Prototype Instrument Development for Measuring Directionality of Aerosol Light Scattering

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Prototype Instrument Development
for Measuring Directionality of Aerosol Light Scattering
by
Esther Koh Monroe

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of Washington University in partial fulfillment of the
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Investigation of aerosol interactions with electromagnetic waves provides insights into the scattering particles. Aerosol phase function, an angular distribution of scattered light, is a value required to calculate parameters used in direct radiative forcing (DRF) models in the atmosphere. Currently no direct measurement of phase function is available hence it is estimated from ground observations such as backscatter fraction $b$ and subsequently relating the parameters via Henyey-Greenstein (HG) approximation. This method has shown to introduce errors in radiation transfer models. HG phase function, in particular, does not account for particle microphysical properties such as shape and refractive index. Given the need for more accurate characterization of aerosol phase function, we developed and tested a prototype instrument for measuring directionality of aerosol light scattering. The study focused on increasing detection sensitivity and mechanical stability by understanding the relationships among instrument components in detail. The prototype was approximately 18” x 12” x 10” in dimensions, designed to be portable for use outside the laboratory and
had a final detectable scattering angle range of approximately $12^\circ < \theta < 167^\circ$. The design incorporated a low-angle elastic scattering detection setup by Ferri [11] and a custom-made elliptical mirror to capture scattered light across the x-plane, a plane perpendicular to the optical axis of the beam. Each of the 512 channels on linear photodiode detectors was matched to a range of polar scattering angles. The angle resolutions were approximately 0.03° and 0.5° for forward and side scattering respectively. The prototype was deployed in the NASA-NOAA FireX-AQ campaign in McCall, Idaho to measure the scattering phase function of aerosols emitted from biomass fuel samples from the Nethker wildfire site. Assuming a constant intensity at $\theta < 12^\circ$, we found the asymmetry parameter $g$ of aerosol field samples to be 0.73 ± 0.02. Subsequent analysis on scanning mobility particle sizer (SMPS) and transmission electron microscopy (TEM) showed that the samples had 500 nm mean diameter. The results demonstrated the feasibility of building a portable optical device for directly measuring aerosol phase function. Main challenges encountered during the field operation were off-axis aberration, focal position and volume control, and inflexible detection system. Following the field campaign, the aforementioned challenges were analyzed and potential near-term solutions were explored using computational modeling in Synopsys® LightTools.
Chapter 1

Introduction to Aerosol Light Scattering

1.1 Significance of light scattering by aerosols

Aerosols are stable clusters of solid or liquid particles suspended in gas. Their dimensions vary from nanometers to tens of microns and typically originate from either natural sources such as sand or from anthropogenic sources like biomass burning [5]. Aerosol particles influence direct radiative forcing (DRF) in the atmosphere by scattering or absorbing solar and thermal radiation, by serving as cloud condensation nuclei and by altering earth’s surface albedo [44]. To this date, aerosol effects on atmosphere’s radiative distribution are not well understood and have proven difficult to model due to their spatial and temporal variability [5]. Along with aerosol optical depth and single-scattering albedo, knowledge of forcing effect by aerosol particles, specifically the direction, is integral to constructing an accurate radiative transfer model [45]. Asymmetry parameter \( g \), a value that can be derived from ground-based in-situ measurements [3], is commonly used to represent directionality of particle scattering in the model. A backscatter fraction \( b \) is one example that can be measured directly using an integrating nephelometer with a backscatter shutter. Its value is then related to \( g \) using Henyey-Greenstein (HG) approximation. HG phase function only depends on a single parameter \( g \) [45] and does not account for particle microphysical properties [33] which has shown to increase errors in estimating phase functions for cases where coarse-mode particle distribution or pronounced backscattering peaks are present [1]. To improve model accuracy, researchers have utilized light scattering techniques as potential ways to directly measure the
particle phase function. Particle interactions with electromagnetic waves produce spectral patterns that carry information about the scattering particles. Ongoing research attempts to apply the existing light scattering theories to retrieving particle properties that can impact how particles scatter light and ultimately impact the radiation budget. However, there still exist uncertainties in obtaining particle morphology based on scattering profiles. The so-called 'backward problem' is prevalent in applications using light scattering methods like satellites and lidar [8]. To simplify transfer model calculations, certain assumptions are made on aerosol properties, one of which regards all particles to be spherical. This approach has shown to result in spectral radiance errors [56]. When it comes to empirical measurement of particle shapes, most analyses have relied on manual scanning under an electron microscope which can be time consuming [5]. Hence, there is a need for more accurate and convenient characterization of particle morphology. Several laboratory studies have attempted optical analysis of mineral dust [6] and ice crystals [8]. However, direct, in-situ measurements of particle shapes are still fairly limited [8]. This thesis, in particular, addresses the challenges associated with building a prototype for measuring directionality of aerosol light scattering. It focuses on short-wave scattering in the visible range by an ensemble of particles and the effects of multiple-scattering are not considered. Chapter 1 aims to cover basic particle light scattering concepts pertinent to the study.

1.2 Principles of light scattering by particles

1.2.1 Basic phenomena

When a photon hits a molecule, the energy sets off oscillating dipole which is seen as 'scattering' when the energy is re-emitted. A monochromatic beam can propagate without changing its frequency upon hitting a particle that scatters some of its energy. Such scattering is termed 'elastic' and it will serve as an assumption for this thesis. It is also assumed that the particles are randomly oriented hence the photons are free to scatter in all directions and that scattering happens without measurable wave interference. Lord Rayleigh used a dimensional analysis to show that the intensity of scattered light in the above scenario is proportional to $1/\lambda^4$ or $\nu^4$ [54].
Light scattering falls into one of three regimes: Rayleigh, Mie and Geometric. A dimensionless parameter $x$ defines which domain the particles exist (Equation (1.1)) where $d_p$ is a particle diameter in $\mu$m, $\lambda$ is source wavelength in $\mu$m, $k$ is circular wavenumber and $r_0$ is particle’s characteristic length [5]. If $x << 1$, Rayleigh scattering occurs, if $x >> 1$, light follows geometric scattering rules. When $x$ is approximately 1, scattering behavior can be described using Mie theory given that the particle is spherical, uniform, and isotropic.

$$x = \frac{\pi d_p}{\lambda} = kr_0$$

(1.1)

Figure 1.1 [28] shows typical aerosols found in each regime. The region relevant to this thesis is shaded yellow. A solid line is drawn at 532nm and an approximate region this thesis addresses is marked yellow. Under most circumstances, the study will involve Mie Scattering with potential Rayleigh scattering from surrounding gas.

In physical world, scattering by particles are dependent on other properties such as size, refractive index, wavelength of radiation, particle size distribution and density. A particle morphology also determines how the particle scatters light, whose angular dependence
increases with particle size. Since the scattering is spatially unique for complex particles, discrete measurements at each angle is required. This presents additional challenges to both empirical measurements and theoretical calculations. Often, spherical forms are assumed due to simplicity and ease of modeling as analytical solutions exist for Maxwell equations with spherical boundary conditions [8]. For particles with complex geometries or with inhomogeneous optical properties, Lorenz-Mie-Debye theory has no exact analytical solutions and requires a numerical calculation. It is also not always clear what \( r_0 \) in Equation 1.1 is for a given particle. T-matrix method proposes that \( r_0 \) is defined as a sphere radius of either equal volume or surface area [5].

1.2.2 Stokes vector and Mueller matrix

Stokes parameters - \( I, Q, U, V \) - are mathematical representation of electromagnetic waves where \( I \) refers to the total light intensity, and the signs of \( Q, U \) and \( V \) each indicate the linearity, angle and circularity of polarization. A Stokes vector (Figure 1.2) which is a vectorized form of Stokes parameters, can be used to relate the incident light to the scattered light via a 4-by-4 scattering matrix \( S \), wave number \( k \), and the distance to the detector \( r \) [8].

\[
\begin{pmatrix}
I_{sca} \\
Q_{sca} \\
U_{sca} \\
V_{sca}
\end{pmatrix} = \frac{1}{k^2 r^2} \begin{pmatrix}
I_{inc} \\
Q_{inc} \\
U_{inc} \\
V_{inc}
\end{pmatrix} S
\]

Figure 1.2: Stokes vector [8]

Scattering matrix (Figure 1.3), often referred to as Mueller matrix, captures the sample or environment properties that potentially alter the beam polarization state and its amplitude. Examples include surfaces, polarizers or optics. Mueller matrix is a function of beam wavelength and propagation direction [46] and each element is intensive with respect to particle size, shape and refractive index [8]. The matrices are known for some common polarization elements.
The first element, \( S_{11}(\theta) \), of the resulting 4-by-4 matrix gives the intensity of total scattered light at an angle \( \theta \) when the particles are assumed to take up random orientations (Equation 1.2).

\[
I = \frac{1}{k^2 r^2} S_{11}(\theta) I_0
\]  

(1.2)

1.2.3 Q-space analysis

Q-space analysis plots the measured intensity against scattering wave vector \( q \) (Equation 1.3) in log-log scale to derive particle size and shape using power laws. The theory effectively connects the real space (r-space) to reciprocal space (q-space) via Fourier transform [14], [36]. In Figure 1.4, an incident scalar wave hits a particle located at \( \vec{r} \) where the incident field will be \( E(\vec{r}) = E_0 exp(i \vec{k} \cdot \vec{r}) \).

Assuming elastic scattering and paraxial approximation, a magnitude of \( q \) is then:

\[
q = 2k\sin\frac{\theta}{2}
\]  

(1.3)

and the resulting amplitude of the scattered wave is [14]:

\[
E_{sca}(\vec{q}, \vec{r}) \sim E_0 exp(i \vec{q} \cdot \vec{r})
\]  

(1.4)
Scattering wave vector, $\vec{q}$, is a difference between an incoming wave vector $k_i$ and scattered wave vector $k_s$. The solution also assumes a superposition of waves from individual scatterers to equal the total scattered intensity. This implies an absence of multiple scattering within the object and that uniform illumination is present. Such leads to Rayleigh-Debye-Gans (RDG) approximation. Writing the density function ($n(\vec{r})$) of scatterers as Dirac delta function, a sum of individual amplitudes can be written in integral:

$$E_{sca}(q) \sim E_0 \int \exp(i\vec{q} \cdot \vec{r}) n(\vec{r}) d\vec{r}$$

(1.5)

For cases where one can expect scattering from ensemble of particles, the vector nature of $q$ is disregarded. The patterns in q-space are best understood using a structure factor, $S(q)$, which is squared complex amplitude of Fourier transformed r-space structure normalized by the number of scatterers (Equation 1.6) \[14\]. This simplification is justified as intensity itself does not depend on wave’s electromagnetic properties or complex index of refraction.

$$S(q) = |V^{-1} \int \exp(i\vec{q} \cdot \vec{r}) n(\vec{r}) d\vec{r}|^2$$

(1.6)
The above structure factor for spherical particles (Equation 1.7) can be approximated by Guinier plot in q-space and a study of resulting pattern can reveal information about particle size and shape as measured in ensemble that satisfies the above q-space assumptions [14].

\[ P(q) \approx 1 - \frac{(qR_g)^2}{3} \] (1.7)

1.3 Directionality of particle light scattering

In single column radiation models, upscatter fraction \( \beta \) is the amount of solar radiation scattered back into space and for diffuse radiation, it is averaged over the earth’s hemisphere. Equation 1.8 shows an expression for average upscatter function (\( \overline{\beta} \)) [23].

\[ \overline{\beta} = \int_0^\pi \overline{\beta}(\theta_0)\sin(\theta_0)d\theta_0 \] (1.8)

\( \overline{\beta}(\theta_0) \) is light scattered at solar zenith angle \( \theta_0 \) and is an integral of aerosol phase function \( P(\theta, \phi) \) over the scattering solid angles, normalized to \( 4\pi \). This implies that \( \overline{\beta} \) used in aerosol radiative forcing calculation can be derived when \( P(\theta) \) is known [23]. The aerosol phase function, often parameterized as asymmetry factor \( g \) is an important parameter in improving the accuracy of radiative forcing calculations along with aerosol optical depth and single-scattering albedo. The scattering phase function \( S_{11} \) is an angular distribution of scattered light and it varies with particle size, refractive index, and shape [1]. Phase function quantifies scattered light as a function of incident beam directions, \( \theta' \) and \( \phi' \), as well as polar and azimuthal scattering angles, \( \theta \) and \( \phi \) [52]. Where there is no preference for scattering direction, the phase function becomes dependent only on scattering angle, \( \theta \). The phase function for a single, spherical particle, illuminated with unpolarized light, is a function of scattering cross section \( (C_{sca}) \), incident beam vector \( (k) \), particle size \( (r) \), and amplitudes of surrounding electric fields [5] (Equation (1.9)):

\[ P(\theta) = \frac{2}{C_{sca}kr^2}(|S_1(\theta)|^2 + |S_2(\theta)|^2) \] (1.9)
The total scattered intensity at a given $\theta$ is defined in Equation 1.10 where $I_0$ is the incident irradiance [5].

$$I(\theta) = C_{sca} P(\theta) I_0 \quad (1.10)$$

Where $S_{11}$ cannot be directly measured, Mie theory can be used to derive the phase function for spherical, homogeneous particles but its application to non-spherical, inhomogeneous particles is limited. As there is currently a lack of reliable field measurement tools for aerosol phase function, estimation of the phase function relies on other ground measurements. One example of direct measurement is backscatter fraction $b$, which corresponds to all scattered intensity from back-half of the hemisphere. $b$ can be measured using an integrating nephelometer with a shutter. The value is generally wavelength-independent for 442-676 nm, though past studies have shown that for a single angle measurement, the value was subject to roughly 10\% error in estimation [20]. Other factors such as particle dielectric properties and morphology also have potential to bias backscatter ratio [21]. For accumulation mode particles with well characterized size distribution, it is possible to relate $b$ to $g$. This relationship is not always clear however, and can be multi-valued for non-spherical particles, making model intercomparisons difficult.

Asymmetry parameter $g$ is a parameterization of the estimated aerosol phase function. One could calculate $g$ by integrating the cosines of the scattering angles, weighted by the normalized phase function. It is essentially a probability density function across all solid angles [5], [45] (Equation (1.11)):

$$g = \frac{1}{2} \int_0^\pi p(\theta) \cos(\theta) \sin \theta d\theta = \int_{4\pi} p(\theta) \cos(\theta) d\Omega \quad (1.11)$$

where $d\Omega = \sin \theta d\theta d\psi$ [43]. $g$ obtains a value between -1 and 1 to measure the fraction of scattering intensity along forward direction which generally increases with particle size. For example, $g = 1$ indicates a complete forward scattering and -1 an entirely backscattering. When $g = 0$, scattering is isotropic and symmetric about $\theta = 90$ [6]. An asymmetry parameter also approaches zero as the particle radii gets smaller compared to the wavelength and a preferential scattering hemisphere starts to decrease [15]. Negative $g$ is rare for ambient measurements since it only happens for small metallic particles [33]. Common sources of $g$
are ground ambient measurements and Mie theory calculations for homogeneous spherical particles or T-matrix and coupled dipole-dipole methods for more complex particles [33].

1.3.1 Henyey-Greenstein (HG) phase function

Henyey-Greenstein phase function ($P_{HG}$) is an analytical approximation of the aerosol phase function, found using a single parameter $g$ (Equation 1.12), and does not account for particle microphysical properties such as size, refractive index and shape [23].

$$P_{HG} = \frac{1 - g^2}{(1 + g^2 - 2g\cos\theta)^{\frac{1}{2}}}$$

Past studies have attempted to connect $b$, and $g$ using HG phase function and calculated expected root mean square error (RMSE) within defined atmospheric conditions [23]. Lack of direct measurement tools have led to wide use of HG phase function in determining aerosol scattering directionality. However, some studies have found that $P_{HG}$ can introduce errors in radiation transfer calculations. A study by Boucher (1997) indicated that $P_{HG}$ cannot accurately produce back scattering peak thus overestimates Mie phase function at $\theta$ between 90 and 150. For same $g$, notable disagreements were found between monodirectional upscatter fraction $\beta(\mu_0)$ from Mie theory and that calculated using HG phase function [1]. Using $P_{HG}$ in $\beta$ calculation has been reported to systematically underestimate the climate forcing effect compared to Mie theory. For example, a 10% error on $g$ can decrease climate forcing by 12% or more [24]. The error in estimation seemed to increase for coarse-mode size distributions, though the prediction improved when concerning more absorbing particles. Also, wider spread of particle sizes reduced discrepancies between $P_{HG}$ and Mie phase functions for accumulation-mode particles but not for coarse-mode [1]. This poses challenges in accurately modeling aerosol effects in atmosphere where scattering dominates such as urban environments with radiation wavelength over 300nm [34] and locations with a higher concentration of coarse-mode particles generated from soil, sea-salt, volcanic ash and plants.
Chapter 2

Instrumentation for Angular Light Scattering Measurement

2.1 Review of instruments

The efforts to quantify particle angular light scattering have been present for a while. In 1949, R G Beuttell and A W Brewer built a visibility meter to measure scattering coefficient of the atmosphere, starting an era of optical instrumentation for atmospheric aerosols. They proposed two designs, one of which applied parallel light to a sample and another which applied a cosine-law diffuse light \[32\]. A cosine detector had to be used with parallel beam to form an arrangement termed ‘reciprocal nephelometer’ \[31\]. Sinclair and La Mer designed the OWL in the same year and it was one of the earliest instruments measuring aerosol phase function. The device had sample flow through a cylindrical cell while a telescope rotated in relation to the incident beam to measure angles sequentially. Despite its mechanical crudeness, the results when compared to Lorenz-Mie theory, were highly accurate. In 1973, Gucker et al. used a high-speed photometer coupled with oscillogram to capture 7-173° and 187-353° scattering angles of single polystyrene latex particle \[9\]. Perry et al. (1978) designed an optical device capable of measuring all elements of Mueller matrix, making a stride towards analyzing nonspherical particles \[19\]. A photometer developed by Bartholdi et al. (1980) provided a basis for an early optical scheme of integrating nephelometers and polar nephelometers, which since became standard tools for measuring aerosol backscatter fraction \[17\]. The device captured 2.5 to 177.5° scattering angles with 60 photodiodes by illuminating an annular strip of an ellipsoidal reflector focal point. Similar designs leveraging
elliptical optic have since been implemented frequently. A polar nephelometer designed by Kaller (2003) had a beam pass through an ellipsoidal mirror through holes on the side to illuminate sample at one focal point. The instrument only detected 0.16° at a time using photomultiplier tube connected to a stepper motor and ran in a chopper mode around 200 Hz to minimize stray light[26]. There were also attempts to use paraboloidal mirrors in place of elliptical ones. Gayet et al. (1996) published a comprehensive study of a polar nephelometer whose design used a paraboloidal reflector (Figure 2.1) and discrete optic fibers connected to photodiodes [27].

Figure 2.1: Configuration of polar nephelometer using a paraboloidal mirror [27]

The instrument analyzed clouds with water droplets or ice crystals, ranging from few micrometers to 500 µm in size. A circular array of 33 photodiodes detected 3.49° to 169° scattering angles. The reported scattering volume was large (0.2 cm³), resulting from 5 mm laser beam width and 10 mm inlet tube diameter. Basic optical configurations of these nephelometers have remained similar and more advances have evolved around laser, electronics and software used in instrument. One example of such development is incorporation of charge-coupled detectors (CCD) to capture 2-D scattering radiant intensity as an alternative to reading a one-dimensional spectral profiles. In 2006, Aptowicz et al. published a study on two-dimensional angular optical scattering (TAOS) patterns of various particle
morphologies. The apparatus used an ellipsoidal reflector to collect 75-135° polar scattering angles and all 360° azimuth angles. A 1024 x 1024 intensified CCD was able to measure a flowing stream of particles in real-time (Figure 2.2) [10].

Despite the advances in these nephelometers, a common challenge of ‘truncation angles’ still exists. Ideal nephelometers would integrate over the entire 4π steradians, but in reality most available nephelometers have to forgo detection of roughly 7° of near-forward and near-backward scattering angles due to non-idealities in instrument design and imperfect particle sampling. Errors in angular response, for example, are consequences of detected intensity that is not cosine weighted while varying wavelength response happens when integrating scattering over a wavelength range. Sampling losses due to impaction and settling also contribute to errors [31]. Resulting truncation error $T$ can be as large as a factor of 2 for particles where size parameter $x > 1$ [23]. This is due to the increased fraction of forward scattering as the particle size increases. A survey of available nephelometers reveal that current designs have difficulties separating the illuminating beam from forward scattering light. Figure 2.3 shows a significant increase in stray light as the angle approaches 0° [2]. Radiant intensity was detected at polar angles 23.1, 29.0, 35.0, 41.0, 47.2, 53.5, 60.0, 66.7, 73.7, 81.1, 88.9, 97.2, 106.3, 116.4, and 128.3 degrees. $T$ is often estimated using Mie theory which cannot be effectively applied to nonspherical particles. For particles with mean volume diameters between 0.2 and 0.4 µm, the error is within 10%. However, as the particle size
increases, the errors can be as high as 20-50%. To reduce the error, diffraction theory can be used to calculate < $10^\circ$ truncation angles [23] as a majority of scattered light off of absorbing particles is due to diffraction and roughly 50% for nonabsorbing particles [31].

Figure 2.3: Detected radiant intensity of stray light as a function of polar angles [2]

Given the significance of truncation angles on scattering measurement, some studies focused on improving instrumentation design to reduce truncation angles. The Integrating Sphere Integrating Nephelometer (ISIN) by Varma and Moosmuller (2003) is a reciprocal nephelometer using a 20 cm diameter integrating sphere, coated in near-Lambertian barium sulfate inside. The illuminating laser is modulated by a chopper and spatially filtered to reduce scattering off the wall. Truncation reduction tubes attach at both sphere apertures while a straight, vertical flow path minimizes sample loss (Figure 2.4). The instrument was able to measure angles between 1° and 179°, increasing detectable particle size by seven-fold with same truncation error [31].

Past studies on low-angle elastic light scattering (LAELS) techniques can also be used to maximize detection of near-forward angles. In 1997, Ferri published a paper on LAELS for particle sizing using a charge-coupled detector [11]. LAELS incorporates a beam stop at a focal plane of lens collecting the scattered light. Another lens is placed between the collecting lens and CCD sensor to conjugate the two planes (Figure 2.5). This achieves one-to-one mapping between each pixel and scattering angle, $\theta$. Magnification (M) of such
system is $Q_2/P_2$. The system was able to obtain approximately two decades in angular range from $0.1^\circ$ to $10^\circ$ which helped to capture most of forward scattering.

Overall, a rise in demands for more reliable, faster and cheaper instrumentation for characterizing particle light scattering has been observed. Though much progress has been made in detection and processing of particle angular light scattering patterns, challenges still remain in transforming the lab-based instruments into one that is deployable where its value
will be greatly increased. Currently, there is no truly portable and scalable field instrument to directly measure aerosol phase function and the value is often derived from other ground measurements, introducing errors in the aerosol forcing estimation [45]. Difficulties in designing reliable optical instrument stem from factors such as optical aberrations, elaborate alignment and calibration procedures required throughout the operation, and high costs. The instrument presented in this study attempts to investigate and tackle some of these challenges.

2.2 Introduction to optical instrumentation design

Optical interrogation provides a non-invasive and fast measurement of various aerosol properties. Quantum theory shows that photon energy is inversely proportional to the wavelength of radiation and that all light measurements have unique spectral, spatial and temporal distributions [53]. The optical techniques focus on quantifying light scattering phenomena as explained in Section 1.2. Fundamentally, optical analysis of particles focuses on irradiance (W/m²) which is power per unit area or radiant intensity (W/sr), which is power per unit solid angle [47]. With respect to light scattering detection, radiant intensity, which refers to the amount of light emitted by a source in particular direction is often a subject of interest. As light travels through space and interacts with matter, it follows a set of principles. Some most relevant to scattering detection are:

- The law of reflection states that the angle of reflection is equal to angle of incidence. It is a property that most planar mirrors with smooth, reflective surface obey and is termed ‘specular’ reflection. When the surface is not uniform, the reflection is ‘diffuse’. A combination of the two is called a ‘spread’ reflection [53]. This serves as a foundation for reconstructing the points where the scattered rays travel from.

- Refraction can also occur when light hits a particle as it passes between two dissimilar medium. Snell’s law (Equation 2.1) relates a degree of refraction to an incident angle and refractive indices of the materials. Refraction becomes important for partially transmitting or optically dense aerosol samples.

$$n_1 \sin(\theta_1) = n_2 \sin(\theta_2)$$  (2.1)
• Diffraction happens as a result of light bending at the edge of an aperture (Equation 2.2). D is the aperture diameter of the object light is passing through and λ is the beam wavelength. Diffraction can pose issues in misaligned optics by generating unwanted stray light and increasing noise.

$$\theta = \frac{\lambda}{D}$$  (2.2)

• Lambert’s Cosine Law states that intensity of light per unit surface is a cosine of the incident angle. This law can be used for calculating exiting radiance or total area detected assuming the surface is Lambertian. When the signal is measured off-normal, a correction on intensity per observed solid angle is required in accordance with the Cosine Law.

Optical detection obeys the above rules to retrieve particle properties such as size, refractive index and morphology from acquired scattering profile. Due to the complexity of particle-light interactions, defining target measurement parameters can be helpful in designing the appropriate instrumentation for chosen applications. Generally speaking, a set of optics is used to focus an illumination beam onto a sample, collect and direct the scattered light to a detector. The specifications and arrangements of these optics as well as the detectors depend on estimated sample properties and operation conditions. Design of an optical system begins with understanding functional roles of all main design components that impact the light detection accuracy and sensitivity. Examples of such components include optical focal lengths, field of view, wavelength and environmental conditions. A list of general considerations for building optical instrumentation is provided in Figure 2.6 [58] and criteria which this study particularly focuses on are highlighted yellow.

2.3 Measurement principles of static light scattering

Static light scattering (SLS) tracks changing light-particle interactions as a function of spatial locations rather than time as in the case of dynamic light scattering. It detects specific intensity at each scattering angle θ as described by Figure 2.7. \( d\sigma \) is an arbitrary area from which light is emitted at θ with respect to a normal vector n. \( S \) is an axis to the cone of solid
Checklist of General Design Features Typically Included in Specifications and Constraint Definitions for Optical Instruments

- Performance requirements such as resolution, MTF at specified spatial frequencies, radial energy distribution, encircled or beamsquared energy at specific wavelengths, or numerical aperture
- Focal length, magnification (if system is afocal), magnification and object-to-image track length (if system has finite conjugates)
- Angular or linear field of view (in specified meridians if anamorphic)
- Entrance and exit pupil sizes and locations
- Spectral transmission requirements
- Image orientation
- Sensor characteristics such as dimensions, spectral response, element size and spacing, and/or frequency response
- Size, shape, and weight limitations
- Survival and operating environmental conditions
- Interfaces (optical, mechanical, electrical, thermal, etc.)
- Thermal stability requirements
- Duty cycle and useful life requirements
- Maintenance and servicing provisions (access, fits, clearances, torquing, etc.)
- Emergency or overload conditions
- Center of gravity (CG) location and lifting provisions
- Human–instrument interface requirements and restrictions (including safety aspects)
- Electrical requirements and restrictions (power consumption, frequency, phase, grounding, etc.)
- Material selection recommendations and limitations
- Finish/color requirements
- Corrosion, fungus, rain, sand/dust, and salt spray erosion protection requirements
- Inspection and test provisions
- Electromagnetic interference restrictions and susceptibility
- Special markings or identifications
- Related consumables

Figure 2.6: General design considerations for optical instruments
angle $d\omega$. The technique has more applications in anisotropic scattering, where scattering intensity varies with angle and where constructive or destructive interference of rays can happen. The resulting theta-dependent distribution of scattered intensities is considered a phase function. When a particle radius of gyration $R_g$ is sufficiently small compared to the wavelength, the particle scatters isotropically. However, as the particle increases in size or becomes more irregular, the light scattering pattern increasingly depends on the scattering angle and SLS technique can be useful in characterizing the scattering object. SLS is applicable to both single-particle and an ensemble of particles detection. In either case the intensity of scattered light is measured according to Equation 1.9 but multiple scattering in a highly concentrated ensemble would have to be accounted for. The total intensity varies with respect to instrument configuration, sample scattering volume, and the distance between scattering particle and the detector, which follows the inverse square law where irradiance is inversely proportional to the square of the distance between scattering sample and a detector [53]. Equation 1.10 implies that a deviation of phase function from 1 will impact the detected intensity assuming all else is constant. The resulting pattern is associated with particle morphology and past studies have attempted to identify the relationship between the normalized intensity and particle non-sphericity [14]. In the following chapters, we focus on minimizing the variability of instrumentation constant through robust instrument design and optimizing scattering volume. These developments will allow a direct measurement of $P(\theta)$, our ultimate aim.
Chapter 3

Development of Prototype Instrument

3.1 Design principles and objectives

The prototype presented in this thesis is a static light scattering detection device that quantifies the spatial distribution of scattered light. Each detector channel sees a particular polar scattering angle that can mathematically be related to the particle’s phase function as noted in Section 1.3. The prototype (Figure 3.2) is a modification to an existing design proposed by Heinson [6], built using the same physical parts from the previous set-up. The original device was confined to a fixed laboratory experiment with minimal automation capabilities (Figure 3.1). The device was operated during a field campaign from which we identified areas for improvements which are further outlined in Chapter 5. The main objective of the improved design was to increase the portability, robustness and sensitivity of the instrument. The design incorporates a low-angle elastic scattering detection setup by Ferri [11] and a custom-made elliptical mirror to capture scattered light across x-plane, a plane perpendicular to the optical axis of the beam. Figure 3.3 shows an overall functional diagram of the instrument. Chapter 3 explains the details and functionalities of each components and relevant design principles.
Figure 3.1: A picture of previous laboratory-based setup

Figure 3.2: A picture of the prototype in development
3.2 Components

3.2.1 532nm laser

532nm OBIS LX/LS laser (530-50 LS, Coherent) was used as an illumination source. It is a TEM$_{00}$ (Gaussian) laser with a 0.7 ± 0.05mm beam diameter at $1/e^2$. An average warm-up time is less than 5 minutes [60]. Relevant considerations when selecting a laser for optical system include temporal and spatial coherence, pulse generation and wavelength [59]. For the purpose of this study, shifts in temporal coherence is treated as negligible assuming that light mostly travels through medium or optics with small refractive index. Both spatial coherence and pulse stability of the above OBIS laser were considered adequate for current stage of instrument development. The wavelength was chosen with regards to expected average particle diameter to be detected and cost of laser. It is helpful to note that the amplitudes of scattered light increases with driving frequency [54] which has an inverse relationship with the wavelength.
3.2.2 Wave plates

Optical wave plates alter the polarization state of an incoming light. Made from birefringent materials such as crystal quartz, waveplates have different indices of refraction depending on the beam orientation. The difference makes the light to travel at different velocities along the fast and slow axes of the wave plate, leading to a phase change. Polarized light, represented by elements Q, U, and V in Stokes vector, can be used to infer all elements of scattering matrix. The phase function, $S_{11}(\theta)$, is the total scattered intensity, $I$, when the incident light is unpolarized or circularly polarized [6]. In this study, a zero-order, quarter-wave plate (WPQ10M-532, Thorlabs) was used to convert a linearly polarized laser light to a circularly polarized beam.

3.2.3 Dielectric mirror

Fused silica broadband dielectric mirror (BB1-E02, Thorlabs) was used to divert the incoming laser beam 90 degrees. The outer lens diameter is 25.4mm and its design wavelength is 400 to 750nm. At 532nm, the reflectance efficiency is close to 99 percent for both S- and P-polarization with angles of incidence ranging from 0 to 45 degrees. The mirror serves a critical role in directing and focusing the laser beam at a correct spot for sample detection as well as dumping. It is a standalone structure in the system therefore is vulnerable to mechanical shocks. Two main factors affecting mirror performance are optical figure and position of the reflecting surface [58]. Of the two, the position tolerance is more significant in instrument operation as a deviation with respect to optical axis will lead to lower signal-to-noise.

3.2.4 Elliptical reflector

Elliptical reflector (E115NV) with custom dimensions (Figure 3.4) was manufactured by Optiforms (Temecula, CA) [6] and used for simultaneously collecting all side scattering by the sample. Based on the properties of an ellipse, a ray from the primary focal point (F1) will be directed to its secondary focal point (F2). In the prototype, an aerosol sample is illuminated at the primary or 'near' focal point and scattered light between angles 12.01
Figure 3.4: Dimensions of elliptical reflector. (a) is cross-section of the physical reflector and (b) is geometry of the base ellipse.

to 167.99 degrees can theoretically be captured at the secondary or 'far' focal point by a detector.

### 3.2.5 Side scattering enclosure

One major drawback of the previous design was an open set-up for collecting side scattering (Figure 3.1) which required use of two distinct lasers to visually locate an exact position of aerosols flowing from the pump. Such alignment method proved to be time-consuming and less precise. An open design also meant that the optics were more susceptible to mechanical shocks and stray light. To reduce set up time and potential noise sources, an aluminum enclosure for elliptical mirror was designed and machined in-house (Figure 3.5). Enclosing an entire side scattering set-up offers three major benefits: easier alignment, blocking of unwanted stray light and higher mechanical stability. The physical dimensions of the enclosure matched the geometry of the ellipse with special attention to the distances between the mirror and each of the optical focal points. The critical control points for enclosure design were:
• Distance from the mirror edge to the first focal point
• Distance from the mirror edge to the second focal point
• Mirror situated perpendicular to the optical axis and straight without any tilt
• Exit iris aligned with both focal points
• Tube inlet/outlet holes vertically aligned at the center of the mirror

The hole for sample inlet was placed at the near focal point (F1), which was 17.09mm away from the back center of the elliptical mirror. The inner diameter of copper sample tube was shrunk from a 1/4” to a 3/16” for a more focused, smaller scattering volume at F1. A 1mm hole was drilled at the far focal point (F2) to function as both the aperture and field stop. Further details of the iris is provided in Section 3.2.6. The distance between the center of the sample inlet and the iris was 114.62mm. The width of the box was as close as possible to 51.03mm - a physical aperture diameter of the elliptical reflector - to reduce the laser beam path length through the enclosure. 1” holes were drilled and tapped on each sides to thread the lens tubes in for easy alignment. An achromat doublet lens (discussed in Section 3.2.6), which collects scattered light and relays it to the pinhole mirror (discussed in Section 3.2.7), was placed 43.4mm away from F1, corresponding to its back focal length, to focus its minimum spot size on the sample. Not shown in the picture, inside walls as well as the external sides of the enclosure were covered with non-reflective black laser cloth to prevent scattering of stray light. To avoid leakage of aspired particles, a vacuum system was installed through the enclosure lid to pump out excess particles. Overall, the enclosure contributed to reduced alignment error via built-in couplings, more mechanical stability, and a lower probability of signal interference by excess sample aerosols floating inside the instrument.

3.2.6 Stops and iris

The iris at far focal point (F2) of the ellipse is an aperture stop in that it controls the amount of light accepted by the side detector. It passes the collected light by the ellipsoid reflector to the detector while blocking ‘unwanted’ direct scattering by the particles. The aperture works to reduce the spherical aberration when positioned at the intersection of marginal rays
Figure 3.5: A picture of the side scattering enclosure

and their caustic envelope [54]. In principle, the field of view (FOV) of the detector is set by its subtended visual angle [39] but in an enclosed system, it is ultimately decided by a field stop. Current prototype is considered one-aperture system where the iris diameter will determine the angle range the detector can see. It is important that scattered rays focus at F2 to avoid mechanical vignetting. Placing baffles between between the exit aperture and detector can also help reduce stray light from reaching the detector.

3.2.7 Achromatic doublets

Achromatized lens is a combination of two lenses to mitigate the effects of spherical aberrations. This two-lens system allows for tighter focusing of collected rays which contributes to more accurate light scattering detection. Achromatic doublets (3.6) in the instrument serve functions of Fourier lens (AC254-050-A-ML, Thorlabs) and imaging lens (AC254-035-A-ML, Thorlabs). Both lenses were 25.4mm in diameter and coated for visible spectral region (400 to 700 nm). First achromat lens was aligned along the optical axis, 43.4mm away from the aerosol sample point. The distance was equal to the back focal length ($f_b$) of the lens rather than the focal length since a physical measurement of $f_b$ was more attainable [41]. A diameter of the first achromat determines the forward polar scattering angles that can be
Figure 3.6: Achromatic doublet [41]

captured by the lens. The rays that reach the concave surface of the lens emerge as parallel which then can be relayed to the pinhole mirror for reflection. Accurate placements of these optics are needed to ensure that information obtained via optical fourier transform is not lost. Reflected rays from the pinhole mirror are collected by the second achromatic doublet which focuses the image at its focal length (35mm). The rays then diverges infinitely until it reaches a detector. A calculation of angular Field of View (AFOV) is required to determine where the detector has to be positioned so its length maps the lens image plane or, in other words, the individual scattering angles (Equation 3.1) [40].

\[ AFOV(^\circ) = 2tan^{-1}\left(\frac{h}{2f}\right) \]  

(3.1)

\( f \) is the focal length of the lens and \( h \) is the horizontal dimension of the detector. The units are in mm. The equation shows that a shorter focal length gives a wider FOV. This set-up optimizes detectable angle resolution as the angles can be spread across all available photodiode channels. For minimum spherical aberrations, the flat side of achromatIC doublets should be facing the desired focus direction [54].

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3.2.8 Pinhole mirror

Pinhole mirror has a through-hole at the center to let the laser beam pass while reflecting the scattered light by particles. Hole diameter and angle of incidence can be chosen based on applications. The mirror (AL-45-500, Lenox Laser) used in the prototype has 500 µm hole at 45deg incident angle. Its diameter and thickness are 25.4 mm and 6.35 mm, respectively. Given the aperture tolerance of 10 percent, the laser beam waist at focus should be smaller than 550 µm to decrease the probability of unwanted deflection. There exists trade-off between hole diameter and detection angles where a larger hole results in loss of scattering angles closer to 0 deg. Due to resource constraint, the pinhole mirror from the original setup was again used in the prototype which contributed to a low signal-to-noise ratio in forward scattering detection. The beam waist of the laser is 0.7 mm which is large relative to the pinhole diameter of 500µm. Consequently, a clean dumping of the beam was not possible and the beam reflection from the hole contributed to higher background noise in small angle detection. The reflected beam was intense enough to saturate forward scattering detector and caused blooming. As a result, the presumed range of angle loss is greater than the previously assumed theoretical range between 0° and 0.7° [6].

3.2.9 Neutral density filters

Neutral density filters can be used to attenuate the incoming laser beam before it reaches the forward scattering detector to avoid saturation. Depending on where it is placed, the filter will also attenuate the scattered light that the system wishes to detect and reduce the signal intensity. In current LabView software controlling the detectors, there is no effective way to separate the main beam from scattering signal hence the signal with lower intensities look washed out on a digital display, posing challenging during calibration and measurements.

3.2.10 Linear photodiode detectors

The dimension of the photodiode detectors (Kansas State Electronics Lab) were reduced to approximately one fourth of the original ones with the same photodiode arrays (Hamamatsu
S3902-512Q). Each array had 512-by-1 pixels and each pixel was 50\(\mu m\)-by-0.5\(mm\) in size. The total active area was 25.6\(mm\) by 0.5\(mm\). Its readout time was approximately 8.192\(ms\) with integration time of 20\(ms\). These times were sensitive enough for aerosol applications in this study. Since the detectors did not have shutters, the diodes were always on integration mode, which meant off-axis, stray light must be blocked well. The system came with a proprietary LabView software, for which no VIs were available for future modifications, and a master controller. Of the two detectors, the one marked ‘Sensor 1’ was the master hence had to be on for Sensor 2 to be operational. This later posed a challenge in measurements due to amplitude difference between side and forward scattering (Section 4.7.3).

3.3 Instrument setup, calibration and operation

3.3.1 Alignment of optics

Centering all system components along the optical axis is essential to minimizing unwanted light reflections and diffraction thus maximizing signal-to-noise ratio. Previous alignment process [6] was complex and lengthy which was a hindrance to deployment of the instrument. Alignment of forward detection setup largely relied on empirical techniques of ‘walking the beam’ [55] where the optical mounts were adjusted based on real-time changes in beam as observed by human eye. The goal was to let the beam pass through consecutive apertures, which are the near focal point (F1) of the ellipse and center of pinhole mirror. Similarly, the sample inlet, ellipsoid reflector, iris and side detector all had to be aligned visually using two lasers [6], the accuracy of which can be subject to large error. To decrease the complexity of optical alignment, some components were mechanically coupled, in particular for side scattering. Also, optical cages were placed along the beam path for quick placement and removal of alignment discs. Better alignment of laser and the sample were done by threading either frosted glass alignment disks (DG05-1500-H1-MD, Thorlabs) or negative crosshair reticles (R1DS3N, Thorlabs) onto the lens tube connected to side scattering enclosure. This method helped reduce a potential tilt in angles and a shift about the optical axis within the alignment hole diameter as well as enabled a faster alignment of optics. Lens tubes had connection points to the main optical board to minimize their vibrations. A precise alignment of optics was done by adjusting xyz-translation stage or angle knobs. The following procedure
only serves as a general guideline as each alignment round would require varying degrees of changes.

1. Place the side scattering enclosure and fix it to the main optical board. This serves as a reference point for other optics.

2. Remove the Fourier Lens from its post holder but not from the xzy-stage.

3. Place an alignment disc at the end of a lens tube facing the planar mirror.

4. Turn on the main laser.

5. Place a neutral density filter between the laser and the planar mirror.

6. Adjust the planar mirror as needed to pass the laser beam through the alignment disc and the center of pinhole mirror.

7. Place an alignment disc at the end of a lens tube attached to the imaging lens.

8. Align the imaging optics.

9. Remove both alignment discs.

10. Place the Fourier lens back to its place and align. Confirm its back reflection does not fall outside the original beam center.

### 3.3.2 Calibration process

There were two main calibration methods from previous studies attempted in this study. First was matching single slit diffraction pattern in q-space to known theoretical patterns [6] by placing a slit where scattering sample will be located. This method required the optics to be rearranged post slit measurement which defeated the purpose of calibration as components were subject to shifts during the adjustment. It also was confined to calibrating forward scattering. Next option was to compare the scattering signal of water with Mie theory [6]. Distilled water aerosolized with compressed nitrogen gas was used for laboratory validation. Expected Mie plot was generated using Philip Laven application [http://www.](http://www.)
Similar approaches can be applied for other particles like polystyrene spheres with known scattering patterns. Though the above methods were valid, exploration for more convenient, accurate and faster method is needed for future studies.

3.3.3 Operation and Data analysis

To operate the instrument, non-conductive tubing had to be first connected to both the sample tube and an aerosol source. For measurements at atmospheric pressure, a use of vacuum pump was required in order to generate a constant flow of aerosols into the system. Having a needle valve or a sonic nozzle attached to the pump helped control the sample flow rate based on expected concentrations and not generate scattering volume that was optically too dense. Once all the tubes were connected and checked for correct flow, the laser was turned on and stabilized. LabView software was downloaded onto a PC that Kansas State provided with the detectors. Prior to each runs, a new file was created then a measurement was taken to record the offset from the electronics. Afterwards, a background signal with just laser on was recorded before starting the sample flow. All measurements had to be taken manually by pausing the live view of detectors and saving the data. The saved data could then be exported as .txt file and analyzed in Python. The final signal was determined by subtracting background signal from sample scattering measurements. Since discrete data files were generated for forward and side detectors, normalization of measured intensities were needed.
Chapter 4

Assessment of Prototype during NASA-NOAA Field Campaign

4.1 Details of FireX-AQ field campaign

Datasets used in the instrument system evaluation are from NASA-NOAA FIREX-AQ field campaign, conducted in McCall, Idaho from 8 August to 22 August 2019. The instrument was located at a ground site and we were able to analyze fuel samples collected at Nethker fire site, 30 miles northeast of McCall. During the field measurement, two different types of fuels, collected on 19 August 2019, were burnt for analysis in a iron lung [13]. The pictures of the samples are included in Section 4.5 below. The fuels were combusted inside the iron lung and the generated particles were pushed into the instrument at a flow rate of 1.2 liters per minute using an external pump. Since the samples were analyzed immediately following the burn, the results would more likely represent aerosols produced by flaming rather than smoldering combustion. A simultaneous measurement of particle size distribution was performed with a scanning mobility particle sizer spectrometer (SMPS) co-located at the ground site.

4.2 Instrument setup for field measurements

The prototype was packaged and transported via a ground vehicle to McCall and partially reassembled on site. Due to movements during transportation and unexpected field operating conditions, parts of the instrument had to be modified and realigned to better suit the
environment. In particular, the detectors had to be adjusted from the initial position to be further away from the aperture which led to all scattering angles being captured by a narrower set of detector channels. Dimensions of the new set up were noted for the purpose of data analysis following the campaign. A geometric ray tracing was performed for both forward and side scattering given the known system dimensions. It was assumed that the beam was aligned to the edge of forward detector and to the center of side detector. Figure 4.1a shows the scattered light from the sample (near focal point of the ellipse) starting at far right being transmitted through the achromat doublet lens then to the pinhole mirror at which the beam is diverted at 90° to the imaging lens and eventually to forward scattering detector. Figure 4.1b traces the ray from the edge of the ellipse to side scattering detector to determine the distance between the optical axis and the point at which ray reaches the detector. An approximate range of detected scattering angles was calculated using a basic trigonometry and Gaussian lens formula [51]. Here, each detector measured 25.6\,mm in length, each having 512 photodiode channels. Under the above set up, 0 to 11.68° would correspond to 18.982\,\text{mm} of 25.6\,\text{mm} long detector or approximately 74 percent of the channels. 380 data points, which are 74 percent of 512, were analyzed for forward scattering. Likewise, about 58 percent of the side detector channels were used for analysis. Since the beam was aligned at the center, 149 channels from each side of the optical axis were considered valid. The remaining data points were excluded from the analysis. The final angle resolutions were approximately 0.03° and 0.5° along the forward and side directions, respectively.

4.3 Noise and Sensitivity testing

Once the optics were fixed in place, a magnitude of systematic noise, resulting from either laser scattering off of gas molecules or stray light reflections, was evaluated by recording the changes in intensity measurements relative to the increase in laser power when the sample aerosols were not present. Figure 4.2 is a plot of absolute intensities measured by side detector at 1, 2, 20 and 30\,mW laser input. For all intensities, the values were close to 0\,V, indicating a low system noise. A slight linear increase in the noise happened at 30\,mW but the value was still below the reported offset of 0.01\,V caused by dark currents from the electronics. Meanwhile, the observed baseline for forward detector was approximately 100 times higher than that of side. This implies that the side scattering light path may be subject
(a) 2-D ray tracing of forward scattering. The dotted blue lines represent the ray paths. In particular, the dotted orange line shows a ray reaching Sensor 1 from the imaging lens. The units are degrees for angles and mm for distances.

(b) 2-D ray tracing of side scattering. An arc on the left is an elliptical mirror and Sensor 2 is at far right, centered about the optical axis. The dotted blue line shows where the ray scattered from the most outer edge of the ellipse falls on the detector. The units are degrees for angles and mm for distances.

Figure 4.1: Geometric ray tracing
to less light exposure compared to forward scattering path as expected by the presence of side scattering enclosure. This exercise helped check that detected signals were not simply a product of stray light inherent in the system.

4.4 Calibration using 300 nm PSL spheres

The prototype was tested with 300 nm polystyrene latex (PSL) beads aerosolized at 20 psi using Collison 6-jet nebulizer (CH Technologies Inc., NJ, USA) and nitrogen gas. Theoretical values of its structure factor were plotted in Python to compare against the experimental data. The details of Python code can be found in Appendix B. For a sphere with radius R, the structure or form factor can be found using Equation (4.1) which is derived from Equation 1.6 in polar coordinates:

\[ S(q) = \left[ 3(sin u - u \cos u) / u^3 \right]^2 \]  \hspace{1cm} (4.1)
where $u = qR$ and the phase shift parameter ($\rho$) is less than 1. Here $q$ is scattering wave vector defined in Equation 1.3. In Figure 4.3, measured $S_{11}(\theta)$ of 300 nm PSL in $q$ space is compared to the theoretical values. The theory (blue) and the experiment (red) values matched well in $q$ space. It is important to underline the fact that due to a small particle size, the scattering intensity from the sample was low. As a result, analyzing the data proved to be especially challenging for values close to the detection limit. For each round of experiments, background with only laser was recorded then subtracted from the measured signal. Unity-based normalization was applied to final intensities to have values between 0 and 1 to allow comparison across samples. An overlay of the measured intensity and the theoretical curve showed a good agreement indicating that the instrument was calibrated. The estimated value of $g$ was approximately $0.3 \pm 0.1$ which was reasonable given a value close to 0 is expected based on presumed symmetric, Rayleigh scattering by 300 nm PSL spheres.
4.5 Measurement of field samples

Following the system calibration, two different types of biomass fuel from Nethker fire site were analyzed. For the samples investigated in this study, $S_{11}(\theta)$ below 12° was assumed to be constant since Rayleigh scattering dominates for the reported average radius of gyration ($R_g$). Figures 4.4 and 4.5 show the $S_{11}(\theta)$ of each sample plotted in $q$ space, zoomed in to inflection point between Rayleigh regime to Guinier regime [14]. The SMPS measurement showed that Sample 1 had a mean particle diameter of 509 nm and a total sample concentration of $1.32 \times 10^6$ particles/cm$^3$. Sample 2 had a mean diameter of 484 nm and a total concentration $1 \times 10^6$ particles/cm$^3$. In both cases, the particle size approximately followed a normal distribution function. Within these observed size ranges, $S_{11}(\theta)$ for $\theta$ less than 12° can be assumed to be constant. Integrating the intensities within the range $12^° < \theta < 167^°$, yielded a $g = 0.73 \pm 0.02$ for Sample 1. The value of $g$ for Sample 2 failed to converge and had a large error. Overall, higher variations among sample data were seen in both samples as expected from heterogeneous fuel source. For example, with Sample 2, a drop of kerosene was required to start the flame which could have contributed to complex particle characteristics.

![Figure 4.4: A picture of Sample 1 and its measured phase function $S_{11}(\theta)$ in $q$-space](image)
Figure 4.5: A picture of Sample 2 and its measured phase function $S_{11}(\theta)$ in q-space

An investigation of the time-dependent dynamics of the phase function revealed the changes in slopes during the initial entering of aerosols into the system. Figure 4.6 depicts five consecutive scans of side detector, where a measurement was taken every five seconds to monitor the changes in intensities. Plot is in log-log scale. On average, the measurements reached steady state after approximately 15 seconds. This pointed at the appropriate wait time before scanning to acquire data representative of the particles under study. On average, dry aerosols at 500 nm have reported a $g$ value between 0.64 and 0.83 [45] which the above experimental results agreed with. A summary table of published $g$ values from past field studies is given in Figure 4.7 [50].
4.6 Transmission Electron Microscopy (TEM) imaging of the collected samples

The collected samples were analyzed with transmission electron microscope (TEM). Figure 4.8b shows morphology of the aggregates commonly found during the analysis whose average diameter (484nm) agreed with SMPS. The observations revealed heterogeneity of the
sample and a high sample concentration delivered by the iron lung leading to aerosol aggregations. With closer examination (Figure 4.8a), individual particle monomers appeared to be non-spherical. Sample heterogeneity was not unexpected for aerosols produced from flaming biomass combustion during which black, organic carbon, and inorganic compounds are produced. Contamination by dust particles during operation and sample analysis was also possible. This was particularly challenging for field-based experiments where control over the environment was limited. Future work will involve designing a precise and reliable sample delivery mechanism as well as characterizing mixed aerosol samples.
Figure 4.8: TEM images of the collected sample aggregates.
Chapter 5

Investigation of Instrument Design Modifications

5.1 Computational modeling

Chapter 5 focuses on modeling and simulation of potential design modifications and comparing the results to identify which variables in the system have significant impacts on how the instrument functions and their relationships. Rather than simply providing a dimension, this chapter aims to serve as a guide for future designs, which can vary depending on intended applications. Main goals going forward are decreasing alignment sensitivity while increasing the signal-to-noise ratio. Basic principles of static light scattering introduced in Section 2.2 as well as design principles from Chapter 3 will be revisited to suggest how sensitivity of scattering detection can be improved with proposed design changes. Specifically, the challenges identified during FireX-Aq campaign (Section 4.7) will be explored. Synopsys® LightTools 9.0.0 was used for simulations in this study. The details of base simulation parameters can be found in Appendix A. A simulation model was simplified to emphasize the impacts of chosen design variables on instrument sensitivity and proportionally scaled for easier display. To start, a collimated illumination source - a 532nm laser - was placed at the origin. The beam diameter was set to 0.7mm based on the manufacturer specification and was assumed to be uniformly distributed. A planar mirror facing the laser source was rotated 45° about the y-axis to divert the incoming beam at 90°. One forward illuminance receiver and one angular radiance receiver were added to the mirror surface to check for radiant intensity and angle of reflections. A default sample at near focal point was a 2.38mm radius sphere
5.2 Challenges during prototype development

5.2.1 Off-axis aberration

The original laboratory setup lacked mechanical couplings among the parts which rendered optical alignment challenging as well as leaving them vulnerable to external shocks. Prior to FireX-Aq, the side scattering enclosure was built to improve the stability of ellipsoidal reflector as well as to minimize stray light. Also multiple lens tubes and optical cages were used to integrate achromat doublets. Overall, the modification contributed to making the instrument more sensitivity and portable. However, laser, planar mirror and pinhole mirror still left rooms for mechanical shifts. Despite the reduction in size and weight, the detectors were too heavy for available optic mounts. The linear shape of the detectors also meant the location of optical axis was not as obvious as in circular objects. In both cases, resulting misalignment led to off-axis aberration. Either deviations from axial symmetry or inclusion of rays outside the paraxial region can result in aberrations and affect the signal-to-noise ratio or more precisely, the accuracy of the signal. Some of well known effects of off-axis optics include coma, astigmatism \[27\] and distortion \[48\]. Most aberrations in the prototype are monochromatic and are products of departing from first-order theory \[54\]. Though incorporation of achromat doublets did contribute to reducing spherical aberrations, both astigmatism and field curvature were harder to correct for when the system was off-axis. For example, a laser misalignment with respect to its boresight angle, \(\theta_b\), can set off a cascade of unwanted light reflection throughout the system. Given a light source is being focused by a lens with focal length, \(f\), the total shift in its pointing angle, \(\Delta\theta\), can be approximately \(\Delta\theta \times f\) \[59\]. Such shift especially has a negative impact on near-forward angles as unclean dumping of laser beam at the pinhole mirror causes diffraction which pollute the scattering signal for angles close to zero. More detailed analysis on impacts of optical misalignment on scattering intensity was done in LightTools. As previously mentioned in Chapter 3, a
planar mirror in the instrument is a potential cause of major ray deviations especially in turbulent environment as it can shift in all dimensions. Varying magnitude of error in laser pointing direction was studied under the following cases: 1) 0.05mm shift in optical axis, b) 0.05mm shift in y-direction, c) 0.5° tilt in alpha which is about the optical axis, d) 0.5° tilt in gamma or about the x-axis. Each of the figures show polar representation of far-field detection of scattering radiant intensity. The exact patterns will change based on beam profile. The laser is traveling from 270L to 90L and the angles in between correspond to side scattering angles. Figure 5.1 shows a case of perfect alignment and Figure 5.2 represents four different scenarios of either position or angle shifts. There are two major implications of this exercise. First, within the tested parameters, the shifts about the non-optical axis (Figure 5.2b and 5.2d) have less influence on the scattering profile than those about the optical axis. It was found that a movement of mirror along the y-axis does not change the observed patterns as long as the mirror stays within the plane of beam incidence. Meanwhile, tilts in mirror angles about the optical axis (Figure 5.2c) had most noticeable impacts on scattering. Next, the observations imply that a free-standing planar mirror poses a high risk for altering the detected signal. Thus, improvements in stabilizing optical mounts and joints should be considered.

![Figure 5.1](image)

Figure 5.1: Scattering intensity distribution at Sensor 2 under a perfect alignment.

Another point of aberration was at the iris through which the scattered rays exit from the side scattering enclosure. As can be seen in Figure 3.5, the aluminum panel on which the iris is drilled was machined too thick to allow the rays to pass without vignetting. Initially, the thickness was intended to accommodate the screws on the side of enclosure. To mitigate
the issue, the back panel was countersunk with available tools on the site. Future prototype would require the exit aperture thickness to be as small as possible.

5.2.2 Lack of focal position and volume control

If the aberrations were concerned with the accuracy of the signal, controlling the sample around the set focal point was linked to the detection sensitivity and limit. The instrument employs a push system for delivery of sample to the near focal point of the ellipse. This led to a high probability of aspired aerosols from the tube moving freely about the focal point before they were evacuated from the system via vacuum tube. Not all aerosols could be confined to a desired focal point which increased unwanted scattering. In this case, relatively large tubing size posed a higher chance of creating non-ideal scattering volume. Figure 5.6 shows...
the approximate spread of the scattering sample where the optical axis runs horizontal to the picture. Investigation of relationships between the focal volume and signal was done.

A radiant intensity, $d\phi$ detected at a given scattering angle, $\theta$ is a function of illuminating flux density, $S$, scattering volume, $dV$, solid angle, $d\omega$ and volume scattering function, $\gamma(\theta)$ (Equation 5.1). Here, the volume scattering function is determined by sample properties and only depends on the scattering angle for an ensemble of irregular particles. It is also extensive and varies proportionally with the sample volume [30].

$$d\phi = S\gamma(\theta)d\omega dV$$  \hspace{1cm} (5.1)

Under the current sample delivery system, aerosols are delivered in a gas stream via a tube to the ellipse near focal point. The laser does not illuminate a single point, rather a transverse plane whose length is the tube diameter. As a result, the effective scattering volume will be a function of beam width and inlet tube diameter [27]. Such setup leads to unwanted scattering by aerosols located outside the focal region. An actual cubic volume element ($dV$), also called voxels, and its cross section seen by the detector will vary along the ellipse axis interacting with the laser beam [26]. This study was done to evaluate how the sample volume, $S$, and cross-section may affect the detected scattering intensity and stray light in the prototype. To reduce unrelated errors, a model was setup without the planar mirror. Also, a dummy plane was created 50 mm down the optical axis from the sample to monitor the distribution of forward scattering. Optical density of the sample was simulated by setting the rays to follow a probabilistic 50% split between reflectance and transmittance. To begin, spheres of three different radius - 2.38 mm, 0.35 mm, and 0.05 mm - were compared. 2.38 mm is a radius of sample tube currently used, and 0.35 mm is a half the minimum laser beam width. For the smallest diameter, a 0.05 mm was chosen as a standard industry miniature 304 Stainless Steel tube is made down to 0.004 inch or 0.1 mm inner diameter (McMaster-Carr). A total of 100,000 rays were traced. Figures 5.3, 5.4 and 5.5 show the detected irradiance for both forward and side scattering when the sample radius was 2.38 mm, 0.35 mm, and 0.05 mm respectively. The figure axes are scaled to display the patterns in more details.
The spot size of forward scattered rays under a current setup (Figure 5.3a) was approximately 10 mm in radius, larger than the other two cases which covered about 4 mm of the detection surface. At the same time, the peak intensity was roughly 100 times lower with the high intensity area, marked red, spread wider. A larger percentage of detected side scattering occurred on the plane of incident beam and attenuated towards the forward angles as a result of increased absorption cross-section and longer distance the beam has to travel before exiting the scattering volume. As the sphere radius decreased (Figure 5.4, 5.5), the ‘image circle’ of forward scattering decreased with increasing peak intensity, indicating higher beam penetration rates. Meanwhile, the symmetry of side scattering profile improved with decreasing sphere radius as it approached Rayleigh scattering. The above results will
likely change with different sample refractive index or optical depth as incidences of out-of-focus scattering will decrease for samples with bigger absorption coefficient and higher percentage of incoming beam will transmit without scattering. The simulation implies that ideal sample volume should be chosen as a function of wavelength and refractive index as well as the mechanical limitations of the optics. In addition, using calibration gas with large index of refraction like Freon-12 can be helpful when the particle number concentration is hard to measure [2].

5.2.3 Inflexible detection system

Photodiode arrays offer benefits in their simple optical configuration and large dynamic range. However, they are subject to low sensitivity and low spatial resolution [11]. The first disadvantage of linear photodiodes used in the prototype was an absence of optical axis based on which the detectors can be aligned with the system. No systematic method for confirming the detector alignment was available other than calibrating with known particles. It was also noted that the diode chips were not sitting straight which further prevented geometry-based alignment. Another challenge with detection system was its software. The provided LabView software did not allow discrete measurements of forward and side scattering. The main controller required an operation of forward sensor to activate the side. Due to a large difference in detectable intensities, the side scattering signals were hard to see on screen when forward detector was actively measuring. Hence, the forward detector was covered with black
taped during side scattering measurements which increased operator errors across multiple runs and delayed measurement. Lastly, lack of automation in software meant no data could be taken overnight when an operator was not present, resulting in missed opportunities.

5.3 Near-term design improvements

5.3.1 Elliptical reflector dimensions

Impacts of decreasing the distance between the two ellipse foci on spot size at far focal point was studied. A smaller waist at the exit iris would reduce unwanted reflection and shorter distance between the two foci could contribute to increased detected intensity (Equation ??). Shorter distance also meant that the near focus point will be farther away from the lens edge, which would be favorable for optical alignment. The challenge lies in the fact that the above goals are in conflicts as shorter distance will lead to increased probability of vignetting and
reflections. It would also affect ellipse eccentricity, which refers to relative deviation from a circular conic section and ultimately influences a total range of detectable angles. As eccentricity approaches 0, the angle range decreases. Optimization simulations were run to evaluate the aforementioned trade-offs. Main variables used in the study were conic constant \( \kappa \), curvature, aperture diameter, distance from near focal point to the lens surface \( Z \), and distance between the two foci \( Z_1 \). The mirror inner diameter was calculated assuming a constant lens thickness of 0.51\( \text{mm} \). In order to eliminate the effects of scattering sample volume, the sphere radius was set to 0.05\( \text{mm} \) based on the finding in Section 5.2. The sample optical properties were not changed. The ellipse eccentricity \( e \) was determined using Equation 5.2 where \( a > b \):

\[
e = \sqrt{1 - \frac{b^2}{a^2}}
\]  

(5.2)

A conic constant \( \kappa \) is \(-e^2\). For the current elliptical reflector (E115NV-R), \( e \) was 0.77 where the major axis \( a \) and minor axis \( b \) are 2.9293 and 1.8681 inches respectively as shown on Figure 3.4. Resulting conic constant was approximately -0.5933. When \( \kappa \) has a value between -1 and 0, an ellipse with major axis on z-axis is created. The Mirror Formula \[54\] (Equation 5.3) was used to calculate the curvature radius \( R \), which affects ellipse major and minor semi-axis along with eccentricity.

\[
\frac{1}{s_0} + \frac{1}{s_i} = -\frac{2}{R}
\]

(5.3)

where \( s_0 \) is the distance between the mirror apex and secondary (far) focal point and \( s_i \) is the distance between the apex and primary focus. Based on the dimensions given by the manufacturer, \( s_0 \) is 131.71\( \text{mm} \) and \( s_i \) is 17.09\( \text{mm} \) for current ellipsoid design. Then, the curvature radius of the elliptical mirror is -30.254\( \text{mm} \) and curvature, which is a reciprocal of the curvature radius, is 0.033. Each optimization parameters was given bounds based on physical models. The lower and upper bounds for \( \kappa \) were -1 and 0, and 0 to 100 for aperture diameter. The step sizes for \( \kappa \) and diameter were 0.01 and 1, respectively. No limits were set for lens curvature as it was already a function of \( \kappa \). \( Z \) and \( Z_1 \) bounds were set with respect to the sample coordinate \((0, 0, 0)\). \( Z \) ranged between 0 and 100 while \( Z_1 \)
could take a value between -114.62 and 0. The ‘merit function’, which was an output to be optimized, calculated the spot size at the center of side scattering receiver to determine the best parameter set within 20 model iterations. Initially, optimization parameters were set to prototype dimensions and ran to establish a base case. Optimization of the base case returned a total drop in merit function by less than factor of 10. Spot size was 5.1213 mm. Subsequent simulations were done to investigate the relationships among design variables and the spot size when $Z_1$ was reduced. Below table summarizes the parameters from select simulation runs. All units are in mm. The distance between each focus and the lens surface was determined at the center of curvature.

<table>
<thead>
<tr>
<th>Parameters</th>
<th>E115NV-R</th>
<th>Simul$_1$</th>
<th>Simul$_2$</th>
</tr>
</thead>
<tbody>
<tr>
<td>Radius of curvature</td>
<td>30.26</td>
<td>29.41</td>
<td>29.41</td>
</tr>
<tr>
<td>Curvature</td>
<td>0.033</td>
<td>0.034</td>
<td>0.034</td>
</tr>
<tr>
<td>Conic constant ($\kappa$)</td>
<td>-0.5933</td>
<td>-0.5935</td>
<td>-0.5897</td>
</tr>
<tr>
<td>Aperture diameter</td>
<td>51.03</td>
<td>58.69</td>
<td>59.47</td>
</tr>
<tr>
<td>Distance between foci ($Z_1$)</td>
<td>114.62</td>
<td>100.63</td>
<td>86.57</td>
</tr>
<tr>
<td>Near focus to lens surface ($Z$)</td>
<td>17.09</td>
<td>17.12</td>
<td>17.15</td>
</tr>
<tr>
<td>Spot size</td>
<td>5.1213</td>
<td>5.5375</td>
<td>7.1735</td>
</tr>
</tbody>
</table>

(a) Increase in lens aperture diameter  
(b) Decrease in lens curvature

Figure 5.7: Iterative simulation results of ellipse geometry

The study revealed that a decrease in $Z_1$ is initially most compensated by an increase in lens aperture diameter (Figure 5.7a) but at decreasing rate as the distance is reduced further.
Meanwhile, both lens curvature (Figure 5.7b) and the spot size increased only slightly when the $Z_1$ was cut by less than 10 percent ($Simul_1$). As the decrease in distance approached 20 percent ($Simul_2$), the spot size increased significantly though the lens properties did not change substantially from their initial values. Based on the observations, approximately 10 percent decrease in $Z_1$ will help improve signal intensity and alignment with minimal loss in detectable angles and potential image distortion. Subsequent increase in $Z$ will also decrease a possibility of beam reflection off of mirror edge.

5.3.2 Sample delivery system

Based on the observations in 5.2, an improved aerosol flow system focused on delivering more representative measurement volume can be a solution to obtain higher signal-to-noise ratio. A divergence of sample aerosols can be minimized by feeding particle-free sheath flow along with it, similar to a design presented in Figure 5.8 [2]. There will be separate flow control systems for sample and sheath flow. Both inlet and exhaust tubes can be attached to the side scattering enclosure to eliminate possibilities for misalignment. Main difference will be feeding of samples from the bottom rather than the top to allow for visual alignment checks with an enclosure lid off. This set-up would have to be checked for potential loss in large particles due to impaction and gravitational settling. Impaction can be minimized by keeping the total tube length as short as possible with no bends.

5.3.3 Detection system

Both optics and electronics compose instrument detection system that affects its sensitivity. To start, current prototype can be improved via addition of collimator optics or baffles to further reduce stray light and increase signal sensitivity [2]. Aspheric lenses, for example, can reduce spherical aberration. The converging Gaussian beam after passing through the aspheric lens will display outer fringe patterns whose central spot can be spatially filtered using pinholes (Figure 5.9) [41].

The emerging beam can then be collimated to be relayed onto a detector. While addition of optics can eliminate noise associated with beam, it is advantageous to minimize the number
of potentially refractive optical surfaces as they cause ghost reflections. Ghost reflections generally happen inside and near the optics field of view and increase with smaller aperture. Baffles can help decrease the multi-bounce path of these reflections. A placement of baffles inside the side enclosure box can block direct reflection from the sample as well as the unwanted stray light. Alternating lens tube diameters can also serve as baffles. Imperfect collimation and dumping of laser beam, in particular, plays a major role in increased stray light in near-forward angles via diffraction. By threading reticles along with spatial filters at both ends of lens tube passing the side enclosure box, stray light reflection and back reflections can be kept out of detector sights.

Electronics, along with optics, play an important role in building a sound detection system. Linear photodiode arrays offer a great range of accessible angles and linearity but low sensitivity and spatial resolution. Also its fixed arrangement limits its use to isotropic samples [11]. For future prototypes, charge-coupled detectors (CCD) can be good alternatives to photodiodes due to their high angular resolution determined by pixel sizes and ease of alignment. However, implementation of CCD brings new sets of considerations such as image sharpness and non-linearity. Specific camera specifications to be evaluated include 1) time resolution, especially the phosphor decay time which determines how fast the previous frame dissipate, 2) spatial resolution based on active sensing area, and 3) image intensifiers. The
input window material and photocathodes used in the intensifier can be chosen based on wavelength and desired quantum efficiency. Quantum efficiency changes as a function of radiant sensitivity, $S$ and wavelength (Equation 5.4) \[38\]. When longer wavelength is used, Rayleigh scattering off the gas molecules in the atmosphere can be reduced but the detector QE may decrease given no change in $S$.

\[
QE(\%) = \frac{S \times 1240}{\lambda} \times 100 \tag{5.4}
\]

There also exist corrections to be made on the acquired raw intensity data from detectors. Where the scattering volume has a certain turbidity $\tau$ and path length $L$ along the illuminating laser path, a constant attenuation factor equal to $e^{\tau L}$ can be calculated for $\theta$, though the difference is negligible. A correction on acceptance solid angle for each $\theta$ using cosine law can be upto 5% for the largest angle. Here, the actual solid angle seen by the detector has $\cos^3 \theta$ multiplied to the nominal solid angle.\[11\].
Chapter 6

Conclusions

The study developed and evaluated a prototype instrument for measuring directionality of aerosol light scattering. Details of optical and mechanical components were discussed with emphasis on their functional roles. The device used an ellipsoidal reflector to collect aerosol light scattering from 12 to 167° polar angles. Forward scattering between 0.7 and 11.68° was measured simultaneously using Fourier and imaging lens, though saturation of detector from beam diffraction resulted in low signal-to-noise in near-forward angles. Addition of side enclosure increased mechanical robustness and decreased alignment sensitivity of the device, rendering it more suitable for field use. Tests using 300 nm polystyrene latex spheres showed a good agreement with theoretical structure factor of homogeneous spheres in q-space. Estimated $g$ was $0.3 \pm 0.1$. The instrument was deployed during NASA-NOAA FireX-AQ to measure two different fuel samples from the Nethker fire site. Our findings estimated that the aerosols emitted from biomass burning had $g = 0.73 \pm 0.02$. Main challenges identified during the study were 1) off-axis aberration largely from optical misalignments, 2) lack of focal position volume control due to a wide sample inlet, and 3) inflexible detection system which increased time spent on alignment and measurement. The effects of first two challenges on scattering signal were studied using computational simulations in Synopsys LightTools. Based on the observations, we proposed a new elliptical reflector with shorter distance between two foci and larger lens aperture diameter to increase signal intensity and reduce alignment sensitivity. We also suggested using a smaller sample tube that is securely fixed on near foci and utilizing charge-coupled detector camera to enable faster alignment. Overall, avoiding the use of standalone components and employing cage systems proved where possible improved portability of the device. Future research on this instrument will focus on easier, faster alignment and calibration of the optomechanical system as well as
higher sensitivity to aerosols at ambient concentrations to be used in field sampling. With this tool, aerosol phase function can be measured directly, eliminating the need to estimate $S_{11}$ from $b$ and HG phase function. Such improvement can contribute to increased accuracy of radiative transfer model in the atmosphere.
Appendix A

Details of LightTools computational simulations

Illumination  A disk light source was placed at the origin. Disk radius was set at FWHM assuming no beam divergence.

- Wavelength: 532nm
- Coordinates (X,Y,Z,α,β,γ) = (0, 0, 0, 0, 0, 0)
- Radiometric power: 20mW
- Beam radius: 0.35mm
- Spectrum: Gaussian, FWHM 0.7mm, wavelength tolerance ± 2nm
- Aim region: Sphere; upper and lower angles at 0 degrees to create a collimated beam
- Calibration tolerance: 5%
- Angular distribution: Uniform

Planar mirror  Surface receivers were added to the front lens surface.

- Lens diameter: 25.4mm
- Lens thickness: 0.15mm
- Optical property: 100% reflectance simple mirror
**Ellipsoid reflector**  The below dimensions reflect the optics used in the FOAM prototype presented in this thesis.

- Diameter: circular, 51.03 mm
- Thickness: 0.51 mm
- Lens front surface shape: conic
- Conic constant: -0.5933
- Curvature: 0.033

**Aerosol sample**  Below parameters were for base case aerosol sample.

- Coordinates (X,Y,Z,α,β,γ) = (50, 0, 0, 0, 0, 0)
- Sphere radius: 2.38 mm
- Optical property: Split Rays. 50% Reflectance and 50% Transmittance

**Detectors**  Far-field dummy planes with surface receivers were created for illuminance analysis of scattered rays.

- Dimensions: rectangular, 65.215 mm by 65.215 mm
- Units: radiometric power

**Ray trace input**

- Total rays to trace: 100,000
- Relative ray power threshold: 0.0001
- Random number generator type: Sobol
- Dispersive mode for spectral
Appendix B

Python script for 300\text{nm} PSL sphere data analysis

Below code was modified for the purpose of transcribing it onto a paper.

""
Created on Sun Aug 11 16:57:28 2019

@author: Esther Koh
""

import pandas as pd
import numpy as np
import matplotlib.pyplot as plt
from pandas import DataFrame

#data import as pd
main = pd.read_csv("file:") #file name
#fwd = pd.read_csv("file:") #file name
#side = pd.read_csv("file:") #file name
#file names edited

#subsetting

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laserfwd = main.iloc[:,2]
fwdsample = main.iloc[:,3:5]
fwdavg = fwdsample.mean(axis=1)
fwdavgplot = fwdsample.mean(axis=1) #for plotting
laserside = main.iloc[:,8]
sidesample = main.iloc[:,9:11]
sideavg = sidesample.mean(axis=1)

beam_corr = 1 #laser beam attenuation
laserfwd *= beam_corr
laserside *= beam_corr

#correction factor
fwdcorr = 0.001#correction factor
print('minside=', min(sideavg))
sidecorr = 0

fwd1 = (fwdsample['psl300fwd1']- laserfwd)+ fwdcorr
fwd1 /= max(fwd1) #Normalizing intensity
fwd2 = (fwdsample['psl300fwd2'] - laserfwd) + fwdcorr
fwd2 /= max(fwd2)
fwdavg = fwdavg - laserfwd + fwdcorr
fwdavg /= max(fwdavg)

sd1 = (sidesample['psl300sd1']- laserside) + sidecorr
sd1 /= max(sd1) #Normalizing intensity
sd2 = (sidesample['psl300sd2'] - laserside) + sidecorr
sd2 /= max(sd2)
sideavg = (sideavg - laserside) + sidecorr
sideavg /= max(sideavg)

#Forward scattering angles
fwd_x=np.linspace(0,11.68,380) #380 detector channels registering signal
#Q-space; \( q = \frac{4\pi}{\lambda} \sin(\theta/2) \times (\pi/180) \)

\( w = 0.532 \) # laser wavelength

\( \text{fwd}_xq = \left(4 \times \text{np}.\text{pi}/w\right) \times \text{np}.\sin((\text{fwd}_x/2) \times (\text{np}.\text{pi}/180)) \)

#Side scattering angles

\( \text{side}_x = \text{np}.\text{linspace}(12.01, 167.99, 298) \) #298 channels detecting a relevant signal

#converting to q-space

#Q-space; \( q = \frac{4\pi}{\lambda} \sin(\theta/2) \times (\pi/180) \)

\( \text{side}_xq = \left(4 \times \text{np}.\text{pi}/w\right) \times \text{np}.\sin((\text{side}_x/2) \times (\text{np}.\text{pi}/180)) \)

#non-dimensionalize (qR space)

\( D = 0.3 \) #diameter (um)

\( R = D/2 \) #volume equivalent dia.(um)

print('R=', R)

\( Rg = \text{np}.\text{sqrt}((3/5)\times(\text{np}.\text{square}(R))) \) #assume a perfect sphere

print('Rg=', Rg)

\( \text{fwd}_xqR = \left(4 \times \text{np}.\text{pi}/w\right) \times \text{np}.\sin((\text{fwd}_x/2) \times (\text{np}.\text{pi}/180)) \times R \)

\( \text{side}_xqR = \left(4 \times \text{np}.\text{pi}/w\right) \times \text{np}.\sin((\text{side}_x/2) \times (\text{np}.\text{pi}/180)) \times R \)

##structure factor calculation

\( u = \text{side}_xq \times R \)

\( \sinu = \text{np}.\sin(u) \)

\( \cosu = \text{np}.\cos(u) \)

\( \text{bit1} = u \times \cosu \)

\( \text{bit2} = u \times u \times u \)

\( S = \text{np}.\text{square}((3*(\sinu-\text{bit1})/\text{bit2})) \)

\( ug = \text{side}_xq \times Rg \)

\( \sinug = \text{np}.\sin(ug) \)

\( \cosug = \text{np}.\cos(ug) \)

\( \text{bit1} = ug \times \cosug \)

\( \text{bit2} = ug \times ug \times ug \)

\( Sg = \text{np}.\text{square}((3*(\sinug-\text{bit1})/\text{bit2})) \)
#forward detector index selection
a_fwd = 80
b_fwd = 235 #235 is the cut off
fwd_x = fwd_x[a_fwd:b_fwd]
fwd_xq = fwd_xq[a_fwd:b_fwd]

#side scattering channel indices (298 channels in total)
a_side = 107
b_side = 405

## laser noise data (1, 2, 20, 30 mW)
noise = pd.read_csv("file:") #file name
#fwd = pd.read_csv("file:") #file name
#side = pd.read_csv("file:") #file name

# subsetting
onemw = noise.iloc[:,0]
twowm = noise.iloc[:,1]
twentymw = noise.iloc[:,4]
thirtymw = noise.iloc[:,7]

# scale = 0 # scaling the side scattering intensity to connect with fwd

## I v. theta plot

ax1 = plt.subplot(211)
ax1.set_xlabel('Theta ($\Theta$)', fontsize = 15)
ax1.set_ylabel('S$_{11}$ (A.U.)', fontsize = 15)
#ax1 = plt.gca() # on the same-axis
#plt.plot(fwd_x, fwd1.values[a_fwd:b_fwd]*-15,'g')
#plt.plot(fwd_x, fwd2.values[a_fwd:b_fwd]*-15,'g')
#plt.plot(fwd_x, fwdavg.values[a_fwd:b_fwd]*-15,'r')
fwdavgplot /= max(fwdavgplot)
fwdsample[\'graph\'] = fwdavgplot#np.random.randint(1,3,size=(512,1))
plt.plot(fwd_x, fwdsample.graph[a_fwd:b_fwd]*2,'r',linewidth = 2,linestyle = "dashdot")
plt.plot(side_x, sd1.values[a_side:b_side]*0.1,'g')
plt.plot(side_x, sd2.values[a_side:b_side]*0.1,'g')
plt.plot(side_x, sideavg.values[a_side:b_side]*0.1,'r',linewidth = 2)
# plt.plot(side_x, S,'y')
# plt.plot(side_x, Sg,'b')

""
plt.plot(side_x, onemw.values[a_side:b_side],label='1mW')
plt.plot(side_x, twomw.values[a_side:b_side],label='2mW')
plt.plot(side_x, twentymw.values[a_side:b_side],label='20mW')
plt.plot(side_x, thirtymw.values[a_side:b_side],label='30mW')
plt.legend(loc='upper right')
"""

## xq plot
ax2 = plt.subplot(212)
ax2.set_xlabel('q $(m^{-1})$',fontsize = 15)
ax2.set_ylabel('S$_{11}$ (A.U.)',fontsize = 15)
ax2 = plt.gca() # on the same-axis
# plt.loglog(fwd_xq, fwd1.values[a_fwd:b_fwd],'k')
# plt.loglog(fwd_xq, fwd2.values[a_fwd:b_fwd],'k')
# plt.loglog(fwd_xq, fwd3.values[a_fwd:b_fwd],'k')
plt.loglog(fwd_xq, fwdavg.values[a_fwd:b_fwd],'g')
fwdsample['graph'] = max(sideavg)
# plt.loglog(fwd_xq, fwdsample.graph[a_fwd:b_fwd],'r', linewidth=2.5)
# plt.loglog(fwd_xq, fwdsample.const[a_fwd:b_fwd],'r', linewidth=2.5)

plt.loglog(side_xq, sd1.values[a_side:b_side],'g')
plt.loglog(side_xq, sd2.values[a_side:b_side],'g')
# plt.loglog(side_xq, sd3.values[a_side:b_side],'g')

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plt.loglog(side_xq, sideavg.values[a_side:b_side], 'r', linewidth = 2.5)

#structure factor plot
plt.loglog(side_xq, S, 'y')
plt.loglog(side_xq, Sg, 'b')
plt.show()

""
plt.loglog(side_xq, onemw.values[a_side:b_side], label='1mW')
plt.loglog(side_xq, twomw.values[a_side:b_side], label='2mW')
plt.loglog(side_xq, twentymw.values[a_side:b_side], label='20mW')
plt.loglog(side_xq, thirtymw.values[a_side:b_side], label='30mW')
#plt.legend(loc='upper right')

""

""
##xqR plot
ax2 = plt.subplot(313)
ax2 = plt.gca() #on the same-axis
plt.loglog(fwd_xqR, fwd1.values[a_fwd:b_fwd], 'k')
plt.loglog(fwd_xqR, fwd2.values[a_fwd:b_fwd], 'k')
#plt.loglog(fwd_xq, fwd3.values[a_fwd:b_fwd], 'k')
fwdsample['graph'] = max(sideavg)
fwdsample['graph'] *=0.8 #scaling to connect the graph to side
plt.loglog(fwd_xqR, fwdsample.graph[a_fwd:b_fwd], 'r', linewidth=2.5)
#plt.loglog(fwd_xq, fwdsample.const[a_fwd:b_fwd], 'r', linewidth=2.5)

plt.loglog(side_xqR, sd1.values[a_side:b_side], 'g')
plt.loglog(side_xqR, sd2.values[a_side:b_side], 'g')
#plt.loglog(side_xqR, sd3.values[a_side:b_side], 'g')
plt.loglog(side_xqR, sideavg.values[a_side:b_side], 'r', linewidth = 2)
plt.show()
# asymmetric parameter

```python
fwdintegral = np.cos(fwd_x)*fwdavg.values[a_fwd:b_fwd]
fwdg = sum(fwdintegral) - np.cos(fwd_x)*fwdcorr)
print('fwdg =', fwdg )

sideintegral = np.cos(side_x)*sideavg.values[a_side:b_side]
sideg = sum(sideintegral-np.cos(side_x)*sidecorr)
# the above correction factor subtracted from g
print ('sideg =', sideg)

g = 0.5*(fwdg+sideg)
print(g)
```
References


Prototype Instrument Development for Measuring Directionality of Aerosol Light Scattering, Monroe, M.S. 2020