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Holographic interferometry for aerosol particle characterization

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ABSTRACT

Using simulations based on Mie theory, this work shows how double-exposure digital holography can be used to measure the change in size of an expanding, or contracting, spherical particle. Here, a single particle is illuminated by a plane wave twice during its expansion: once when the particle is 27λ in radius, and again when it is 47λ. A hologram is formed from each illumination stage from the interference of the scattered and unscattered, *i.e.*, incident, light. The two holograms are then superposed to form a double exposure. By applying the Fresnel–Kirchhoff diffraction theory to the double-exposed hologram, a silhouette-like image of the particle is computationally reconstructed that is superposed with interference fringes. These fringes are a direct result of the change in particle size occurring between the two illumination stages. The study finds that expansion on the scale of $\sim 6\lambda$ is readily discerned from the reconstructed particle image. This work could be important for improved characterization of single and multiple aerosol particles *in situ*. For example, by illuminating an aerosol particle with infrared light, it may be possible to measure photothermally induced particle expansion, thus providing insight into a particle's material properties simultaneous with an image of the particle.

1. Introduction

Recent advances in the resolution of commercial optoelectronic sensors, such as Charged Coupled Device (CCD) arrays, have revived interest in holography as a contact-free imaging technique [1–5]. In the context of aerosol science, such digital holography is able to provide single, and multiple, particle size and shape without the collection or immobilization of a sample [6]. In short this is done by allowing a particle's scattered light to interfere with unscattered light across the sensor. The resulting interference pattern, which constitutes a digital hologram, is then computationally processed to render a silhouette-like image of the particle.

Historically, such *in situ* aerosol characterization is often attempted by measuring a particle's (angular) scattering pattern, and then inferring the particle size and shape from specific features in such a pattern [7]. Unfortunately, the patterns for most particles of interest are so complicated that this interpretation is not practical, or at best, is only able to classify particle shape into general categories based on pattern-symmetry, for example [8]. Fundamentally, the lack of an unambiguous relationship between a particle and its pattern is due to the absence of scattered-wave phase information in the scattering measurement. However, such phase information is readily available in holography and accounts for its ability to produce clear particle images.

The availability of this phase information allows one to do more with the hologram than imaging alone. Specifically, this paper will describe how digital holography can be used to measure the change of a particle's size as it

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expands or contracts. This could be useful, *e.g.*, to quantify the optical absorption of particles undergoing photothermal expansion, or the contraction of particles undergoing evaporation and/or emission of volatile components. The basic concept involves the interference of *two* scattered waves, each computationally reconstructed from the same double-exposed hologram. Using simulations based on Mie theory, this study will demonstrate the concept for a single spherical particle. While holographic interferometry is not a new concept, a unique aspect to this work is that it deals with *wavelength* sized particles, rather than the geometrically large objects that have been considered in similar studies.

2. Imaging

Before explaining how the particle expansion/contraction measurement is done, it is necessary to first briefly describe how particle images are produced from a digital hologram. Consider the arrangement in Fig. 1, which shows a particle illuminated by a linearly polarized plane wave traveling along the z-axis. Such a wave approximates a collimated laser-beam. Located a distance ℓ from the particle is the sensor, called the hologram plane $\mathcal{S}$, which consists of an array of $N \times N$ square pixels of size Δ. Here, the location of each pixel is given in the (ξ, η, ℓ) coordinate system. Next, at a distance z from the particle, is the image plane where the particle image is eventually formed computationally. In this configuration, much of the incident wave passes by the particle largely unperturbed due to its small size. To good approximation, the hologram plane then resolves the interference pattern produced by the unscattered and scattered light, which is denoted I^{holo}. The reader should note that this arrangement is referred to as "in-line" since the scattered and unscattered light share the same optical path.

A particle image is obtained from I^{holo} by treating the hologram as a transmission diffraction-grating illuminated by the same incident plane wave. Here, the diffraction process is modeled computationally using the Fresnel-Kirchhoff diffraction theory [9]. Let the position of a pixel in the hologram be $\mathbf{r}' = \xi\hat{\mathbf{x}} + \eta\hat{\mathbf{y}} + \ell\hat{\mathbf{z}}$ and a point in the image plane be $\mathbf{r} = x\hat{\mathbf{x}} + y\hat{\mathbf{y}} + z\hat{\mathbf{z}}$. Then, the diffracted wave *amplitude* K in the image plane under the Fresnel-approximation is [9,10,11]

$$K(x, y) = \alpha \iint_{\mathcal{S}} I^{\text{holo}}(\xi, \eta) \frac{\exp(ik\mathfrak{r})}{\mathfrak{r}} \cos\,\theta \, \mathrm{d}\xi \, \mathrm{d}\eta, \tag{1}$$

where α is a constant, $k = 2\pi/\lambda$, $\mathfrak{r} = |\boldsymbol{\mathfrak{r}}| = |\mathbf{r} - \mathbf{r}'|$, and θ is the angle between $\hat{\boldsymbol{\mathfrak{r}}}$ and the normal to $\mathcal{S}$ along the forward direction. One can think of K as an approximation of the particle's near-field scattered wave amplitude.

From I^{holo}, the particle-free background, or reference intensity I^{ref}, across the sensor is subtracted to yield a contrast hologram I^{con}. In practice, this subtraction is important as it removes most imperfections in the illumination-beam profile and substantially improves the particle image. Next, using $\mathfrak{r}\,\cos\,\theta = d$, where $d = z - \ell$, and the binominal expansion in $\mathfrak{r}$ given that the distance d between the hologram and image plane is much larger than the hologram size $N\Delta$, Eq. (1) simplifies to

$$K(x, y) = \alpha \iint_{\mathcal{S}} I^{\text{con}}(\xi, \eta) \exp\left\{ \frac{ik}{2d} \left[(x - \xi)^2 + (y - \eta)^2 \right] \right\} \mathrm{d}\xi \, \mathrm{d}\eta, \tag{2}$$

where additional constant factors have been absorbed into α. If Eq. (2) is evaluated in the image plane, *i.e.*, $z = 2\ell$, then the absolute square of the resulting wave amplitude, $|K|^2$, forms the image of the particle. Notice, however, that one is not obliged to take the absolute square, in which case phase information is available; this will be important in the following.

There are advantages to this technique over conventional imaging. For example, collection of particles followed by optical or electron microscopy is often not suitable as this can distort particle shape, including aggregation and fragmentation. In another approach, which is *in situ*, particles are imaged directly with a telemicroscope consisting of relay lenses, microscope objective (MO), and a pulsed laser to freeze particle motion [13,14]. In that work $\lambda = 532$ nm and a MO with a Numerical Aperture (NA) of 0.4 is used, thus a theoretical resolution of $\sim 0.7\ \mu$m could be obtained. However, such high NA also means that the depth-of-field, Z, is small, *i.e.*, $Z = 2\lambda/(\text{NA})^2 \simeq 6\ \mu$m using the figures above. Consequently, particles in a sample stream must be controlled to within $\sim 6\ \mu$m for the image to be in-focus. Unfortunately, this precision is highly challenging in practice on *flowing* aerosols, *e.g.*, see [8]. Moreover, if multiple particles are present in close proximity, it would be unlikely

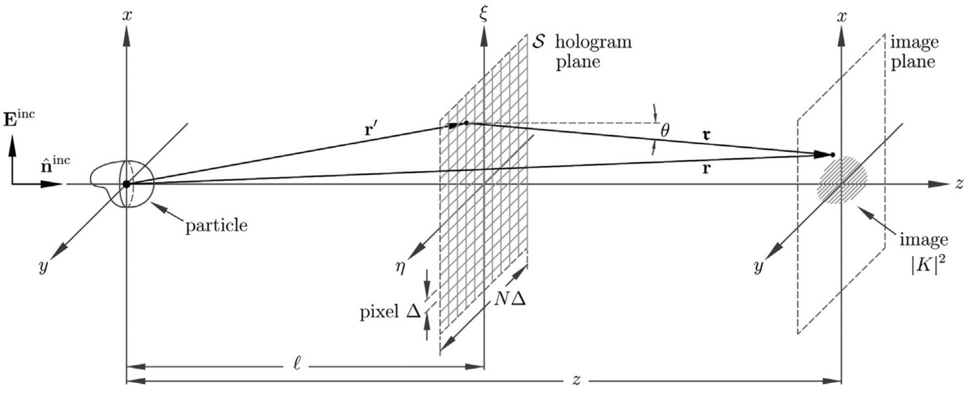

Fig. 1. Image formation for digital holography. See text for further explanation.