Scattering of light by mineral-dust particles much larger than the wavelength

T. Nousiainen*[,1], O. Muñoz[2], H. Lindqvist[1], P. Mauno[1], and G. Videen[3]

[1] *Department of Physics, P.O. box 48, FI-00014 University of Helsinki, Finland.*

[2] *Instituto de Astrofísica de Andalucía, CSIC, c/ Camino Bajo de Huétor 50, 18080 Granada, Spain.*

[3] *Army Research Laboratory, 2800 Powder Mill Road,Adelphi, MD 20783, USA.*

We use a modified ray-optics code RODS (Ray Optics with Diffuse and Specular interactions) and laboratory-measured Mueller matrices to study light scattering by dust particles much larger than the wavelength. The rough-surface treatment of the RODS model allows us to reproduce the measured scattering matrices very well, except for the phase function. Surface roughness is found to decrease the asymmetry parameter and increase the single-particle albedo.

INTRODUCTION

Mineral dust is an important component in the Earth's atmosphere, its impacts ranging from direct radiative effects to fertilizing oceans and rain forests, and acting as freezing nuclei for ice clouds. In addition, mineral-dust particles are found in great abundance in, e.g., the Martian atmosphere and regoliths of many Solar-system bodies.

Accurate optical modeling of these particles is very challenging especially when they are larger than the wavelength [1]. One of the complicating factors is the presence of wavelength-scale surface roughness that cannot be explicitly accounted for in traditional ray tracing. Earlier, surface-roughness effects have been studied, e.g., by [2] and [3], using an *ad hoc* Lambertian modification to ray optics. Here we test whether a physically more rigorous RODS model (Ray optics with Diffuse and Specular interactions) introduced by [4] could account for the surface-roughness effects realistically. To this end, a laboratory-measured Mueller matrix of Libyan sand, provided by [3], is used as a reference to which RODS simulations based on the actual, measured size distribution are compared. The Libyan sand sample has an effective radius $r_{\text{eff}} = 125\ \mu$m and effective variance $\nu_{\text{eff}} = 0.15$, guaranteeing that all dust particles are in the ray-optics domain at visible wavelengths.

MODELING APPROACH

The RODS model consists of geometric-optics and diffraction parts. The latter is solved in a Fraunhofer approximation. The geometric-optics part is augmented such that the target shape can be covered with a layer of external scatterers with given single-scattering properties. There are a number of ways the external scatterers can be characterized in the model; here we apply a method where we specify their phase matrix, referred to as an input matrix. In addition, the optical depth of the layer, τ_0, needs to be specified. Here we consider it a free parameter. The single-scattering albedo of the external scatterers, ϖ_0, is set to unity;

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[*] Corresponding author: Timo Nousiainen (timo.nousiainen@helsinki.fi)

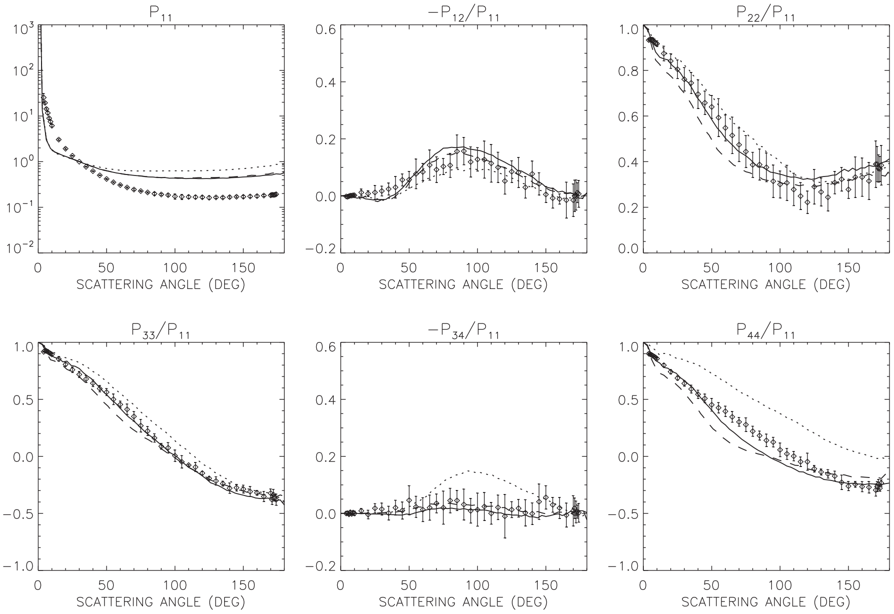

Figure 1. Comparison of simulated and measured Mueller matrices for Libyan sand. The simulated matrices based on different input matrices are plotted with dotted (feldspar), dashed (calcite), and solid (generic clay) lines with $\tau_0 = 1.0$ and $\mathrm{Im}(m) = 3.0 \cdot 10^{-4}$. The measurements are indicated with diamonds and error bars. The P_{11} elements have been renormalized to unity at 30° scattering angle.

we consider the external scatterers to be in the lower part of the resonance domain where even moderately absorbing materials would have high ϖ_0.

Three different input matrices are considered. First, we use a Mueller matrix based on a feldspar sample measured by [5]. Instead of the actual measurements that do not cover the whole scattering-angle range, however, we use the modeled phase matrix based on the $n = 3$ shape distribution of spheroids fitted to the feldspar sample by [6]. In addition, we compile two input matrices from the computations for flaky particles presented in [7]. One is based on calculations for calcite flakes, where the birefringence is fully accounted for. The other is based on the isotropic analog for the calcite flakes and acts here as a proxy for generic clay flakes. The calculations are here integrated over a lognormal size distribution with the geometric mean radius $r_g = 0.35$ μm and the geometric standard deviation $\sigma_g = 1.8$. These two input matrices are motivated by the observation that flake-like particles often cover the surface of large dust particles [7]. All three input matrices represent phase matrices for polydisperse small particles with effective radii between 0.5 and 1.0 μm. The asymmetry parameters associated to the input matrices are $g_0 = 0.725$ for feldspar, $g_0 = 0.808$ for calcite flakes, and $g_0 = 0.815$ for the generic clay.

RESULTS

The simulations are carried out at the wavelength of 633 nm. We use two different optical depths, $\tau_0 = 0.5$ and 1.0, for the layer of external scatterers in addition to a case with no

Table 1. The asymmetry parameter for varying τ_0, $\mathrm{Im}(m)$, and the input matrix.

$\mathrm{Im}(m)$	Asymmetry parameter g						
	$\tau_0 = 0$	$\tau_0 = 0.5$			$\tau_0 = 1.0$		
		Feldspar	Generic clay	Calcite	Feldspar	Generic clay	Calcite
$3 \cdot 10^{-4}$	0.895	0.804	0.830	0.828	0.741	0.782	0.777
$1 \cdot 10^{-4}$	0.801	0.728	0.749	0.747	0.677	0.709	0.706
$3 \cdot 10^{-5}$	0.716	0.653	0.672	0.670	0.611	0.639	0.636

external scatterers. The real part of the refractive index is fixed at 1.55, while the imaginary part is varied between $\mathrm{Im}(m) = 3 \cdot 10^{-4}$ and $3 \cdot 10^{-5}$. The shapes of the model particles are based on the Gaussian random sphere geometry [8] with shape parameters $\sigma = 0.2$ and $\nu = 3.3$ taken from [3].

When the RODS model is run without external scatterers, good fits to the measured Mueller matrix cannot be obtained. When the external scatterers, mimicking the small-scale surface roughness, are introduced, the agreement between simulations and measurements improves drastically. As shown in Figure 1, other elements except the phase function (P_{11}) can be matched very well. The agreements are as good or even better than those attained by [3] using the Lambertian modification, and here we can achieve that by using a physically rigorous model and realistic shapes for model particles.

Different input matrices perform differently and the agreement to measurements depends also on $\mathrm{Im}(m)$ value used, but overall the matrix based on the feldspar sample performs worse than those based on the flaky shapes. Here the Generic clay matrix provides the best fit.

The impact of τ_0, $\mathrm{Im}(m)$, and the input matrices on the asymmetry parameter g and single-particle albedo ϖ are summarized in Tables 1 and 2. Obviously, g increases with increasing $\mathrm{Im}(m)$. Further, g increases with decreasing τ_0. The input matrices also affect g, but their influence is smaller. Not surprisingly, the input matrices with largest asymmetry parameters, g_0, also lead to largest g values for the whole particles. It is noteworthy, however, that the g values for layered particles can be smaller than the g_0 value of the external scatterers or the g value for unlayered particles, signifying the impact of multiple scattering on g.

Likewise, ϖ increases with decreasing $\mathrm{Im}(m)$ or increasing τ_0. The latter is partially connected to the fact that the single-scattering albedo of the roughness elements, ϖ_0, has been set to unity. Still, even if the surface elements were composed of the same material as the host particles, their smaller size parameters would make their single-scattering albedos higher than that of the host particle. This signifies the potential impact of surface roughness on the single-particle albedo of large dust particles.

CONCLUSIONS

The RODS method seems a promising way of modeling the optical properties of dust particles large compared to the wavelength. In particular, all phase matrix elements of the reference Libyan sand sample except the phase function can be reproduced very well based

Table 2. As Table 1 but for the single-particle albedo.

Im(m)	Single-particle albedo ϖ						
	$\tau_0 = 0$	$\tau_0 = 0.5$			$\tau_0 = 1.0$		
		Feldspar	Generic clay	Calcite	Feldspar	Generic clay	Calcite
$3 \cdot 10^{-4}$	0.639	0.668	0.659	0.660	0.691	0.676	0.677
$1 \cdot 10^{-4}$	0.777	0.789	0.786	0.786	0.799	0.793	0.793
$3 \cdot 10^{-5}$	0.905	0.907	0.906	0.906	0.909	0.908	0.908

on realistic model shapes. The problems with the phase function seem to be concentrated on the forward angles, implying that the size distribution of the sample may be in error, or else the surface roughness also affects the diffraction part of the phase function in ways that cannot be realistically accounted for with the present method.

Such amounts of surface roughness that allow good fits between the measurements and simulations ($\tau_0 \approx 1.0$) affect both the asymmetry parameter and the single-particle albedo quite considerably and systematically. This implies that large-particle contributions based on smooth model particles might lead to systematic errors in radiative-transfer applications such as dust radiative-forcing calculations or remote sensing.

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