

LASER-BASED SPRAY DIAGNOSTICS IN COMMERCIAL FUELS USING A NEW ALL SOLID-STATE LASER SYSTEM

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ABSTRACT

We present a new approach for spray diagnostics in practical fuel sprays without the need of using model fuels. Commercial fuels for internal combustion engines have large absorption cross-sections in the UV and the blue. This absorption hinders measurements and yields fluorescence that is difficult to quantify and interferes with fluorescence from conventional tracers. Therefore, tracers have been chosen that allow excitation in the red where the fuels are transparent. A new all-solid-state Nd:YAlO₃ laser system was developed that emits pulse bursts at 671 nm. The individual laser pulses have lengths of ~30 ns and a minimal temporal delay of 30 µs and are therefore suited for time-resolved measurements of spray formation, transport and evaporation. In this work we characterize two possible candidates for LIF tracers: Rhodamine 800 and Atto 680 in terms of relative fluorescence quantum yield and absorption cross-section in a Diesel environment as well as temperature dependence of the LIF intensity. First results of laser-sheet dropsizing experiments with Rhodamine 800 as a fluorescence tracer in a model Diesel spray are presented.

INTRODUCTION

Laser-sheet dropsizing has become a common non-intrusive method to determine two-dimensional droplet size distributions in practical sprays [1]. It provides high temporal and spatial resolution and has been applied to many different spray systems [2-4]. Its imaging capability allows the fast measurement of average droplet sizes within extended cross-sections through the spray. This is especially important in environments where the optical access is rapidly blocked by window fouling or where transient phenomena are of interest. It can furthermore be applied to dense sprays where conventional techniques like Phase-Doppler measurements fail because of the high density of particles. A thin laser light sheet illuminates the spray and fluorescence and scattering signals from the spray are detected simultaneously. The Mie-scattering signal I_{Mie} is assumed to be proportional to the local mean droplet surface $\langle d^2 \rangle$, the LIF signal I_{LIF} is proportional to the local mean volume $\langle d^3 \rangle$ of the droplets with diameter d . The ratio of these two signals is used to determine the local Sauter Mean Diameter (SMD)

$$SMD = \frac{1}{N} \cdot \sum_{i=1}^N d_i^3 \bigg/ \frac{1}{N} \cdot \sum_{i=1}^N d_i^2 = k_{SMD} \cdot \frac{I_{LIF}}{I_{Mie}} \quad (1)$$

The observation of sprays in internal combustion engines is of particular interest because the exact knowledge of spray breakup and evaporation is crucial for the optimization of the combustion process. Commercial fuels contain a variety of fluorescing compounds. Sazhin et al. have studied the spectral properties of Diesel fuel [5] and have found large absorption cross-sections for near-UV wavelengths. This absorption leads to strong fluorescence that can be measured to qualitatively visualize the liquid fuel distribution. The resulting fluorescence signal, however, can usually not be quantified because it originates from a large number of different species that have individual dependencies on temperature, pressure and collisional quenching and that can be inhomogeneously distributed because of variation in their evaporation and transport properties. In order to overcome these limitations organic molecules with well-characterized spectroscopic properties are typically added to the fuel as fluorescent tracers.

Usually tracers are preferred that can be excited within a wavelength range where the fuel is transparent, in order to avoid laser attenuation, energy transfer and interfering fluorescence signals. Commercial fuels have low absorption cross-sections in a wavelength range starting at ~600 nm and reaching into the near infrared. Therefore, a novel laser light source was developed to access this spectral range. It operates at 671 nm and allows LIF-measurements in sprays of commercial fuels. Possible fluorescent tracers that can be excited and selectively detected within at this wavelength have been identified and evaluated.

BACKGROUND

Laser dyes are commonly used as fluorescent tracers [6]. They usually have high fluorescence quantum yields and are available for excitation throughout the visible spectrum, reaching into the UV and NIR. They have negligible vapor pressure and usually decompose before boiling. It is commonly assumed, that for slow evaporation the dye accumulates completely inside the droplet during the evaporation process [7] thus giving a measure of the initial spray volume. No signal from the gas phase is expected. As a representative of this group Rhodamine 800 [8] has been analyzed in this work.

Fluorescent dyes and markers used in bioscience are optimized for quantum yield to enable the LIF detection of single molecules [9]. Fluorescence markers have been developed that can be excited at wavelengths > 600 nm. These efficient fluorescence tracers, therefore, became possible candidates for spray diagnostics in practical fuels as well. As a representative of this group Atto 680 (ATTO-TEC, Siegen, Germany) has been analyzed in this work.

At moderate tracer concentrations ($< 10^{-5}$ mol/l) the fluorescence intensity is

$$I_{LIF} \propto c \sigma \frac{A_{21}}{Q_0 m \exp(-E_a/kT)} \quad (2)$$

where c is the tracer concentration, σ the absorption cross-section, A_{21} the spontaneous emission rate, and $Q_0 m \exp(-E_a/kT)$ the temperature-dependent quenching caused by the quencher at concentration m [10]. E_a is the activation energy of the quenching process.

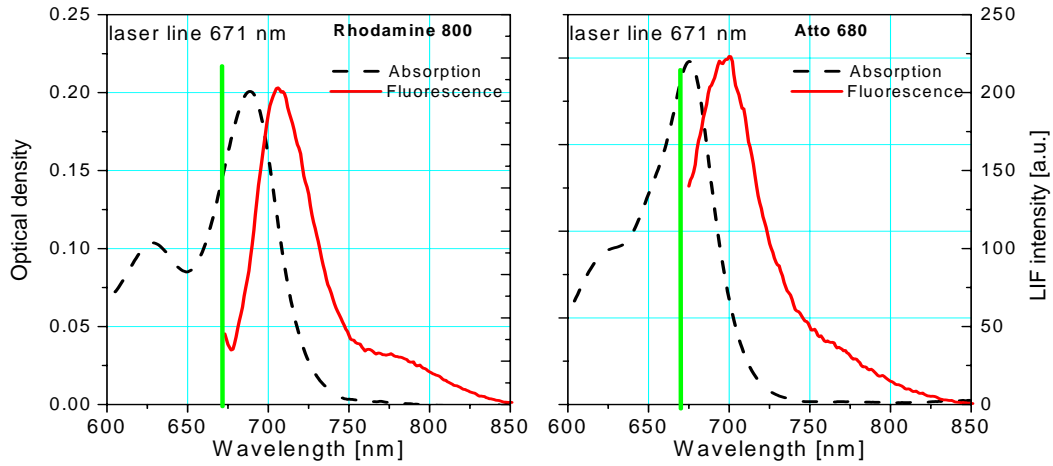


Figure 1: Absorption and fluorescence spectra of Rhodamine 800 and Atto 680 solved in commercial Diesel fuel.

Fig. 1 shows absorption and emission spectra of the two dyes dissolved in commercial Diesel fuel. Both tracers are hydrophilic and therefore solve poorly in Diesel, which consists primarily of unpolar hydrocarbons. Therefore the dyes were first solved in hexanol and then a small amount of the hexanol/dye solution was mixed to the Diesel.

LASER SYSTEM

A compact all-solid-state laser has been developed in order to allow high-repetition-rate imaging of LIF from the tracers dissolved in the liquid phase. The laser system has been designed to meet the requirements of the fluorescence experiments such as an emission wavelength close to 671 nm, a pulse energy greater than 1 mJ and the ability of generating pulse bursts with a minimum time interval between the pulses around 30 μ s (corresponding to a repetition rate of about 33 kHz) [11]. For the observation of spray formation or evaporation no more than 5 – 10 laser pulses are required. Therefore, the laser is optimized to generate pulse bursts with a total duration no longer than 300 μ s. Therefore, quasi-continuous diode lasers can be used for laser pumping, which strongly reduces cooling requirements and costs. It was decided to use the $^4F_{3/2} - ^4I_{13/2}$ laser transition of the Neodymium ion, that provides an emission wavelength at ~ 1340 nm which is then frequency-doubled to 671 nm. The advantages to the competing concept of using an optical parametric oscillator, which is pumped by the frequency doubled, green beam of a Nd-laser emitting on the strongest transition $^4F_{3/2} - ^4I_{11/2}$, can be summarized by a higher expected overall efficiency, less complexity, and a higher reliability.

The laser configuration consists of an acousto-optically Q-switched oscillator and a 5-pass amplifier (Fig. 2), each of which is transversally pumped by two quasi-cw AlGaAs-diode laser stacks. Each of the linear array bars is equipped with an aspherical cylindrical microlens that collimates the fast axis of the diode laser beam. The pump radiation

emitted by the diode laser stacks is focused by a pair of cylindrical lenses into the crystals of the oscillator and the amplifier, generating a nearly 500- μm thick horizontal layer of inversion in the center planes of the crystals.

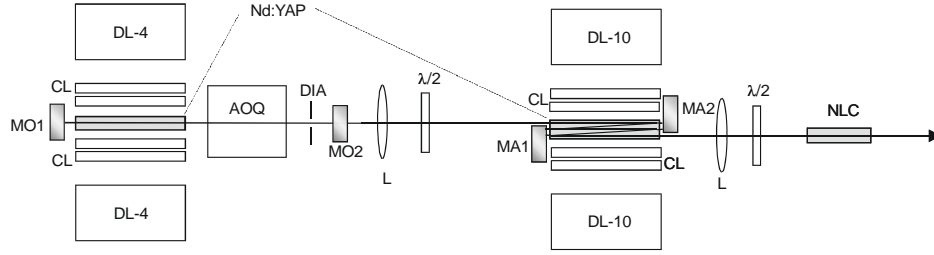


Figure 2: Set-up of the laser system. MO1,2: mirrors of oscillator; MA1,2: mirrors of amplifier; DL-4,-10: qcw diode laser stacks of oscillator and amplifier (resp.); CL: cylindrical lenses; AOQ: acousto-optical Q-switch; DIA: diaphragm; L: lens; $\lambda/2$: retardation plate; NLC: nonlinear crystal.

The active material selected for the laser is Nd:YAlO₃ (Nd:YAP, Yttrium-Aluminum Perovskite). This crystal is characterized by a large laser cross-section of $1.2 \times 10^{-19} \text{ cm}^2$ at an emission wavelength of 1341 nm, naturally polarized radiation, and high optical and mechanical quality.

The stable resonator of the laser oscillator is formed by a flat, highly-reflecting mirror and a concave outcoupling mirror with 1 m radius of curvature and a transmission of 30% at the laser wavelength. An electronic circuit generates the delay and the steering pulses for the acousto-optical Q-switch, by which stable laser pulse bursts are produced with uniform, equal amplitudes during a pumping period of 250 μs . Under operating conditions with pulse-to-pulse intervals of 33 μs the energy of a single pulse is 0.44 mJ, while the pulse width is 30 ns. For optimum coupling of the oscillator beam to the amplifier an $f = 500$ mm lens and a $\lambda/2$ retardation plate are used. A 5-fold path in the amplifier crystal is generated by means of two plane mirrors. At a maximum applied pump current of 110 A an amplification of nearly 10 has been achieved which resulted in an energy for a single pulse of 4.4 mJ. A beam quality factor M^2 of 1.2 was measured, which represents an optimum prerequisite to achieve high conversion efficiency in the subsequent frequency-doubling process. The amplified laser beam is focused into a 10 mm long KTP crystal by an $f = 250$ mm lens and a $\lambda/2$ retardation plate. A maximum pulse energy of up to 1.3 mJ has been generated at a wavelength of 671 nm. Due to the phase-matching properties of the nonlinear crystal the red output beam has a slight ellipticity of about 1:2.5, which is advantageous with respect to generating a light sheet for imaging experiments.

SPRAY EXPERIMENT

Absorption spectra of different tracer-doped fuel samples were taken with a *Varian CARY 500 Scan* spectrometer and temperature dependent LIF emission spectra were measured with a *Varian CARY Eclipse* fluorescence spectrometer. For the fluorescence spectra the temperature of the sample could be varied from 15 – 60°C.

Two-dimensional LIF images of a model spray have been recorded with the experimental setup shown in Fig. 3. For these experiments the Nd:YAlO₃ laser system was applied. The tracer concentrations were $2 \times 10^{-6} \text{ mol/l}$ Rhodamine 800 and $2 \times 10^{-6} \text{ mol/l}$ Atto 680, respectively in Diesel with 5% hexanol. A standard medical nebulizer was used to generate a homogenous spray with droplets of an average diameter of $\sim 8.5 \mu\text{m}$. The laser beam was formed into a thin light sheet ($d \approx 0.5 \text{ mm}$) by a cylindrical lens ($f = 750 \text{ mm}$). The LIF signal was imaged onto an ICCD camera (LaVision, *Streak Star*) by a Nikon $f = 105 \text{ mm}$, $f_{\#} = 2$ macro lens. Scattered laser light was suppressed by a long-pass edge filter (*Omega Optical XF3095 ALP 700*). The Mie signal was simultaneously imaged in the same way onto a second ICCD camera.

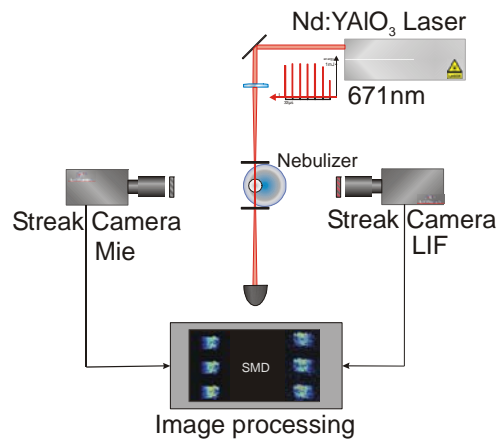


Figure 3: Experimental setup for planar LIF and simultaneous Mie scattering measurements.

RESULTS

The temperature-dependence of the fluorescence of both dyes was quantified by heating a sample of Diesel mixed with 5% hexanol and 2×10^{-6} mol/l Rhodamine 800 and 2×10^{-6} mol/l Atto 680, respectively, and taking fluorescence spectra at temperatures between 15 and 60°C. Fig. 4 shows the normalized integrated fluorescence intensity at different temperatures.

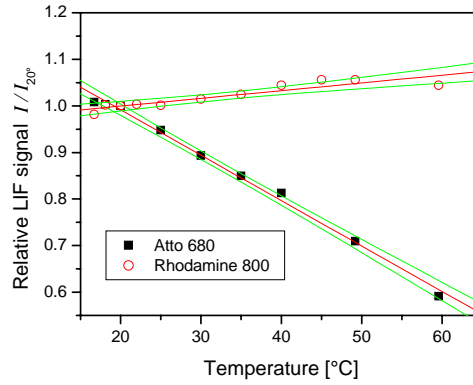


Figure 4: Temperature dependence of the integrated fluorescence intensity. The intensity is normalized to its value at $T = 20^\circ\text{C}$.

Atto 680 fluorescence strongly decreases with increasing temperature. After taking the last spectrum at 60°C the Atto 680 sample was cooled to 22°C, the fluorescence intensity increased again but did not reach its original value. This leads to the assumption that the Atto 680 molecules decompose at higher temperatures or the solution changes its structure (i.e. separation of tracer and fuel). The fluorescence of Rhodamine 800 showed little variation with increasing temperature. No indications for decomposing of Rhodamine 800 or deterioration of the solution were found. Both dyes have been tested for fluorescence quenching by oxygen [7] by purging the samples, first with nitrogen, then with oxygen. Fluorescence spectra were taken before and after the purging with oxygen no significant changes in the intensity were found. This leads to the assumption that Rhodamine 800 is the better choice as a fluorescence tracer for spray experiments, its absorptivity and fluorescence intensity are linear in the relevant range of concentrations, the temperature-dependence of its fluorescence quantum yield is negligible within the analyzed temperature range (15 – 60°C) and its fluorescence is not quenched by oxygen.

First experiments with LIF and Mie-scattering in an actual Diesel-spray have been performed with the described novel frequency-doubled Nd:YAlO₃ laser system. Fig. 5 shows typical single-shot Rhodamine-LIF and Mie-scattering images of the spray generated by a medical nebulizer. The droplet velocity as well as the evaporation rate in the observed spray were too slow to visibly change the signal on the observed timescale ($\Delta t = 66 \mu\text{s}$). The LIF signal is separated from the scattered laser light by a long pass filter with a total transmission of <1% for wavelengths < 700 nm. The LIF images were corrected for remaining scattered light and fluorescence background from the Diesel fuel. Fig. 5 shows a signal proportional to the local SMD in the nebulizer-spray, obtained by simultaneous measurement of LIF and Mie signal. The signal was averaged over 200 single shots. The images were corrected for spatial distortions caused by the camera optics and the image intensifier and were carefully superimposed. Phase-Doppler measurements in the same spray at three different positions were used to calibrate the SMD signal. The relative variation in droplet sizes measured by the Phase-Doppler technique across the cross-section through the spray matches well the results of the laser-sheet dropsizing measurement.

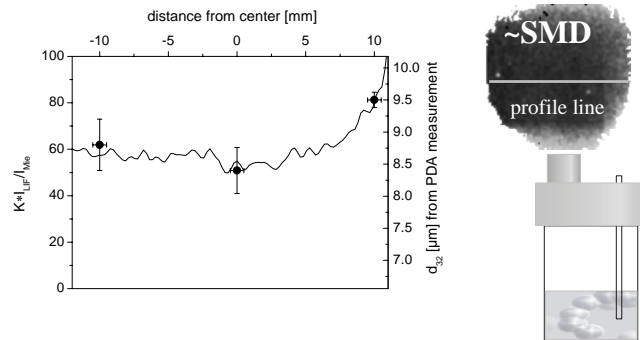


Figure 5: Profile of average droplet sizes (SMD) extracted from the laser-sheet dropsizing measurement in the Rhodamine 800-doped Diesel spray generated by a medical nebulizer. The left plot shows a cross-section through the relative SMD field at ~ 10 mm downstream of the nozzle. The dots represent droplet size measurements with a Phase Doppler Anemometer. The error bars indicate the statistical uncertainty in the droplet diameter PDA measurement. The size of the observed field in the imaging measurements (right) is $20 \times 20 \text{ mm}^2$. The position of the profile is marked.

The analyzed spray showed no evaporation within the observation time of 132 μs . However, spray development and respectively droplet movement and velocity can be visualized by comparing the scattered light signal from different pulses within one burst. Figure 6 shows an example. During the image-processing procedure both frames are corrected for background and spatial distortions, then the frame recorded at t_0 (first laser pulse in burst) was subtracted from the frame recorded at $t_0 + 132 \mu\text{s}$ (last laser pulse in burst). A constant value was added to the result.

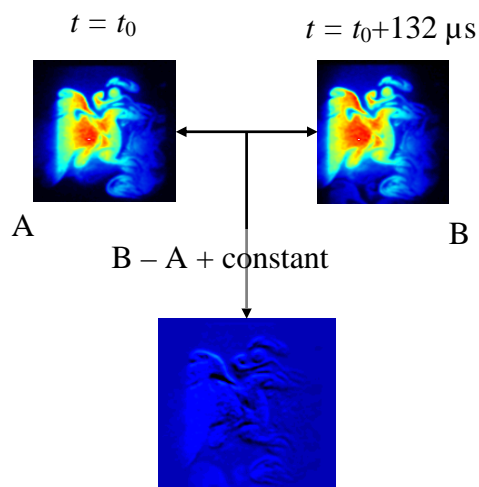


Figure 6 Spray motion analysis.

The lighter areas in the resulting image show where droplets are moving to, darker areas show where droplets move away. The model spray investigated here shows low droplet velocities ($\sim 5 \text{ m/s}$). Therefore, no large velocity variations are visible except on the borders of the spray.

SUMMARY AND CONCLUSIONS

Two different fluorescent dyes, Rhodamine 800 and Atto 680 have been characterized and evaluated in respect to their applicability as fluorescence tracers in fuel sprays with excitation with a new all solid-state pulse-burst laser source in the red. Absorption and emission spectra were taken from both tracers at different concentrations. Atto 680 showed an overall higher fluorescence quantum yield, but also showed a non-linear dependence of the tracer concentration that is not yet fully understood.

The temperature-dependence of the fluorescence intensity of both tracers was analyzed. Atto 680 showed a decrease in fluorescence intensity with increasing temperature by $\sim 1\%/K$, whereas Rhodamine 800 is temperature-independent in the temperature range from 16-60°C.

In spite of its low quantum yield compared to Atto 680, Rhodamine 800 shows several properties a fluorescence tracer for LIF applications in Diesel fuel should have: Its absorptivity and fluorescence intensity are linear in the relevant range of concentrations, the temperature-dependence of its fluorescence quantum yield is negligible within the analyzed temperature range and its fluorescence is not quenched by oxygen. Atto 680, however, shows a strong decrease in quantum yield with increasing temperature. Considering also the high costs and low availability of the fluorescence marker Atto 680, Rhodamine 800 seems to be the better choice – as long as its LIF-signal is strong enough for the specific application.

A new laser system was developed in order to meet the requirements for LIF/Mie-dropsizing experiments in practical fuel sprays using the tested fluorescence tracers. The diode-pumped Nd:YAlO₃ laser system produces bursts of 5 – 10 short pulses at 671 nm with time intervals of 33 μs between the pulses and is therefore capable of resolving spray formation, transport and evaporation processes in a fuel injection spray.

Two-dimensional LIF images of a Diesel spray generated by a medical nebulizer have been taken, using both dyes as fluorescence tracers. With both tracers the selective detection of the tracer is possible in practical sprays of commercial Diesel fuel. First qualitative dropsizing experiments in Diesel-sprays containing Rhodamine 800 as a fluorescence tracer using the new Nd:YAlO₃ laser system have shown promising results. In future experiments in Diesel sprays in hot environments both tracers will be tested further for applicability in LIF. With simultaneous imaging of the scattered laser-light, quantitative LIF/Mie-dropsizing in practical Diesel injection sprays should be possible.

Furthermore, other possibilities will have to be evaluated, to improve the solubility of the discussed tracers in unipolar fuels. The use of detergents or the modification of the tracer molecules are promising techniques in that direction.

ACKNOWLEDGEMENTS

The financial support by the Deutsche Forschungsgemeinschaft within SFB359, by the German Federal Ministry of Research, contract 13N7184/3, and by the DaimlerChrysler AG, contract F20513, is gratefully acknowledged.

NOMENCLATURE

A_{21}	spontaneous emission rate (s^{-1})
c	tracer concentration ($mol\ l^{-1}$)
d	droplet diameter (m)
E_a	activation energy of the quenching process (J)
I	Signal intensity (photon counts)
k	Boltzmann constant ($=1.3806503 \times 10^{-23}\ m^2\ kg\ s^{-2}\ K^{-1}$)
k_{SMD}	constant calibration factor (μm)
m	quencher concentration ($mol\ l^{-1}$)
N	total number of droplets (dimensionless)
Q_0	specific quenching rate (s^{-1})
SMD	Sauter Mean diameter (μm)
t	time (μs)
T	temperature (K)
λ	wavelength (nm)
σ	absorption cross-section (cm^2)

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