine the suspension concentration

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Abstract

In this study, the light scattering of a colloid droplet on an elliptical Gaussian beam was analyzed to determine the concentration of the suspended particles in a suspension droplet. The light scattering was studied for a fixed scattering angle in the backscatter region. The data was acquired with a high time resolution by using photo multiplier and high-speed signal analyzer. The measurements of the different suspension concentrations of the droplet were performed in order to provide the sufficient data for the construction of an empirical model giving the correlation between the light scattering signal and the suspension concentration. The measurements were carried out with a monodisperse droplet generator. The correlation between the light scattering signal and the suspension concentration in the droplet was established by two methods, relying on an artificial neural network and relying on statistical properties of the light scattering signal.

Keywords: Suspension droplets, machine learning, spray drying

Introduction

The characterization of the suspension droplets plays a role in various processes, including pharmaceutical 3D printing of tablets, spray drying for powder production, and spray painting optimization. This study aims to develop a method for determining the concentration of a suspension droplet. In particular, it focuses on investigating light scattering effects in an elliptical Gaussian beam, where the beam size is smaller than the suspension droplets.

A commercial device [1][2] utilizing the time-shift technique was used to determine the size and velocity of droplets in sprays. This technique uses a time delay in the signals resulting from light scattering from droplets on the focused Gaussian elliptical beam. A software modification made it possible to acquire and to save the light scattering signals from a single droplet with a high-speed digitizer.

In order to get an idea of how the light scatters on a suspension droplet, we examined the individual suspension droplets with different concentrations. For this purpose, droplets were generated with an almost constant droplet size using a commercial monodisperse droplet generator [3]. The light scattering signals from the individual suspension droplets with different concentrations were categorized for a single droplet size. We used a milk-water mixture with different mixing ratios to simulate the suspension with different concentrations.

Using two different signal processing approaches, the concentration of the suspension droplets could be detected. We tested these approaches in measurements with a monodisperse droplet generator.

Tropea et al. [4][5] first presented a two-dimensional light scattering model for the light scattering signal of a single suspension droplet on a light sheet. While experimental results showed good agreement with this model at high suspension concentrations, it was unsuitable for lower concentrations. In addition, the Gaussian beam was approximated to a laser line; this is only sufficient if the droplet is much larger than the Gaussian beam thickness.

Rosenkranz et al. [6] developed a model based on ray tracing for two-dimensional simulation of light scattering on a Gaussian beam for suspension droplets. This model demonstrated the potential for measuring suspension concentration by analyzing the ratio of the internal scattering signal amplitude to the reflection peak, which is proportional to the optical mean free path and, consequently, to the

suspension concentration. Experimental validation was performed using milk-water mixtures at concentrations of 15%, 50% and 100%, corresponding to optical mean free path $E(L)$ of 10μm, 5μm and 2μm, respectively.
Li et al. [7] used a Monte Carlo ray tracing model for the three-dimensional simulation of light scattering on a Gaussian beam for the suspension droplet. It was observed that for volumetric suspension concentrations between 0.05% and 0.25% and $E(L)$ between 467μm and 80 μm, the ratio of the intensities of the second-order refraction scattering peak of a suspension and pure droplet is proportional to the suspension concentration. For higher volume concentrations, 2% and $E(L) = 10$ μm, the ratio of the intensities of internal and reflection scattering was proposed to characterize suspension concentration in a way similar to that of Rosenkranz et al. [6]. Additionally, it was noted that this intensity ratio depends on droplet diameter due to variations in reflective peak amplitude.
Li and Tropea [8] further investigated light scattering in a Gaussian beam to measure suspension droplet concentrations containing spherical nanoparticles using two approaches. The first approach utilizes the ratio of the signal scattering from the nano-particles and the reflective scattering from the undisturbed droplet. The second approach relies on the attenuation of the portion of the light scattering signal generated by second-order refractive scattering. Li and Tropea have developed a procedure for identifying peaks in the light scattering signal and investigated suspension concentrations from 0 to 10%. The article has proposed methods for the measurements of the suspension concentration and sizes of the nano-particles and droplets.
This paper is structured as follows: Section 1 introduces the theoretical background of light scattering in suspension droplets and discusses previous work in the field. Section 2 describes the experimental setup, including the droplet generation system and the light scattering measurement procedure. Section 3 presents the artificial neural network (ANN) approach for analyzing the light scattering signals and determining the suspension concentration. Section 4 explores an alternative statistical analysis method based on the shape factor of the light scattering signal. Finally, Section 5 summarizes the key findings and outlines directions for future research.

1 Light scattering of a suspension droplet on a Gaussian beam

The light scattering behavior of individual suspension droplets in a Gaussian beam has been investigated in previous studies [4] [5] [6] [7] [8], where corresponding light scattering signal models were developed. Unlike transparent droplets, suspension droplets contain dispersed liquid or solid particles within another liquid, making their light scattering characteristics more complex. In this case, light scattering includes not only the reflection and refraction light scattering effects, but also light scattering from small particles within a liquid, here called "scattering centers" (see Figure 1). In the present study, the

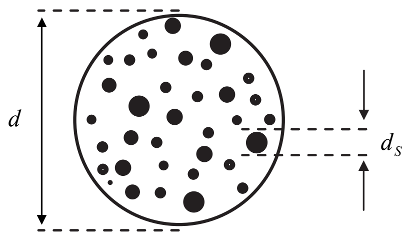
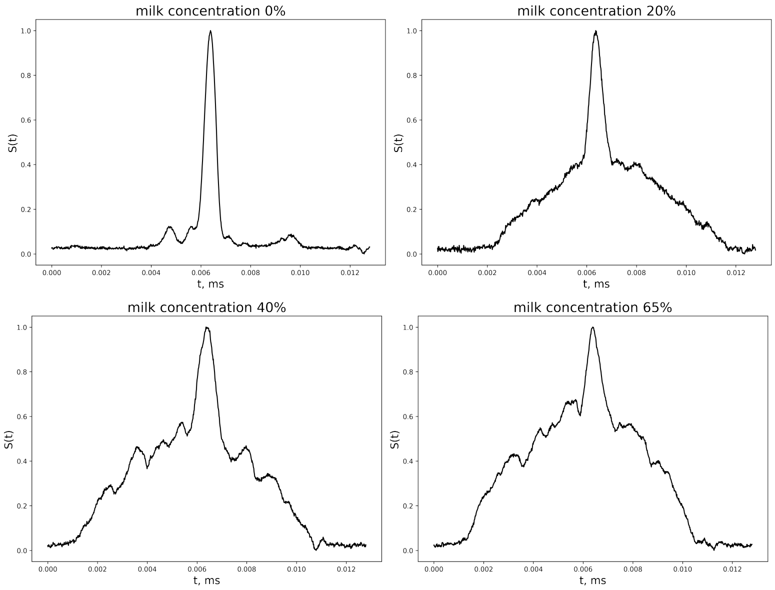


Figure 1. *Schematic illustration of a suspension particle. d_s is the size of a scattering center and d is the droplet size.*

Gaussian beam is smaller than the droplet diameter, allowing individual scattering orders to be temporally separated as the droplet traverses the beam. The droplet is "scanned" by the beam while passing through it. Transparent droplets exhibit reflection and refraction [9] [10], while in suspension droplets, internal light undergoes multiple scattering events due to reflections and refractions at the scattering centers. This process results in a more uniform overall scattering distribution. Depending on the concentration and absorption properties, the scattering centers can lead to a significant weakening of the refracted scattered light, eventually suppressing refraction-based scattering contributions. Suspension droplets with a low concentration of internal scattering centers can be considered as semi-transparent, such that higher-order refraction contributions can still be observed. With increasing concentrations, the refractive scattering orders will decrease in intensity, eventually leaving only reflected light. The high concentration of scattering centers suppresses higher order refraction. In contrast to higher order refraction, surface reflection at the scattering centers remains independent of concentration. Experimental observations of milk-water mixtures with varying concentrations (see Figure 2) reveal distinct differences in the structure of the recorded light scattering signals.

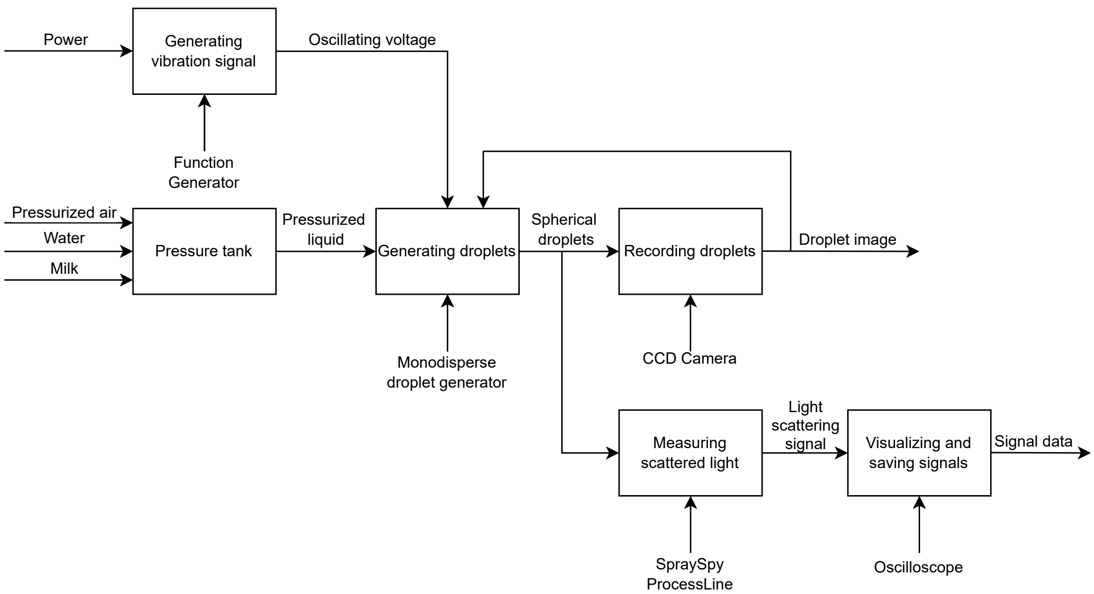
Figure 2. *Examples of training samples.*



2 Experimental setup

To measure the concentration of the suspension droplet and thus generating the necessary data, a specific experimental set up was used. Therefore, this chapter focuses on how the suspension droplets were created and the light scattering signals were measured. A schematic of the setup, following the IDEF0 functional modeling standard, is presented in Figure 3.

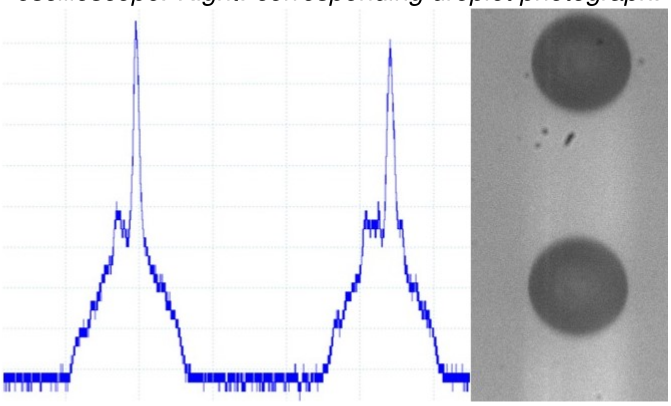
Figure 3. *IDEF0 diagram of the experimental setup, illustrating functions (blocks), inputs (left arrows), outputs (right arrows), controls (top arrows), and required resources (bottom arrows).*



The measurement process begins with the preparation of the suspension by mixing water and milk at a predefined weight percentage. The mixture is then introduced into a commercial monodisperse

droplet generator [3] at a constant pressure of 240 kPa. Under vibrations at frequencies ranging from approximately 37 kHz to 42 kHz, the fluid stream disperses into a chain of monodisperse spherical droplets. The exact frequency depends on parameters such as concentration and pressure and must be adjusted to maintain a stable droplet chain.
Light scattering from the droplets is measured using a commercial system [1], which includes a 405 nm laser source and two photodetectors (designated as A and B) positioned at a fixed scattering angle of 165 deg.
The scattered light signals are converted into voltage signals via a transimpedance amplifier and recorded by a digital oscilloscope [11]. Each measurement consists of 32 frames, each lasting 20 ms and sampled at 312.5 MS/s. A single frame contains approximately 1000±150 individual light scattering signals. Since two detectors are used, the oscilloscope records signals from two independent channels, designated as Channel A and Channel B. The raw measurement data is openly accessible [12], and ensemble-averaged light scattering spectra (LSS), each derived from 1000 ± 150 individual LSS, have also been published [13].
A CCD camera independently monitors the droplet chain while light scattering data is acquired. The captured images ensure droplet sphericity and allow for adjustments to the droplet generator parameters. The camera also provides visual measurements of the droplet diameter, which can be further analyzed using image processing techniques. Representative images of the droplets and their corresponding oscilloscope signals are shown in Figure 4.

Figure 4. *Left: light scattering signal of a 19% concentration suspension read directly from the oscilloscope. Right: corresponding droplet photograph.*

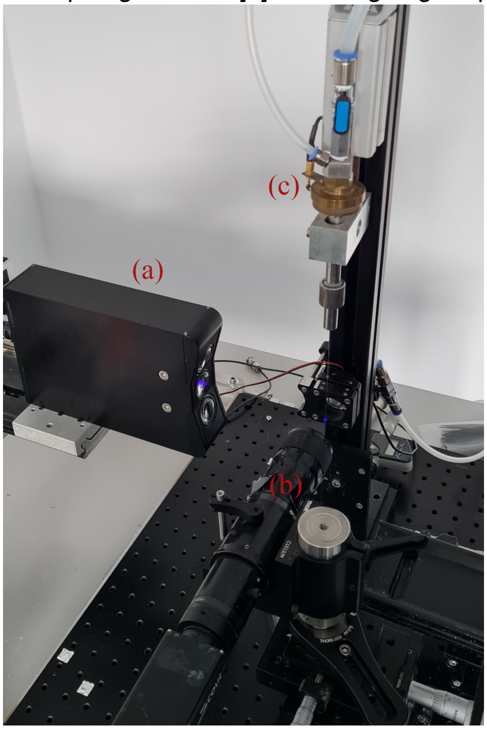


To ensure consistency across measurements, the droplet chain is positioned at the point of maximum light scattering amplitude within the measurement plane. This guarantees a stable measurement position when adjusting suspension concentration. The complete experimental setup is depicted in Figure 5.
Measurements were conducted for suspension concentrations ranging from 0% to 100%. For concentrations between 0% and 25%, measurements were taken in 1% increments, whereas for concentrations above 25%, a step size of 2.5% (rounded to 3%) was used. The finer resolution at low concentrations accounts for the significant variations in optical properties in this range, where the droplets exhibit semi-transparent behavior. The droplet diameter, averaging approximately 150 µm, was verified by comparing CCD camera images with a scaled reference template (Figure 6). This verification can also be performed digitally using image processing techniques.

3 Light scattering signal analysis by ANN

In this section, we present a signal processing method that uses an ANN for signal analysis to determine the suspension droplet concentration. The light scattering signal (LSS) from each individual droplet was extracted from the frames by using a scipy algorithm [14]. The duration of the each light scattering signal corresponded to 1000 time samples. The examples of the light scattering signals are shown in Figure 2. The signal amplitude for each LSS are normalized by a maximum to ensure that the ANN algorithm recognizes the LSS according to the signal shape.
The architecture of the ANN algorithm is typical for the signal analysis approaches based on the convolutional neural networks. It has a block structure (see Figure 7), where each of ten blocks includes a convolutional layer, a batch normalization layer [15] and an activation ReLu function [16]. Each block,

Figure 5. *Experimental setup with the measuring head (a) [1], the CCD camera (b) and the monodisperse droplet generator [3] with outgoing droplet chain (c).*



except for the terminal one, is followed by a max pooling layer. Each convolution layer is applied with the following parameters: kernel size = 3, padding = 2, dilation = 1, stride = 1. Each max pooling layer is applied for the kernel size = 2, padding = 0, dilation = 1. The final stage of the ANN algorithm is a fully connected dense layer with the output processed by the Sigmoid function to obtain the results from 0 to 1, which corresponds to the milk concentration. The algorithm is implemented in Pytorch package [17] in Python software.
For the milk concentrations below 25% an amount of 1000 LSS are selected for the ANN training, for milk concentrations above 25% an amount of 1350 LSS is selected for the ANN training. Thus, the total set of 138700 LSS is used. 80% of the LSS from this amount are used for the training, whereas 20% of the remaining ones are used for the validation. For the testing of the ANN algorithm another 150 LSS for each concentration are selected. Therefore 17550 LSS are used for the testing. The cost function during the training is the root-mean-square deviation between the predicted and real milk concentration. During each training iteration a batch size of 100 snapshots is used. Figure 8 demonstrates loss value during training session for training and validation subsets.
As an optimiser AdamW is used [18]. Initial learning rate is $5{\cdot}10^{-4}$ and it is decreased every tens epoch by the factor of 2. The total duration time of each training on a GPU NVIDIA 1050 (4 GB) was approximately 100-110 minutes and includes 75 training epochs. Figure 8 shows that the convergence is achieved after 40 training epochs with the prediction precision of approximately 3% for the validation subset.
The results of the ANN algorithm testing for the various milk concentrations are shown in Figure 9. The color indicates the absolute prediction error as a percentage. As can be seen from Figure 9, the prediction accuracy is typically better than 5% for the milk concentration of 40%. Figure 10 shows the standard deviation of the absolute error, which for the present data set growth linearly with the milk concentration and is typically better than 2% for the concentration lower than 40%. In addition, Figure 11 shows signals that have significant prediction errors.

4 Light scattering signal analysis by statistics

In this section, an alternative approach for concentration measurement is described. The approach is relying on statistical properties of light scattering signals of the suspension droplets on the Gaussian beam and thus expected to be useful for complex situations. The previous studies [6] and [7] have demonstrated that the ratio of the amplitudes of the internal scattering signal and the reflection peak is proportional to the suspension concentration. Li and Tropea [8] have elaborated a procedure to identify

Figure 6. *Presentation of scaled template with inserted droplet. Each tick on the calibration scale is 50 μm.*

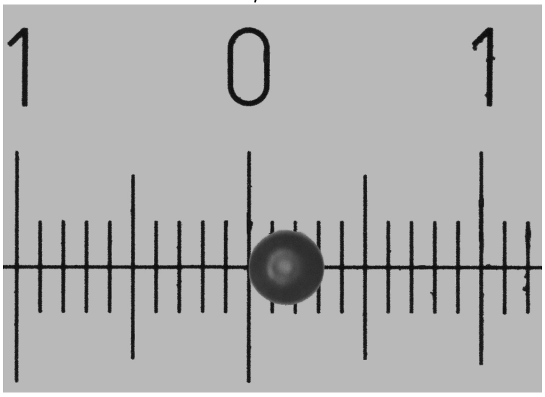
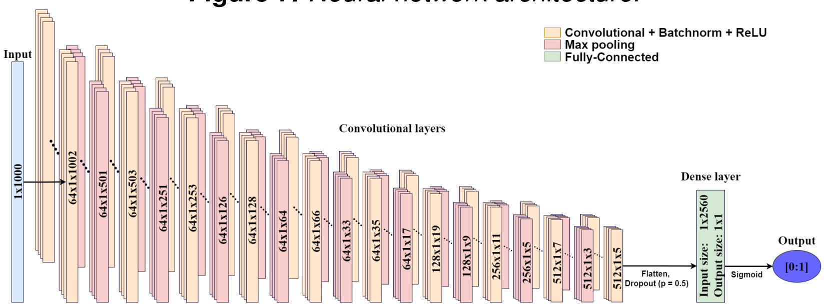


Figure 7. *Neural network architecture.*



the amplitude of the reflection peak for spherical nano-particles of constant size in a single spherical droplet.

We have applied a classification machine learning to find statistical properties of the LSS correlated with the suspension concentration. Statistical moments of the LSS from first to fourth (mean, standard deviation, skewness, kurtosis) were evaluated and the machine learning has identified that the mean and standard deviation are the two parameters of importance for signal classification. Further analysis has revealed that these two parameters can be combined in a single parameter by using their ratio. One can note that there is a clear analogy of this ratio with the peak intensity ratio used in [8]. In our case, the standard deviation is measuring the dispersion of the signal and characterizing the reflection peaks and the mean is characterizing the internal scattering. In the current study we are calling the ratio of the mean and standard deviation of the LSS a shape factor, H, in analogy with the shape factor of the boundary layer velocity profiles, which is also essentially the ratio of the first- and second-order statistical moments. Current definition of the shape factor is as follows:

$$H = \frac{S^*_{mean}}{S^*_{std}}$$

where $S^*(t)$ is a part of the scaled ensemble-averaged time signal $S(t)$ above a certain threshold level. A typical threshold level of 5% of the maximum amplitude was used in the current case. The threshold should be high enough to provide immunity against possible noise in the signal and low enough to include the entire meaningful part of the signal.

For digitized time signals the mean and standard deviation are calculated as:

$$S^*_{mean} = \frac{1}{N}\sum_{i=1}^{N} S^*_i$$

$$S^*_{std} = \sqrt{\frac{1}{N-1}\sum_{i=1}^{N} S^{*2}_i}$$

N is the total number of digitized signal samples above the threshold.

Figure 8. *Loss value during training session for train and validation subsets.*

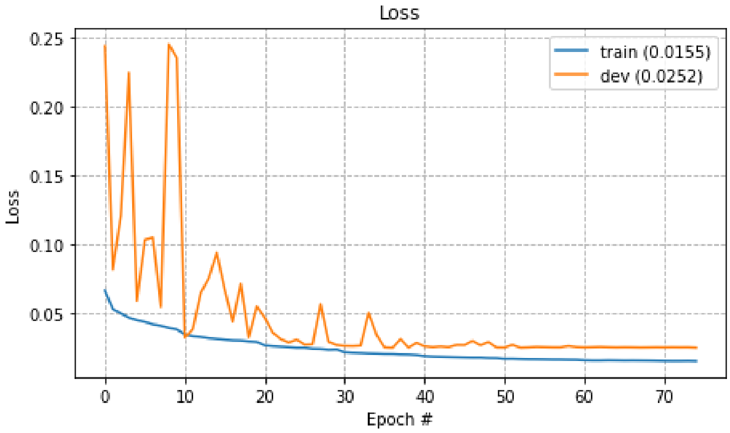


Figure 9. *Prediction vs. milk concentration. The color indicates the absolute prediction error as a percentage.*

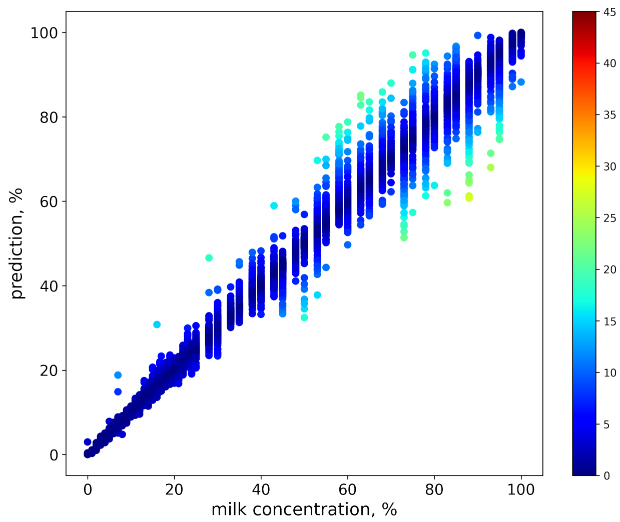
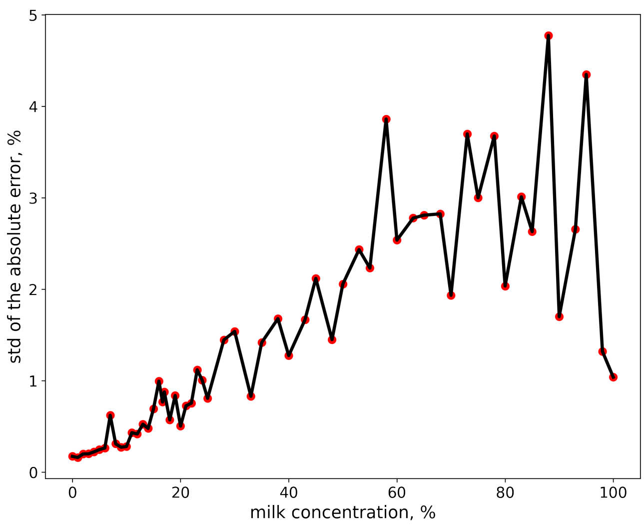
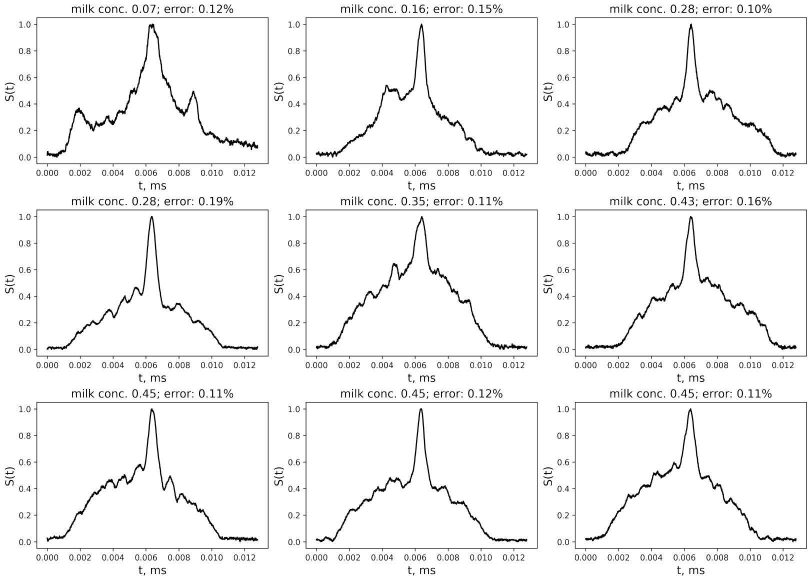


Figure 10. *Standard deviation of the absolute error.*



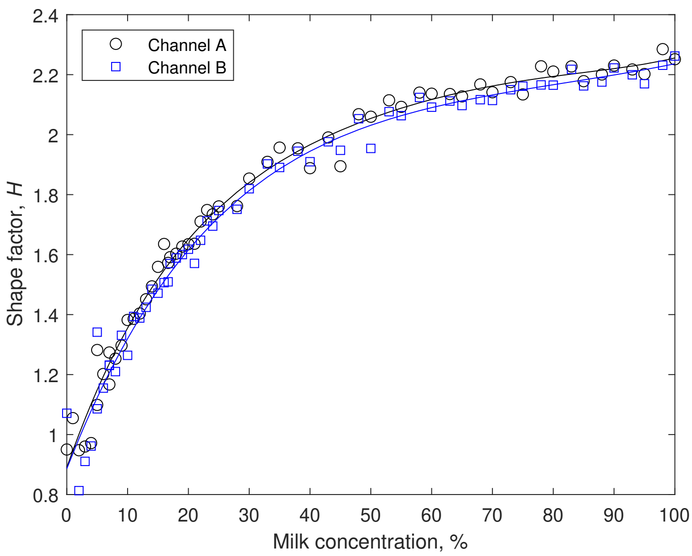
The advantage of the current statistical approach is that it is robust and potentially can be used for

Figure 11. *Signals having significant prediction error.*



complex signals from sprays with variable droplets and particles. Figure 12 shows the shape factor, H, evaluated from our data as a function of the milk concentration. The experimental data points for channels A and B are shown by the markers and the lines show two fifth order polynomials used to fit the data sets. The figure shows that there is a clear functional relationship between the shape factor and the concentration, which can be used as a calibration function for concentration measurement.

Figure 12. *Shape factor versus concentration*



5 Conclusion and future work

In this work, we have demonstrated a method of using light scattering from a single Gaussian beam to detect the concentration of suspension droplets in a spray by calculating the shaping factor of the light scattering signals. To begin with, we used the light scattering signals from the droplet of the same size. Then we applied the method to the measurement with a suspension spray. To get the concentration we don't need to calculate the droplet size and droplet velocity and can only use a single detector. This method needs to be analyzed with other liquids and sprays to get the limits of the performance of this method. We will do that in the next study.

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