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# Measurements of refractive index and size of a spherical drop from Gaussian beam scattering in the primary rainbow region

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## ABSTRACT

The primary rainbow observed when light is scattered by a spherical drop has been exploited in the past to measure drop size and relative refractive index. However, if higher spatial resolution is required in denser drop ensembles/sprays, and to avoid then multiple drops simultaneously appearing in the measurement volume, a highly focused beam is desirable, inevitably with a Gaussian intensity profile. The present study examines the primary rainbow pattern resulting when a Gaussian beam is scattered by a spherical drop and estimates the attainable accuracy when extracting size and refractive index. The scattering is computed using generalized Lorenz–Mie theory (GLMT) and Debye series decomposition of the Gaussian beam scattering. The results of these simulations show that the measurement accuracy is dependent on both the beam waist radius and the position of the drop in the beam waist.

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## 1. Introduction

The measurement of drop size, velocity and refractive index has become commonplace in the process industry for quality control and is widespread in academic research. The physical principles used for optical drop characterization can be classified into direct imaging, intensity or intensity ratio, interferometry, time shift and Raman scattering [1]. Direct imaging techniques can be used for drops of size above the diffraction limits of the optical system [2]. Using a high-speed camera the drops can also be tracked in time to yield drop velocity [3,4]. The shadow Doppler technique also uses imaging for particle size and a laser Doppler interferometer for velocity measurement [5]. The time-shift technique, also known as pulsed-displacement technique, can measure size, velocity, and refractive index of transparent particles [6,7]. However, the most widespread and most accurate technique for obtaining size and velocity of (homogeneous spherical) drops remains phase Doppler interferometry [8].

Nevertheless, the size and the refractive index (hence the temperature) of a drop can also be determined using the primary rainbow [9]. The rainbow technique is applicable to spherical drops, but also measurements with ellipsoidal drops have been demonstrated [10,11]. The rainbow scattering patterns from spheroidal oblate drops were measured by Marston and coworkers [12,13].

Furthermore, using a relation between aspect ratio and curvature of the rainbow fringes, the aspect ratio of oblate drops aligned with the optical axis was measured [14]. The radial refractive index gradient of particles and temperature gradient of a liquid cylinder were also studied using the rainbow technique [15,16] and the Möbius shift, i.e. the deviation between the rainbow angle for spheroidal and spherical droplets, was investigated using a vectorial complex ray model [17,18]. The same model was used to study the rainbow scattering for oblate drops [19]. For simulating rainbows of drops, a physically based model was derived, which is based on ray tracing extended to account for dispersion, polarization, interference, and diffraction [20]. For a deformed liquid jet, the shift of the rainbow position in the horizontal and vertical directions was investigated by experiment and simulation [21].

A generalization of the rainbow technique, known as global rainbow thermometry (GRT), was introduced by van Beeck et al. [22] to measure the mean size and temperature of an ensemble of spray drops. The technique has been compared with the phase Doppler technique [23] to assess its accuracy and to evaluate which effective drop diameter is measured. The GRT has been used for the measurement of a liquid–liquid suspension [24] and spray drops in a large containment vessel [25]. Additionally, the sensitivity of GRT to non-sphericity of the drop was investigated by simulation and experiment [26,27]. Based on a one-dimensional spatial filter in the Fourier domain, a new optical configuration of the one-dimensional rainbow technique was recently proposed [28]. This one-dimensional phase rainbow refractometer has been

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developed for the accurate measurement of instantaneous refractive index and drop size [29]. However, the GRT operates on light scattered from a large ensemble of drops, consequently not offering high spatial resolution. The advantage is that the random orientation of any non-spherical drops in the ensemble will lead to a reduction of any influence that non-sphericity of single drops has on the results. One possibility to achieve a higher spatial resolution is to focus the illuminating beam stronger and the present contribution investigates the influence of this focusing on the resulting primary rainbow pattern.

Lorenz–Mie theory provides a rigorous solution to describe the scattering of a linearly polarized plane wave by a homogeneous spherical particle [30]. It forms the basis of a number of techniques for optical particle characterization in various fields [31,32]. However, classical Lorenz–Mie theory fails to accurately describe the scattering characteristics of particles illuminated by a shaped beam. For this situation Gouesbet and coworkers [33] developed the generalized Lorenz–Mie theory (GLMT), which rigorously describes the interaction between an arbitrarily shaped beam and a regularly shaped particle. During the last three decades, the GLMT for spherical particles has been developed to a state of maturity [33–41] and extensions to the GLMT allow prediction of light from spheroidal particles [42–46]. Nevertheless, existing programs within the framework of GLMT for light scattering from spheroidal particles with arbitrary orientation still have limitations for certain sizes [45,46]. For scattering from spherical, non-spherical, or coated particles, the GLMT can be complemented by computations using the Debye series [47–53]. The rainbow scattering of a spherical particle, a non-spherical particle, and a circular cylinder were studied by using Debye series [47–49,51,53]. In the present study, both GLMT and decomposition using the Debye series will be used.

This paper is organized as follows: Section 2 presents the computational procedure used to explore the influence of the Gaussian beam on the position of the rainbow, using a combination of the Airy function, Debye series decomposition, and GLMT. Section 3 discusses the inversion of the primary rainbow pattern ( $p=2$ ) using GLMT, to yield size and relative refractive index. Section 4 presents conclusions.

## 2. Influence of Gaussian beam shape on the primary rainbow pattern

A spherical coordinate system  $(r, \theta, \phi)$  centered on the particle is used and the relative refractive index and wavelength are denoted as  $m$  and  $\lambda$  respectively. A further Cartesian coordinate system  $(x, y, z)$ , also centered on the particle with the  $z$ -axis along the incident beam propagation direction, is used to describe the beam placement. Offsets of the beam will be examined in the  $y$  direction. As a first step, the influence of the beam waist radius on the position of the rainbow is investigated. This is performed by examining the rainbow arising only from scattered rays of second-order refraction ( $p=2$ ), as obtained using the Debye decomposition of the Mie coefficients, as described in [50]. The Gaussian beam axis is placed at the position of the Descartes ray [12], i.e. the incident ray exiting the particle at the rainbow angle according to Airy theory [10]. For instance, for a particle of radius  $r=50\mu\text{m}$  and relative refractive index of  $m=1.33$ , the Descartes ray enters the particle at an offset of  $y=\pm 43.12\mu\text{m}$ . Fig. 1(a) shows the computed scattered light intensity in the primary rainbow region for water drops of different radii using a wavelength of  $\lambda=0.65\mu\text{m}$  and a beam waist radius of  $\omega_0=100\mu\text{m}$ , whereby the curves have been normalized using their respective maximum amplitude. Fig. 1(b) shows how the position of the first peak changes with beam waist radius. As shown in Fig. 1(a), all intensity distributions for different particle sizes exhibit a common inflection point at the scattering angle  $\theta=137.5^\circ$ , which is the rainbow angle according to geometric op-

tics. Furthermore, it can be seen that as the beam waist increases, or the particle becomes smaller, the position of the first peak in the primary rainbow approaches the value corresponding to illumination by a plane wave. This limit is reached when the beam waist radius is approximately equal to the radius of the drop.

Using the Debye series decomposition, the light intensity distribution in the primary rainbow region shown above, also called the rainbow pattern, was computed using only the second-order refracted rays. However, the generalized Lorenz–Mie theory [40] provides a more complete result, including contributions from other scattering orders. Using GLMT, the rainbow pattern arising from illumination with a Gaussian beam ( $\omega_0=100\mu\text{m}$ ) centrally positioned at the Descartes ray is shown in Fig. 2 for three different drop radii ( $50\mu\text{m}$ ,  $150\mu\text{m}$ , and  $200\mu\text{m}$ ). For small drops, a ripple structure exists, arising from interference of second-order refracted rays and reflected rays [10].

If a ripple structure exists, and the intensity curve is passed through a low-pass Gaussian filter, then curves similar to those shown in Fig. 1(a) are obtained. The filter cut-off frequency is iteratively chosen, such that the first two peaks are still easily distinguishable. For larger drops and for the same Gaussian beam illumination, the intensity of the reflected ray decreases and the amplitude of the ripples also decrease, as observed in the results for the larger drops shown in Fig. 2. For ratios of drop-to-beam-waist radius above 2, the ripple structure is virtually non-existent.

## 3. Drop characteristics computed using generalized Lorenz–Mie theory

According to geometrical optics, the rainbow angle only depends on the refractive index of the drop. According to Airy theory, the relation between the geometrical rainbow angle and the angles of the first two peaks in the rainbow diagram is given by [11]:

$$\theta_{rg} = \frac{\theta_1 - C\theta_2}{1 - C} \quad (1)$$

where  $\theta_1$  and  $\theta_2$  are the angles of the first two peaks of the filtered rainbow pattern. The parameter  $C$  is a constant, which will be given below.

If one can measure  $\theta_1$  and  $\theta_2$ , then the refractive index can be calculated using the relation between the geometrical rainbow angle and the refractive index, given as:

$$\theta_{rg} = \pi + 2\sin^{-1}\sqrt{\frac{4-m^2}{3}} - 4\sin^{-1}\sqrt{\frac{4-m^2}{3m^2}}. \quad (2)$$

The relation between the drop radius ( $r$ ), the refractive index and the peak angles (i.e.  $\theta_1$  and  $\theta_2$ ) is given by:

$$r = \frac{\lambda}{8} \left( \frac{\alpha_1 - \alpha_2}{\theta_1 - \theta_2} \right)^{3/2} \left[ \frac{3(4-m^2)^{1/2}}{(m^2-1)^{3/2}} \right]^{1/2} \quad (3)$$

where  $\alpha_1=1.0874$ ,  $\alpha_2=3.4668$  and  $C=\alpha_2/\alpha_1$ . In the following analysis, the rainbow pattern is calculated using GLMT and Eqs. (1)–(3) are then used to extract the refractive index and size of the drop, solving Eq. (2) iteratively for  $m$ .

First, the influence of the waist radius of a Gaussian illumination beam is investigated. According to geometrical optics, the primary rainbow arises from the Descartes ray. For example, for a water drop with  $r=100\mu\text{m}$ , the center position of Descartes ray is  $y=\pm 86.24\mu\text{m}$ . Computations of the rainbow pattern for a Gaussian beam centered on the Descartes ray for different beam waist radii are performed first. The computed rainbow patterns are then used to compute the drop size and refractive index. Table 1 summarizes the results. For a water drop with  $r=1000\mu\text{m}$  and a Gaussian beam waist of radius  $\omega_0=80\mu\text{m}$ , the relative error of the computed water drop radius is  $-5.63\%$ . As the beam waist radius

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