Observations and calculations of two-dimensional angular optical scattering patterns of a levitated bi-sphere

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We use single bi-sphere particles levitated in an electrodynamic balance to record two-dimensional angular scattering patterns at different angles of the coordinate system of the aggregate relative to the incident laser beam. Due to Brownian motion, the particle covers the whole set of possible angles with time and allows to select patterns with high symmetry for analysis. These are qualitatively compared to numerical calculations. An experimental procedure is proposed for studying restructuring effects occurring in mixed particles upon evaporation.

INTRODUCTION

Our interest in measuring two-dimensional angular optical scattering (TAOS) patterns of single aggregate particles made out of spheres originate from previous work [1], where we were using optical resonance spectroscopy for sizing evaporating solid, non-spherical particles. We observed that the shift of optical resonances with time, when the particle is solid, exhibits distinct discontinuities which we ascribed to sudden rearrangements processes within the evaporating solid. While this explanation is plausible, it warrants a more detailed investigation. A simple aggregate built from polystyrene latex microspheres and a solid with sufficient high vapor pressure could provide a model system for studying such processes. Besides optical resonance spectroscopy, analyzing the TAOS patterns with the help of numerical simulations should allow a more detailed analysis of the rearrangements taking place in aggregate particles upon evaporation.

Here we present a test of the basic concept by measuring the simplest aggregate particle possible, a bi-sphere, and compare the observed TAOS patterns with calculated ones using the code developed by Mackowski [2].

Our experimental setup resembles in a lot of aspects the one used in the pioneering work of Bottiger et al. [3], but the analysis of the scattered light differs in one essential aspect. Namely, instead of recording rotational averaged scattering matrices, we record the TAOS pattern for a fixed, distinct orientation. Previously, Holler et al. [4] have done similar experiments for sphere clusters consisting of multiple spheres, but they observed these clusters in an aerosol flow, while we levitate a single cluster and observe the same cluster for a prolonged period of time. This allows us to study the scattering at different angles between the coordinate system of the cluster and the incoming laser beam.

EXPERIMENTAL

The experimental setup has been described elsewhere in [5]. Briefly, Fig. 1 shows a schematic of the experimental setup. A particle is levitated in an electrodynamic balance with CCD1 recording a microscopic image of the particle illuminated with a weakly focused laser beam from below. CCD2 is a fast, progressive scan CCD (60 fps, shutter speed: 1/250 s) to record

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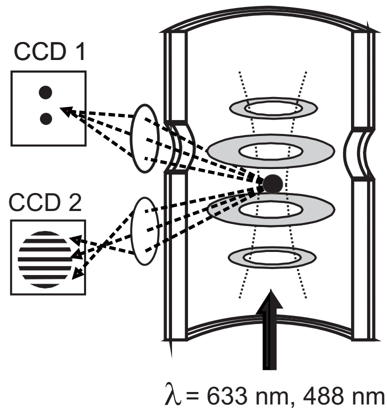

Figure 1. Schematic of the experimental setup. CCD1 records a microscopic image of the particle, CCD2 records the TAOS pattern due to scattered laser light with a wavelength of either 633 nm or 488 nm.

the corresponding TAOS pattern. The observed angles range approximately from 85° to 95° in both, polar and azimuthal angle, but no effort was made for these test experiments to calibrate the angles using the scattering of a single sphere of known radius and refractive index. A single-particle generator (Hewlett-Packard 51633A ink jet cartridge) is used to inject a single liquid droplet (volume 140 pl) from solutions made from suspensions of polystyrene latex particles purchased from Polyscience, Inc., diluted with MilliQ water. The number of primary particles within the droplet follows Poisson statistics and depends on the concentration of the suspension. The water evaporates and leaves an aggregate of spheres levitated in the electrodynamic balance. By adjusting the concentration of the suspension, the probability for obtaining a single sphere, a bi-sphere or tri-sphere may be optimized. By replacing the water with a dilute aqueous solution of, for example, a low-vapor-pressure dicarboxylic acid and subsequent drying of the particle in the electrodynamic balance, mixed particles consisting of polystyrene spheres and crystalline dicarboxylic acid may be produced. Upon evaporation of the dicarboxylic acid, restructuring of the remaining polystyrene spheres is expected to occur.

The particle is subject to rotational Brownian motion while trapped in the center of the electrodynamic balance [6]. In addition, with larger particles (radius > 5 µm), slight asymmetries in the balance together with the anisotropic polarizability of the bi-sphere particles induce an almost stationary rotation of the particle around one of the axes of symmetry. We observed a typical timescale of this motion of about 1 round per 10 seconds. If this effect is undesirable, the electrodynamic balance can be switched to a quasi-electrostatic mode, which allows the free Brownian motion about all axes [6]. For a particle of 5-µm radius, rotational Brownian motion leads to a mean rotation of 22 degrees per second. When movies of the Brownian motion are recorded with CCD1 and CCD2, single frames can be selected showing high symmetry because the polar and azimuthal angles of the aggregate are parallel or perpendicular to the plane of polarization of the illuminating laser beam. Two examples are shown in Fig. 2.

RESULTS

The upper row in Fig. 2 shows the experimental TAOS pattern in panel (a), the corresponding TAOS calculations [2] in panel (b) and the microscopic image of the particle in panel (c). The particle is a bi-sphere cluster with the nominal radii of the single spheres being 5.0 µm. The lower row shows the same particle but in a different orientation: (d) is again the measured TAOS pattern, (e) shows the calculated TAOS pattern and (f) the microscopic image. The microscopic images clearly reveal the orientation of the bi-sphere perpendicular to the line of view of CCD1 by showing 4 separated glare spots in the upper row image panel (c). Note that the glare spots seem to form at the corners of a square, as expected for a bi-sphere consisting of two spheres with the same radius. When rotated by 90° only 2 glare spots are visible as seen in the lower row image panel (f). These orientations yield distinct TAOS patterns, the one in the lower row (d) almost resembling the typical Mie pattern of a single sphere.

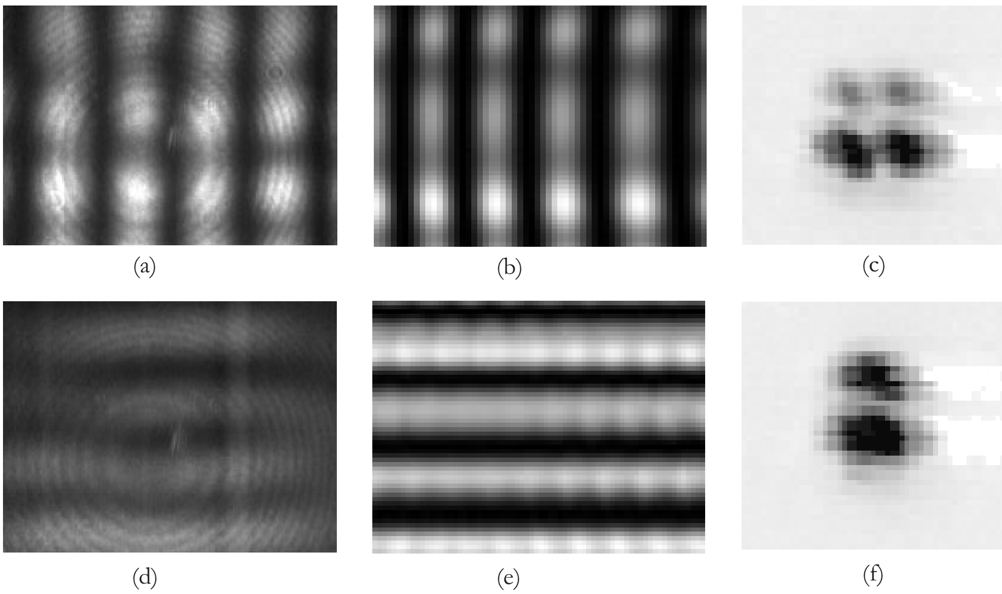

Figure 2. Observed (a, d) and calculated TAOS patterns (b, e) with microscopic images of a bi-sphere particle with single-sphere radii of 5.0 µm. Incident laser wavelength 488 nm.

A randomly picked TAOS pattern shows typically some distortion relative to the patterns shown in Fig. 2. Two examples are shown in Fig. 3. We do not know azimuthal and polar angles a priori and did not try to find these angles through comparison with calculations. But, in principle, comparison with a number of calculations performed over a sufficiently narrow grid should allow to estimate the angles of the laser beam relative to the bi-sphere coordinate system.

CONCLUSIONS AND OUTLOOK

Our setup allows us to measure the TAOS patterns of single, levitated aggregate sphere particles. Characterization of the particles is possible by comparing the observed and cal-

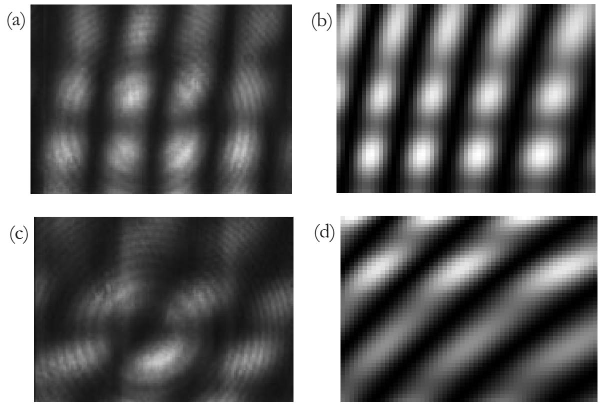

Figure 3. Observed TAOS patterns (a) and (c) which appear to be tilted relative to the ones shown in Fig. 2. Patterns originate from scattering of the same particle as in Fig. 2. For comparison, panels (b) and (d) show calculated TAOS patterns with angles α, β [2] of 0°, 15° and 0°, 45°, respectively.

culated TAOS patterns. In the future, we intend to study mixed particles, composites of polystyrene spheres and crystalline succinic acid, with the succinic acid crystallites evaporating slowly with time. This should allow us to analyze the restructuring of the composite particle by comparing its TAOS patterns with the corresponding calculations.

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