

Validation of light scattering models with advanced 4π scatterometry

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Abstract

Within the ERC Advanced project ‘SAEMPL’ we have developed a novel orientation-controlled scatterometer – an instrument for precise full Mueller matrix measurement of light scattered by a mm- to μm -sized sample manipulated by ultrasound. The orientation of the sample is controlled by the custom-built acoustic levitator to allow non-destructive light scattering measurement in any solid angle, providing a full 4π measurement result. Additionally, high speed camera monitoring the sample is used for a photogrammetric 3D shape reconstruction of the sample. By providing robust experimental data for well-characterized samples we enable verification of the state-of-the-art light scattering models. Moreover, non-destructive non-contact measurements are also important for characterizing unique and valuable samples, such as cosmic dust and planetary samples.

1. Introduction

Computational Electromagnetics methods are used for modeling the light scattering response of single particle to more elaborate targets. These methods are computationally expensive and limited with respect to the size of the scatterer, compared to the wavelength [1]. On the other hand, scattering experiments, along with the sample characterization, are more convenient when done with particles moving in a flow, particle agglomerates, or particulate surfaces [2-5]. The aim of our project was to bridge this gap by going beyond the state of the art in both, theory and experiment.

One of the first scatterometry setups for small particle characterization was built in Arizona [6]. The system was used to characterize 110 nm diameter latex spheres. A more recent system was built in Amsterdam [7], and further developed at the IAA cosmic dust laboratory in Granada [2], to measure

scattering properties of irregularly shaped mineral samples. The main difference between these systems and our instrument is that they measure the statistical average of a flow of ‘running’ particles. On the other hand, the levitator keeps sample still, while absence of interfering sample holder in our experimental setup allows non-destructive non-contact measurements of well-characterized sample in all orientations [8].

2. Theoretical pipeline

For theoretical computations we utilize SAEMPL software suite, where light-scattering characteristics of the sample are modeled using novel multiple scattering methods for close-packed random media, such as a geometric optics method SIRIS4 [9] and the radiative transfer with reciprocal transactions R^2T^2 [10]. The R^2T^2 method solves the ensemble-averaged Foldy-Lax equation involving the ladder and maximally crossed diagrams as well as the near field corrections. The near field corrections are implemented in terms of the incoherent volume element containing all the scattering diagrams that do not cancel out in the near-zone [10]. The incoherent scattering parameters of the volume elements are solved exactly by the fast superposition T-matrix method [11]. The latter enables us to extend the applicability of the radiative transfer to close-packed random media [10].

3. Design of the scatterometer

The developed scatterometer comprises 4 parts, based on their function: the light source, the sample levitator, the analyzer, and the monitoring camera:

- The light source is a laser-stabilized arc lamp producing white light, which is then filtered by a line filter and polarized by a linear polarizer. A reference photomultiplier tube (PMT) is used for monitoring the

beam intensity, and the signal can be used to calibrate the observed signal power level.

- The acoustic levitator is custom built and features 400 transducer elements, grouped into 28 phase controlled channels. The resulting two hemispherical phased arrays produce an asymmetric acoustic trap which holds the sample in place at an adjustable orientation (heading, pitch and roll).
- The analyzer comprises a PMT with an integrated solid state high speed shutter, a motorized quarter wave plate, a motorized linear polarizer, and a motorized shutter. It is mounted on a large, motorized rotation stage, allowing it to scan the scattered light with an angular accuracy of 15'.
- The camera monitoring system allows to verify the stability of the ultrasonic levitation and orientation control. It features a high speed camera and near infrared LED illumination.

4. Validation results

With cross-validation purposes application of the software suite to the defined close-packed random media is followed by comparison with the experimental results. Among suitable planetary analogue samples for the experimental study we have defined, among others, macroscopic agglomerates formed by ballistic hit-and-stick deposition. The agglomerates consist of monodisperse SiO₂ spheres of known radius. The light-scattering characteristics of the formed of SiO₂ spheres agglomerates are thoroughly measured with the scatterometer. The shape and the packing density of the agglomerates are then obtained to independently enter into the theoretical pipeline along with the used in the measurements incident light wavelength and the refractive index of the sample material. The results obtained by studying the agglomerates composed of monodisperse SiO₂ spheres demonstrate, that application of the software suite for analyzing optical properties of close-packed random media matches well with the experimental results obtained using the described orientation-controlled scatterometer.

Acknowledgments

The ERC Advanced Grant No. 320773.

References

- [1] Mishchenko, M.I., Travis, L.D., Lacis, A.A., Scattering, absorption and emission of light by small particles. Cambridge University Press (2002).
- [2] Muñoz, O., Moreno, F., Guirado, D., Ramos, J. L., López, A., Girela, F., Jerónimo, J. M., Costillo, L. P., Bustamante, I., Experimental determination of scattering matrices of dust particles at visible wavelengths: The IAA light scattering apparatus. *JQSRT* 111(1), 187-196 (2010).
- [3] Videen, G., Kocifaj, M. (Eds.), Optics of cosmic dust (NATO Science Series). Kluwer Academic Publishers (2002).
- [4] Peltoniemi, J. I., Gritsevich, M., Hakala, T., Dagsson-Waldhauserová, P., Arnalds, Ó., Anttila, K., Hannula, H.-R., Kivekäs, N., Lihavainen, H., Meinander, O., Svensson, J., Virkkula, A., de Leeuw, G. Soot on snow experiment: bidirectional reflectance factor measurements of contaminated snow. *The Cryosphere* 9, 2323–2337, 2015.
- [5] Zubko N., Gritsevich M., Zubko E., Hakala T., Peltoniemi J.I., Optical measurements of chemically heterogeneous particulate surfaces. *JQSRT* 178, 422-431 (2016).
- [6] Hunt, A. J., Huffman, D. R., A new polarization-modulated light scattering instrument. *Review of Scientific Instruments*, 44(12), 1753-1762 (1973).
- [7] Volten, H, Munoz, O, Rol, E, de Haan, JF, Vassen, W, Hovenier, JW, Muinonen, K, Nousiainen, T. Scattering matrices of mineral aerosol particles at 441.6 nm and 632.8 nm. *Journal of Geophysical Research: Atmospheres*, 106, 17375-17401, 2001.
- [8] Maconi G., Penttilä A., Kassamakov I., Gritsevich M., Helander P., Puranen T., Salmi A., Hægström E., Muinonen K. (2018): Non-destructive controlled single-particle light scattering measurement. *JQSRT*, 204, 159–164.
- [9] Martikainen J., Penttilä A., Gritsevich M., Lindqvist H., Muinonen K. (2018): Spectral modeling of meteorites at UV-vis-NIR wavelengths. *JQSRT*, 204, 144–151.
- [10] Muinonen K., Markkanen J., Väisänen T., Peltoniemi J., Penttilä A. (2018) Multiple scattering of light in discrete random media using incoherent interactions. *Optics Letters* 43(4):683–686.
- [11] Markkanen J. and Yuffa A. J. (2017) Fast superposition T-matrix solution for clusters with arbitrarily-shaped constituent particles. *JQSRT* 189:181–189.