Development of an angle-scanning spectropolarimeter: Preliminary results

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A fixed-angle spectropolarimeter capable of measuring the Mueller matrix of particle deposits and conventional optical elements over the 300–1100 nm spectral range has been built, calibrated and extensively tested. A second generation of this instrument is being built which can scan from 0° to near 180° in both scattering angle and sample orientation, enabling studies of the bidirectional Mueller matrices of nanoparticle arrays, atmospheric aerosol deposits, and nano- and microstructured surfaces. This system will also provide a much needed metrology capability for fully characterizing the performance of optical devices and device components from the near-infrared through the medium wave ultraviolet. Experimental results taken using the first generation fixed-angle arrangement will be presented along with the rationale for building the second.

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1. Introduction

Atmospheric particulate matter (PM) is dominated, to varying degrees, by ammonium salts, mineral dust, diesel exhaust and a mix of complex organic species. Pollen, fungal spores, and particles formed from plant waxes may also be present. The presence of this background aerosol seriously complicates the detection of airborne pathogens. Lethal spore concentrations on the order of (roughly) 1000/liter would constitute a mass concentration of about $10\,\mu g/m^3$, which is comparable to ambient PM concentrations routinely measured over the continental U.S. (Annual averages for which have been found to range from $1.9\,\mu g/m^3$ in the Sierra Nevadas to $23\,\mu g/m^3$ in the Washington D.C. area [1]). Detection of bioaerosols at sufficiently high signal-to-noise ratios and acceptable rates of false positives can only be accomplished with instrumentation of high sensitivity and information content. Spectropolarimetric analysis of scattered radiation holds great promise in this regard, but the detected signal will be that of all particles in a volume probed by a source beam, and thus must be further processed to extract the signature of the true targets of the probe. The optical properties of both biohazard and background (ambient and/or obscurant) particles must be well characterized in order to discern signatures of the targeted constituent from complex aerosol mixtures.

Passive infrared sensors are capable of remote detection of biological aerosols if there is sufficient thermal contrast, but this cannot be guaranteed in all applications [2].

Measurements of linearly depolarized light scattering have met with some success in identifying bioparticles in a background of water droplets [3], but it is unlikely that such a method would be effective if the background aerosols were dust or other nonspherical particles. More promising is the use of the full Mueller matrix obtained in polarimetric measurements which can distinguish between populations of particles with different morphologies [4]. Recent work [5], an example of which is revisited in this article, has supported that thesis. The range of PM samples on which measurements have been made thus far include kerosene soot, mineral dust, various species of pollen and Bacillus subtilis (BG); a well-known surrogate for anthrax spores. The soot, dust and pollen samples represent major constituents of the nonspherical ambient atmospheric aerosol and can be considered major interferants in the bio-aerosol detection process.

The new angle-scanning instrumentation will help address the need for rapidly detecting and identifying biological particles and chemical agents in complex mixtures. The importance, the many techniques employed and the urgent need for improved technology are highlighted by Hurst [6]. Among the many techniques being explored, polarimetry is especially promising and incidence angle dependent polarimetry over a broad wavelength band would be a significant advancement. Experimental studies aimed at measuring polarized light scattered from PM samples have been limited to a few discrete wavelengths in the visible and IR regions. Spectral measurements of polarization properties of such samples over

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a broad wavelength region are scarce primarily due to a lack of instrumentation [7–9].

2. Basic spectropolarimetric system theory

2.1. Stokes parameters and Mueller matrices

There are various mathematical representations available to describe polarized light and its interaction with matter. Mueller calculus and Jones calculus are two widely used analytical frameworks for describing polarized light propagation through optical elements [10]. The Stokes vector formalism is based on measured irradiance as opposed to the Jones vector formalism, which is based on field amplitudes and phase differences. For descriptive purposes it is assumed here that the electric field propagates in the z direction and has amplitude E_0 with components in the x and y directions: $\vec{E} = \vec{E_0}\, e^{i(\vec{k}\,\cdot\,\vec{z} - \omega t)}$. The Stokes vector is defined as:

$$\vec{S} = \begin{bmatrix} S_0 \\ S_1 \\ S_2 \\ S_3 \end{bmatrix} = 1/S_0 \begin{bmatrix} 1 \\ S_1 \\ S_2 \\ S_3 \end{bmatrix} = \begin{bmatrix} E_{0x}^2 + E_{0y}^2 \\ E_{0x}^2 - E_{0y}^2 \\ 2E_{0x}E_{0y}\cos\delta \\ 2E_{0x}E_{0y}\sin\delta \end{bmatrix} = \begin{bmatrix} I_{tot} \\ I_H - I_V \\ I_{45°} - I_{135°} \\ I_R - I_L \end{bmatrix} \tag{1}$$

where E_{0x} and E_{0y} are the electric field components along the x and y axes respectively and δ is the phase difference between these components. The first element of the Stokes vector is the total irradiance while the subsequent three elements are differences in irradiance between three pairs of orthogonal states of polarization. Any medium which interacts with light transforms the incident Stokes vector into a transmitted Stokes vector by means of a 4×4 real matrix called the Mueller matrix, which completely describes the polarization characteristics of the medium. This transformation can be written as:

$$\vec{S}_{out} = \mathbf{M}.\vec{S}_{in} = \begin{bmatrix} m_{00} & m_{01} & m_{02} & m_{03} \\ m_{10} & m_{11} & m_{12} & m_{13} \\ m_{20} & m_{21} & m_{22} & m_{23} \\ m_{30} & m_{31} & m_{32} & m_{33} \end{bmatrix} \begin{bmatrix} S_0 \\ S_1 \\ S_2 \\ S_3 \end{bmatrix} \tag{2}$$

The first element of the Mueller matrix (m_{00}) represents the absolute transmittance or reflectance (depending on the physical arrangement of the experiment) of the medium for unpolarized incident light. The other elements are related to different physical polarization properties such as retardance, diattenuation and depolarization.

2.2. Dual rotating retarder polarimeters

Measurement and analysis of the polarization characteristics of optical beams and elements with which they interact is the science of polarimetry [11]. Light measuring polarimeters measure Stokes vectors and hence the state of polarization, degree of polarization, the elliptical parameters and other polarization characteristics of light beams. Sample measuring polarimeters or Mueller matrix polarimeters, on the other hand, measure Mueller matrices and hence the relationship between the incident and exiting states of polarization for any sample. Sample measuring polarimeters employ a Polarization State Generator (PSG) preceding the sample in order to interrogate the sample with well characterized polarization states and a Polarization State Analyzer (PSA) succeeding the sample to detect specific polarization states exiting the sample. This establishes the relationship between the incident and exiting polarization states, namely the Mueller matrix of the sample. The dual rotating retarder configuration is the most popular architecture used for complete Mueller matrix polarimetry [12,13]. The polarimeter

consists of a combination of a linear retarder and a polarizer in the PSG and PSA [14].

The polarimetric measurement sequence usually starts with all the critical axes aligned with each other. A series of irradiance measurements are then made at unique rotational positions of the generator and the analyzer retarders. The output polarization state vector is given by:

$$\vec{S}_{out}^q = P_2 R_2^q M_{sample} R_1^q P_1 \vec{S}_{in}, \tag{3}$$

where P_1 and P_2 are the Mueller matrices of the polarizers and R_1^q and R_2^q are the Mueller matrices of the retarders in the PSG and PSA respectively. M_{sample} is the Mueller matrix of the sample that is to be determined. The index q denotes different rotational positions of the retarders during the measurement sequence. $\vec{S}_{in}$ and $\vec{S}_{out}$ are the incident and exiting Stokes vectors respectively.

For measuring the complete Mueller matrix, a rotating retarder polarimeter should make Q(≥ 16) irradiance measurements at unique orientations of the PSG and PSA retarders. One way to achieve this would be to rotate R_1 and R_2 in discrete angular steps, maintaining a ratio of at least 1:5 between them in order to cover all the required nondegenerate measurement states.

For the qth measurement, the irradiance (I_q) recorded by the detector is related to the Stokes vector of the beam produced by the PSG ($\vec{S}_q = R_1^q P_1 \vec{S}_{in}$), the Mueller matrix of the sample (M) and the analyzer vector (A_q) which is the first row of the matrix $P_2 R_2^q$, and is given by:

$$I_q = \vec{A_q^T} M \vec{S_q} = \vec{W_q}.\vec{M}, \tag{4}$$

where $\vec{W_q}$ is a 16×1 polarimetric vector defined as:

$$\vec{W_q} = \left(a_{q,0}s_{q,0},\ a_{q,0}s_{q,1},\ a_{q,0}s_{q,2},\ a_{q,0}s_{q,3}, a_{q,1}s_{q,0},\ \ldots.\ a_{q,3}s_{q,3} \right)^T \tag{5}$$

And $\vec{M}$ is a 16×1 column vector constructed using the Mueller matrix elements:

$$\vec{M} = \left(m_{00}\ m_{01}\ m_{02}\ m_{03}\ m_{10}\ldots.\ m_{33} \right)^T \tag{6}$$

Generalizing the above into a matrix equation describing all Q measurements gives:

$$\vec{I} = W\vec{M} = \begin{bmatrix} \vec{W}_0^T \\ \vec{W}_1^T \\ \vec{W}_2^T \\ \vec{W}_3^T \\ . \\ . \\ . \\ \vec{W}_{Q-1}^T \\ \vec{W}_Q^T \end{bmatrix} \begin{bmatrix} m_{00} \\ m_{01} \\ m_{02} \\ m_{03} \\ . \\ . \\ . \\ m_{33} \end{bmatrix} \tag{7}$$

where $\vec{I} = (I_0\ I_1\ I_2\ \ldots\ I_qQ)^T$ is a column vector representing Q irradiance measurements. The matrix W is called the polarimetric measurement matrix (or calibration matrix). The data reduction equation, which calculates the Mueller matrix from the irradiance measurements, is given by

$\vec{M} = W_p^{-1} \vec{I}$, where W_p^{-1} is the pseudoinverse of the matrix W (since W is a 16 x Q matrix where Q > 16).

2.3. Mueller matrix decomposition

The physically realizable Mueller matrix alone does not provide much insight into all the physical polarization properties of a sample. In order to quantify meaningful polarization properties, the Mueller matrix can be decomposed into three unique constituent matrices using polar decomposition, as proposed by Gil et al., and Lu and Chipman [15–19].