Increase of the threshold for stimulated Raman scattering by a spectral broadening of the excitation source

R. F. Hankel, Friedrich-Alexander University of Erlangen-Nuremberg, Germany (June 2012)

Application Note

Introduction

The knowledge of droplet sizes in spray applications is a crucial parameter. To name some examples the evaporation behavior in a fuel spray or the inhalation of an asthma spray depends on the droplet sizes and even more important on the droplet size distribution in the investigated spray. So there are some reasons to gain information about the spray parameters. Moreover the measurements have to be non-invasive in order not to influence the spray parameters by the measurement itself. This leads to an optical measurement technique which in comparison with other techniques does not influence the fluid. In the field of optical metrology there are various possible techniques that can be used for drop sizing. For the current project we looked for a measuring technique which can also be applied to dense sprays. In this case the choice was laser sheet drop sizing (LSDS). This technique is based on the detection of two different signals simultaneously. The first signal scales with the surface of the droplet and is called Mie-scattering. It is elastic scattering, so the signal appears at the same wavelength as the incident photons. The signal intensity is relatively strong, so it can be detected with a common CCD detector. The second signal should be proportional to the droplet volume. If you detect both signals just from one laser shot the ratio of the two signals scales with the droplet diameter and is, after a calibration, a way to determine the size of the droplets. There are two different ways to generate a volume dependent signal. The first one is laser induced fluorescence (LIF) which exhibits a strong signal level. The second one is Raman scattering, an inelastically scattered signal with a low scattering cross section resulting in a weak signal level. Unfortunately, LIF has some drawbacks. Since not every molecule exhibits fluorescence signals, very frequently there is the need of adding tracer molecules to the spray which might show some misleading results. Tracer and spray molecules have different properties in volatility which makes a calibration in a reacting spray elaborate. Moreover, fluorescence signals are strongly quenched as soon as you work in an oxygen atmosphere. This leads to very time consuming calibration steps since the quenching is strongly influenced by temperature and pressure changes. These drawbacks are reason enough to choose Raman scattering as the volume dependent signal. Since almost every molecule is Raman-active no tracer molecules are needed. Furthermore, there is no absorption during the Raman process which makes the signal independent of quenching effects. Since the already mentioned low scattering cross section has to be overcome the application of a detector with high quantum efficiency at the measured wavelength is beneficial.

A way to increase the Raman signal level is simply to increase the amount of incident photons, so the intensity of the excitation source. Unfortunately, the intensity raise is limited since at a certain threshold, known as threshold for stimulated Raman scattering (SRS), the droplets itself can act as a cavity for the laser and Raman wavelengths and increase the signal exponentially. These enhanced signals cannot be used for LSDS anymore, since the proportionality to the droplet volume is not valid anymore. Another point is the need of a high frame rate since a spray is a highly turbulent process. The application of an EMCCD detector is highly reasonable in this case. For the subsequently described measurements an EMCCD spectroscopy detector (Andor Newton DU971N-BV) with an array of 1600 x 400 pixels and a pixel size of 16 x 16 μm² was used. The camera's electron multiplication (EM) register allows the detection of ultra-weak signals with a comparably high signal-to-noise ratio at a high frame rate. The quantum efficiency in the relevant wavelength range is higher than 90%.

Experimental setup

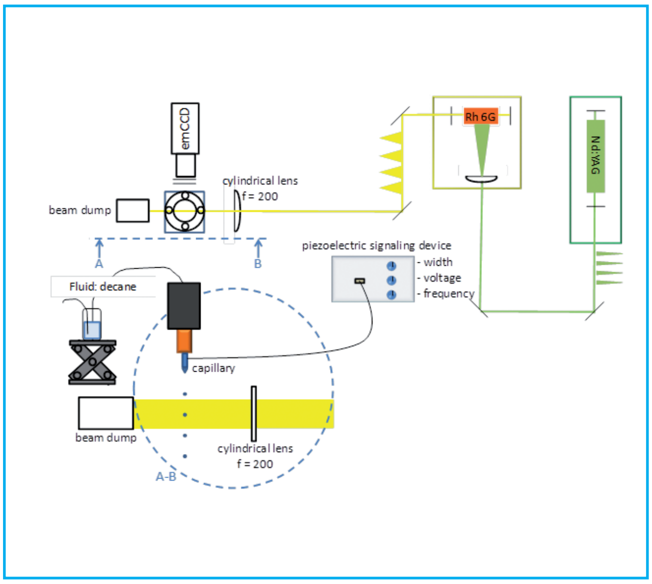

Figure 1: Experimental setup

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For the illumination of the vertical droplet chain (fluid: decane) either a frequency doubled pulsed Nd:YAG laser at 532 nm or a Rhodamin 6G dye resonator (shown in figure 1) was used. The Nd:YAG emits pulses of 10 ns (FWHM) pulse duration at a repetition rate of 10 Hz and a single-pulse energy with a maximum of 350 mJ at 532 nm (FWHM=0.05 nm). For the experiments with the dye resonator the Nd:YAG laser was used to pump the dye what resulted in a central wavelength of 566 nm (FWHM=4 nm) for the dye emission. Both emissions were formed to a light sheet via a cylindrical lens with a focal length of 200 mm resulting in a light sheet thickness of approximately 200 µm in the focal plane. For both lasers the Raman-Stokes signals from a vertical droplet chain were detected using the EMCCD detector equipped with a Nikon objective with a focal length of 50 mm and an F-number of 1.2. In comparison to former experiments the detector was used for imaging purposes and not for spectroscopy. Adverse in this case was the absence of a shutter device in front of the chip, since an exposure of the chip from computer screens and lab control lamps after the measurement was not avoidable and leaded to smearing effects on the resulting image. To increase the solid angle of detection two spacer rings with 50 mm length were put in between the camera chip and the camera lens. With the goal to achieve fast read-out the pixels were binned 2x2. The exposure time of the camera was 10 µs and the sensor was cooled down to -85 °C to reduce dark current.

In order just to detect the inelastically scattered light the camera lens was equipped with several long pass and band pass filters with a high transmission in the desired Raman wavelength regions.

Results

Aim of the present work was to increase the Raman signal intensity but still having the volume dependent signal from the droplets in order to use the resulting signal for LSDS. The approach of using a broadband excitation source instead of a narrowband Nd:YAG laser was tested to achieve that goal. The following figures 2 and 3 show the results of the realized experiments.

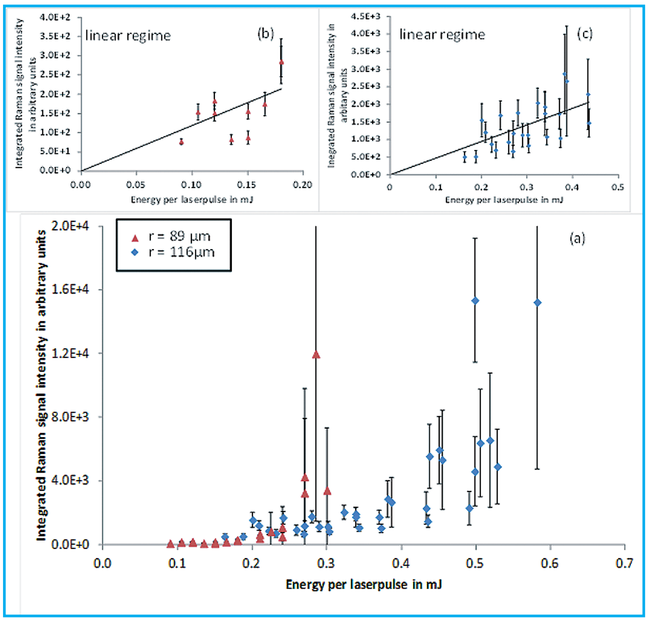

Figure 2: Integrated Raman signal intensity as a function of the excitation energy for the narrowband excitation with a central wavelength of 532 nm and a FWHM of 0.05 nm

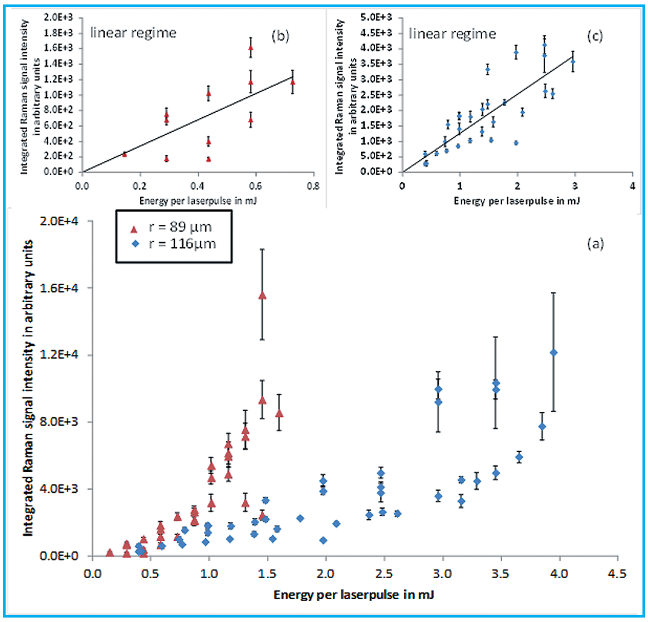

Figure 3: Integrated Raman signal intensity as a function of the excitation energy for the broadband excitation with a central wavelength of 566 nm and a FWHM of 4 nm

Each data point in the diagrams consists of at least 400 single shot measurements. The error bars were formed as standard deviations from the mean values.

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For each excitation source two different droplet sizes were investigated, namely droplets in the size of 89 µm and 116 µm, respectively. The smaller diagrams (b) and (c) in the figures 2 and 3 show details of the corresponding diagram (a), where (b) shows the results for the smaller droplets and (c) shows the results for the bigger droplets. It is highly remarkable in that context that the broadband excitation source leads to an extended linear regime for both droplet sizes since the probability of stimulated effects inside the droplets is reduced significantly.

As soon there was no signal detection possible anymore in the conventional mode of the detector, the EM register was used to amplify the signal to noise ratio. For that reason the EM gain of the detector was adapted to different values depending on the excitation energy. To get an idea of the amplification effect of the register we calibrated the register with constant excitation energy and constant droplet size. The resulting curve is shown in figure 4. For the signal detection only a small region of the multiplication register curve was found to be capable since a small gain factor results in no amplification of the signal to noise ratio and a high gain factor leads to a saturation of the pixels what significantly reduces the dynamic range of the detector.

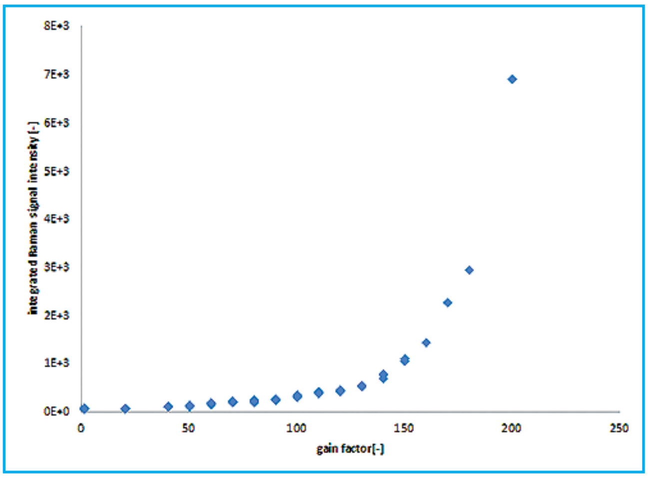

Figure 4: Raman signal intensity as a function of the gain factor of the multiplication register for a droplet size of r=89 µm and pulse energy of 60 µJ

Finally, it can be concluded that pumping of micro droplets with a broadband excitation source leads to an extended region, where the Raman signal intensity is linear with respect to the laser's excitation energy.

This result attenuates the problem of the low scattering cross section of Raman scattering and hence allows the detection of a higher signal to noise ratio for LSDS. The EMCCD detector allows the detection of ultra-low signals with the help of its multiplication register, when signals cannot be detected in the conventional mode anymore.

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