SAND2015-8324PE

Inverse Micro-Scale Modeling:
A Numerically Invertible Reflectance Model for an Optically Thick Deposit

HARD Solids Independent Review

Tom Reichardt and Tom Kulp
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Livermore, CA 94551

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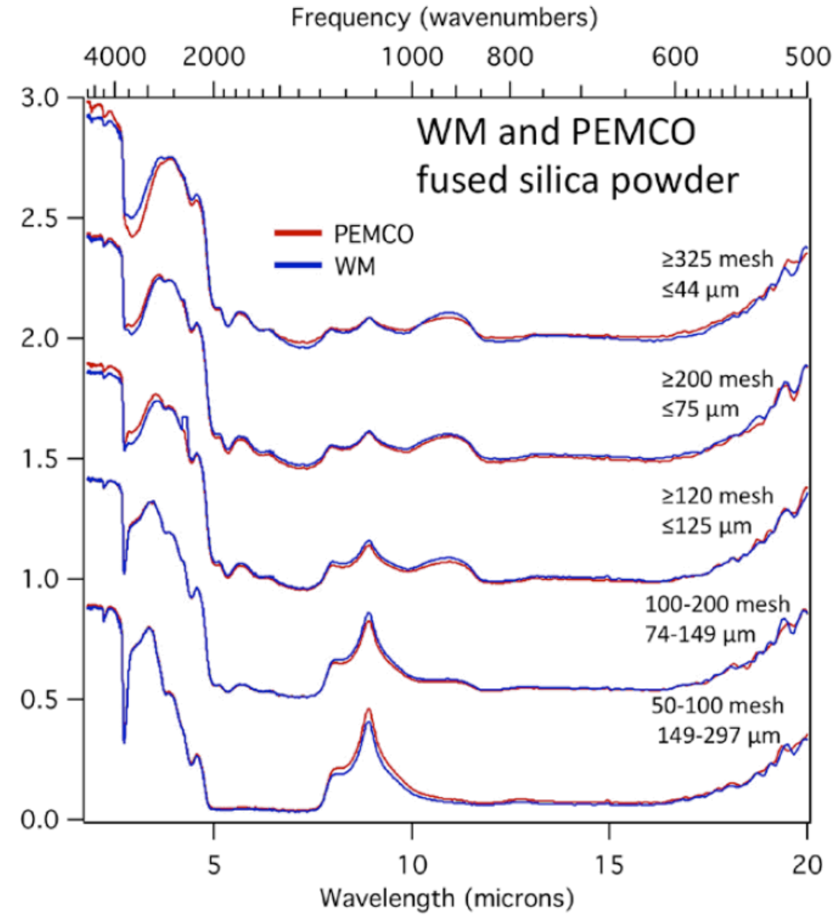
Our Goal, Method, and Focus

- ***Goal***: In a collaboration with the Micro-Scale Forward Modeling Team, develop a morphologically aware spectral library via physics-based modeling
- ***Method***: Develop radiative transfer (RT) model with parametric inputs that can be varied in optimizing model fit to measurements
  - Incorporate fundamental physical properties (*n*,*k*) and account for morphological characteristics – particle size distribution (PSD), shape, and packing density
  - Optimized, the model should demonstrate agreement with reflectance spectra…
  - …while the extracted parameters should demonstrate agreement with independent measurements of related properties



- ***Focus***: Match model to reflectance spectra of silica powders from Micro-Scale Measurements team

A Bit of the Backstory

- We had been applying the Kubelka-Munk (K-M) 2-stream model to chemical deposits of arbitrary optical thickness
  - K-M model widely applied to paints and pigments
  - Provides simple analytical expression (attractive!)
  - K-M model's scattering and absorption coefficients, as well as interface reflectance terms, depend on *n*, *k*, and morphology
  - But this dependence is not explicitly defined…
  - So, as of late 2012, we were stuck using a phenomenological model
- Met with Michael Mishchenko, Jacek Chowdhary, Brian Cairns, et al. (NASA GISS / Columbia Univ.) on 11/14/12

Acknowledgments: Michael's Suggested 5-Step Plan

1) Optically thick deposit, accounting for packing density w/ static structure factor (SSF) + quasi-crystalline approximation (QCA)
   – *Ambartsumian invariant imbedding solution*

2) Variable optical thickness with adding-doubling solution

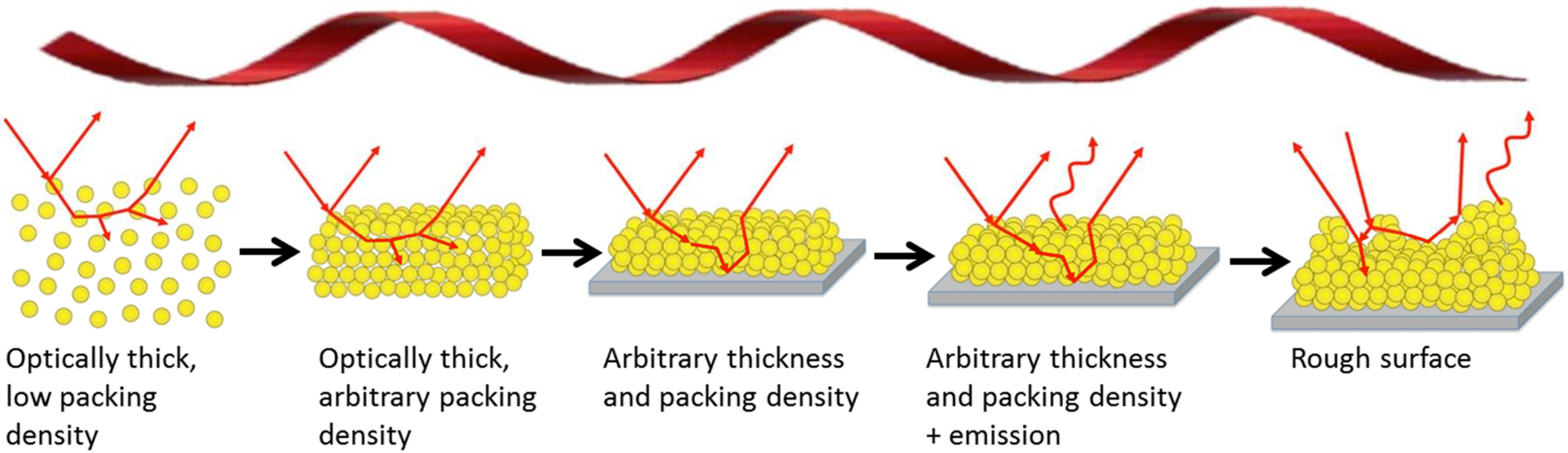
3) Incorporate thermal emission for optically thick deposit

4) Incorporate thermal emission for variable optical thickness

5) Allow for macroscopic undulations of the upper boundary
   – *Monte Carlo modeling of surface scatter*

Demonstrate inversion at every step.

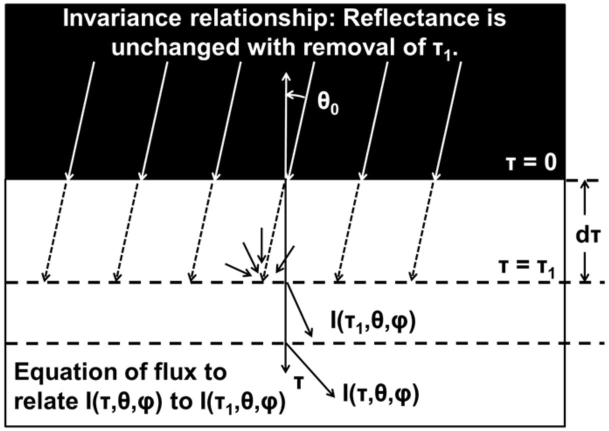
Acknowledgments: Michael's Suggested 5-Step Plan



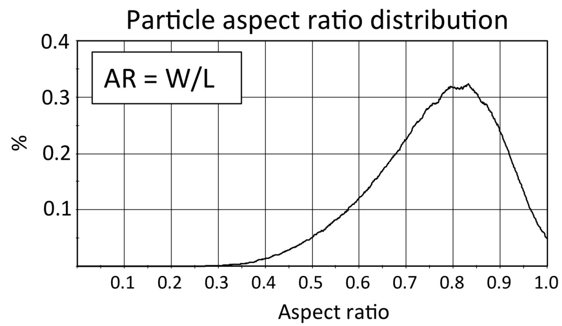
Demonstrate inversion at every step.

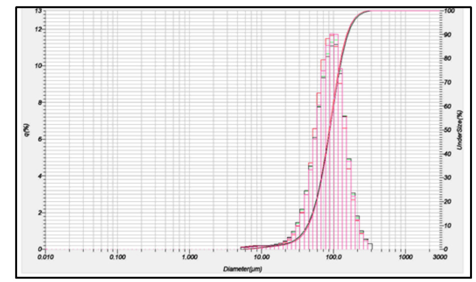
Three Discussion Topics

(1) Baseline Forward Model

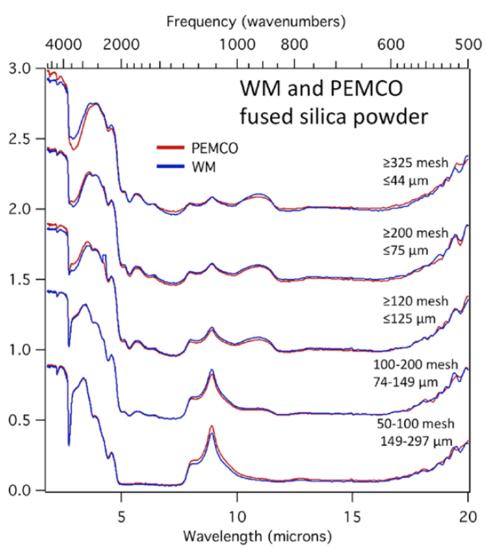


(2) Model Input Parameters





$$\mathbf{x} = \mathbf{x}_1, \mathbf{x}_2 \ldots \mathbf{x}_n$$



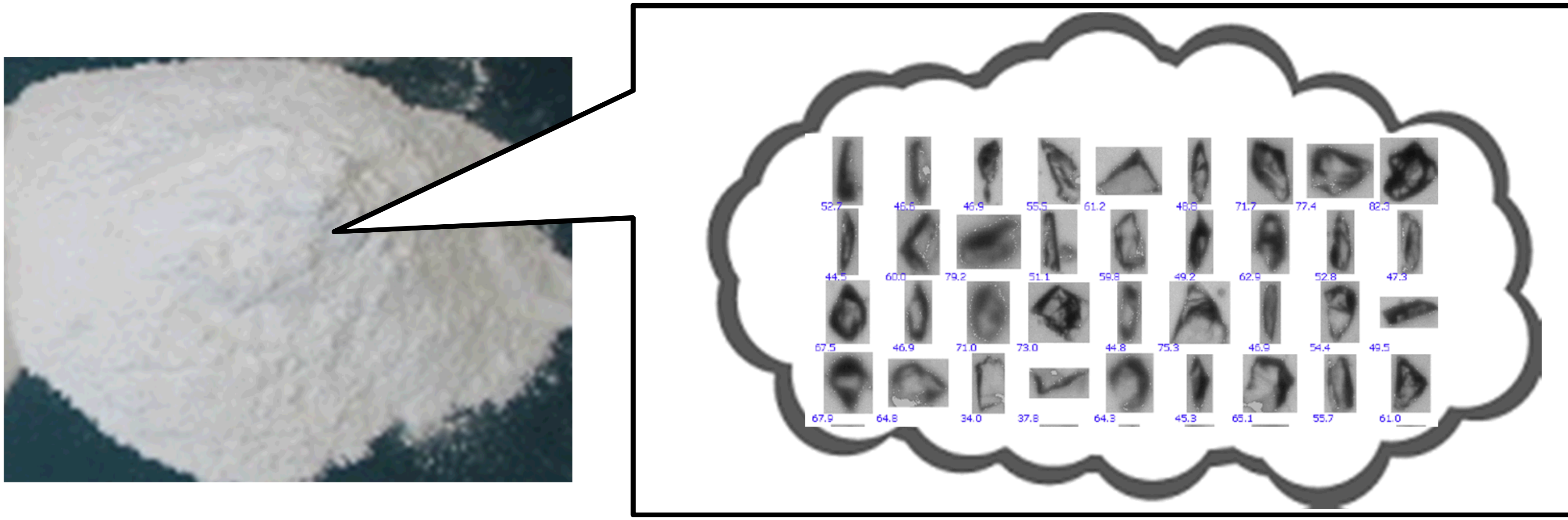
(3) Optimizing Model to Data

DAKOTA

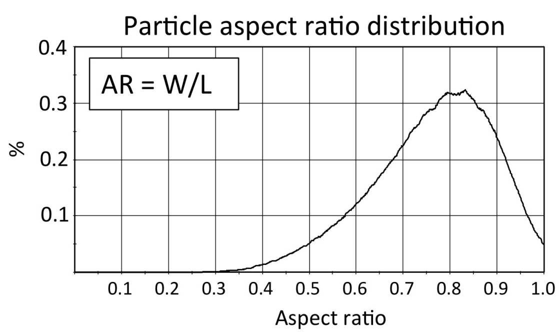
$$f(\mathbf{x}) = \sum_{i=1}^{n} \left| T_i(\mathbf{x}) \right|^2$$

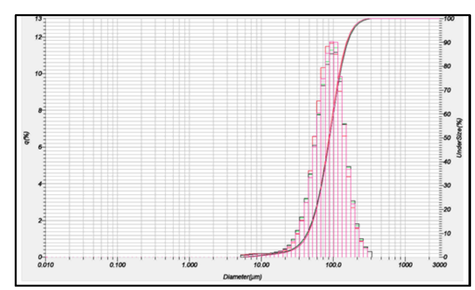
$$\frac{\partial^2 f}{\partial \mathbf{x}^2} = F\left(T_i(\mathbf{x}) \cdot T_i''(\mathbf{x}),\ \left| T_i'(\mathbf{x}) \right|^2 \right)$$

RT Model Attempts to Approximate Physics

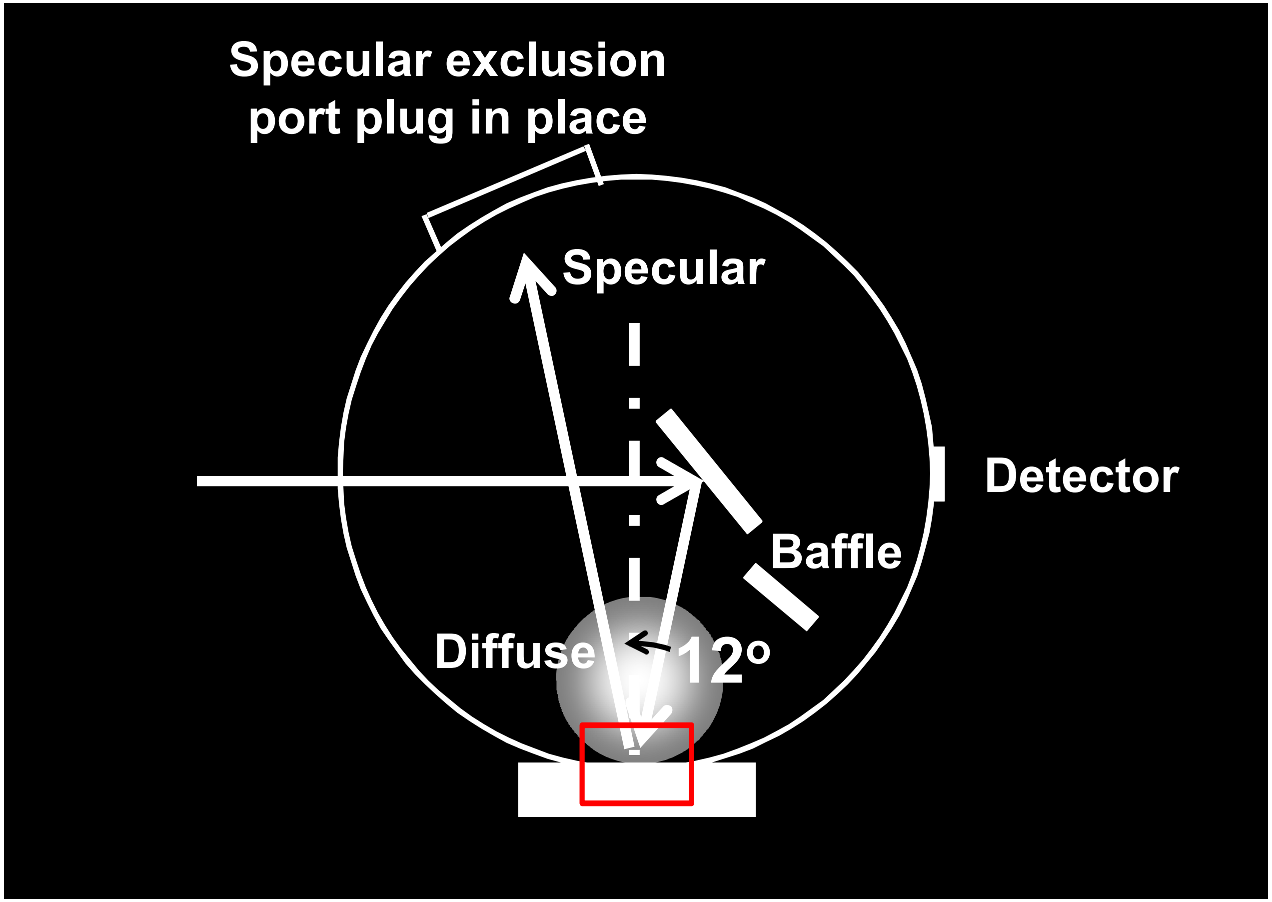


What factors might be "folded into" each model approximation, and how much crosstalk exists between these approximations?





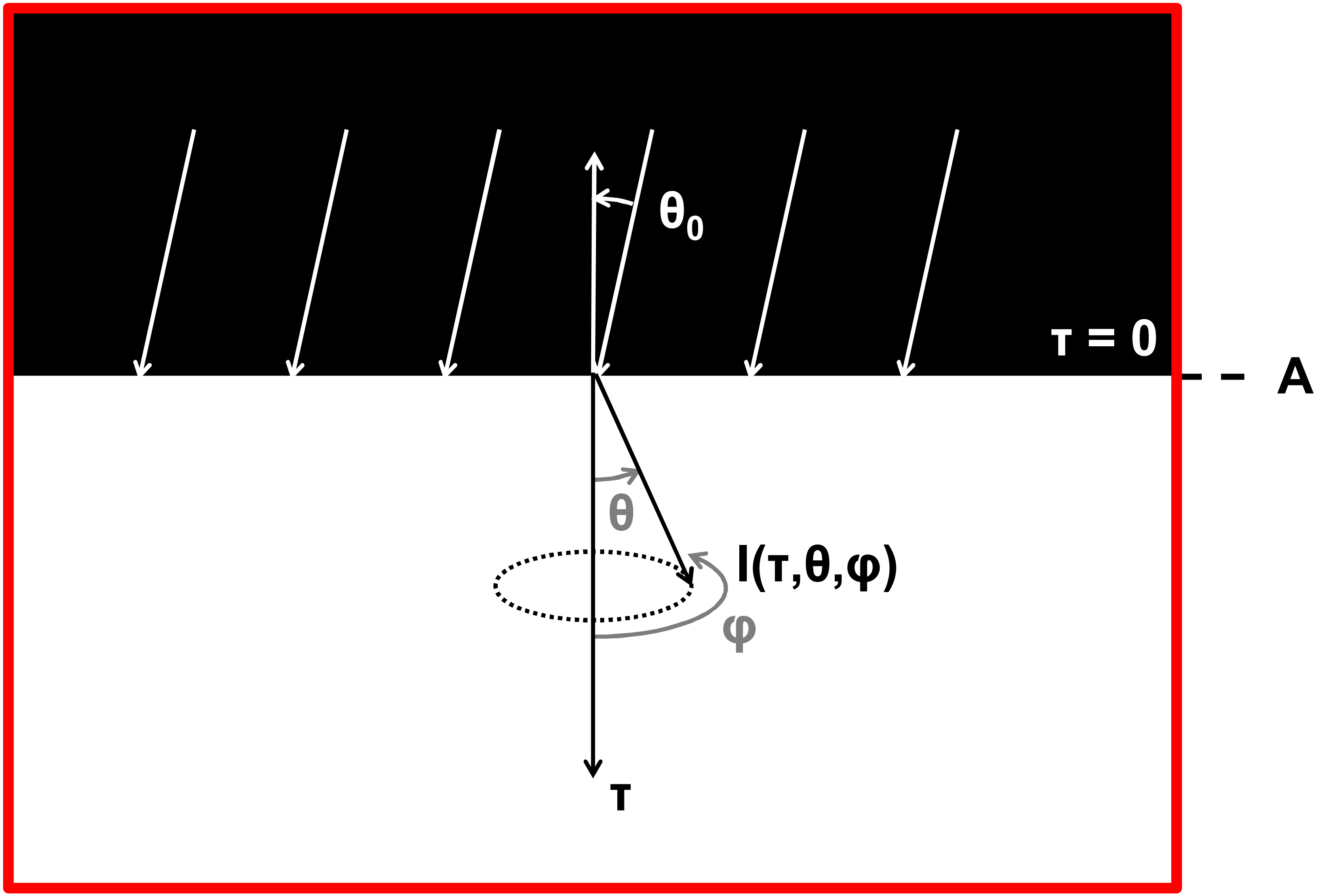
Ambartsumian Nonlinear Integral Equation



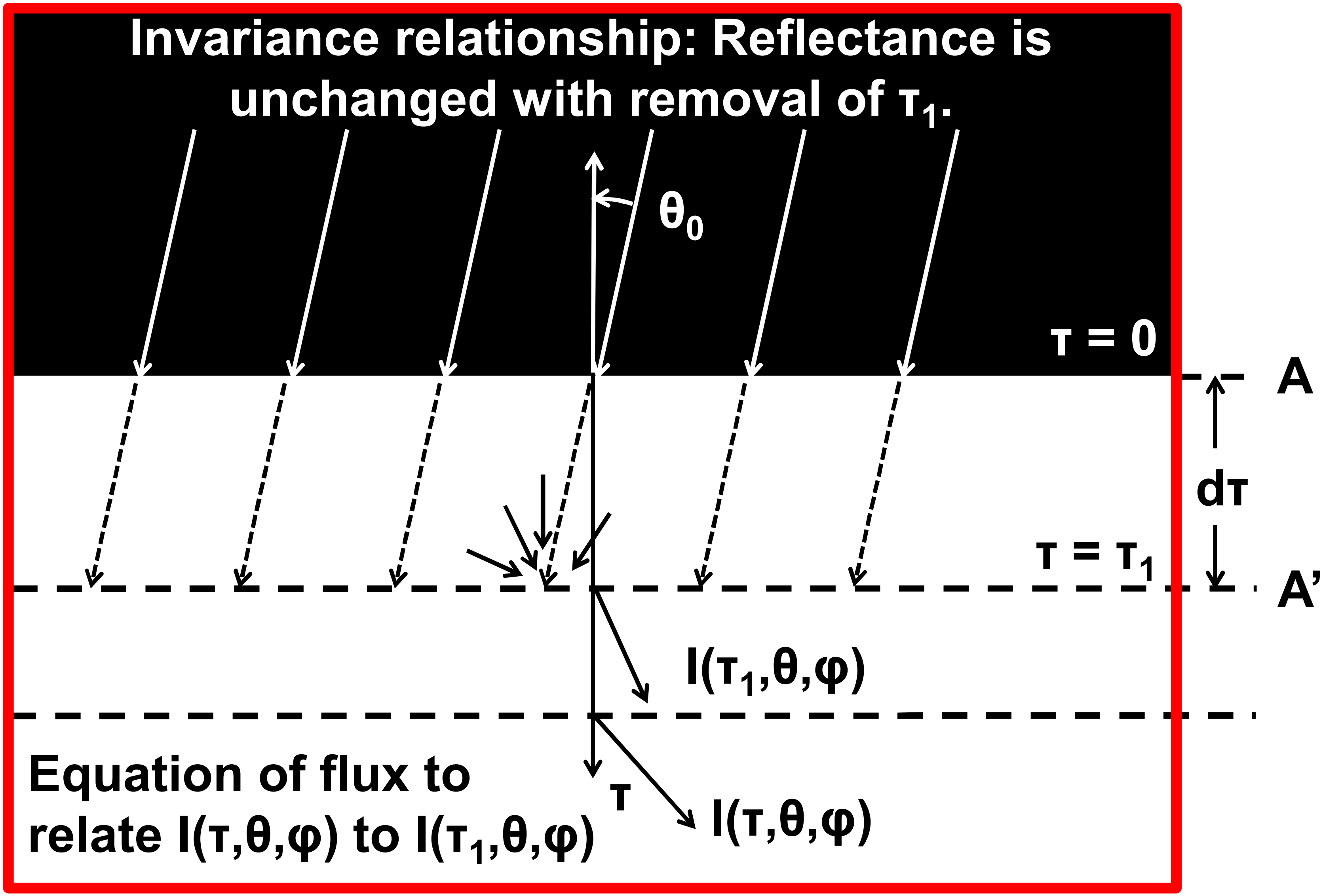
Measuring plane albedo with 12° incidence angle

Ambartsumian Nonlinear Integral Equation

Ambartsumian Nonlinear Integral Equation



Ambartsumian Nonlinear Integral Equation



Ambartsumian Nonlinear Integral Equation

E. G. Yanovitskij, *Light scattering in Inhomogeneous Atmospheres*, Trans. by S. Ginsheimer and O. Yanovitskij, Springer (1997).

$$
\begin{aligned}
(\mu+\mu_0) R^m(\mu,\mu_0) = {} & \frac{\varpi}{4} P^m(-\mu,\mu_0) \\
& + \frac{\varpi}{2}\mu_0 \int_0^1 P^m(\mu,\mu') R^m(\mu',\mu_0)\, d\mu' \\
& + \frac{\varpi}{2}\mu \int_0^1 R^m(\mu,\mu') P^m(\mu',\mu_0)\, d\mu' \\
& + \varpi \mu \mu_0 \int_0^1 \int_0^1 R^m(\mu,\mu') P^m(-\mu',\mu'') \\
& \qquad \cdot R^m(\mu'',\mu_0)\, d\mu' d\mu''
\end{aligned}
$$

$$P^m, \varpi \longrightarrow R^m$$

Ambartsumian Nonlinear Integral Equation

PERGAMON

Journal of Quantitative Spectroscopy & Radiative Transfer 63 (1999) 409–432

Journal of Quantitative Spectroscopy & Radiative Transfer

FORTRAN codes

To retrieve a code, click on the code name and use the "Save As..." option from the "File" menu.

- refl.f - This code computes Fourier components of the reflection function
- interp.f - This code computes the bidirectional reflection function for a given set of scattering geometries
- spher.f - This code computes the Legendre expansion coefficients for polydisperse spherical particles using the standard Lorenz-Mie theory

We describe a simple and highly efficient and accurate radiative transfer technique for computing bidirectional reflectance of a macroscopically flat scattering layer composed of nonabsorbing or weakly absorbing, arbitrarily shaped, randomly oriented and randomly distributed particles. The layer is assumed to be homogeneous and optically semi-infinite, and the bidirectional reflection function (BRF) is found by a simple iterative solution of the Ambartsumian's nonlinear integral equation. As an exact solution of the radiative transfer equation, the reflection function thus obtained fully obeys the fundamental physical laws of energy conservation and reciprocity. Since this technique bypasses the computation of the internal radiation field, it is by far the fastest numerical approach available and can be used as an ideal input for Monte Carlo procedures calculating BRFs of scattering layers with macroscopically rough surfaces. Although the effects of packing density and coherent backscattering are currently neglected, they can also be incorporated. The FORTRAN implementation of the technique is available on the World Wide Web at http://www.giss.nasa.gov/ ~ crmim/brf.html and can be applied to a wide range of remote sensing, engineering, and biophysical problems. We also examine the potential effect of ice crystal shape on the bidirectional reflectance of flat snow surfaces and the applicability of the Henyey–Greenstein phase function and the δ-Eddington approximation in calculations for soil surfaces.

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0022-4073/99/$ - see front matter
PII: S0022-4073(99)00028-X

Electromagnetic Scattering by Particles and Surfaces

leave a message at mmishchenko@giss.nasa.gov indicating your name and mailing address.

The users of the codes are encouraged to visit this page on a regular basis for information on latests developments, warnings, and/or errors found. We would highly appreciate informing us of any problems and errors encountered with these codes. Please e-mail your questions and comments to mmishchenko@giss.nasa.gov.

FORTRAN codes
To retrieve a code, click on the code name and use the "Save As..." option from the "File" menu.

- refl.f - This code computes Fourier components of the reflection function
- interp.f - This code computes the bidirectional reflection function for a given set of scattering geometries
- spher.f - This code computes the Legendre expansion coefficients for polydisperse spherical particles using the standard Lorenz-Mie theory

The codes must be run in the following sequence: spher.f -> refl.f -> interp.f.

Note that the Legendre expansion coefficients for polydisperse, randomly oriented nonspherical particles and sphere aggregates can be computed using O T-matrix codes also available on this website. The expansion coefficients for the standard and double-peaked Henyey-Greenstein phase functions are computed using Eqs. (15) and (19) of the manual. Below we also provide the Legendre expansion coefficients for two nonspherical ice particle models described in the manual.

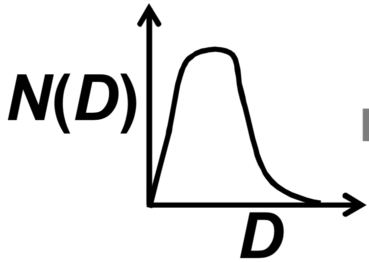
Benchmark results
The following output files were computed by the codes in their current settings and may provide a useful test of the performance of the codes on different computers:

- spher.print
- spher.write
- refl.print
- interp.write

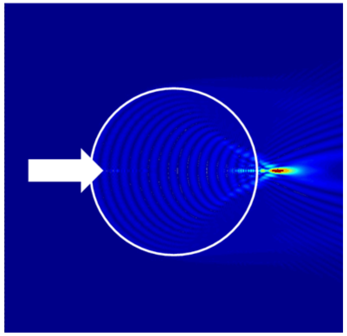
The file refl.write is not given here because of its large size.

Solution Provided by Michael Mishchenko (NASA GISS)

Step #1: n, k +



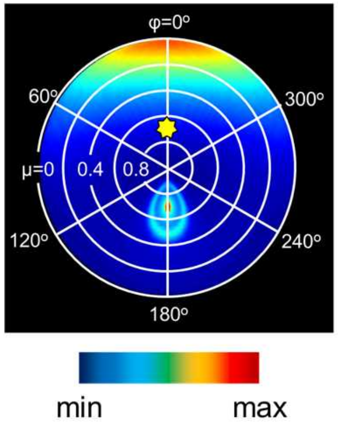
→ P^m, $\tilde{\omega}$



$$(\mu+\mu_0)R^m(\mu,\mu_0)=\frac{\varpi}{4}P^m(-\mu,\mu_0)+\frac{\varpi}{2}\mu_0\int_0^1 P^m(\mu,\mu')\,R^m(\mu',\mu_0)\,d\mu'$$
$$+\frac{\varpi}{2}\mu\int_0^1 R^m(\mu,\mu')\,P^m(\mu',\mu_0)\,d\mu'$$
$$+\varpi\mu\mu_0\int_0^1\int_0^1 R^m(\mu,\mu')\,P^m(-\mu',\mu'')\cdot R^m(\mu'',\mu_0)\,d\mu'\,d\mu''$$

Step #2: P^m, $\tilde{\omega}$ → R^m

Step #3: R^m + μ_0 → BRDF(μ_0)



Calculating P^m, $\tilde{\omega}$

Pergamon

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0022-4073/98 $19.00 + 0.00

CAPABILITIES AND LIMITATIONS OF A CURRENT FORTRAN IMPLEMENTATION OF THE *T*-MATRIX METHOD FOR RANDOMLY ORIENTED, ROTATIONALLY SYMMETRIC SCATTERERS

MICHAEL I. MISHCHENKO† and LARRY D. TRAVIS
NASA Goddard Institute for Space Studies, 2880 Broadway, New York, New York 10025, U.S.A.

Abstract—We describe in detail a software implementation of a current version of the *T*-matrix method for computing light scattering by polydisperse, randomly oriented, rotationally symmetric particles. The FORTRAN *T*-matrix codes are publicly available on the World Wide

$$\begin{bmatrix} \mathbf{p} \\ \mathbf{q} \end{bmatrix} = \begin{bmatrix} \mathbf{T}^{11} & \mathbf{T}^{12} \\ \mathbf{T}^{21} & \mathbf{T}^{22} \end{bmatrix}$$

$\pi D/\lambda > 40$

procedure convenient in massive computer calculations for particle polydispersions, and Ref. 5 presents benchmark *T*-matrix computations for particles with non-smooth surfaces (finite circular cylinders). A general review of the *T*-matrix method can be found in Ref. 7.

In this paper we provide a detailed description of modern *T*-matrix FORTRAN codes which incorporate all recent developments, are publicly available on the World Wide Web, and are, apparently, the most efficient and powerful tool for accurately computing light scattering by randomly oriented rotationally symmetric particles. For the first time, we collect in one place all necessary formulas, discuss numerical aspects for *T*-matrix computations, describe the input and output parameters, and demonstrate the capabilities and limitations of the codes. The paper is intended to serve as a detailed user guide to a versatile tool suitable for a wide range of practical applications. We specifically target the users who are interested in practical applications of the *T*-matrix method rather than in details of its mathematical formulation.

2. BASIC DEFINITIONS

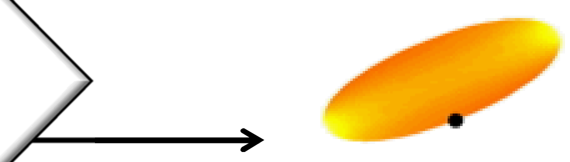
The single scattering of light by a small-volume element dv consisting of randomly oriented, rotationally symmetric, independently scattering particles is completely described by the ensemble-averaged extinction, C_{ext}, and scattering, C_{sca}, cross sections per particle and the dimensionless

† Author to whom correspondence should be addressed.

Applicability of regular particle shapes in light scattering calculations for atmospheric ice particles

Andreas Macke and Michael I. Mishchenko

We ascertain the usefulness of simple ice particle geometries for modeling the intensity distribution of light scattering by atmospheric ice particles. To this end, similarities and differences in light scattering by axis-equivalent, regular and distorted hexagonal cylindric, ellipsoidal, and circular cylindric ice particles are reported. All the results pertain to particles with sizes much larger than a



erably larger than the wavelengths of the incoming solar radiation, especially in the visible spectral region. Therefore, the geometrical optics approximation offers a conceptually simple although time-consuming way to simulate single scattering by almost arbitrarily shaped scatterers.[1–5] Whereas these papers take more and more complex particle geometries such as bullet rosettes, dendrites, or polycrystals into account, in this paper we examine the possibility of representing the scattering proper-

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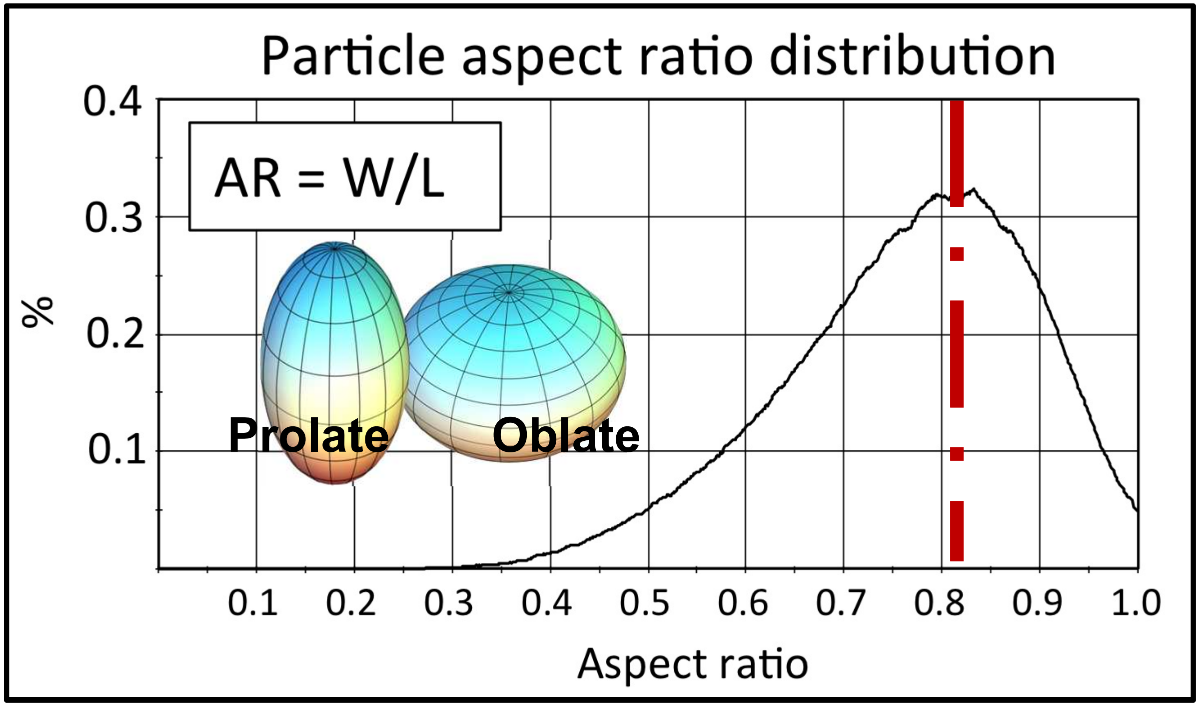
Received 28 August 1995; revised manuscript received 29 January 1996.
0003-6935/96/214291-06$10.00/0

On the other hand, the three (two) semiaxes of an ellipsoid (circular cylinder) allow for a variability of particle shapes that may cover to some extent the natural variability of atmospheric ice crystal habits.

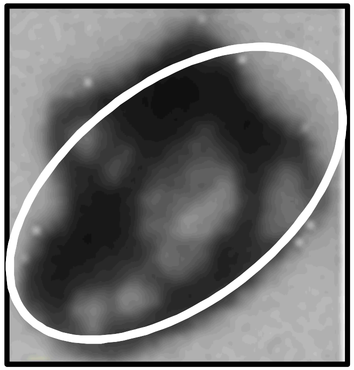
Another motivation arises from uncertainties in our knowledge of real ice particle shapes. The study of observationally derived two-dimensional ice crystal shadow images[6] or replicas[7,8] clearly demonstrates that solid hexagonal columns or plates are a strong idealization of atmospheric ice crystals. However, statistically reliable shape information is difficult to extract from these data, partly because of the strong natural variability. Therefore it appears reasonable to ascertain the use of nonhexagonal but still simple geometries as substitutes for a polydisperse ion of complicated ice particle shapes.

Because of the lack of sharp edges, ellipsoids do not provide strong halos that are characteristic of regular hexagonal particles. However, the absence of these features, as reported in a number of radiance measurements in or above cirrus clouds,[9,10] emphasizes the potential use of nonhexagonal par-

Our Canonical Geometry: Spheroids



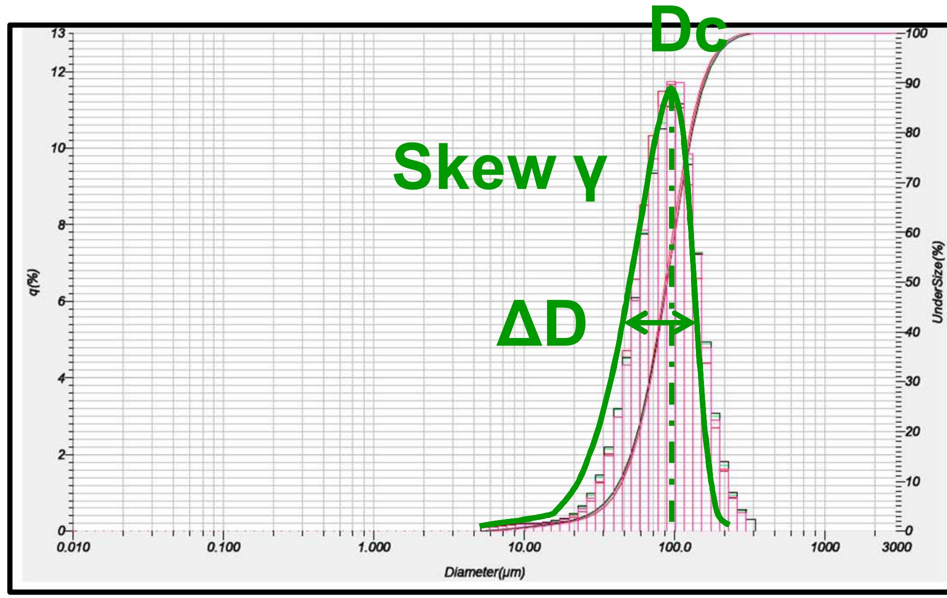
- 2 shape bins: AR = 0.8 (one bin each per prolate, oblate)
- Silica powders not really spheroidal → Treat particle as silica/air mixture



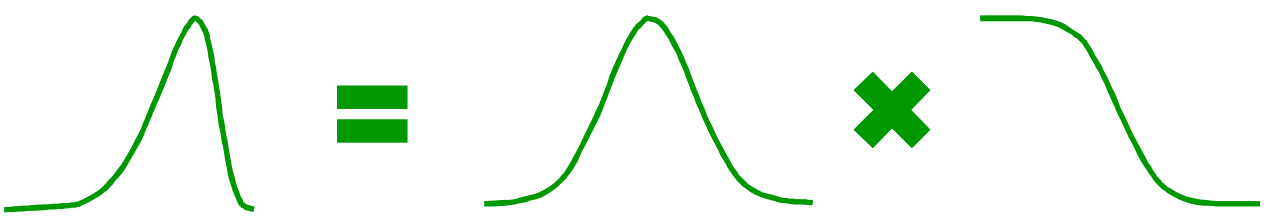
$$\varepsilon_{eff} = \varepsilon_{sil}\left[1 + \frac{3 f_{air}\left(\dfrac{\varepsilon_{air} - \varepsilon_{sil}}{\varepsilon_{air} + 2\varepsilon_{sil}}\right)}{1 - f_{air}\left(\dfrac{\varepsilon_{air} - \varepsilon_{sil}}{\varepsilon_{air} + 2\varepsilon_{sil}}\right)}\right]$$

Particle Size Distribution (PSD)

- Approximate with skewed *volume* log-normal distribution
- Expression with 3 parameters: r_g, σ_g, γ



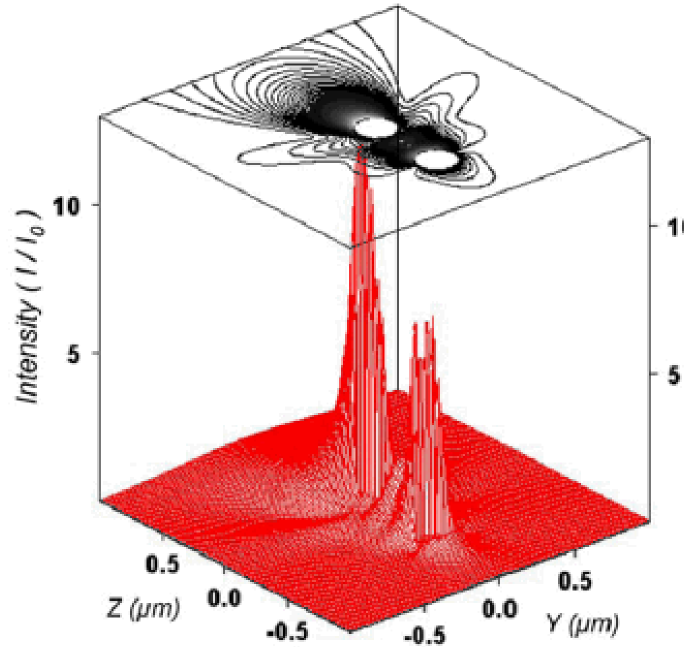
$$n(r) = \text{constant} \times r^{-4} \exp\left[-\frac{\left(\ln r - \ln r_g\right)^2}{2\ln^2 \sigma_g} \right] \times 2\left[1 + \text{erf}\left(\frac{\alpha\left(\ln r - \ln r_g\right)^2}{2\sqrt{2}\,\ln^2 \sigma_g} \right) \right]$$



"Patching" the Scattering Properties

- The radiative transfer equation (RTE) is strictly applicable only for sparse media (packing density < 1%)
- ***S(θ): Static structure factor (SSF)***
  - Acts as a multiplier to the scattering cross section and phase function
  - Analytical expression available for monodisperse spheres

$$p(\theta) = \frac{4\pi}{C_{\mathrm{sca}}} \frac{dC_{\mathrm{sca}}}{d\Omega} S(\theta)$$



J.-C. Auger and B. Stout, "Local field intensity in aggregates illuminated by diffuse light: T matrix approach," Appl. Opt. **47**, 2897-2905 (2008).

"Patching" the Scattering Properties

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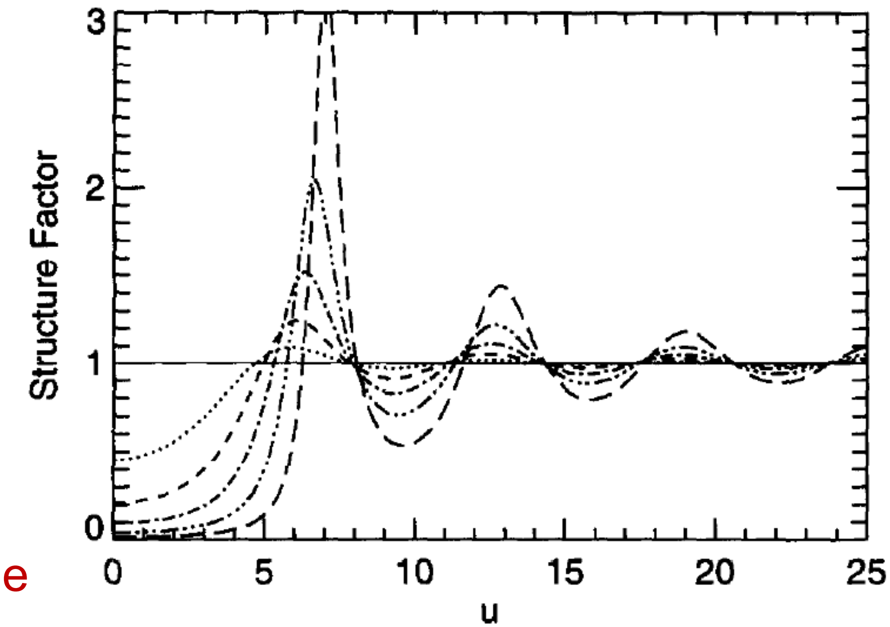
- ***S(θ): Static structure factor (SSF)***
  - Acts as a multiplier to the scattering cross section and phase function
  - Analytical expression available for monodisperse spheres
  - *S(θ) = F(f_{SSF}, u)*
  - $u = \frac{8\pi r_0}{\lambda}\sin\left(\frac{\theta}{2}\right)$

$$= 4\left(\frac{\pi D}{\lambda}\right)\times\sin\left(\frac{\theta}{2}\right)$$

Scattering angle

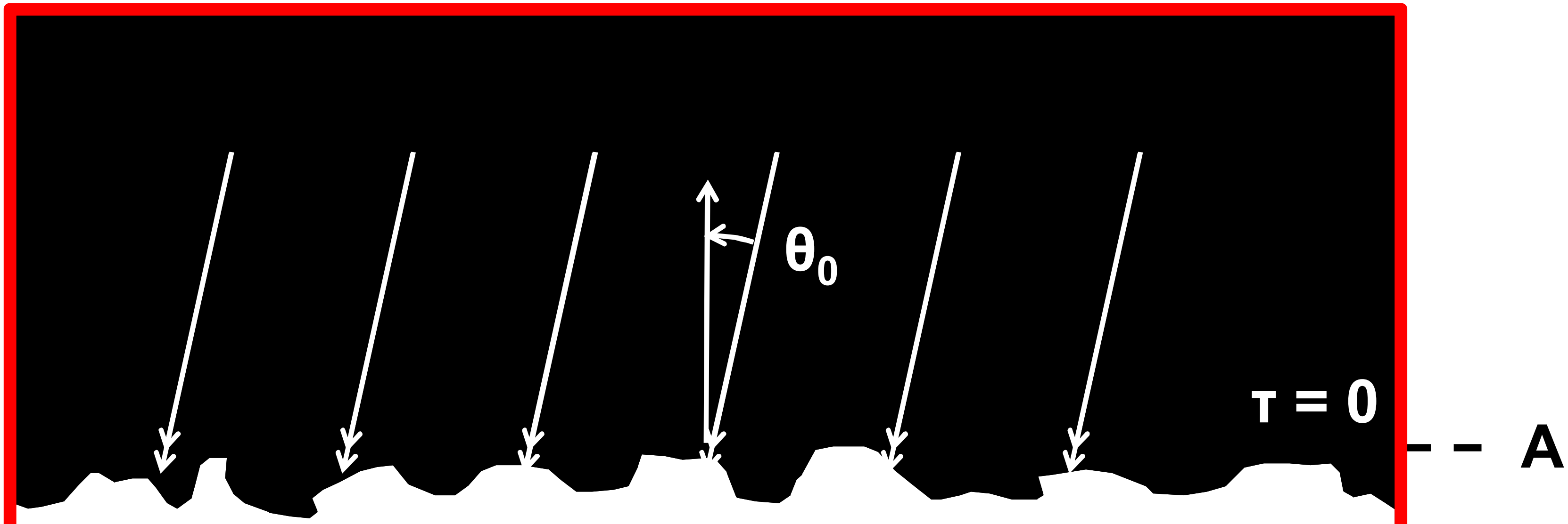
Size parameter

$$p(\boldsymbol{\theta}) = \frac{4\pi}{C_{sca}}\frac{dC_{sca}}{d\Omega}S(\theta)$$



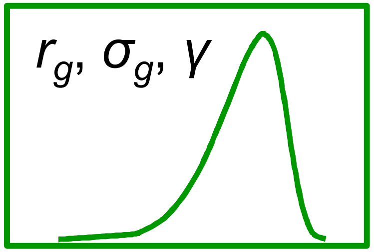
M. I. Mishchenko, "Asymmetry parameters of the phase function for densely packed scattering grains," JQSRT **52**, 95-110 (1994).

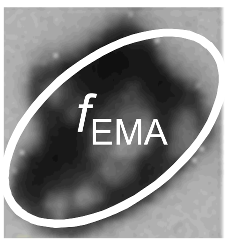
Macroscopic Undulations of Upper Boundary

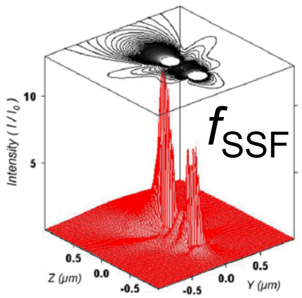


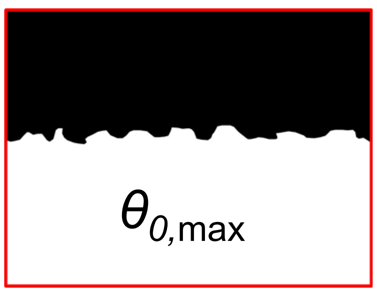
- Range of incidence angles: θ_0 encompasses 0^o to $\theta_{0,max}$
- Rougher surface → Larger $\theta_{0,max}$
- Impacts contribution of first-surface reflection
  - e.g., “surface scattering”
  - Especially significant for high *k*

Optimizing Model to Measurements







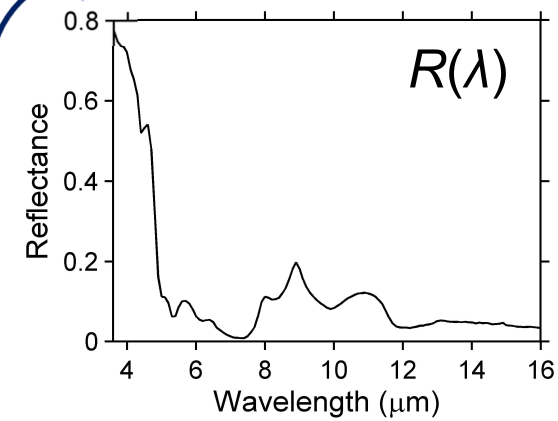


6 parameters

DAKOTA

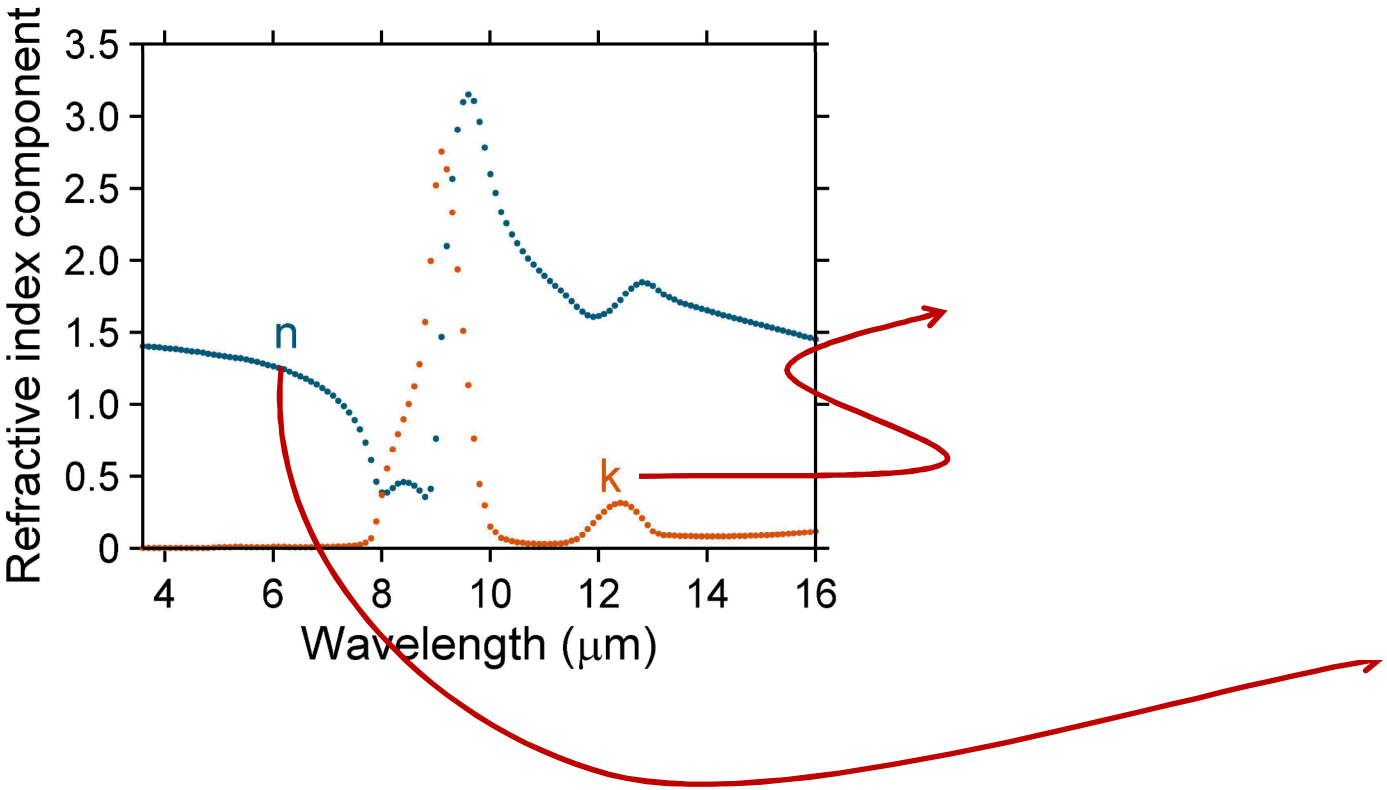
$$(\mu+\mu_0)R^m(\mu,\mu_0)$$
$$=\frac{\varpi}{4}P^m(-\mu,\mu_0)$$
$$+\frac{\varpi}{2}\mu_0\int_0^1 P^m(\mu,\mu')\,R^m(\mu',\mu_0)\,d\mu'$$
$$+\frac{\varpi}{2}\mu\int_0^1 R^m(\mu,\mu')\,P^m(\mu',\mu_0)\,d\mu'$$
$$+\varpi\mu\mu_0\int_0^1\int_0^1 R^m(\mu,\mu')\,P^m(-\mu',\mu'')$$
$$\cdot R^m(\mu'',\mu_0)\,d\mu'\,d\mu''$$

Computational Model



Compare to measured reflectance spectrum

Wavelength Dependence: Literature *n* and *k* for Silica, 3.6-16 µm



Optimization via "nl2sol" in DAKOTA

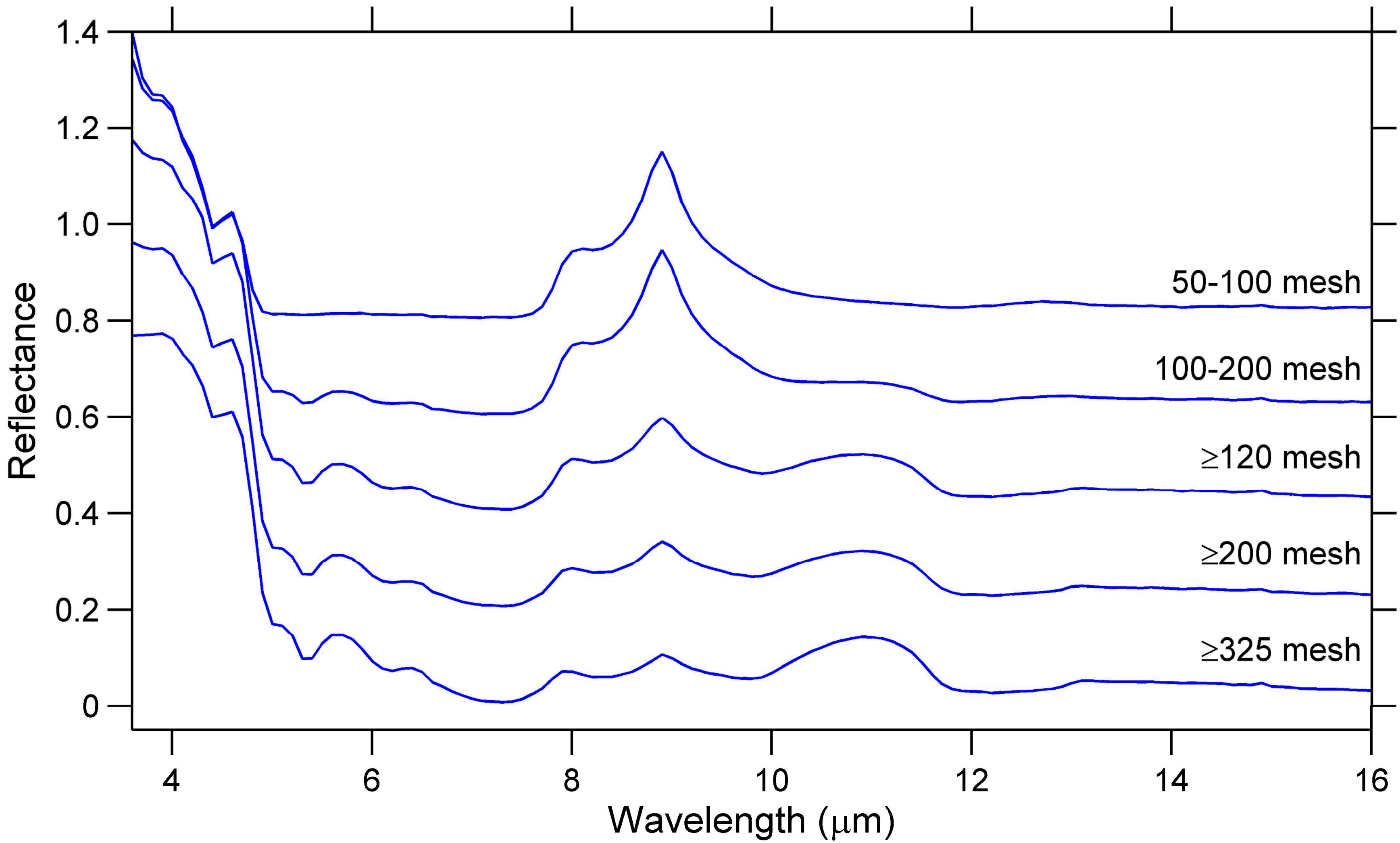
- 125 λs: 3.6-16 μm @ 0.1 μm resolution
- The 125 λ-dependent calculations are divided among 64 processors (4 nodes, 16 cores/node)
- The Jacobian (matrix of 1st-order partial derivatives) is numerically determined through forward difference calculations.
- The Hessian (matrix of 2nd-order partial derivatives) is numerically approximated from special properties of the sum-of-squares:

$$f(\mathbf{x}) = \sum_{i=1}^{n} \left| T_i(\mathbf{x}) \right|^2$$

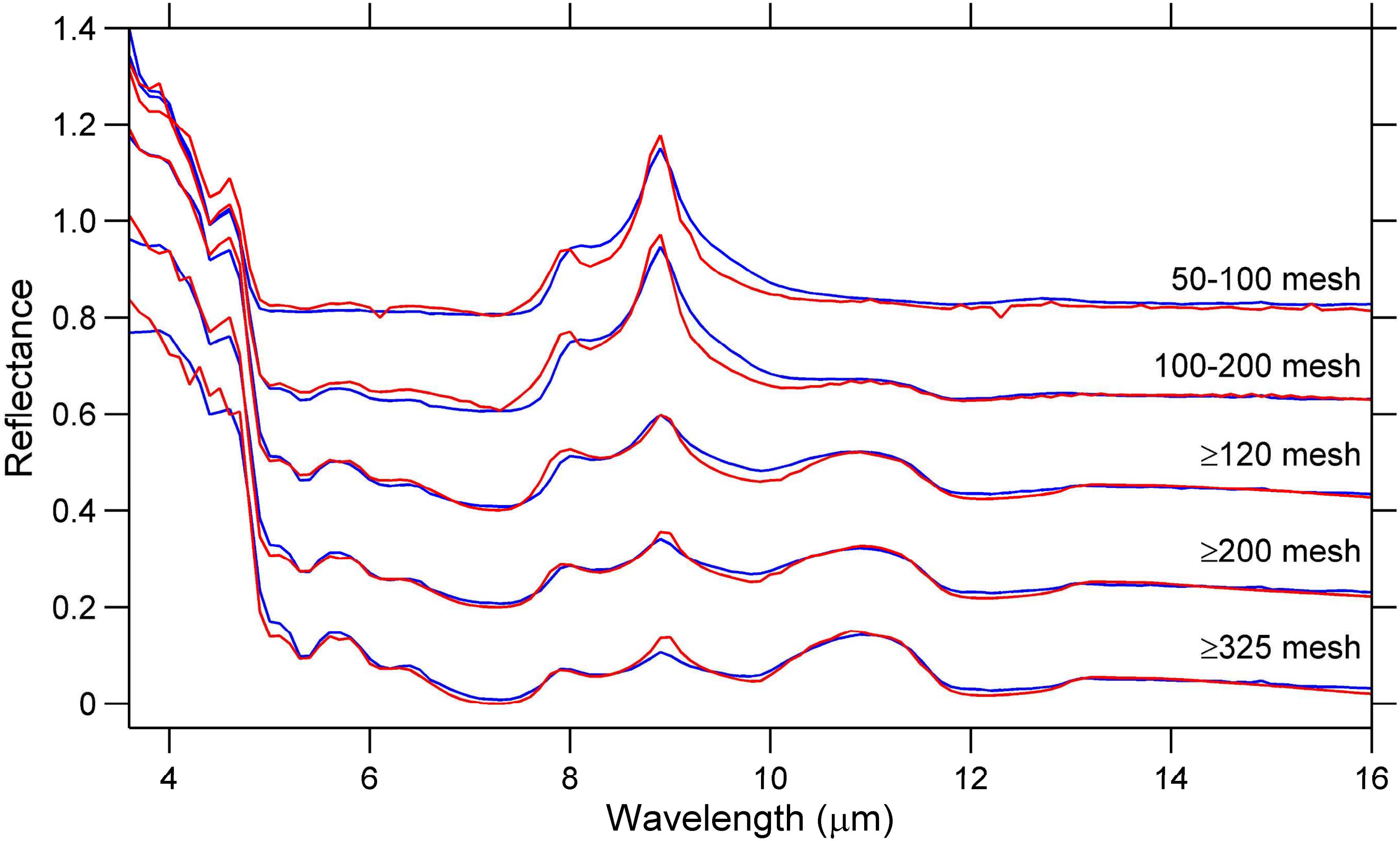
$$\frac{\partial^2 f}{\partial \mathbf{x}^2} = F\left(\cancelto{0}{T_i(\mathbf{x})} \cdot T_i''(\mathbf{x}),\ \left| T_i'(\mathbf{x}) \right|^2 \right)$$

- (But the original guess for **x** has to be reasonably close)

Measured Reflectance Spectra of Silica Model Systems



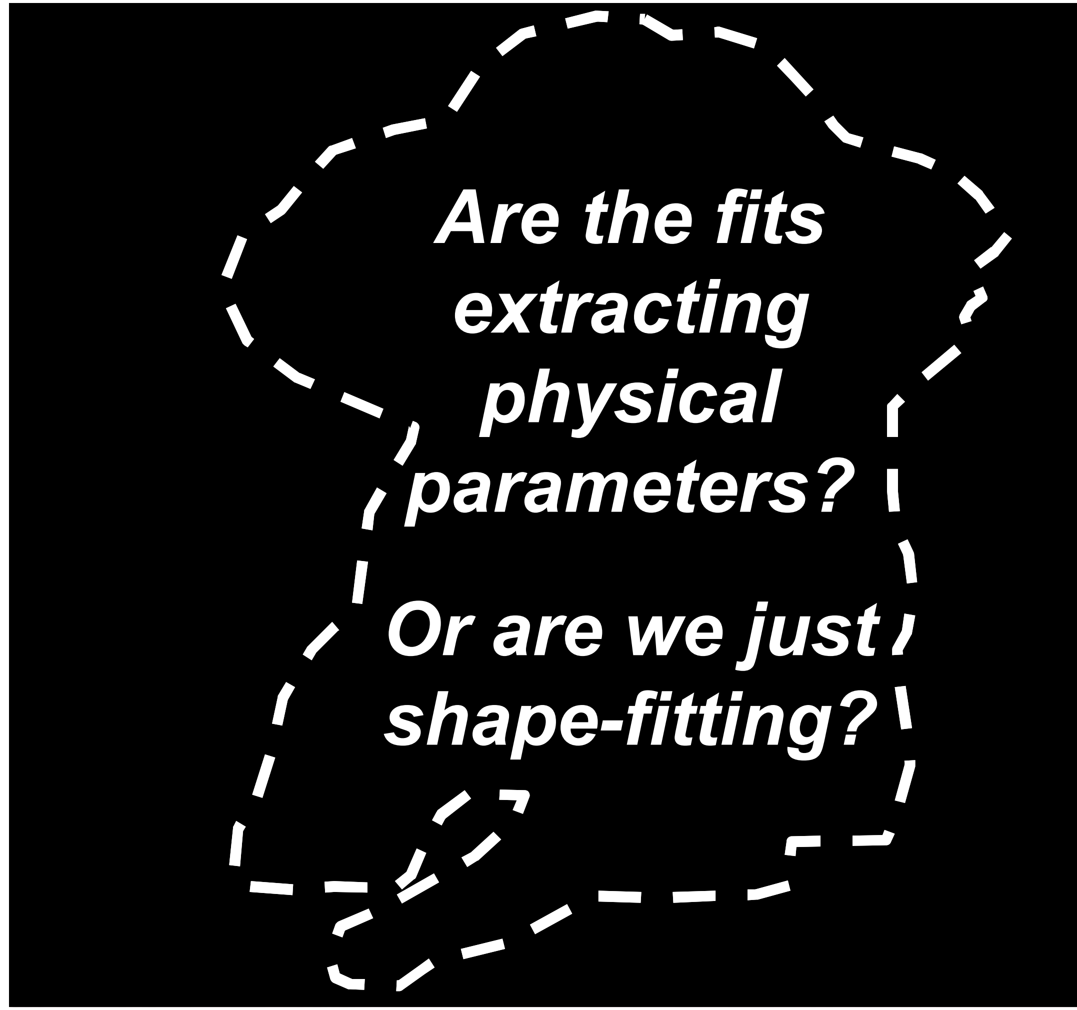
Modeled Reflectance Spectra of Silica Model Systems



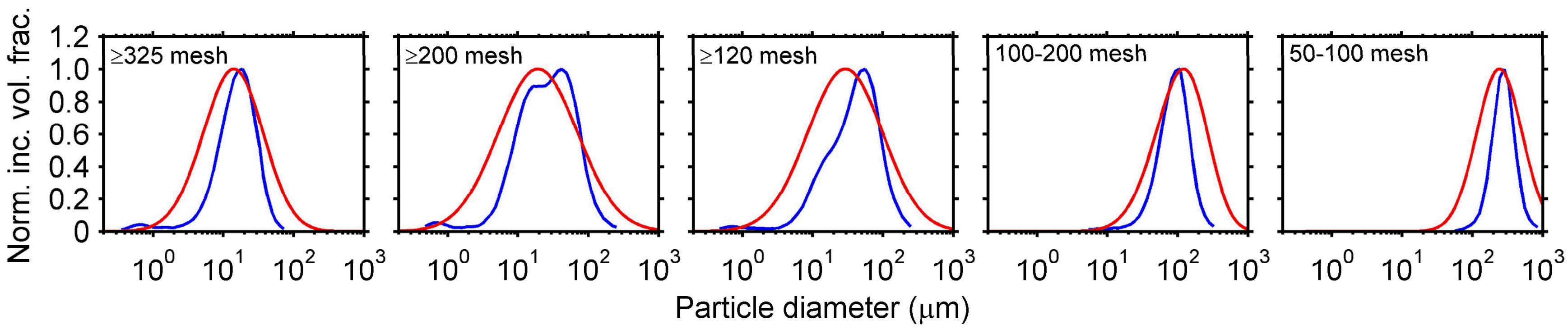
Nice agreement, *but…*

…“give me seven free parameters and I can fit an elephant.”

K. Lumme and A. Penttilä, “Model of light scattering by dust particles in the solar system: Application to cometary comae and planetary regoliths,” J. Quant. Spectrosc. & Rad. Trans. **112**, 1658-1670 (2011).



Measured PSDs vs Reflectance-Extracted PSDs



Mesh	f_{EMA}	f_{SSF}	θ_0
≥325	0.058	0.34	0-31°
≥200	0.046	0.79	0-47°
≥120	0.009	0.60	0-66°
100-200	0.24	0.81	0-89°
50-100	0.29	0.89	0-89°

Playing the Devil's Advocate

- Spectral agreement suffers between 8-10 µm
  - So is the agreement "good enough?"
  - Assuming accurate *n*/*k* values (?), is the disagreement due to the model approximations? To the numerical inversion?
  - Is another factor needed in the model?
- Extracted PSDs are wider than those measured via laser diffraction
  - How good are the laser diffraction measurements?
- Measurement-vs-model comparisons are limited to $\pi D/\lambda \geq 1$ (thus far)
  - Narrower size distributions to be measured and modeled
- Given the variability of f_{SSF}, is it having the expected impact?
- Why is f_{EMA} so much larger for coarse particles than for fine particles?
  - Should $f_{EMA} = \boldsymbol{F}(\pi D/\lambda)$?

Task Summary, Relationships to Other Tasks, and Next Steps

- Numerically invertible model provides the means to account for particle size effects in the reflectance spectra of particulate media
  - Computational model is based on fundamental physical properties (*n*,*k*)
  - Extracted parameters can be compared to auxiliary measurements
  - The PSD agreement is encouraging (we are not just fitting an elephant)…
  - … but we desire improved understanding of the f_{EMA} and f_{SSA} trends.
- Our effort directly links to the other *HARD Solids* tasks
  - ***Laboratory Optical Measurements***: Modeling more materials as *n*,*k* become available
  - ***Laboratory System Measurements***: Considering increased system complexity
  - ***Forward Microscale Modeling***: Numerically inverting more advanced models
  - ***Validation Framework***: Performing Bayesian calibration via Dakota
  - ***System Model***: Providing morphologically aware input spectra
- Next: Increased rigor and system complexity
  - Assist with model validation effort → Begin to define what is "good enough"
  - Implement P_m, $\tilde{\omega}$ in adding-doubling solution for deposits of variable optical thickness (On to Step #2 on Michael's list!)

Thanks! Questions?

Surface Scatter vs. Volume Scatter