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## Spectral polarimetric light-scattering by particulate media: 1. Theory of spectral Vector Radiative Transfer

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### ABSTRACT

Spectral polarimetric light-scattering by particulate media has recently attracted growing interests for various applications due to the production of directional broadband light sources. Here the spectral polarimetric light-scattering signatures of particulate media are simulated using a numerical model based on the spectral Vector Radiative Transfer Equation (VRTE). A microphysical analysis is conducted to understand the dependence of the light-scattering signatures upon the microphysical parameters of particles. We reveal that depolarization from multiple scattering results in remarkable spectral and directional features, which are simulated by our model over a wide spectral range from visible to near-infrared. We propose to use these features to improve the inversion of the scattering problem in the fields of remote sensing, astrophysics, material science, or biomedical.

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### 1. Introduction

Multiple light-scattering refers to electromagnetic scattering by collections of particles from ultraviolet to infrared and occurs in particulate media when light is scattered several times before detection [1–3]. It is prominent when light propagates through dense media such as plumes, clouds, or colloids. This phenomenon has been an object of interest in many scientific fields from remote sensing [4], astrophysics [5] to biomedical [6] and material science [7].

Several inverse methods have been developed to retrieve microphysical properties of particulate media [8]. However, the inversion of single-wavelength scattering signatures is usually not permitted without prior assumptions. Such restrictions are due to the non-uniqueness in the solutions of the scattering problem [9]. Prior knowledge about particles (e.g. refractive index) is usually necessary to retrieve microphysical properties. Polarimetric [10] or multispectral measurements [11–13] have been proposed as a useful tool to improve light-scattering inverse methods since they provide additional information.

Recent experiments have reported the potential use of merging spectral and polarimetric light-scattering for optical diagnostic [14–19]. Due to the ill-posed inverse scattering problem, we propose to merge spectral (*i.e.* broadband or hyperspectral) and polarization information in order to develop novel inversion methods. A numerical model based on the spectral Vector Radiative Transfer Equation (VRTE) is presented to compute the spectral polarimetric light-scattering by particulate media. Furthermore, a microphysical analysis based on our model is conducted to determine the role and dependence of the particles properties (*i.e.* mean size, refractive index, and volume fraction) on simulated signatures. This approach is expected to improve physical insights and remove solution ambiguities in the development of future inverse methods.

### 2. Theory

The radiative transfer theory is applied with great results in astrophysics, remote sensing, physics, geophysics, atmospheric physics, and other areas of science. The Radiative Transfer Equation (RTE) was first introduced

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in 1905 for atmospheric studies [20] and solved in 1943 using the invariance principle for the problem of diffuse reflection of light by a scattering medium [21]. In this theory, particulate media are modeled as semi-infinite plane-parallel multi-layered isotropic scattering media to account multiple-scattering. One essential condition for using the RTE to model multiple light-scattering by particulate media is that the distance  $d$  between particles must be larger than the wavelength  $\lambda$  such as  $d > \lambda$ . In such a way, the scattering events always occur in the far-field region of each particle and prevent coherent effects such as dependent scattering.

Later, this theory has been extended to a larger range of scientific problems, including anisotropic scattering medium and electromagnetic scattering by an ensemble of randomly distributed and oriented particles [22]. The Vector Radiative Transfer Equation (VRTE) [23] is the vector counterpart of the RTE and has been recently derived from first-principles [24].

Let us first consider a particulate media as an ensemble of  $N$  particles in random orientations and positions in a non-scattering host medium. The spectral VRTE is given for a polarized broadband directional light source as:

$$\mathbf{u} \cdot \nabla \mathbf{S}(\mathbf{r}, \mathbf{u}, \lambda) = -\mathbf{k}_{\text{ext}}(\mathbf{r}, \lambda) \cdot \mathbf{S}(\mathbf{r}, \mathbf{u}, \lambda) + \frac{1}{4\pi} \int_0^{2\pi} \int_0^1 d\mathbf{u}' \cdot P(\mathbf{r}, \lambda) \cdot \mathbf{k}_{\text{sca}}(\mathbf{r}, \mathbf{u}, \mathbf{u}', \lambda) \cdot \mathbf{S}(\mathbf{r}, \mathbf{u}, \lambda) + \mathbf{k}_{\text{abs}}(\mathbf{r}, \lambda) \cdot \mathbf{S}_0(\mathbf{r}, \mathbf{u}, \lambda) \quad (1.1)$$

The spectral Stokes vector  $\mathbf{S}(\mathbf{r}, \mathbf{u}, \lambda) = (I, Q, U, V)^T$  are the spectral VRTE solutions which encompass the spectral and polarimetric signatures of a particulate medium of interest as a function of position  $\mathbf{r}$ , direction  $\mathbf{u}$  and wavelength  $\lambda$ . The phase matrix  $P(\mathbf{r}, \lambda)$  of an ensemble of particles is calculated by integrating the Mueller Matrix elements  $M_{ij}(r_p, \mathbf{r}, \lambda)$  over the particle size distribution  $n(r_p)$  of an ensemble ( $i$  and  $j$  range from 1 to 4):

$$P(\mathbf{r}, \lambda) = \begin{bmatrix} P_{11} & P_{12} & P_{13} & P_{14} \\ P_{21} & P_{22} & P_{23} & P_{24} \\ P_{31} & P_{32} & P_{33} & P_{34} \\ P_{41} & P_{42} & P_{43} & P_{44} \end{bmatrix} \quad (1.2)$$

with:

$$P_{ij}(\mathbf{r}, \lambda) = \frac{\lambda}{\pi k_{\text{sca}}^2} \int dr_p M_{ij}(r_p, \mathbf{r}, \lambda) n(r_p) \quad (1.3)$$

Particulate or scattering media are modeled for numerical purposes as semi-infinite plane-parallel layers with constant radiative parameters as shown in Fig. 1. For instance, a particulate medium inside a quartz or glass slab is modeled as a series of vertically inhomogeneous layers containing randomly oriented particles of various geometries. The general description of atmospheric layers is substituted herein by optical material interfaces, such as quartz or glass, with given complex refractive index. The extension of this model to spectral polarimetric light-scattering requires several modifications including spectral dispersion of the interfaces or solving Fresnel equations at every interface for every wavelength.

Our model based on the spectral VRTE solves numerically a system of  $L$  equations, where  $L$  is the number of

discrete wavelengths composing the broadband light source (typically  $L=1000$ ). The spectral operation consists of calculating the scattered Stokes vector corresponding at each wavelength in the spectral range. Azimuthal angles are expressed as a Fourier series and polarization rotations are performed directly in azimuth space. Subsequently, Fourier transform is performed to retrieve the scattering matrix for each Fourier azimuth mode [23,25,26]. Multiple reflections between layers are also taken into account in the spectral polarimetric radiance balance at the top and bottom of each layer. The adding-doubling technique is then deployed for each Fourier mode to model multiple scattering. This technique is numerically stable and is used to model multiple light scattering in an intuitive, efficient, and simple way. A good selection of the Fourier modes and the quadratic angles for every wavelength is crucial for the computation.

Validation of the model has been carried out following a three-step procedure [27]. Briefly, (i) an analytical validation was conducted for Rayleigh scattering from Coulson's table, (ii) a stochastic method was used to validate the model for Mie scattering and (iii) experiments were carried out on polystyrene particles in aqueous solution. As stated above, the spectral VRTE model is valid only for intermediate volume fractions, in dispersed-phase, as a large value of this parameter refers to a large number of multiple scattering events and multiple reflections inside a glass cuvette for instance.

### 3. Methodology

Spectral polarimetric light-scattering simulations are carried out using our spectral VRTE model for an incident broadband  $p$ -polarized collimated illumination. Typical radiative transfer inputs are optical thickness or albedo. However, for broadband calculations, these radiative parameters cannot be employed because of their spectral dependency. For broadband calculations, particulate media must be rather described in terms of microphysical parameters such as volume fraction or refractive index (both for particles and host medium), which are not dependent on the wavelength of illumination. Thus, a particulate scattering medium composed of an ensemble of random spherical particles is modeled with three principal microphysical parameters:

- (1) Particle size distribution, which follows a log-normal size distribution such as

$$n(r_p) = \frac{dN(r_p)}{dr} = \frac{N_0}{\sqrt{2\pi} r_p \ln(\sigma)} \exp \left[ -\frac{1}{2} \left( \frac{\ln(r_p/r_m)}{\ln(\sigma)} \right)^2 \right] \quad (1.4)$$

where  $n(r_p)$  is the particle size distribution as a function of the particle radius  $r_p$ ,  $N_0$  is the total number density of particles,  $r_m$  is the mean particle radius and  $\sigma$  is the standard deviation of distribution.

- (2) Complex optical index, which accounts for the spectral dispersion and absorption of both particles and host

media. Several index models [28] can be employed to compute spectral refractive index of materials.

- (3) Volume fraction, which describes the fraction of particles in the particulate media volume [29] such as:

$$f = \frac{V_{part}}{V_{part} + V_{media}} \quad (1.5)$$

where  $V_{part}$  is the particulate volume and  $V_{media}$  is the host volume media (e.g. air, liquid water).

In what follows, a particulate medium composed of spherical silica particles is considered here. The microphysical parameters for the simulation correspond to a mean radius  $r_m$  of 100 nm, an optical index varying over

the spectral range (e.g.  $n = 1.59 + i0.0$  at 532 nm), and a volume fraction of 1%.

The first and second elements of the spectral Stokes vectors are computed and presented in Fig. 2 against  $\mu = \cos(\theta)$ , the cosine of the scattering angle  $\theta$ . The scattered intensity, related to the first Stokes matrix element  $I/I_0$ , decreases for large wavelengths. This spectral dependence is shown in the backscattering ( $\mu = -1$ ) and forward scattering ( $\mu = +1$ ) directions. This dependence could be understood as the variation of the single-scattering efficiency over a broadband spectrum. The single-scattering efficiency is known to vary with the size parameter (i.e. ratio of the particle diameter to wavelength) [24]. Thus, it also varies when spectral scattered-intensities are computed over a wide spectral range for a fixed particle size (here, particle radius is fixed). The spectral Degree Of Linear Polarization (DOLP), related to the second Stokes matrix element  $Q/I$ , also exhibits a clear spectral dependence especially for reflection angles (between  $\mu = -1$  and  $\mu = 0$ ). More interestingly, a noticeable feature is found around  $\mu = -0.5$ . It corresponds to a maximum depolarization where the spectral DOLP reaches low values. Similar features have been previously reported from experiments [13] and are further analyzed in the next section. A numerical artifact produces unrealistic values at  $\mu = 0$ .

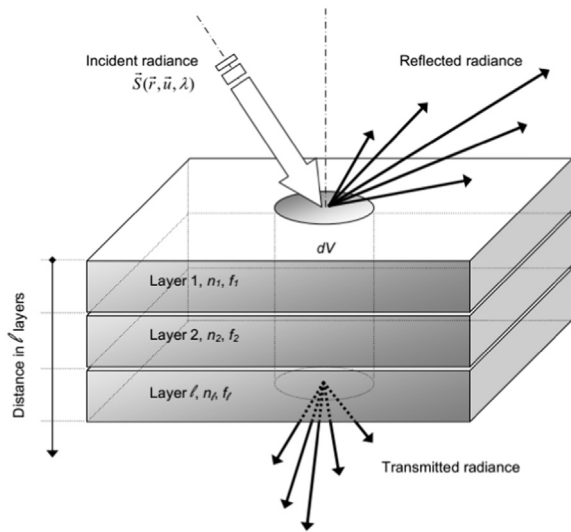


Fig. 1. Schematic of a generic particulate medium modeled as semi-infinite multiple layers.

#### 4. Results

The microphysical parameters of particles are expected to play a major role in the spectral polarimetric light-scattering signatures of particulate media. A microphysical analysis is carried out to determine their impact on spectral polarimetric light-scattering signatures. Such analysis is a prior requirement in the development of specific inversion methods merging spectral and polarimetric information. In the following, spectral polarimetric light-scattering signatures are computed with different mean particle radius, refractive index, and volume fraction in the

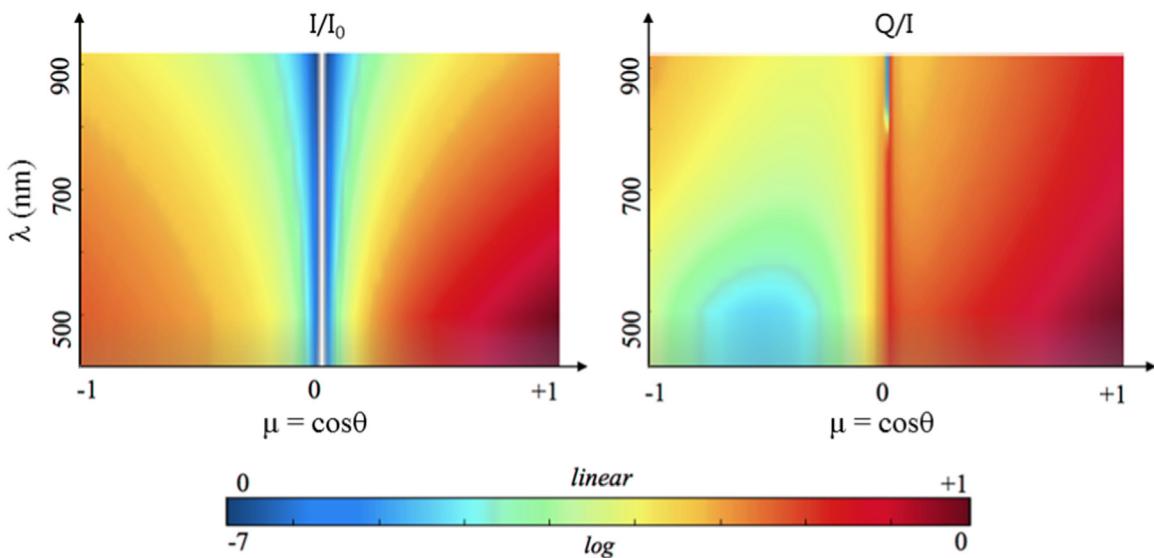
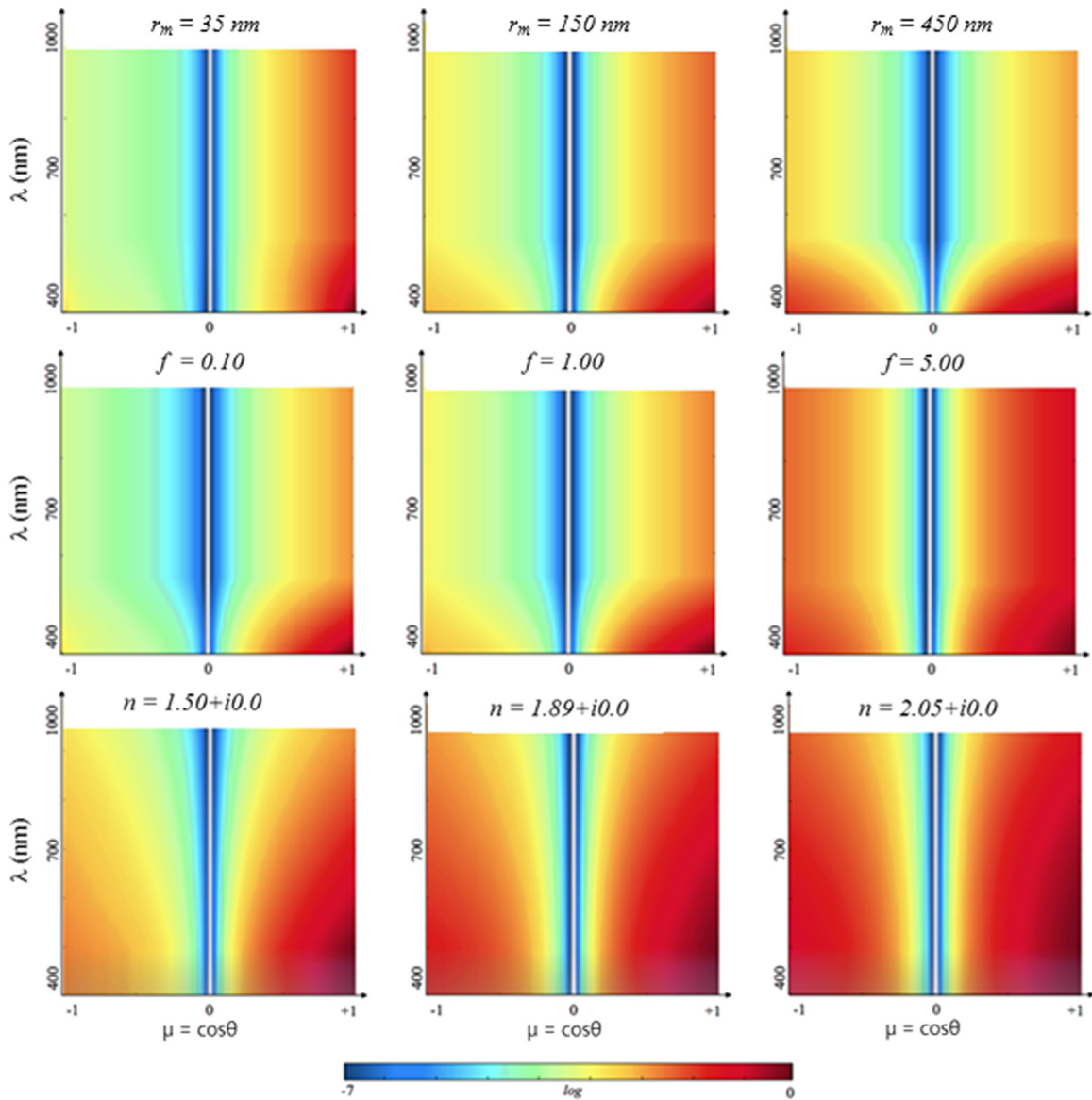


Fig. 2. Simulation of the spectral Stokes vectors ( $I/I_0$  in log-scale and  $Q/I$  in linear-scale) scattered by a particulate medium for a broadband (480–1020 nm, with spectral resolution of 5 nm) p-polarized illumination.



**Fig. 3.** Spectral computations of the first Stokes element ( $I/I_0$ ) in log-scale for an incident broadband (480–1020 nm, with spectral resolution of 5 nm)  $p$ -polarized collimated illumination. Variations of the mean particle size ( $r_m$ ), refractive index ( $n$ ), and volume fraction ( $f$ ) of particles.

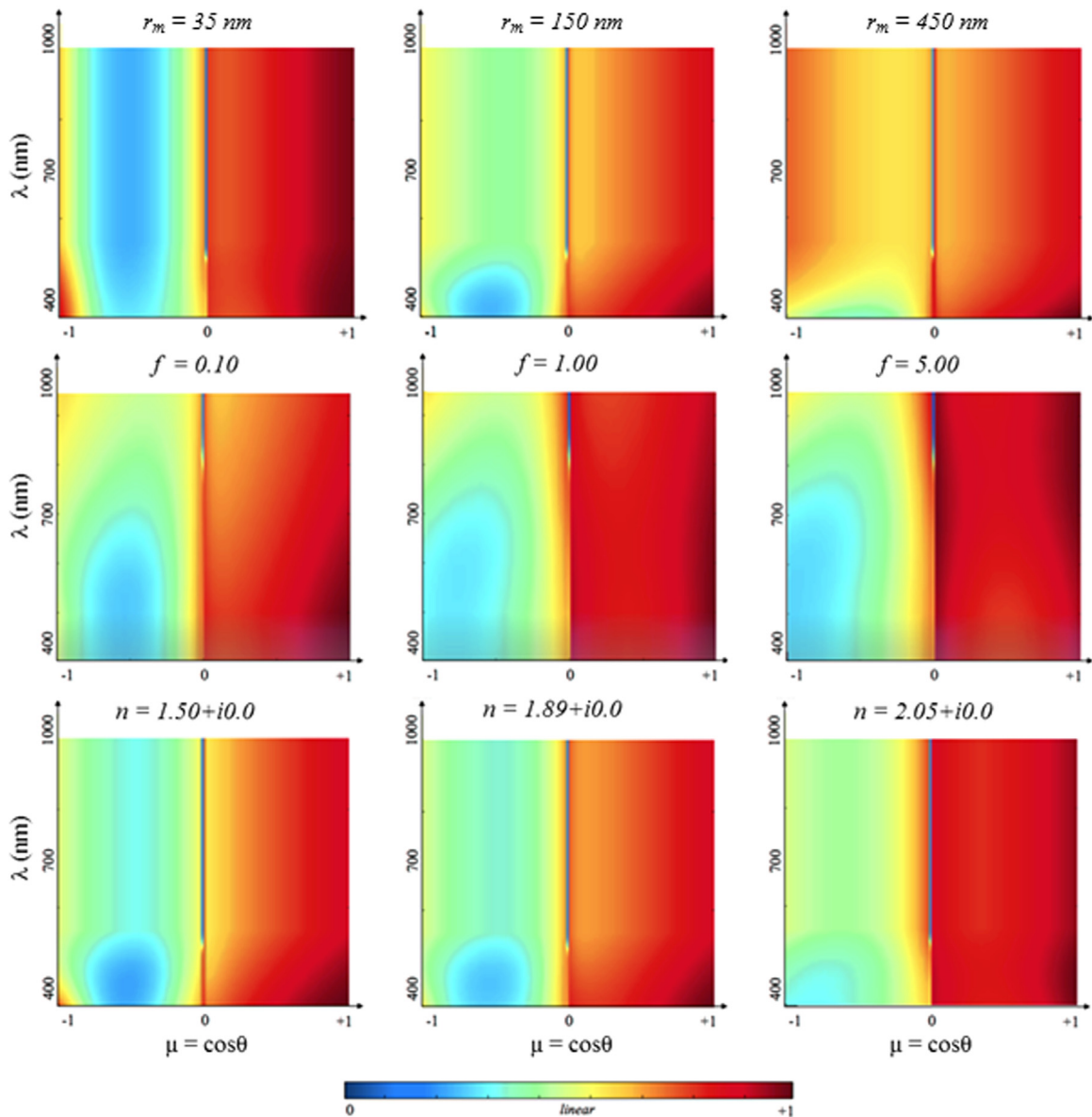
range of validity of the VRTE model. The mean particle radius ranges from 35 nm to 450 nm, the complex optical index is constant over the spectral range with fixed values ranging from  $1.5+i0.0$  to  $2.05+i0.0$ , and the volume fraction varies from 0.1% to 5.0%.

Fig. 3 presents the spectral first Stokes elements ( $I/I_0$ ) resulting from multiple light-scattering by particulate media with varying microphysical parameters. They are plotted in log-scale for an incident broadband  $p$ -polarized collimated illumination and different sets of parameters. For each line, a single parameter varies when others remain constant. When not specified, the mean particle radius for a log-normal size distribution is  $r_m=100$  nm, the volume fraction is  $f=1.0\%$ , and the refractive index is  $n=1.50+i0.0$ .

Fig. 4 presents the spectral second Stokes elements ( $Q/I$ ) resulting from multiple light-scattering by particulate media with varying microphysical parameters. They are plotted for an incident broadband  $p$ -polarized collimated illumination and different sets of parameters. For each line, a single parameter varies when others remain constant, similarly to Fig. 3.

## 5. Discussion

Light-scattering by particulate media depend on the microphysical parameters of particles such as the particle size, refractive index, or volume fraction. Some of these parameters depend on the wavelength and may vary significantly for broadband calculation. Moreover, scattering regimes have been defined [1] and categorized by the



**Fig. 4.** Spectral computations of the second Stokes element ( $Q/I$ ) for an incident broadband (480–1020 nm, with spectral resolution of 5 nm) p-polarized collimated illumination. Variations of the mean particle size ( $r_m$ ), refractive index ( $n$ ), and volume fraction ( $f$ ) of particles.

product of the size parameter  $x$  by the relative refractive index (between particles and host medium). These regimes are ruled by light-phase variations induced when light passes through a particle. For instance, Rayleigh scattering regime applies for small size parameters and intermediate refractive index whereas Mie scattering regime applies for intermediate size parameters and refractive index.

Usual monochromatic light-scattering techniques investigate a single scattering regime. One advantage of spectral or broadband light-scattering computations is to address distinct scattering regimes for a given particulate media. From Fig. 3, a change of scattering regime is identified when the mean particle size or refractive index increases. However, no change is noticed for varying volume fraction. This is explained as the governing parameters of scattering (*i.e.*

size and refractive index) are kept constant for different volume fractions. These results expose the interaction between microphysical parameters of particles and the wavelength of the light. Spectral polarimetric light-scattering clearly provides interesting physical insights about the nature of particles in particulate media.

Due to the interplay between the size and refractive index, using only the spectral scattering intensity may result in serious inversion hardships. The merge of spectral and polarimetric information is proposed by computing the spectral second Stokes vector element ( $Q/I$ ) or the spectral DOLP. Depolarization from multiple scattering is a directional phenomenon (*i.e.* depends on the scattered angle) and can be simulated by our model over a wide spectral range from visible to near-infrared. As shown in

Fig. 4, the incident polarization is conserved for transmission angles (including forward-scattering), whereas significant depolarization appears at reflection angles (including backward-scattering).

Depolarization from multiple-scattering is known to depend on the particle size or refractive index [30] and has been accurately computed by our model. When performing the microphysical analysis, the mean particle size, refractive index, and volume fraction are important factors that drive the spectral and directional depolarization at reflection angles. Spectral polarimetric light-scattering by particulate media appears to present features related to distinctive microphysical parameters.

1. *Mean particle size.* A spectral dependence of the depolarization on the mean particle size is reported. For constant refractive index and volume fraction, minimum DOLP values (i.e. maximum depolarization) appear around  $\mu = -0.5$  and strongly varies with the wavelength and particles size. One can identify a limited spectral domain where the depolarization is maximum. This spectral domain is found variable from small particles to large particles. Thus, this spectral feature would permit to identify the mean particle size of an unknown particulate media.
2. *Refractive index.* A directional dependence of the depolarization on the refractive index is reported. For constant particle size and volume fraction, a maximum depolarization appears around  $\mu = -0.5$  and strongly varies with directions. The shape of the limited spectral domain with maximum depolarization is remarkably different when the refractive index varies. As refractive index increases, directions of maximum depolarization also increase. For instance, at  $\mu = -1$  (in the perfect backscattering), DOLP ranges from 0.5 (i.e. partial depolarization) for  $n = 1.59 + i0.0$  to 0.1 (i.e. near complete depolarization) for  $n = 2.05 + i0.0$ . This directional feature is proposed as a tool to probe the refractive index of particles composing an unknown particulate media.
3. *Volume fraction.* The limited spectral domain with maximum depolarization shows neither spectral nor directional dependence on volume fraction. However, a dependence of the minimum DOLP value on the volume fraction is reported. For a given particle size and refractive index, the minimum DOLP value varies from 0.2 to 0.5 within an increase of the volume fraction. The minimum value of DOLP would allow to retrieve the volume fraction of particulate media.

## 6. Conclusion

One fundamental advantage of merging spectral and polarimetric information for light-scattering is to address distinct scattering regimes and to extract useful information about a particulate media of interest involving multiple-scattering. This study highlights the potential use of spectral polarimetric light-scattering to probe simultaneously several microphysical properties of particles, including mean particle size, refractive index, or volume fraction.

A numerical model based on the spectral VRTE has been developed to demonstrate that depolarization induced by multiple-scattering presents spectral and polarimetric features which could be used to retrieve microphysical properties of interest. The influence of these properties on the spectral polarimetric light-scattering signatures has been analyzed and revealed interesting depolarization features. In a nutshell, the mean particle size has been found to relate to spectral depolarization, the refractive index to directional depolarization, and the volume fraction to the minimum DOLP value.

We propose to use these spectral and polarimetric features to improve inversion methods and identify microphysical properties of an unknown particulate media. This analysis is a first stone for novel optical techniques based on spectral or hyperspectral polarimetric light-scattering.

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