

## STUDYING THE DYNAMICS OF COLLOIDAL PARTICLES WITH DIGITAL HOLOGRAPHIC MICROSCOPY AND ELECTROMAGNETIC SCATTERING SOLUTIONS

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**ABSTRACT.** Digital holographic microscopy (DHM) can measure the 3D positions as well as the scattering properties of colloidal particles in a single 2D image. We describe DHM and our analysis of recorded holograms with exact scattering solutions, which permit the measurement of 3D particle positions with  $\sim 10$  nm precision and millisecond time resolution, and discuss studies of the Brownian dynamics of clusters of spheres with DHM.

### 1. Introduction

Probes of the 3D dynamics of colloidal particles are widely useful in soft matter physics. Measuring the dynamics of multiple interacting particles can give insight into the interaction forces between the particles [1]. In microrheology, studying the Brownian motion of isolated particles in a complex material can elucidate the structure of the material [2]. But most current techniques for measuring colloidal dynamics are either limited to 2D, require an ensemble average, or have undesirably long acquisition times of seconds or more.

Digital holographic microscopy (DHM) can locally probe 3D colloidal dynamics over time scales as short as tens of microseconds. When combined with exact scattering solutions, DHM can measure the positions of particles in a colloidal suspension with nanometer precision. Previously, Lee *et al.* fit holograms of isolated colloidal spheres to the Lorenz-Mie solution and measured particle positions to a precision of  $\sim 10$  nm [3]. Recently, Fung *et al.* fit holograms of clusters of spheres to T-matrix calculations for the first time and showed that the translational, rotational, and vibrational dynamics of colloidal clusters could be probed simultaneously [4].

Here we describe our implementation of DHM, show that we find good agreement between our recorded holograms and best-fit models incorporating exact scattering solutions, and briefly discuss some applications.

### 2. Recording and Analyzing Holograms

In in-line DHM, which is well-suited for studying dilute colloidal suspensions, we capture 3D information in a 2D image by illuminating a sample with a collimated laser, as

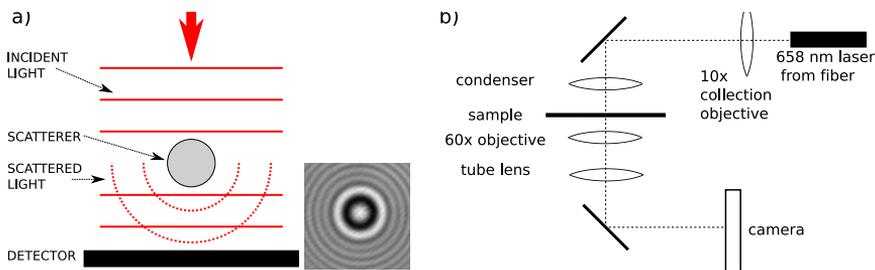
shown in Figure 1a. A small fraction of the incident light is scattered by particles in the sample. We record the interference pattern, or hologram, formed by scattered light and unscattered light, which acts as a reference.

The fringes of a hologram encode information about both the 3D position and the scattering properties of the particles that form them. The intensity  $I$  of a pixel of a recorded hologram can be described as follows:

$$I = |\mathbf{E}_{inc}|^2 + 2\Re[\mathbf{E}_{inc} \cdot \mathbf{E}_{scat}^*] + |\mathbf{E}_{scat}|^2 \quad (1)$$

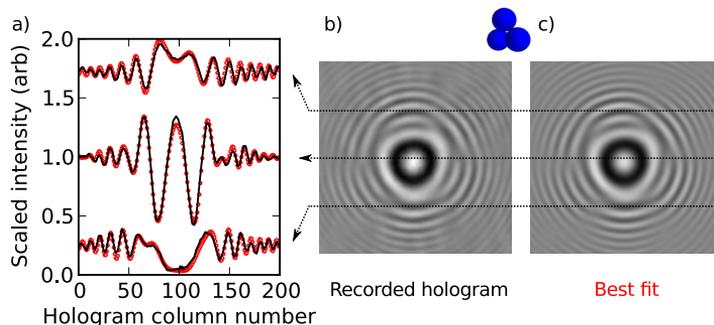
where  $\mathbf{E}_{inc}$  is the amplitude of the incident field at that pixel, and  $\mathbf{E}_{scat}$  is the amplitude of the scattered field. The position of a particle along the direction of incident light propagation is partly encoded in the second term, which is sensitive to the phase difference between the incident and scattered light. At the same time, holograms also record the angular variation of the intensity of the light scattered by particles. This encodes information about the scattering properties of the particles, such as their size, and for non-spherical particles, their orientation. See Fung *et. al.* for a more detailed discussion [4].

We perform DHM with a modified Nikon TE2000 inverted microscope. In brief, we replace the microscope's light source with a fiber-coupled 658 nm diode laser and record holograms with a Photon Focus MVD-1024E-160 CMOS camera. Our apparatus is schematically illustrated in Figure 1b; see Fung *et. al.* for details [4].



**Figure 1.** a) Schematic illustration of DHM. The inset is a typical recorded hologram of a  $1 \mu\text{m}$  polystyrene sphere in water. b) Diagram of apparatus.

We extract physical information from holograms by fitting recorded holograms to a model that depends on physical quantities such as particle coordinates, sizes, and refractive indices. Modeling a hologram according to Equation 1 requires knowing  $\mathbf{E}_{scat}$ , which we obtain from exact scattering solutions to Maxwell's equations. For isolated spheres, we use the Lorenz-Mie solution. For multiple spheres separated by a fraction of the incident wavelength, we must account for near-field optical coupling between the spheres [4]. Therefore, we use the T-matrix code SCSMFO1B, developed by Mackowski *et. al.*, to calculate  $\mathbf{E}_{scat}$  from clusters of spheres [5]. Fitting a time series of holograms allows us to measure colloidal dynamics. See Fung *et. al.* for further details [4]. Figure 2 shows that we find excellent agreement between a recorded hologram of a triangular cluster of three touching polystyrene spheres in water and a best-fit model hologram where  $\mathbf{E}_{scat}$  has been determined with SCSMFO1B.



**Figure 2.** Digital hologram of a cluster of three touching  $1.3 \mu\text{m}$  diameter polystyrene spheres in water. a) Comparison between recorded hologram (solid black line) and best-fit model calculated from T-matrix solution (red symbols), plotted along the three dashed lines shown. b) Recorded hologram. c) Best fit. The blue rendering shows the orientation of the cluster as determined from the best-fit model parameters; the incident light propagates into the page.

### 3. Brownian Dynamics of Colloidal Clusters

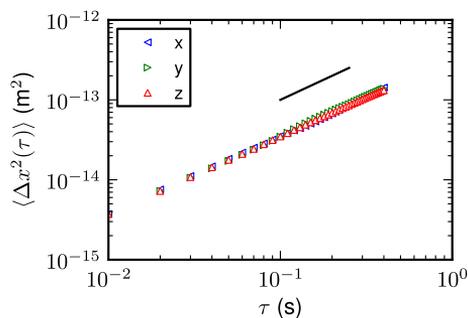
One application of DHM is in studying the 3D Brownian dynamics of colloidal clusters. Clusters made out of micron-sized spheres irreversibly bound by van der Waals forces exhibit translational and rotational Brownian motion. To illustrate, we show the mean squared displacements of the center of mass of a cluster of three spheres as a function of time interval  $\tau$  in Figure 3. The motion in all three dimensions is consistent with diffusion as expected, and by extrapolation to  $\tau = 0$ , we find that we are tracking the cluster center of mass to a precision of 20 nm or better in all three dimensions. Because DHM is sensitive to the orientation of our clusters, we can measure their rotational diffusion as well. See Fung *et al.* for further details [4]. We are also using DHM to study the poorly-understood forces on charged colloidal spheres near liquid-liquid interfaces.

### 4. Conclusions

DHM, combined with exact scattering solutions, is a powerful tool for the study of dynamics in colloidal suspensions due to its high precision and rapid time resolution. Future work will include expanding the range of types of colloidal particles that can be studied with scattering solutions, as well as improving the efficiency of our fitting techniques.

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**Figure 3.** Laboratory-frame mean-squared displacement of the center of mass of a triangular cluster of three  $1.3 \mu\text{m}$  diameter polystyrene spheres. The solid black line indicates diffusive behavior. Error bars are at most comparable in size to the plot symbols.

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