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Development of an experimental setup applying rainbow schlieren deflectometry for visualization and quantification of heat and mass transfer in multiphase systems

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Abstract

An experimental setup applying the rainbow schlieren measurement technique is developed to visualize and quantify the heat and mass transfer in single- and multiphase systems. The method is capable of measuring the projected density gradient field directly by means of measuring the light deflection angle due to inhomogeneities from, e.g., concentration or temperature differences. An optical schlieren-based technique has the advantage of high temporal and spatial resolutions, enabling an in situ and non-invasive observation of a spatial region in real time. The presented research introduces the experimental setup, which applies concepts of earlier publications concerning schlieren-based techniques, and shows a proof of concept for the mass transfer in established liquid/liquid reference systems. Different ways of validating the setup, including simple mass transfer cases, free-convection heat transfer and numerical approaches, will be presented in the future.

1 Introduction

The experimental determination of the spatial distribution of a state variable, such as local temperature, pressure or concentration, in inhomogeneous single or multiphase systems is a challenging yet important task. Optical measurement techniques based on the change of the refractive index due to density differences, namely shadowgraphy, interferometry or schlieren-based techniques, have the advantage of being applicable in situ with high spatiotemporal resolution. This allows non-invasive real time measurements of a state variable of concern, as long as an optical access is possible and the observed media are not opaque (Settles, 2001). Thus they can be applied to a wide range of substances. They come at a relatively low cost compared to, e. g., laserinduced fluorescence, Raman spectroscopy or tomographical approaches such as nuclear magnetic resonance tomography (NMR). One has to keep in mind though, that the experimental results have an integrational character and the quantitative evaluation is restricted to certain symmetric boundary conditions or a tomographic approach is needed (Greenberg et al., 1995).

Schlieren methods have been used for the visualization of various phenomena in the past. An extensive overview concerning the historical background and research milestones, e. g. in flight research, achieved by applying refraction-based optical measurement techniques is given in (Settles, 2001). Settles and Hargather (2017) review recent developments which are the result of technical advances in, e. g., illumination technology, namely LEDs, and digital photography as well as the huge impact of digital image processing and evaluation (Kulkarni and Rastogi, 2016). These advances brought especially quantitative refractive index measurements to attention of an increasing number of researchers and expand the application of schlieren techniques from flow visualization to the quantification of heat and mass transfer in transparent media, see e. g. (Panigrahi and Muralidhar, 2012). Panigrahi and Muralidhar (2013) as well as Settles and Hargather (2017) give an overview regarding recent studies on quantitative heat and mass transfer. While referencing the whole field of publications dealing with schlieren imaging is beyond the scope of the present work, it is evident, that the majority of publications concerning quantitative refraction-based measurements consider gaseous systems (Jain et al., 2016). Additionally, flow and heat transfer studies are much more prevalent than mass transfer studies.

The present paper focuses on the development of an experimental setup applying the rainbow schlieren measurement technique, also referred to as color schlieren. The aim is to visualize and quantify the concentration fields in the continuous phase of disperse immiscible liquid/liquid systems in situ, non-invasive and in real time. The evaluation of local concentrations near the interface is an important aspect for understanding and describing interfacial mass transfer and can give an insight into the occurring transport phenomena. To get a deeper understanding of the mass transfer in liquid/liquid systems, especially in the presence of surfactants, a deeper insight into the prevalent interfacial phenomena is needed. Some qualitative optical investigations in liquid/liquid systems have been conducted, see, e. g., (Sawistowski, 1971) for an overview or (Agble and Mendes-Tatsis, 2000; Arendt and Eggers, 2007; Wang et al., 2011) for more recent results. These qualitative observations led to a deeper insight into interfacial convection and the occuring Marangoni effects, also considering surfactants to some extent.

Expanding the qualitative to a quantitative approach concerning mass transfer measurements, e. g., Al-Ammar et al. (1998) and Pasumarthi and Agrawal (2003) measure the concentration field in a helium jet emitted into ambient air. Srivastava (2013) evaluates the crystal growth in a liquid environment in combination with a tomographic approach to consider asymmetric distributions. Considering the analogy between heat and mass transfer, several authors conducted successful refraction-based temperature field measurements in a multitude of experimental implementations, e. g., in gaseous systems for heated gas jets (Agrawal et al., 1998) or for the free convection from heated bodies, namely horizontal or vertical plates (Alvarez-Herrera et al., 2009; Martínez-González et al., 2017; Hargather and Settles, 2012) in surrounding air. Tanda et al. (2014) and Jain et al. (2016) show interesting results by determining local temperatures for the heat transfer due to free convection from a plate to surrounding water or nanofluids respectively.

The qualitative results obtained for liquid/liquid mass transfer and the increasing success to use refraction-based techniques quantitatively in recent years pose a promising combination for a deeper understanding of the underlying transport phenomena which make the prediction and theoretical and numerical description of liquid/liquid mass transfer a challenging task.

2 Measuring principle

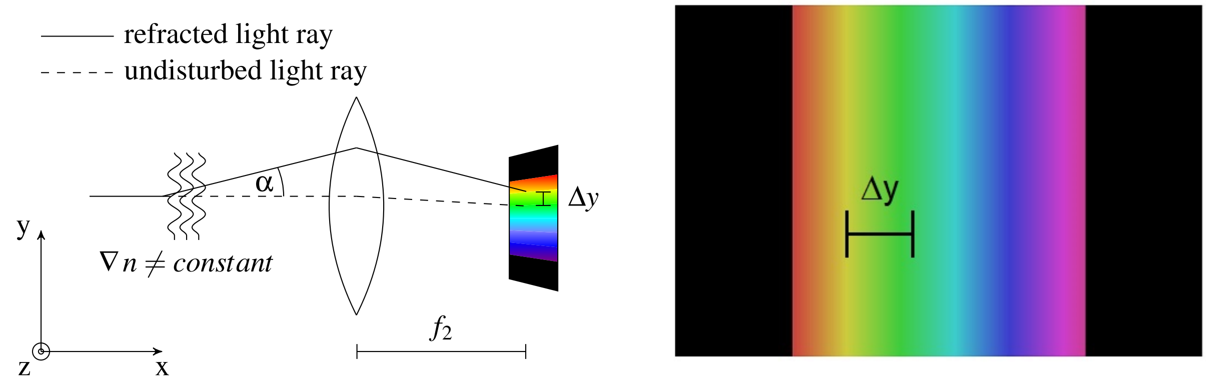
The measuring principle of schlieren deflectometry is based on collimated light rays, which are refracted due to local density differences in the observed media. These differences result from, e. g., inhomogeneous temperature or concentration distributions and cause a variation in refractive index, which the light rays propagate through. The relation between the cumulative deflection angle α and the spatial distribution of the refractive index $n = f(y,z)$ is based on the integration of the refractive index gradient field as shown in equation 1, where L represents the length of the integration path (Goldstein, 1983).

$$\alpha(y,z) = \int_0^L \frac{1}{n} \frac{\partial n(y,z)}{\partial y} dx \tag{1}$$

Figure 1(a) shows the measuring principle schematically. An exemplary light ray propagates through a transparent medium. Inhomogeneities in refractive index lead to a deflection of the light ray. Thus, the light exits the inhomogeneous region with the deflection angle α relative to the undisturbed reference ray, which represents the integrated deflection along its path through the refractive index gradient field. After leaving the measuring volume, the undisturbed parallel light rays are focused by a collecting lens or parabolic mirror onto the focal point of the respective optic, while the deflection of the disturbed light rays depends on their respective refraction angle which results from the propagation through the inhomogeneous region. Consequently, as shown in figure 1(a), the disturbed light rays experience a displacement Δy in the focal plane relative to the focal point.

The rainbow schlieren technique applies filters with a varying gradation of color, as shown exemplary in figure 1(b), for the visualization and quantification of the displacement and thus, the refractive index gradients are accessible. The offset between the focal point and the deflected light ray results in a color change as can be seen in figure 1(a) and 1(b), which can be evaluated by comparing the color values of the undisturbed reference image and the schlieren image pixelwise. By calibration, the difference in hue value can be correlated with the displacement on the filter in the focal plane. Thus, the cumulative refraction angle distribution can be extracted from the experimental data and in combination with equation 1 the refractive index gradient field is accessible experimentally. Details concerning the color scale filter shown in figure 1(b) are presented in section 3.

For a detailed description of the physical principles which found the basis for qualitative and quantitative refraction-based measurement see (Goldstein, 1983; Settles, 2001).



(a) Deflection of a light ray due to an inhomogeneous distribution of refractive index

(b) Color scale filter with exemplary deflection Δy

Figure 1: Schematic of light deflection (a) and a color scale filter of 20 mm color gradient width (b)

3 Experimental setup

The experimental setup as shown in figure 2 applies the rainbow schlieren measurement technique as described in the previous section. The optical components are aligned on a X95 optical aluminum rail as a main axis. For illumination purposes a fiber-coupled broadband LED is used as light source. The fiber optic patch cable is connected to an optical cage system, which centers the light source in vertical and horizontal direction and allows easy alignment of the different optical components by reducing the degree of freedom to one, since only an alignment in axial direction is necessary. The diameter of the fiber determines the size of the light source, which can be reduced further by applying slit or pinhole apertures at the fiber outlet. To reduce optical aberration at the outer regions of the applied lenses, an iris diaphragm is used to control the diameter of the light beam emitted through the apparatus.

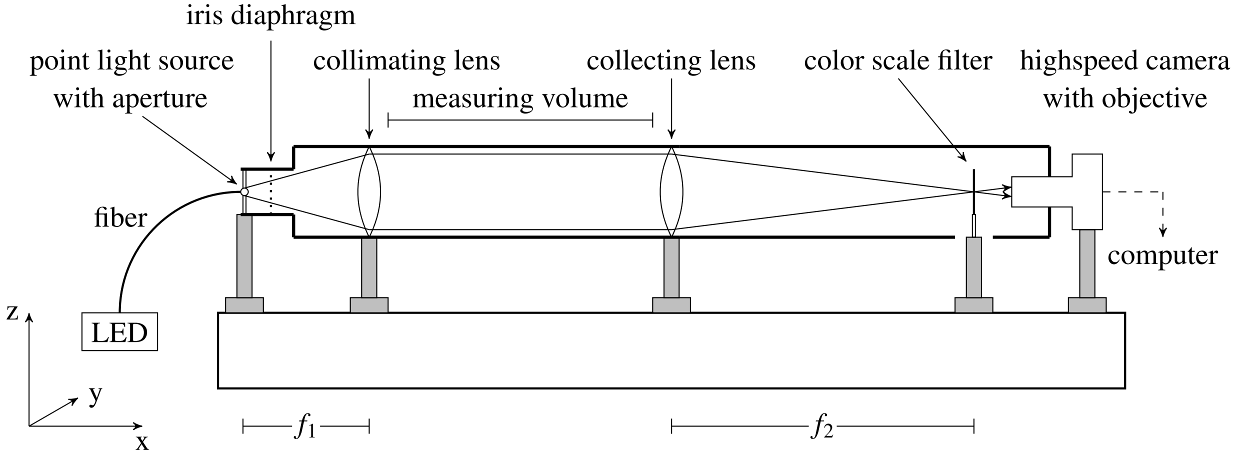


Figure 2: Schematic of the experimental setup

The applied setup uses the described point light source positioned in the focal point of an achromatic lens with a focal length of $f_1 = 250\,\mathrm{mm}$ for collimation. The focal length of the collimating lens f_1 fits the numerical aperture of the fiber (NA= 0.22) and determines the optical magnification of the focal point in conjunction with the focal length of the collecting lens f_2. A second achromatic lens is used for refocusing the collimated light rays. The measuring volume as indicated in figure 2 is located between the collimating and the collecting lens and is flexible in size. Thus, different setups with an optical access can be incorporated into the apparatus. The beam diameter determines the horizontal and vertical dimensions and the distance between the two lenses determines the depth of the measuring volume.

In case of rainbow schlieren, a color scale filter is positioned in the focal plane of the collecting lens.

Inspired by Greenberg et al. (1995) one-dimensional color scale filters with a linear change of hue value h in one direction and constant hue values in the other direction as shown in figure 1(b) are applied. The hue values used in the 1D filters, which have been manufactured by digital print on a diapositive, range from $h = 0$ to 0.9. Since the sensitivity is determined by the gradation of color, different filters with varying gradation are applied by changing the width of the filter between 20 and 0.5 mm, resulting in a color gradation of $4.5 \cdot 10^{-2}$ to 1.8 $^{\text{hue}}/_{\text{mm}}$. The other parameters in the applied HSV color scheme, namely saturation s and value v, stay constant at $s = 0.7$ and $v = 0.8$ respectively.

The application of color scale filters necessitates the application of a light source with a broad spectrum in the visual range and thus achromatic lenses have to be used to reduce chromatic aberrations. The digital printed color scale filter is mounted into a YZ-translation mount for rectangular optics, which is post-mounted and positioned in the focal plane of the collecting lens inside of the optical cage. The translation mount enables the calibration procedure as given by Greenberg et al. (1995) to obtain the correlation between the YZ-position of the focal point on the filter and the resulting color impression $h = f(y,z)$.

For imaging purposes a colored 4 MP CMOS-highspeed camera *Vieworks VC-4MC-C180EO-CM* is used in conjunction with different camera lenses, depending on the focal length of the collecting lens and the intended magnification. From the resulting images the hue value of each pixel can be extracted and compared to the respective values in the undisturbed reference image. Using the calibration curve obtained during the calibration procedure $h = f(y,z)$, the difference in hue values between the experimental and the reference image can be correlated to the displacement on the filter Δy or Δz respectively and thus the quantitative evaluation of the projected refractive index gradient field is possible. As mentioned before, to determine, e. g., the temperature or concentration from the experimental data, specific symmetrical conditions have to apply and a correlation between the refractive index and the state variable of concern is needed as well as the other state variables influencing the refractive index to stay constant.

4 Proof of concept

As mentioned in section 1, the rainbow schlieren technique can be used for a variety of substances. As a proof of concept, measurements in numerous transparent liquid/liquid systems were conducted. Water was usually the polar phase while, e. g., 1-octanol, toluene and cyclohexanol were used as organic phases. Acetic acid and acetone were chosen as transfer components and the surfactants SDS, Triton X-100, DTAB and butyldiglycol (C_4E_2) were considered to study the influence of surfactants on the interfacial mass transfer. Overall, the qualitative effects described in the literature, see section 1 for references, could be visualized with the presented experimental setup with high spatial and temporal resolution. Additionally, the images taken show an improved quality and, moreover, the color distribution can be accessed for quantitative evaluation.

Figure 3 shows the refractive index gradient field of the surfactant butyldiglycol around a stagnant 1-octanol droplet in water. Water and 1-octanol have been presaturated and the transfer component is initially located in the disperse phase, thus enabling mass transfer from the disperse to the continuous phase (d → c). A glass cuvette with planar 50x50 mm surfaces was used as test cell. The octanol drop was generated through a stainless steel cannula (outer diameter 0.52 mm), which is located in the bottom part of the images (vertical black bar) and attached to a syringe pump. Due to the curvature of the drop surface, refraction increases with distance from the center. Therefore, the drop appears mainly black because the refracted light passes the filter plane outside of the colored part of the filter. Due to weaker deflection, the colored gradation of the filter is visible in the center of the drop. The investigations have been done applying a collecting lens with a focal length of 100 mm and a color scale filter with a horizontal gradation of hue $h = 0$ to 0.9 over a length of 5 mm. Image acquisition at 3 fps was sufficient and realized with a 12x zoom lens. In figure 3, four images taken at different times after the drop production are displayed. The refractive index gradient field around the drop can be seen clearly. The light deflection at the air/glass- and glass/water interface before passing the concentration boundary layer of the drop can be neglected since the collimated light rays pass orthogonally through the cuvette's wall. For quantitative analysis, refraction at the end of the cell has to be taken into account. Discussing qualitative results, the deflection when exiting the cuvette can be neglected as well.

After formation of the drop, high concentration gradients next to the interface of the octanol drop occur. This results in high refractive index gradients, accordingly. Therefore, the area close to the drop appears black. During the first few seconds after drop formation, small convection cells develop and combine. These cells can be seen in figures 3(a) and 3(b) at the liquid/liquid interface. After combination of these small cells, the refractive index gradient field, visualized by the colored aureole-like shape, envelopes the octanol drop. This symmetrical diffusion is occasionally interrupted by global eruptions, which lead to

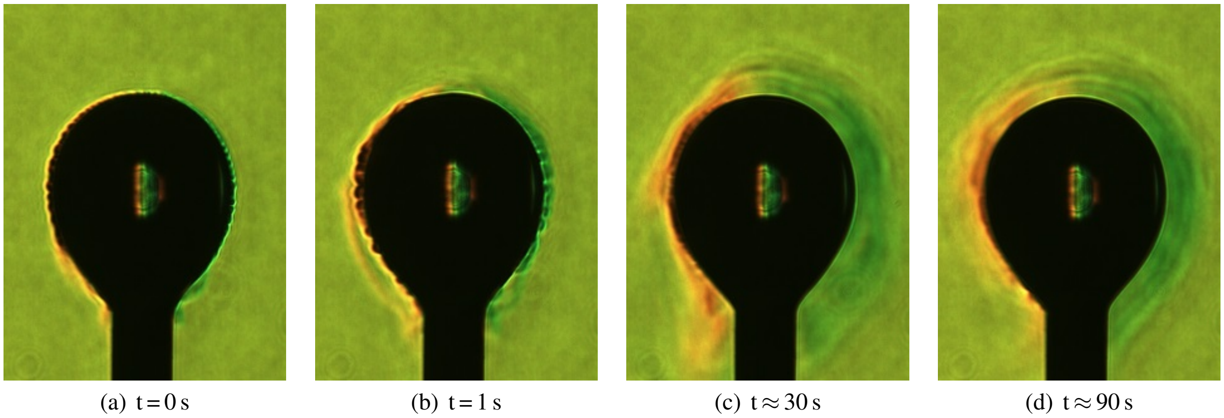

(a) t = 0 s (b) t = 1 s (c) t ≈ 30 s (d) t ≈ 90 s

Figure 3: Rainbow schlieren images of the system 1-octanol/butyldiglycol/water at different times after drop formation was completed, mass transfer direction d → c.

higher concentration gradients near the liquid/liquid interface and higher mass transfer rates accordingly. In the presented system, the aureole-like shape increases in size over time, which represents expected behavior for diffusional mass transfer and indicates an increase of concentration boundary layer thickness. In contrast, other examined systems showed stochastic vibrations or oscillation with high transfer rates of the respective transfer component, e.g. in the system toluene/acetone/water with high mass fractions of acetone in the organic dispersed phase. The described effects represent characteristic behavior in case of the occurence of interfacial phenomena and can be found in the literature, see e. g. Sawistowski (1971) for a profound description. The main driving force for these interfacial effects are differences in interfacial tension, which result in interfacial movement from areas with lower interfacial tension to those with higher interfacial tension and are often referred to as Marangoni instabilities (Sawistowski, 1971). These differences can, e. g., be generated by differences in concentration or temperature.

The Marangoni effect can distort the boundary layer or even lead to oscillation of the drop at the capillary. In figures 3(c) and 3(d) it can be observed that, due to distortion of the boundary layer, concentration gradients increase locally which benefits the local mass transfer. Additionally, these figures show that in case of strong distortion of the boundary layer, mass transfer phenomena at the surface of newly generated drops can reoccur. Especially in figure 3(c) small cells are visible at the liquid/liquid interface at the left side of the drop.

5 Conclusion

This publication proves the applicability of rainbow schlieren measurements for qualitative analysis of concentration fields near liquid/liquid interfaces. The color values can be easily extracted from the images, thus a successful quantitative evaluation of the concentration fields in situ with high spatiotemporal resolution is feasible. The experiments show that qualitative observations are reproducible. Quantitative repeatability has to be examined, e.g., time dependence of local concentrations, because of the highly coincidental character of the transient and inhomogeneous interfacial phenomena. Nevertheless, an non-invasive quantitative experimental access to the concentration field with high spatial and temporal resolution has the potential to greatly benefit the description of mass transfer phenomena and the influence on mass transfer rates connected to their occurence. Additionally, a determination of concentrations near the interface could enable an improved modeling and validate numerical approaches for the quantification of liquid/liquid mass transfer.

For validation purposes, the temperature field around different test bodies and simple mass transfer systems will be evaluated in the near future. Additionally, a ray tracing model will be incorporated into a CFD model for numerical validation and estimation of the errors connected to, e. g., the asymmetric character of the interfacial transport phenomena.

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