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On the interpolation of light-scattering responses from irregularly shaped particles

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ABSTRACT

Common particle characteristics needed for many applications may include size, eccentricity, porosity and refractive index. Determining such characteristics from scattered light is a primary goal of remote sensing. For other applications, like differentiating a hazardous particle from the natural background, information about higher fidelity particle characteristics may be required, including specific shape or chemical composition. While a complete characterization of a particle system from its scattered light through the inversion process remains unachievable, great strides have been made in providing information in the form of constraints on particle characteristics. Recent advances have been made in quantifying the characteristics of polydispersions of irregularly shaped particles by making comparisons of the light-scattering signals from model simulant particles. We show that when the refractive index is changed, the light-scattering characteristics from polydispersions of such particles behave monotonically over relatively large parameter ranges compared with those of monodisperse distributions of particles having regular shapes, like spheres, spheroids, etc. This allows for their properties to be interpolated, which results in a significant reduction of the computational load when performing inversions.

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1. Introduction

Obtaining particle characteristics from scattered light, the so-called inverse problem, is intimately associated with the direct problem of determining light-scattering properties of particles. The direct problem is well-defined and can be obtained by applying the Maxwell equations and satisfying boundary conditions. Traditionally, the inverse problem has relied on matching measured scattering characteristics with those of model particles obtained via the forward problem. To achieve this, it is necessary to have a diverse set of solutions to the direct problem. For this reason, most light-scattering inversions have been performed using relatively simple systems for which we have obtained exact theoretical solutions and from which analytical results can be obtained rapidly. Early work was focused on exact light-scattering characteristics of spheres and cylinders for which exact analytical solutions [12,14] and subsequent computer programs [2] became available. For instance, Bell and Bickel [1] characterized the size of micron-sized cylinders to within nanometers by matching the angular resonances of the polarized light-scattering matrix. Chýlek, Ngo and Pinnick [3] demonstrated characterization of droplets using their resonance characteristics. As models became more sophisticated, researchers could treat more complicated particles, like multiple [8] and osculating [27] spheres, for instance. The morphologies of such particle systems were known; thus, making the fitting to theory straightforward, even if tedious and difficult. In nearly all instances, the fitting utilizes the complex light-scattering structure of single particles.

When the particle shape is unknown, the inverse problem becomes much more complex because light scattering is strongly dependent on particle morphology [10,26]. For many years, the Lorenz-Mie theory of light-scattering from a perfect, homogeneous sphere was used as a simulant for that of unknown particles. Although the justification was often that a sphere represents some

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average particle, in reality, the Lorenz-Mie algorithm published by Bohren and Huffman [2] provided a simple means of making such calculations, which did not exist for other particle systems without significant additional effort (cf., [17]). One significant challenge with light scattering from a sphere is the strong angular resonances that occur. These resonances make it extremely difficult to interpolate the light-scattering properties to retrieve intermediate values; thus, calculations need to be made for each set of parameters.

Spheres have other shortcomings. They cannot always reproduce the great variety of light scattering that is observed, most especially in the polarization elements of the Mueller scattering matrix. Most importantly, interpreting the results is challenging. For example, Mukai et al. [18] used spheres as model particles to retrieve a refractive index of 1.38 for particles in the coma of Comet 1P/Halley. This refractive index suggests dirty ice, but this meets a problem, since such particles have an extremely short lifetime at the heliocentric distance at which the phase curve was measured. Spheroids came to replace spheres, and with the additional free parameter of eccentricity (cf., [5]), it is found that they can reproduce the light-scattering properties from a wider range of particles. However, they too have shortcomings with regard to interpretation, as the retrievals are not unique and do not always reproduce the properties of the real particles [21,22]. To address uniqueness, additional fitting parameters could be used, for instance, considering multiple wavelengths [6]. Unfortunately, it is found that a single distribution of spheroids is not able to reproduce the entire Mueller matrix at multiple wavelengths. One issue is that spheroids tend to produce a blue polarization color; whereas, the feldspar samples used in their tests have red polarization color. Recent work by Espinosa et al. [7] using a new retrieval algorithm suggests that a red polarization color can be achieved in fitting forested plumes. Such fittings have yet to be performed on well-characterized samples. Attempts also have been made using ellipsoids as simulants. Unfortunately, the best fits are achieved with model parameters that are different from the actual particles, making their use in retrievals unreliable [11].

One purpose of this study is to demonstrate that model agglomerated debris particles can reproduce the light-scattering of a sample of olivine having the same size distribution and refractive index. Although previous demonstrations by Zubko et al. [33] and Zubko [34] already have been made, additional confirmation increases the confidence in the use of such particles for remote-sensing inversions. The second purpose for this study is to demonstrate that interpolations of light scattering from polydispersions of irregularly shaped particles can yield accurate results. Such interpolations greatly reduce the computational load required to perform inversions and open the possibility of conducting real-time inversions, for instance, of atmospheric aerosols.

2. Olivine

While the use of irregularly shaped particles as model simulants appears straightforward, it is not without difficulty. Algorithms to calculate the light scattering from such particles are either numerical, like the discrete dipole approximation (DDA) [4,9,25] or they are tedious to code and not widely available [23,24]. Because of the particles' lack of symmetry, performing enough calculations to attain a converged solution for a particular set of parameters adds to the computational cost. For instance, Zubko et al. [30–32] find that 500 or more different particle configurations are needed to calculate average scattering properties for a single set of parameters. The computational resources required to complete the scattering calculations for these 500 configurations depends strongly on the number of dipoles needed to construct the desired particle shape and size parameter. For a particle with a size parameter of 10 ($\sim 4\times10^4$ dipoles), the average scattering properties can be calculated in approximately 5 days using a single 2.2 GHz cpu core; whereas, one of our test cases with a size parameter of 48 ($\sim 2\times10^6$ dipoles) took approximately 80 days using 20, 2.5 GHz CPU cores. Interpreting spectroscopic data requires enough such calculations to meet the desired wavelength resolution, compounding the computational burden. Such resource-taxing requirements apparently have dampened remote-sensing research using irregularly shaped particles, as agglomerated debris particles are currently the only model particles that have been demonstrated to reproduce the light-scattering properties of real samples of irregularly shaped dust particles having the same parameters simultaneously at multiple wavelengths, despite the suggestion that other irregularly shaped particle prescriptions may prove equally successful [33]. Currently, simulant testing has been demonstrated for Feldspar samples [28] by Zubko et al. [33] and Forsterite samples [29] by Zubko [34]. Despite the experimental samples and the agglomerated debris particles having significantly different morphologies, the simulant particles still are able to reproduce the light-scattering Mueller matrix elements of the experimentally measured particle samples when the same measured refractive indices and size distributions are used. This last point is critical because it means that if these simulant particles are used in remote-sensing retrievals, then they would yield the sample parameters.

To complement these previous comparisons, we show in Fig. 1 comparisons of the angle-dependent intensity I and degree of linear polarization P of the olivine sample (Olivine M) measured at wavelength $\lambda = 0.633$ μm that are taken from the Amsterdam Granada light-scattering database [19,20] and results calculated using model agglomerated debris particles. To fit the samples, we use the experimentally estimated refractive index ($m = m_r + im_i = 1.62 + 0.0i$) and fit the measured size distribution to a power-law distribution r^{-n}, where $n = 2.7$ (dashed red line), at $r_{min} \approx 0.1$ μm and $r_{max} \approx 3.22$ μm. We note that due to olivine being a common material in the solar system, its refractive index is well characterized. However, the precise composition of this particular sample was not measured and it was only noted by Muñoz et al. [20] that it was Mg-rich. The total intensity and degree of linear polarization are two pieces of information that can be obtained when natural, unpolarized illumination is used. The total intensity and degree of linear polarization are related to the Mueller matrix elements S_{11} and S_{12}. The other elements of the 16-element Mueller matrix have similar fittings.

What is clear from Fig. 1 is that this model data (dashed red line) fit that of the experiment fairly well. Typical fitting parameters include the magnitudes of maximum and minimum, and the zero-crossing point of the polarization. The greatest discrepancies occur in the polarization in the forward slope and in the backscatter regions. The experimental data cuts off just prior to the backscattering intensity surge that occurs as scattering angle θ approaches 180°. The amplitude of the negative polarization branch in the linear polarization response P is not as deep as the model results, although they remain within 1σ for all but one of the data points. This can be seen especially in the polarization minimum that occurs at $\theta \sim 170°$. Similar overestimation of the negative-polarization magnitude was seen in previous fittings as well [33,34]. We suspect this may be due to stray light entering the detector, since backscattering measurements are low in intensity, and specularly reflected light off dampening material from the laser beam and the forward-scattered light from the particles may still be significant and would have near neutral or positive polarization, which would reduce the magnitude of this minimum. We also note that the error bars are significantly greater in this region, due primarily to the low intensities of the scattered light. In ad-