



# Simultaneous measurement of monocomponent droplet temperature/refractive index, size and evaporation rate with phase rainbow refractometry

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## ABSTRACT

The accurate measurements of droplet temperature, size and evaporation rate are of great importance to characterize the heat and mass transfer during evaporation/condensation processes. The nanoscale size change of a micron-sized droplet exactly describes its transient mass transfer, but is difficult to measure because it is smaller than the resolutions of current size measurement techniques. The Phase Rainbow Refractometry (PRR) technique is developed and applied to measure droplet temperature, size and transient size changes and thereafter evaporation rate simultaneously. The measurement principle of PRR is theoretically derived, and it reveals that the phase shift of the time-resolved ripple structures linearly depends on, and can directly yield, nano-scale size changes of droplets. The PRR technique is first verified through the simulation of rainbows of droplets with changing size, and results show that PRR can precisely measure droplet refractive index, absolute size, as well as size change with absolute and relative errors within several nanometers and 0.6%, respectively, and thus PRR permits accurate measurements of transient droplet evaporation rates. The evaporations of flowing single n-nonane droplet and mono-dispersed n-heptane droplet stream are investigated by two PRR systems with a high speed linear CCD and a low speed array CCD, respectively. Their transient evaporation rates are experimentally determined and quantitatively agree well with the theoretical values predicted by classical Maxwell and Stefan–Fuchs models. With the demonstration of evaporation rate measurement of monocomponent droplet in this work, PRR is an ideal tool for measurements of transient droplet evaporation/condensation processes, and can be extended to multicomponent droplets in a wide range of industrially-relevant applications.

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## 1. Introduction

Droplets in sprays have tremendous applications in a wide variety of fields, including spray drying in the food industry, pharmaceutical powder production, and the mixing of liquid fuels in automotive and aeronautical combustion systems. In these applications, droplets dynamically evolve in morphology upon breakup from ligaments due to hydrodynamic instability. Besides, droplets can also interact with a surrounding medium through coupled heat and mass transfer processes, such as heating, cooling, condensation and evaporation [1,2]. In order to gain a comprehensive understanding

of droplet dynamics, measurements of transient droplets properties, such as size and temperature, as well as their changes during these dynamic processes, are of essential importance. Hence, a range of optical methods have been proposed based on the complex interactions between light and droplets [3–6] to measure such properties non-intrusively. With regard to morphology characterization, the non-spherical or even irregular droplets are usually recorded by 2D direct microscopic imaging [7,8] or 3D digital holographic imaging [9,10]. In terms of size measurement, in addition to the above two imaging approaches, several interferometric methods have been proposed for spherical droplets. PDA (Phase Doppler Anemometry) is a commercialized instrument for size and velocity measurement for single spherical droplets [11]. Interferometric Particle Imaging (IPI) [12–14], which was originally proposed as Interferometric Laser Imaging for Droplet Sizing (ILIDS)

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[15], can measure sizes of droplets in a planar region by analyzing the fringes spacing created by the interference of the droplet's glare points. Morphology-Dependent Resonance (MDR) of a spherical droplet, which is also called Whispering-Gallery Modes (WGM) [16,17], is a chaotic phenomenon characterized by angularly pulsed peaks with large intensities, and results from constructive interference of multiple light waves that circulate around a sphere's rim via total internal reflections, and return in phase. This only occurs at particular resonant conditions when the optical path length of a round trip inside the droplet is an integral multiple of the exciting light's wavelength. These periodical peaks can be used to measure the droplet size with ultra-high accuracy. As for droplet temperature, it is usually measured with spectroscopic imaging, such as thermal infrared imaging, or Laser-Induced Fluorescence (LIF) which can also be employed to measure species concentration in multicomponent droplets [18–20].

The evaporation rate of a droplet, as characterized by the well-known  $D^2$ -law, is an issue of great importance, and has been intensively documented [1,21,22]. Experimental droplet evaporation rates are usually determined by attaching a droplet onto a fiber, and tracking its change of size with time [23–25]. This is a prevailing approach and can be operated at elevated temperatures and pressures. For this approach, the studied droplets tend to be relatively large with initial sizes up to a millimeter. These droplets are much larger than those typically found in combustion systems, which are in the order of tens of microns or less. The attachment to a fiber affects the evaporation rate through the conduction of heat, but also through the modification of the droplet's external area and shape by surface tension, which in turn affect the exchange of heat with the surrounding gas. Moreover, the effects of droplet relative velocity with respect to its surrounding, which could be up to dozens and or even hundreds of meters per second in certain engines, cannot be taken into account because the droplet is statically hanged. The suspended method cannot be applied to droplet streams with different spacing parameters [26]. Another approach is to employ sessile droplets to measure the evaporation by monitoring the surface regressions, e.g., via direct imaging or interferometry [27], or even from the vapor gradient by phase measurement using interferometry [28]. Single droplets can also be fixed to study their evaporation by other non-contact methods, such as optical levitation [29–31]. Some experimental measurements of transient evaporation rate of individual moving fuel droplets [14,32–35] and mono-disperse droplet streams [17,36–38] have also been reported. The experimental strategy can be classified into two categories. One is a Lagrangian strategy to track the droplet absolute size from multiple samplings. For example, Marié et al. [35] measured the fast evaporation process of a falling diethyl ether droplet by combining a high-speed Digital Holographic Particle Tracking Velocimetry (DH-PTV) system to track the size and rainbow refractometry to measure the temperature. This strategy is suitable for relatively large size changes, which requires a fast evaporating droplet or long observation times. The other strategy is to directly measure the size change (mass loss) caused by evaporation of a single droplet. Actually, the size change can be regarded as the mass transfer of the evaporating/condensing droplet with its surrounding, and thus is of crucial significance in the characterization of heat and mass transfer of fuel droplet evaporation. MDRs of scattering [17,30] or fluorescence [17,29] are usually used for precise identification of evaporation rate via size changes. Recently, PHase Interferometric Particle Imaging (PHIPI) [14] was proposed to measure the absolute diameter, as well as the diameter changes (evaporation rate), of single droplets by analyzing the time-resolved dynamic light scattering in the forward direction.

Rainbow refractometry [38–43] is a method to simultaneously measure the refractive index and diameter of a droplet by analyzing

the light scattering around the rainbow angle. This technique has been applied to determine other droplet parameters on which the refractive index depends, including temperature [44–49], and species composition and concentration [31,50–52]. It was first proposed in the form of the Standard Rainbow Technique (SRT) [39] to measure individual spherical droplets or identical droplet streams only, because the rainbow pattern is highly sensitive to droplet sphericity and small non-sphericity can bring about severe distortions [53–55]. In order to capture light scattering of a single droplet in a spray and to avoid interference from other droplets, one droplet in the probe volume is isolated using a small pinhole and a short exposure time. Later, the technique was adapted to measure the average refractive index and size distribution of a dense droplet field with dispersed sizes, which was realized by enlarging the pinhole size and extending the exposure time. This particular approach is called Global Rainbow Technique (GRT) [40]. The rainbow signal in GRT integrates rainbows of a large number of droplets, with the individual ripple structures being averaged out. This results in a smooth Airy rainbow that counterbalances the sensitivity to droplet sphericity. Both SRT and GRT are point probe measurements defined by the pinhole. Recently, One-dimensional rainbow refractometry, which extends the measurement volume to a one-dimensional line section, has been proposed and implemented with two different configurations in both spatial [41] and Fourier domains [42]. One-dimensional rainbow refractometry can be operated in both SRT [38,42,56] and GRT modes [41,42]. Moreover, it is found that the phase of rainbow is extremely sensitive to minute changes in droplet size and refractive index. The phase shift of the ripple structures in rainbows of single droplets or mono-dispersed droplet streams can be recorded with one-dimensional rainbow refractometry as droplets flow along the probed line section, and thus has been applied to simultaneously measure droplet refractive index, size and their changes. The ripple phase shifts with respect to nano-scale changes was also noticed by Sankar et al. [36] and Han et al. [57]. This approach is named Phase Rainbow Refractometry (PRR) [38], since it uses the phase information of the rainbow signal, in addition to the amplitude and frequency information that are used in traditional SRT and GRT. We briefly introduced PRR with a proof-of-concept validation but without detailed description [38].

Here we present a comprehensive investigation of the development of the PRR technique as well as its application to simultaneously measure droplet temperature via refractive index, diameter, and diameter changes and thereafter evaporation rate of spherical evaporating droplets. We derive the measurement principle of PRR (Section 2.1), and then the data processing algorithm to retrieve the droplet refractive index, size and size changes (Section 2.2). In Section 3 the PRR technique is tested and verified by simulated rainbow images using rigorous Generalized Lorenz–Mie Theories (GLMT). Finally, we present two experimental implementations of PRR for fuel droplets, demonstrating the technique's capabilities for both single droplets and continuous droplet streams (Section 4).

## 2. Phase rainbow refractometry (PRR)

### 2.1. Principle and derivation

The primary rainbow of a homogeneous and spherical droplet with a refractive index  $n$  respective to its surrounding medium can be exactly described by light scattering theory [58–60], and expressed as an infinite series which integrates all the scattering processes. Reformulating the scattering coefficients, the Lorenz–Mie light scattering can be decomposed into different processes analogous to the view point of geometric optics and equivalently noted as Debye series with different orders  $p$  [61,62], e.g., reflection ( $p = 0$ , which is computed together with diffraction ( $p = -1$ ) for

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