Laser sheet drop sizing using Raman and Mie scattering

A Malarski, B Schuerer and A Leipertz
Lehrstuhl für Technische Thermodynamik, Friedrich-Alexander Universität Erlangen-Nürnberg, Am Weichselgarten 8, 91058 Erlangen, Germany
E-mail: malarski@ltt.uni-erlangen.de

Abstract. The knowledge of droplet size distribution is very important in different medical, pharmaceutical and technical applications. Researchers in all these fields are interested in a temporally and spatially resolving two-dimensional measurement technique. A new approach for the two-dimensional determination of the droplet size is the use of two different imaging measurement techniques whose signal intensities have different dependences on the particle size. The combination of laser Raman scattering with Mie scattering is presented here. Whereas Raman scattering intensities are dependent on the measurement volume and Mie scattering on the surface, the ratio of the signal intensities of the inelastically and the elastically scattered light is therefore proportional to the droplet size. The challenge by the application of this combination of measurement techniques, the advantages and the evaluation strategy will be described exemplary shown by measurements of a single droplet chain.

1. Introduction
The use of liquid sprays in gas atmospheres is prevalent in different industrial and everyday applications, for example as injected fuel spray in internal combustion engines, as aerosols in inhalers or during the spray drying in food industries. Mostly the droplet size distribution is a key quality in such systems and not easy to obtain. Non-intrusive measurement techniques based on laser-induced processes were developed within the last decades, e.g., Phase Doppler Anemometry (PDA) as a method for droplet sizes and velocity are commercially available, details are described elsewhere [1]. This method allows the real-time point-wise monitoring of sprays and therefore a statistical evaluation of the whole spray, therefore the mapping of sprays is time consuming. Planar imaging methods like laser sheet drop sizing (LSD) were developed. LSD is based on the measurement of the intensity ratio of laser-induced fluorescence (LIF) and Mie scattering. The dependence of the intensity on the droplet diameter of the Mie scattered light for spherical droplets larger than twice the wavelength is quadratic. For an idealized behavior the fluorescence intensity scales with the cubic of the droplet diameter. Hence, in principle the ratio of fluorescence intensity and Mie intensity is proportional to the surface volume or the Sauter mean diameter (SMD) [2]. However, this holds only for transparent, evenly illuminated particles. As fluorescence is based on light absorption processes this condition cannot be fulfilled. Signal integration over a large number of particles can neglect this effect. Beside this there are other undesirable aspects when using fluorescence as the measurement principle. A fluorescent media or a fluorophor is needed, the signal intensity can be disturbed by quenching effects and the cubic dependence of the signal on the droplet diameter is only valid for a carefully chosen absorption grade and absorption length of the droplets [2].
Raman scattering does not suffer from these problems. As the signal intensity is volume dependent and nearly every medium is Raman active it can preferably be used instead of fluorescence. The major drawback is its weak signal density compared to the Mie intensity. So at first high excitation intensity is needed and secondly efficient filter optic has to be used. The successful simultaneous application of Raman and Mie imaging in a particle laden liquid system was already published by some of the authors [3]. The application of LSD both using fluorescence and Raman scattering has to be calibrated by a standard. Within this paper the calibration strategy for LSD with Raman and Mie scattering is demonstrated using a chain of mono-disperse droplets. Additionally the limitation of the technique is described.

2. Theory
The principle of LSD is the use of two imaging methods based on different scattering types whose intensities have a different functional correlation with the droplet diameter. Mie scattering is elastically scattered light and the Mie theory describes for spherical particles larger than two times the laser wavelength $\lambda$ a quadratic dependence of the signal intensity on the droplet diameter. Thereby the droplet emits not isotropic but a characteristic double spot pattern is visible with high enough resolution [2]. The other measurement technique has to be based on an inelastic scattering because their signal intensities are mostly volume dependent. Due to its large scattering cross section and therefore high signal intensities, LIF is used so far. As the signal wavelength is red shifted according to the excitation wavelength the signal separation can be performed by appropriate band pass filter optics. However LIF is only applicable in fluorescent or tracered media and the laser wavelength has to be set to absorption bands of the fluorescent which are often in the ultraviolet region. Raman scattering on the other hand is applicable in every medium except noble gases and the excitation wavelength is freely eligible. The major drawback is the weak signal intensity compared to other processes. New developments in the detection of an optic field, both by efficient filters and high quantum efficient cameras made the two-dimensional detection of Raman scattering with laser light sheet excitation possible [4]. The signal intensity is dependent on the species molar quantity of the species in the measurement volume and scales therefore with the cubic of the droplet diameter. So the ratio of the Raman to Mie signal intensity should be proportional to the mean diameter of spherical droplets.

However, the application of laser measurement techniques in droplets suffers from morphology dependent resonances (MDR) [5]. These resonances enhance the signal in droplets whose circumference is multiple of the wavelength [6]. The signal intensity dependence therefore becomes unsteady. Additionally, when MDRs of the input and the output wavelength occur in the same droplet, stimulated Raman scattering (SRS) can appear [7]. Differently from SRS in the liquid bulk which is mainly emitted in the direction of the incoming laser beam, it is nearly isotropic in droplets [5, 6]. Whereas MDRs are not avoidable, SRS has to be suppressed. Therefore it is necessary to know the different influences on the SRS threshold, the highest radiant flux density where no nonlinear scattering processes occur. Different authors investigated these influences, and thus only a brief summary is given here. First, the particles size itself influences the threshold level, different connections were reported. Acker et al. [6] mention a preference of SRS in larger droplets, Lin and Campillo [7] measured an increase of the threshold with rising droplet diameter. Second, the Raman line width detected has a great influence, the broader the transition detected the higher the threshold [7]. Additionally the gain factor for SRS itself which is a function of the scattering cross section shows the strong species dependence, too [8]. Third, the quality factor of the cavity that can reach values larger than $10^7$ [6] and can be reduced by two methods. The minimization of the refraction index difference between the droplets and the environment increases the loss of radiation and therefore decreases the quality factor. So for high pressure or near critical applications the appearance of SRS is eliminated [6]. Another method is the use of absorbing dyes whose absorbance in the laser wavelength region is high and in the signal wavelength region low. Furthermore, the dye may not fluoresce at the Raman wavelength [9]. Over all it has to be checked whether the excitation intensity is low enough to
avoid SRS in the given system. Mewes et al. for example used a microsecond pulse laser to provide enough excitation intensity for their Raman signal without exceeding the SRS threshold [10]. Although the absolute droplet size of single droplets is not accessible by this measurement method due to the MDRs, the mean size of a droplet ensemble is available. The necessary requirement is the averaging over a droplet size range with several resonance conditions. Schweiger showed that the Raman signal is then on a higher level but nevertheless proportional to the droplet size [11].

3. Experimental setup

A sketch of the experimental setup is shown in figure 1.

![Figure 1. Scheme of the experimental setup](image)

Both scattering processes have been excited by a pulsed frequency-doubled Nd:YAG laser (532 nm, 10 Hz, 8 ns) formed via lenses to a light sheet with approximately 200 µm thickness in the focus plane. A vertical droplet chain was illuminated in the middle of the light sheet. The scattered light was first separated by a dichroic beam splitter that reflects light within the 532 nm region and allows the Raman shifted light to transmit. The Mie scattered light was attenuated by a neutral density filter with a transmission of 0.01 % to set the signal level to the same magnitude like the Raman signal strength and was separated by a band pass filter with a center wavelength of 532 nm and a full width half maximum (FWHM) of 10 nm. In order to suppress elastically scattered light in the Raman signal path a combination of a high pass filter and a notch filter was used, for details see Malarski et al. [3]. The band pass filter with a center wavelength of 630 nm and a FWHM of 10 nm was set to the CH peak of the carbon hydroxides that emerge with Raman shifts around 2900 cm$^{-1}$. The signals were imaged onto two identical intensified charge coupled device (ICCD) cameras equipped with CCD-chips of 384 x 576 pixels with a single pixel size of 23 x 23 µm. Both cameras were equipped with Nikon® objectives with a focal length of 50 mm and a F-number of 1.2. The averaged laser pulse power was measured discontinuously before and after each measurement series.

Monodisperse droplets were generated by a Microdrop® droplet generator. It consists of glass capillaries with different outlet diameters driven by a piezo. The regulation of the droplet size is done via capillary diameter and pulse width, voltage and frequency of the electrical supply. The size variance is between 5 % for droplets between 70-90 µm and 8% for smaller particles. So it is guaranteed that the calibration is done with an averaging over a proper amount of resonance positions. For the experiments described in this paper droplets of isooctane as a fuel substitute were generated. As the laser light flux density exceeded the SRS threshold, liquid Sudan® M Red 462 (BASF) was
solved in isooctane. This increased the SRS threshold, e.g. with a concentration of 25mg dye per liter isooctane by a factor of nine in comparison to measurements with pure isooctane.

4. Results and discussion

Each measurement series consists of 190 pairs of single-shot images showing 3 to 5 illuminated droplets depending on the adjustment of the droplet generator. For the evaluation a mean background image (measurement without laser excitation) was subtracted and the intensities of the droplet signals within a region of interest (ROI) were added.

First, the concentration of the dye had to be defined, therefore the SRS threshold was determined for different concentrations. For the determination of the threshold the method described by Lin and Campillo [7] was used. The Raman intensity was measured as a function of the laser power and thereby the change from linear to nonlinear functional dependence marks the SRS threshold. The measurement series for a given droplet size is shown in figure 2.

![Figure 2. SRS threshold as a function of dye concentration](image)

The dotted line is a linear fit to the data until the concentration of 25mg/l. It is remarkable that from a certain dye concentration on the SRS threshold does not increase anymore. This is not caused by the higher dye concentration and can be explained by laser induced plasmas whose occur above a laser power of 70 mJ/pulse in the used setup.

A typical scatter plot of the achieved values and the calculated Raman/Mie intensity ratio is shown in figure 3 on the left hand side. As expected the intensities scatter around a mean value. But measurements with the same droplet size and different excitation intensities below the SRS threshold showed a very good reproducibility. This independence of the excitation intensity is a very important condition because the illumination of sprays is non-uniform due to scattering on the droplets and absorption processes. On the right hand side the mean value of the intensity ratio is shown in dependence on the number of evaluated measurements. It shows that 70 measurements will be enough to get the correct value. The mean values of more than 70 measurements show only small deviations.
Figure 3. Plot of the Raman and Mie intensity and their ratio (left) and of the mean intensity ratio as a function of evaluated measurements (right)

The evaluation of the Raman/Mie intensity ratio showed a linear behavior for the investigated droplet sizes. The measurements were made with the Sudan M Red concentration of 25mg/l. The results averaged over 380 to 760 images are shown in figure 4.

Figure 4. Raman/Mie intensity ratio as a function of droplet size

The dotted line shows a linear fit to the data. For a comprehensive calibration, especially for droplet sizes occurring in a fuel spray, the measurements of more droplet sizes are necessary. It has to be emphasized that the droplet size distribution in fuel sprays is in the range of 10-90 µm. So the laser power has to be adjusted carefully according to the change of the SRS threshold with decreasing droplet size.
5. Conclusions
It was shown that the principle of LSD is transferable from the LIF/Mie intensity ratio evaluation to Raman/Mie intensity ratio measurement. The main challenge is the correct adjustment of the excitation intensity. Due to the weak signal strength of Raman scattering a high laser power is desirable, however it is limited by the stimulation of nonlinear scattering processes. One recommended solution is the use of microsecond instead of nanosecond pulse lasers [10] due to the lower peak power at the same excitation energy. Here another method was used instead because of the available laser facility. By use of a dye the quality factor of the cavity in the droplets was reduced and therefore the SRS threshold was increased. A homogenization of the Gaussian intensity profile of the laser beam may further enhance the maximum of the applicable laser power. Additionally it was verified that the averaging over a certain droplet size range gives reproducible data for the intensity ratio. In spray measurements this averaging is done locally instead of the temporal averaging used in the shown experiments. Therefore a single-shot imaging should be possible if the resolution is raw enough. It has to be warranted that several droplets are mapped on each image pixel. If the resolution should be higher image averaging has to be executed and the temporal information will be lost. Apparently a compromise between temporal and local resolution has to be found.

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