Angularly-resolved elastic scatter from single particles collected over a large solid angle and with high resolution

Kevin B Aptowicz and Richard K Chang
Center for Laser Diagnostics, Dept. of Applied Physics, Yale University, New Haven, CT 06520-8284, USA
E-mail: richard.chang@yale.edu

Abstract. Elastic light scattering from a single non-spherical particle of various morphologies has been measured simultaneously with a large angular range (90° < θ < 165° and 0° < ϕ < 360°) and with high angular resolution (1024 pixels in θ and 512 pixels in ϕ). Because the single-shot laser pulse is short (pulse duration of 70 ns), the tumbling and flowing particle can be treated as frozen in space. The large angle two-dimensional angular optical scattering (hereafter referred to as LA TAOS) intensity pattern, I(θ, ϕ), has been measured for a variety of particle morphology, such as the following: (1) single polystyrene latex (PSL) sphere; (2) cluster of PSL spheres; (3) single Bacillus subtilis (BG) spore; (4) cluster of BG spores; (5) dried aggregates of bio-aerosols as well as background clutter aerosols. All these measurements were made using the second harmonic of a Nd:YAG laser (0.532 µm). Islands structures in the LA TAOS patterns seem to be the prominent feature. Efforts are being made to extract metrics from these islands and compare them to theoretical results based on the T-matrix method.

Light scattering of small particles (size relative to the incident wavelength λ_{inc}) and their aggregates are of great interest to the electromagnetic computational community and to the homeland security concern. Surprisingly, particles and their aggregates are also of interest to the interstellar chemists. Light scattering from particles is a strong linear process and thereby experimentalists can use short pulse techniques to freeze the particle motion (translational and rotational of a particle or aggregate). Furthermore, the elastic scattering involves no change in wavelength and is coherent as it is dependent on the phase shift of different portions of the particle. The angular pattern of elastic scattering has the potential of providing the morphology information (size, shape, surface roughness, index of refraction [m(ω)] and inhomogeneities of the refractive index), for single particles and their aggregates.[1, 2, 3]

The inversion technique, which is far from being realized at the present, is to deduce the morphology information from the scattering angular distribution. For a spherical particle, Lorenz-Mie theory can be extended to large size parameter ka where k is 2π m(ω) / λ and a is the particle radius. For the simplest case, the sphere is assumed to be isotropic and homogenous and therefore its morphology can be totally defined by two parameters: radius (a) and the refractive index m(ω). Using a particle levitator and a tunable laser, and utilizing the rich optical resonance spectra implicit in Mie theory solutions, the radius and the refractive index of small spheres (~1 µm diameter) have been backed out with relative errors of 5×10^{-5}.[4, 5] However, extracting this information at a single wavelength in a ‘flow-through’ system has proven to be quite challenging. These systems have a lower signal-to-noise ratio which results in, when curve-fitting to Mie theory, multiple fits each with a
different radius and refractive index. The “flow-through” system designed by F.T. Gucker was capable of measuring the refractive index to within 0.7% and the radius to within 0.5%.[6]

For non-spherical particles and aggregates, some variation of the T-matrix formalism has been made. To calculate the angular scattering pattern, the computer processing time places a limitation on the size parameter of the primary particles and the number of primary particles forming the aggregates. Given the limitations of computation time, a few calculations on aggregates comprised of non-spherical primary particles have been attempted.[7, 8] Only recently have there been any experimental results over a large angular range that could be used to compare with the computational results. The main difficulty with such a comparison is the inability of the experimentalist to know the exact structure and orientation of the aggregate. Indeed, if the aggregate it made up of non-spherical primary particles, the orientation of each primary particle as well as the cluster needs to be determined. In addition, the internal structure of the aggregate (the relative location of the primary particles) also has to be known and feed into the numerical simulation.

In this paper we will try to summarize the current activities at Yale University involving the angular pattern of elastic scattering and its potential of extracting some information of the morphology of the particle. This work was done in collaboration with Ronald G. Pinnick, Steven C. Hill, and Richard L. Tober (U.S. Army Research Laboratory); Anish Goyal and Thomas Jeys (MIT Lincoln Laboratory); Burt V. Bronk (U.S. Air Force Research Laboratory at Edgewood Chemical and Biological Center) and Orazio I. Sindoni (Edgewood Chemical and Biological Center).

The LA TAOS (Large-Angle Two-dimensional Angular Optical Scattering) technique is sensitive to a particle’s morphology. To investigate the potential of this technique to discriminate between particles, LA TAOS patterns from a multitude of samples were measured. In addition, SEM (Scanning Electron Microscope) pictures were taken of these aerosols to assist in correlating particle’s structure to its LA TAOS pattern.

Figure 1 is a diagram of the setup employed for capturing LA TAOS patterns with a single laser pulse of 70 ns duration. The output nozzle of the aerosol generator forces the particles to travel downstream with a linear trajectory and form a focused column of particles with a beam diameter of 0.6 mm. The particles flow through the first focal point of an ellipsoidal reflector. At this stage, a triggering scheme (to be discussed in the next paragraph) fires a Q-switched Nd:YAG laser which is frequency doubled to produce light at 532 nm. The laser pulse (duration 70 ns) then illuminates the particle in the trigger volume leading to the scattering event. The ellipsoidal mirror reflects a solid angle greater than $2\pi$ of the scattered light to the second focal point. After propagating through a spatial filter located at the second focus, the scattered light is detected with an ICCD detector, which is also triggered by the triggering scheme.

![Figure 1. Set-up for capturing LA TAOS patterns.](image-url)
A trigger volume is defined by the intersection of two nearly perpendicular and tightly focused cw diode lasers ($\lambda = 635$ nm and 685 nm), as shown in Figure 2. When a particle traverses the trigger volume, the scattered light in the near forward direction is detected by two photomultipliers (PMTs with interference filters at 635 nm and 685 nm, respectively) at the same time. An AND gate output is produced and triggers the Q-switched Nd:YAG laser to fire as well as the ICCD detector to begin capturing an image.

**Figure 2.** Design of the cross-beam trigger system. Only when a particle is traversing the focal volume of the mirror does it scatter light from both diode beams and trigger the pulsed laser source.

**Figure 3.** The LA TAOS patterns for a single polystyrene sphere and a single Bacillus subtilis (BG) spore with their corresponding SEM micrographs, respectively.
As was shown by Kaye et al., an ellipsoidal mirror can be used to collect scattered light from single flowing aerosols over a huge angle range. However, aberrations due to the extremely short depth of field can distort the pattern and eliminate the finer features. By incorporating the cross-beam trigger system in the setup and using a pulsed laser source, the particle can be well localized at the focal point and ‘frozen’ with regard to translation and rotation. This has lead to LA TAOS patterns with angular resolution on the order of a quarter of a degree and labeling accuracy of those angles on the order of one degree. Using a spherical coordinate system, with the z-axis being defined by the laser propagation direction, scattered light is usually taken over the angular range $\theta = 165^\circ$ to $90^\circ$ and $\phi = 0^\circ$ to $360^\circ$.\[11\]

The left LA TAOS pattern in Figure 3 shows the expected rings associated with a single polystyrene latex (PSL) sphere of 1 µm in diameter. The right LA TAOS pattern is of a single randomly oriented *Bacillus subtilis* (BG) spore. The incident wavelength for both LA TAOS figures displayed above is at 532 nm. The only thing the experiment could not control for sequential shots is the orientation of the longer axis of the BG spores relative to the z-direction that is defined to be the laser propagation direction. Note that the LA TAOS pattern for the elongated BG spore is easily identifiable from that of the single PSL sphere.

![LA TAOS patterns from single clusters with single laser shots with pulse duration of 15 ns.](image)

**Figure 4:** LA TAOS patterns from single clusters with single laser shots with pulse duration of 15 ns. Therefore, it is assumed that the translation and rotation of the particles do not take place during this short time.

The LA TAOS pattern and the SEM micrograph of a cluster of polystyrene spheres made up of primary spheres (d = 1 µm) are shown on the left-hand side of Figure 4. On the right-hand side the LA TAOS pattern and the SEM micrograph of a cluster of BG spores (length = 1 µm, width = 0.5 µm)
are shown. Because of the surface roughness caused by the discrete nature of the primary particles, one sees a speckled pattern rather than the smooth rings found on the LA TAOS pattern of a single polystyrene sphere. The surface roughness of the BG cluster also gives the LA TAOS pattern a more textured look compared with that from a single BG spore. Such fine structures of speckled or textured patterns can only be observed in the back-scattered hemisphere ($\theta = 180^\circ$ to $90^\circ$). In the forward-scattered hemisphere ($\theta = 90^\circ$ to $0^\circ$), such fine resolution cannot be obtained. Dr. Jean-Claude Auger is currently tackling the problem of computing the theoretical LA TAOS patterns of a polystyrene sphere cluster and a BG cluster using the T-matrix method. He has already succeeded in computing the scattering from a cluster of spheres.[8] In addition, he has simulated the scattering from a single spore with a good degree of accuracy. The question now remains how to orient and position these single spores in a cluster to emulate those seen in the lab.

**Figure 5.** LA TAOS patterns and SEM images of a hodge-podge of aerosol particles. The center of the pattern would relate to the direct backward scattered light. The outer circle of the image corresponds to light scattered perpendicular to the laser axis. The black bar located on the right side of all the patterns is the beam block mount.
In addition to clusters of *Bacillus subtilis* spores and PSL spheres, LA TAOS patterns of other clusters were detected as shown in Figure 5. These clusters were generated using a Ink Jet Aerosol Generator which dries droplets composed of a suspension to form dry aggregates.[12] It should be noted that the aerosols generated with this generator are not always closely-packed structures. Indeed, many of them appear to be cracked shells or dimpled spheres. To understand why these structures occur, the physics at the surface of the drying droplets generated by the IJAG must be understood. Initially, as the droplets flow through the drying column, the droplets shrink isotropically. During this stage, a shell is formed at the air/water interface as particles pile up due to the receding boundary. This is due to the fact that the rate of the evaporation is much faster than the rate of diffusion.[13] The process continues until a thick shell is formed, after which further drying results in the shell either becoming deformed or cracked. Some examples of these shells are shown in the SEM pictures of Figure 5. The ammonium sulfate represents a cracked shell. The sodium chloride, which was expected to have a cubic-like structure, looks more similar to an igloo where small cubes form a spherical surface. Finally the tobacco particles look to be buckled spheres with noticeable dimples.

One prominent feature in all of the LA TAOS patterns is the high-frequency island-like structure. Currently, efforts are being made to extract information from these islands. Using image processing routines in Matlab® the island structures can be accentuated as shown in Figure 6. After a series of image processing steps, islands in the initial TAOS pattern are identified and shown as solid white islands in the final image. Note that the program removes islands that are too close the boundary of the collected LA TAOS pattern. The next step is to build up statistics on these islands for a particular aerosol type. Metrics that we are currently being investigated are the island’s area, orientation, and eccentricity, as well as the density of islands.

![Figure 6](image)

*Figure 6.* Output from image processing routine used to analyze islands structure found in the LA TAOS patterns. The islands found in the pattern are shown in the final image as solid white islands.
Also, for LA TAOS to be useful for bio-aerosol enrichment, the response time for analysis of the patterns must be in the microsecond regime. Using CCD technology with complex pattern recognition techniques, the processing time is more on the order of milliseconds, if not longer. Thus, a push is being made to use photo-multiplier tube (PMT) arrays (8x8) with supporting digital signal processing circuitry to discriminate between different aerosols. The image processing analysis of the islands structures will help us determine where to locate the cathodes of these PMTs.

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