TIME INTEGRATED DETECTION AND APPLICATIONS OF FS-LASERPULSES SCATTERED BY SMALL DROPLETS

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Abstract  A characteristic of femtosecond pulse scattering on small particles is the temporal separation of the pulses, which is due to the different optical path lengths through the particle. Therefore the particle converts the incident pulse in a train of pulses, with the pulse heights and temporal separation depending on particle size, refractive index and observation angle. Because of the temporal pulse separation, signal parts from different scattering orders are no longer coherent. Therefore interference structures, which are in the case of continuous illumination the cause of scattering lobes, disappear. The interference inside a single scattering order, as for the rainbow, exists as long as the pulses from different paths overlap. The detection does not require high temporal resolution but is time integrated. This offers the possibility to improve or extend techniques which are disturbed by additional scattering orders, as the rainbow technique for small particles. A second characteristic of femtosecond pulses is the broad electromagnetic spectrum of the illumination. For continuous laser illumination with one spectral line, a spherical particle generates optical resonances for various wavelength-diameter ratios (morphology dependent resonances, MDRs). These optical resonances increase the optical cross-section of the particle and generate strong oscillations in the diameter-intensity relation. Because the power of a pulse is distributed over several spectral lines, which generally do not fulfill the resonant conditions at the same time, the influence of optical resonances is suppressed strongly. We demonstrate an electrodynamic trap with a novel geometry of electrodes making it possible to follow the evaporation of water droplets over seconds and prove the monotonic nature of the recorded intensity as a function of diameter. The illumination with femtosecond laser pulses forces a monotonous relation between intensity and diameter without any ambiguity. This opens the way for particle sizing using the scattered intensity.

1. Laser pulse and continuous wave scattering on single droplets

Illuminating single droplets with electromagnetic waves yields an angular intensity distribution depending on frequency, refractive index and coherence length. Investigating this interdependence for microscopic scale droplets leads to new possibilities in particle characterization. A spatially extended droplet with corresponding size and refractive index will generate multiple scattering orders when interacting with electromagnetic waves. Incident waves are partly reflected by, and partly refracted into the droplet. Internal waves split further because of ongoing reflection and refraction. If the droplet is imaged, different scattering orders are observed on the sphere’s surface as a juxtaposition of point sources. In the special case of an incoming plane wave the Lorenz-Mie Theory yields the total scattered field, which can be expanded into Debye-series. Individual complex terms of the series are interpreted as individual scattering orders (Albrecht et al. 2003). In analogy to summing up the contributions of individual complex terms, the angular intensity distribution in the far field of the droplet is a result of interference: Electromagnetic field vectors of individual scattering orders add up (Fig. 1-a). The assumption of a monochromatic,
incident plane wave is valid for continuous wave (CW) lasers or for pulsed lasers with pulse lengths far larger than the diameter of a droplet. Fourier decomposition can be used to expand the scope of the Lorenz-Mie Theory to inhomogeneous waves scattered from a spherical, homogeneous, isotropic droplet. In the special case of ultrashort laser pulses temporal effects become important, because the laser pulse length is in the order of microns (a 10fsec pulse corresponds to a 3µm pulse length). For laser pulses far shorter than the diameter of the scattering object this results in temporally separated scattering orders described by the Debye-expansion (Bech and Leder 2004; Damaschke et al. 2002; Mees et al. 2001).

Because the coherence length corresponds to the pulse length, no far field interference takes place between the individual scattering orders. In this case the angular intensity distribution of the droplet is not a sum of electromagnetic field vectors, but of the intensities of individual scattering orders detected at specific scattering angles (Fig. 1-b).

2. The Rainbow

The term rainbow is used to describe angular scattering regions in which more than one solution exists for a single scattering order. The second-order refraction in water droplets forms the well-known rainbow observed in nature. The interference between two different partial rays of the second-order refraction results in intensity maxima and minima in the angular intensity distribution. The angular distances between the maxima contain information about the diameter of a spherical, homogeneous droplet. Furthermore, the absolute angular position of the maxima is a function of the refractive index, which can be used to quantify the droplet temperature (van Beeck 1997).

The angular intensity distribution of the second-order refraction forms the rainbow due to different path lengths for its two geometric solutions which interfere with each other in the far-field. Unfortunately far field interference with the surface reflexion and higher scattering orders is superimposed on the rainbow structure, obscuring the second-order intensity distribution.
(Damaschke et al. 1998). For droplets with a diameter smaller than 20\(\mu\)m the so-called ripple-structure prevents accurate particle characterization other than under precisely controlled laboratory conditions (Onofri 2004). Numerical simulation indicates an adequate suppression of the ripple-structure if similar droplets are illuminated with femtosecond laser pulses (Fig 2-a,b). The pulse length can not be chosen arbitrarily, because a coherence length of the order of microns is necessary to extract information with the Rainbow method. Eliminating coherence completely, for example by using a white light source, would remove the intensity modulation resulting from the two-ray-solution of the second-order refraction. Numerical simulation indicates an adequate suppression of the ripple-structure if similar droplets are illuminated with femtosecond laser pulses (Fig 2-a,b). The pulse length can not be chosen arbitrarily, because a coherence length of the order of microns is necessary to extract information with the Rainbow method. Eliminating coherence completely, for example by using a white light source, would remove the intensity modulation resulting from the two-ray-solution of the second-order refraction. For experimental verification monodisperse water droplets (Brenn et al. 1996) were created with a piezoelectric droplet generator (Bakic et al. 2006). The surpression of the ripple-structure observed experimentally follows closely the numerical predictions (Fig. 2-c,d). Due to the costly generation of femtosecond laser pulses, an experimental alternative to shorten the coherence length sufficiently in CW-mode while preserving an acceptable laser intensity was proposed and verified (Peil et al. 2006).

The numerically obtained data in Fig. 3 clarifies the surpression of interference structures in the scattering function for time integrated detection by showing the angular distribution of local maxima of the scattering function for a spherical water droplet illuminated with \(t_p = 50\text{fs}\) laser pulses and for diameters up to \(d_p = 50\mu\text{m}\) (\(n = 1.333\)). The angle \(\alpha_s = 0^\circ\) represents the direction of the incoming laser beam and \(\alpha_s = 180^\circ\) is therefore the backscatter direction. For continuous wave illumination the diagram would display a large number of additional maxima due to interference between different scattering orders and also due to morphology dependent resonances. With fs-laser pulses the scattering function is however smoothed, even for droplet diameters down to approximately \(d_p = 5\ \mu\text{m}\) at certain scattering angles. Clearly visible are the positions of the primary and secondary rainbow and the

![Fig. 2: a,b) Numerically obtained angular intensity distribution in the far field of a 94\(\mu\text{m}\) droplet illuminated by a continuous source and 200fs laser pulses. Pulsed illumination suppresses the ripple-structure. c,d) The corresponding measured angular intensity distribution confirms previous calculations.](image)
change in angular position of the supernumerary bows due to different diameters. It is of extraordinary importance for rainbow refractometry, that the first and second supernumerary bows remain visible. Their exact position can be used as the basis for in-situ refractive index measurement.

Ultrashort pulse lengths correspond to an extremely wide spectral bandwidth. Propagation of a pulse through a transparent but dispersive medium therefore results among others in a duration broadening of the pulse and a frequency chirp. These effects scale with the traveled distance in the medium and are to be considered and corrected for experimental setups including lenses. For small particle characterization the necessary pulse length declines together with the distance traveled within the medium. For a \( d_p = 5 \mu m \) particle the traveled distance is only a few times the wavelength and is therefore neglected. The separation of reflection and second-order refraction and therefore the smoothing of the ripple structure is not affected because the reflection scattering order does not penetrate the particle.

The implementation of Rainbow refractometry fails for larger particles due to the sensitivity of the rainbow position to non-sphericity. However, small particles \( (d_p < 30 \mu m) \) are highly spherical because of the strong influence of surface tension. Extended GLMT was used to estimate the deviation in the angular position of the first rainbow maximum for small prolate and oblate droplets under pulsed illumination (Xu 2007). On the basis of Gouesbet et al. pioneering work on GLMT describing the interaction between a Gaussian beam and a sphere (Gouesbet 1988), Xu et al. has extended this theory to the case of a spheroid illuminated by arbitrary laser beams (Xu et al. 2007).

The whole scheme of calculation is comparable to that for a sphere (Mees et al. 2001). It can be shown that half-axis deviations of \( \pm a/b = 1+1E-3 \) are necessary for an angular deviation less then \( \Theta = 0.1^\circ \) for both oblate and prolate spheroids which corresponds to a deviation in temperature of up to 10 K. To attain the angular deviation of \( \Theta = 0.01^\circ \), which matches a deviation in temperature of only 1 K, a necessary half-axis deviation as low as \( \pm a/b = 1+1E-4 \) must be assumed.

### 3. Morphology Dependent Resonances

A second characteristic of femtosecond pulses is the broad electromagnetic spectrum of the illumination. For continuous laser illumination with one spectral line, a spherical particle generates optical resonances for various

![Fig. 3: Angular distribution of local maxima of the scattering function for a range of diameters \( (d_p = 50 \text{ fs}, \lambda = 780 \text{ nm}, n = 1.333, \) perpendicular polarization \( ) \). The primary rainbow is located between 140° and 180° and the secondary rainbow between 90° and 130° degrees](image)
wavelength-diameter ratios (morphology dependent resonances, MDR): These optical resonances increase the optical cross-section of the particle and generate strong oscillations in the diameter-intensity relation. Because the power of a pulse is distributed over several spectral lines, which generally do not fulfill the resonant conditions at the same time, the influence of optical resonances is suppressed strongly.

Fig. 4 shows the numerically obtained total scattered intensity over particle diameter for (a) continuous wave and (b) pulsed illumination. If individual scattering orders are separated due to short pulse lengths, the scattering function is monotonic with relation to the droplet diameter. The absence of MDR results from the pulse energy of ultrashort laser pulses being distributed over a considerable spectral width.

As seen in Fig. 3, the complex interaction of scattering orders results in the scattering function not being monotonic to an equal degree for all scattering angles. Hence, for applications using the intensity as a measure of particle size, the detection angle must be carefully chosen. In the case of illumination with 50fs pulses and a scattering angle of $\eta_s = 70^\circ$, numerical results indicate an intensity-diameter relation which is monotonic down to a diameter of approximately $d_p = 5\mu m$. While MDR can be used to determine evaporation rates of liquid droplets, their suppression can be useful for determining small changes in absolute size through intensity based size measurement with micron accuracy. Due to its controllable beam qualities such as polarization, wavelength and in particular energy density and focussing, a laser source is of course preferable to a spatially extended white light source, which obviously also distributes the radiated energy over a considerable spectral width and has a coherence length of microns.

For investigating Morphology Dependent Resonances in continuous wave and in femtosecond pulse illumination the setup shown in Fig. 5a was used. A water droplet generated by a HP 51604A inkjet cartridge was levitated in air without wall contact using an electrodynamic trap. The electrodynamic levitation was chosen because it is suitable for droplets of a radius between $d_p = 3\mu m$ and $d_p = 300\mu m$. For our experiment an electrodynamic trap with a recently developed geometry of electrodes (Fig. 5b) was used providing 360° optical access horizontally and 43° vertically. This

Fig. 4: Numerically obtained intensity distribution in the far field for a range of diameters of a spherical, homogeneous droplet ($n=1,333$, $\eta=70^\circ$). a) for cw illumination ($\lambda = 780nm$) the contribution of MDR’s becomes apparent on a nanometer scale. b) For pulsed illumination ($t_{puls} = 200fsec$, $\lambda = 780nm$) the intensity distribution is free of MDR’s and scattering lobes.
new geometry consists of four tube-like electrodes: two inner and two outer (Fig. 5b). This allows the angular range of detection to be easily varied during the experiment. Moreover, for future experiments the trap provides a channel for a gas stream, which allows for control of the evaporation rate of droplets and thus the frequency of morphology dependent resonances in time. The vertical position of the droplet is controlled during evaporation by a control loop consisting of a segmented photodiode on which the droplet is imaged by a lens, and computer control of the applied AC and DC voltage. The droplet was illuminated by the Ti:Sa laser at a wavelength of 784nm which can be driven in continuous wave mode with an output power of 800mW or in pulsed mode with 75MHz repetition rate and 11nJ energy per puls. The time dependent intensity of the light scattered by the droplet was recorded with a rate of 10kHz using a photodiode on which the droplet is imaged by a lens, collecting the scattered light in the range between 55° and 97° in the horizontal plane.

Fig. 6a displays the measured intensity distribution for a droplet captured inside the electromagnetic trap illuminated by the same Ti:Sa laser in CW operation mode. The pronounced oscillations correspond to the numerically obtained intensity distribution in Fig. 4a. The scattering lobes are significantly smoothed by pulsed illumination (Fig. 6b). Furthermore morphology dependent resonances are substantially reduced as predicted (Fig. 4b). Fourier transform analysis of the intensity distribution from pulsed illumination confirms that minor oscillations in Fig. 4b can be found at $\nu = 50\text{Hz}$ and $\nu = 200\text{Hz}$, which are the frequencies of the electric grid and the variation of the electromagnetic field inside the levitator (Bakic et al. 2008)

4. Conclusion

The angular distributions of local maxima of the scattering function have been presented for small particles under ultrashort pulse illumination. For ultrashort pulse illumination much of the local maxima are distinct, exhibiting only the maxima of the supernumerary bows of the primary rainbow and freed from disturbing interferences with higher order refractive contributions. This indicates the possibility of precise in-situ measurements with the Rainbow refractometry for highly spherical
small particles. Moreover, we experimentally measured the intensity scattered by water droplets of diameter less than \( d_p = 60 \mu m \) integrated over a certain solid angle under the illumination of 200fs pulses. In this way a substantial reduction of morphology dependent resonances for microscopic water droplets has been successfully demonstrated. The use of the electrodynamic trap together with the Ti:Sa laser allows us, for the first time, to successfully observe the scattering of femtosecond laser pulses on water droplets not only during a short period of milliseconds like in a droplet stream, but also over the temporal evolution of the evaporating droplet. As the diameter of droplets in sprays changes in general, this can be seen as an important step towards spray diagnostics using femtosecond laser pulses. The new geometry of the electrodes of the electrodynamic trap does not restrict the horizontal observation angle for the scattered light. Therefore we were able to center the detector in our case easily at the scattering angle of 76°, being in the range where the scattering function can be made monotonic down to a droplet diameter of less than \( d_p = 35 \mu m \), according to theory.

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**Fig. 6: a,b) Enlarged representation of the temporal intensity distributions of Fig.8 emphasizing the contribution of MDR’s for a) cw and b) pulsed illumination (t_{puls} = 200fsec, \( \lambda = 780nm \)) with regard to levitated droplets ( \( n = 1,333 \) ) and a receiver angle of 70°.**


