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Experimental light scattering by small particles: First results with a novel Mueller matrix scatterometer

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ABSTRACT

We describe a setup for measuring the full angular Mueller matrix profile of a single mm- to μm -sized sample, and verify the experimental results against a theoretical model. The scatterometer has a fixed or levitating sample, illuminated with a laser beam whose full polarization state is controlled. The scattered light is detected with a combination of wave retarder, linear polarizer, and photomultiplier tube that is attached to a rotational stage. The first results are reported.

Keywords: Scatterometry, polarimetry, single particle scattering, Mueller matrix measurement, ultrasonic levitation.

1. INTRODUCTION

Measuring scattering properties of different targets is important for material characterization, remote sensing applications, and for verifying theoretical results. Furthermore, there are usually simplifications made when we model targets and compute the scattering properties, e.g., ideal shape or constant optical parameters throughout the target material. Experimental studies can help us in understanding the link between the observed properties and computed results.

Experimentally derived Mueller matrices of particles can be used as input for larger-scale scattering simulations, e.g., radiative transfer computations. This method allows us to bypass the problem of using idealized model for single-particle properties. There are publicly available studies of the scattering properties of particles, e.g., the Granada light scattering database.¹ With our scatterometer, we aim to offer similar material for single, small (down to μm -scale) targets. While other sources usually offer ensemble- and orientation-averaged particle properties, we will be able to measure individual particles with controlled or known orientation.

2. SCATTEROMETER

We have developed a multi-angular Mueller matrix scatterometer capable of measuring the scattering profile of mm- to μm -sized targets. A more complete description of the equipment can be found in the paper by Maconi et al.,² and here we give a shorter overview.

The measurement can be done in several colors in the visual band. The measurement head comprises a combination of wave retarder, linear polarizer, and photomultiplier tube. One or several of these heads are attached to a rotational stage, so that a range of scattering angles can be covered. The schematic drawing of the measurement head and the stage is shown in Fig. 1.

The whole system is divided into two chambers that are divided by partition walls. The walls are covered with non-reflecting fabric with only a small pinhole for the incident beam to avoid excess reflections. All the optical components shaping the incident beam are seated in the right chamber (*b* in Fig. 2), and the scatterer and the detectors are located in the left chamber (*a* in Fig. 2). Furthermore, there is a second pinhole on the leftmost wall for the forward beam to be guided out from the measurement chamber and into a beam trap.

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Figure 1. Schematic overview of the levitating target in the middle, one measurement head, and the rotational stage.

Figure 2. Image of the current development version of the scatterometer. The sample is standing on a static conic pedestal in this image, and not levitated with the ultrasonic device.

The incident beam is generated with a tunable multimode Argon-krypton laser, with 12 selectable wavelengths ranging from 465 to 676 nm. The laser is placed outside the chambers to avoid excess heat and vibration, and the light is brought in with an optical fiber. In chamber *b* (Fig. 2), the beam is expanded, diffused, and shaped after the fiber output to create a speckle-free, collimated constant-intensity beam. The polarization state of the beam is controlled by rotation-controlled linear polarizer and wave retarder. The incident beam is monitored by a

photomultiplier tube (PMT) detecting the secondary reflection intensity from the system before the measurement chamber.

The incident beam encounters the scattering target in chamber *a* (Fig. 2). The target will be controlled with an advanced ultrasonic levitator. The levitator comprises several individually-controlled amplifiers and outputs to generate a complex ultrasound field. The shape of the field can be modified to generate a stable trap or even vortex forces in the trap. We aim to be able to control the target orientation in the trap with the ultrasound field, and the target position and orientation can be monitored using a high-speed camera attached to the system. The ultrasound trap is still under development, and the first tests presented here are executed with the target seated on a thin solid cone pedestal, or with a more simple single-amplifier levitator. An example of a 3-mm sphere levitating in the trap is shown in Fig. 3.

Figure 3. Single-amplifier ultrasonic levitator with a 3 mm glass sphere trapped in the upper node of the standing ultrasound wave.

The detectors, Hamamatsu micro-PMTs, are mounted radially towards the target on a rotational stage. The stage is controlled by a rotation motor with an accuracy of $15'$. The current 150-mm radius allows measuring all azimuthal angles except for $\pm 4^\circ$ around the backward scattering direction. With several detectors we can do a simultaneous multi-angle measurement, and by rotating the stage we can increase and sample more densely the angle range. The small physical dimensions of the new micro-PMTs allow us to seat many detectors side-by-side without compromising in the distance to the target too much.

3. FROM MEASUREMENTS TO MUELLER MATRIX

To control the polarization state of the incident beam we have a linear polarizer and a wave retarder. Similarly, before the PMT we have a wave retarder and a linear polarizer. In Stokes-Mueller presentation, our incident beam state can be expressed with the four Stokes vector components $\mathbf{I} = (I, Q, U, V)$, and any interaction with the beam by the left multiplication with the corresponding 4×4 Mueller matrix $\mathbf{M}$. The polarizer $\mathbf{M}_p$ and the quarter wave plate (wave retarder) $\mathbf{M}_q$, turned to angle θ from the horizontal plane, can be expressed with

Mueller matrices as

$$\begin{aligned} \mathbf{M}_p(\theta) &= \frac{1}{2} \begin{bmatrix} 1 & \cos 2\theta & \sin 2\theta & 0 \\ \cos 2\theta & \cos^2 2\theta & \sin 2\theta \cos 2\theta & 0 \\ \sin 2\theta & \sin 2\theta \cos 2\theta & \sin^2 2\theta & 0 \\ 0 & 0 & 0 & 0 \end{bmatrix}, \text{ and} \\ \mathbf{M}_q(\theta) &= \frac{1}{2} \begin{bmatrix} 1 & 0 & 0 & 0 \\ 0 & \cos^2 2\theta & \sin 2\theta \cos 2\theta & \sin 2\theta \\ 0 & \sin 2\theta \cos 2\theta & \sin^2 2\theta & -\cos 2\theta \\ 0 & -\sin 2\theta & \cos 2\theta & 0 \end{bmatrix} \end{aligned} \quad (1)$$

Therefore, the complete optical chain when measuring a particle (its scattered Stokes vector $\mathbf{I}_{sca}$) at direction α with Mueller matrix $\mathbf{M}$, is

$$\mathbf{I}_{sca} = \mathbf{M}_p(\theta_4) \mathbf{M}_q(\theta_3) \mathbf{M} \mathbf{M}_q(\theta_2) \mathbf{M}_p(\theta_1) \mathbf{I}_{inc}. \quad (2)$$

The PMTs can measure only intensity, not the complete Stokes vector. Thus, if not taking into account any possible symmetries in the Mueller matrix of the scatterer, we need 16 combinations of the rotation angles $\theta_1, \dots, \theta_4$ to measure all the 16 elements of the Mueller matrix $\mathbf{M}$.

At the first stage, we have omitted the wave retarders from the optical chain. Therefore we are able to measure the upper left 2×2 submatrix of the $\mathbf{M}$ with the following setup:

$$\begin{aligned} I_1 : \theta_1 = 0^\circ, \theta_4 = 0^\circ \\ I_2 : \theta_1 = 90^\circ, \theta_4 = 90^\circ \\ I_3 : \theta_1 = 0^\circ, \theta_4 = 90^\circ \\ I_4 : \theta_1 = 90^\circ, \theta_4 = 0^\circ, \end{aligned} \quad (3)$$

whereafter the Muller matrix elements can be derived from the measurements as:

$$\begin{aligned} M_{11} &= I_1 + I_2 + I_3 + I_4 \\ M_{12} &= I_1 - I_2 + I_3 - I_4 \\ M_{21} &= I_1 - I_2 - I_3 + I_4 \\ M_{22} &= I_1 + I_2 - I_3 - I_4. \end{aligned} \quad (4)$$

4. RESULTS

We have conducted the first calibration measurements with the scatterometer. The first set of measurements are done with one wavelength at 514 nm, with one detector that is rotated over the sample, and with linear polarizers only. The sample, which is a N-BK7 clear glass sphere from Edmund Optics, diameter $d = 5$ mm and refractive index $n = 1.5$, is mounted on a thin black cone (static sample).

We verify the first measurement set by comparing the angular scattering profile against the theoretical results computed using Mie theory. The Mie results are averaged over a small angular window to accommodate the actual acceptance angle of the detector. For the clear sphere, the symmetries require that $M_{11} = M_{22}$ and that $M_{12} = M_{21}$. In Figs. 4 and 5 we can see that the symmetries hold quite well. Also, comparing to model (Mie) results show quite nice agreement, taking into account that the cone holding the sphere will affect the results, especially in the off-diagonal elements M_{12} and M_{21} .

The second set of calibration measurements are done using $d = 3$ mm N-BK7 glass sphere that has matte surface, i.e., the surface has microroughness giving it a 'frosted' appearance. This treatment has the consequence that the sphere will have depolarization ($M_{11} > M_{22}$), and that comparing to Mie is not meaningful. Otherwise, the set-up is as in the first set of measurements, except that we will measure both static (on a pedestal) and levitating (using a simple ultrasonic levitator) samples. The results in Figs. 6 and 7 show that the levitation

Figure 4. Measurements and model for 5-mm clear static sphere, diagonal elements M_{11} and M_{22} . The measured elements overlap, so the measured M_{11} curve is behind the measured M_{22} .

Figure 5. Measurements and model for 5-mm clear static sphere, relative off-diagonal elements M_{12} and M_{21} . The measured elements overlap, so the measured M_{12} curve is behind the measured M_{21} .

is stable enough so that the Mueller matrix diagonal elements compare well to those of the static sample. For off-diagonal elements in Fig. 7, the cone pedestal is already having such a effect in the measurements that the comparison with the static sample is not meaningful. However, the two off-diagonal elements agree quite well with each other.

Figure 6. Measurements for 3-mm matte-surface static or levitating sphere, diagonal elements M_{11} and M_{22} .

Figure 7. Measurements for 3-mm matte-surface levitating sphere, off-diagonal relative elements M_{12} and M_{21}

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