Modeling the Reflectance of Particulate Media

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Presented at IARPA MORGOTH'S CROWN Workshop
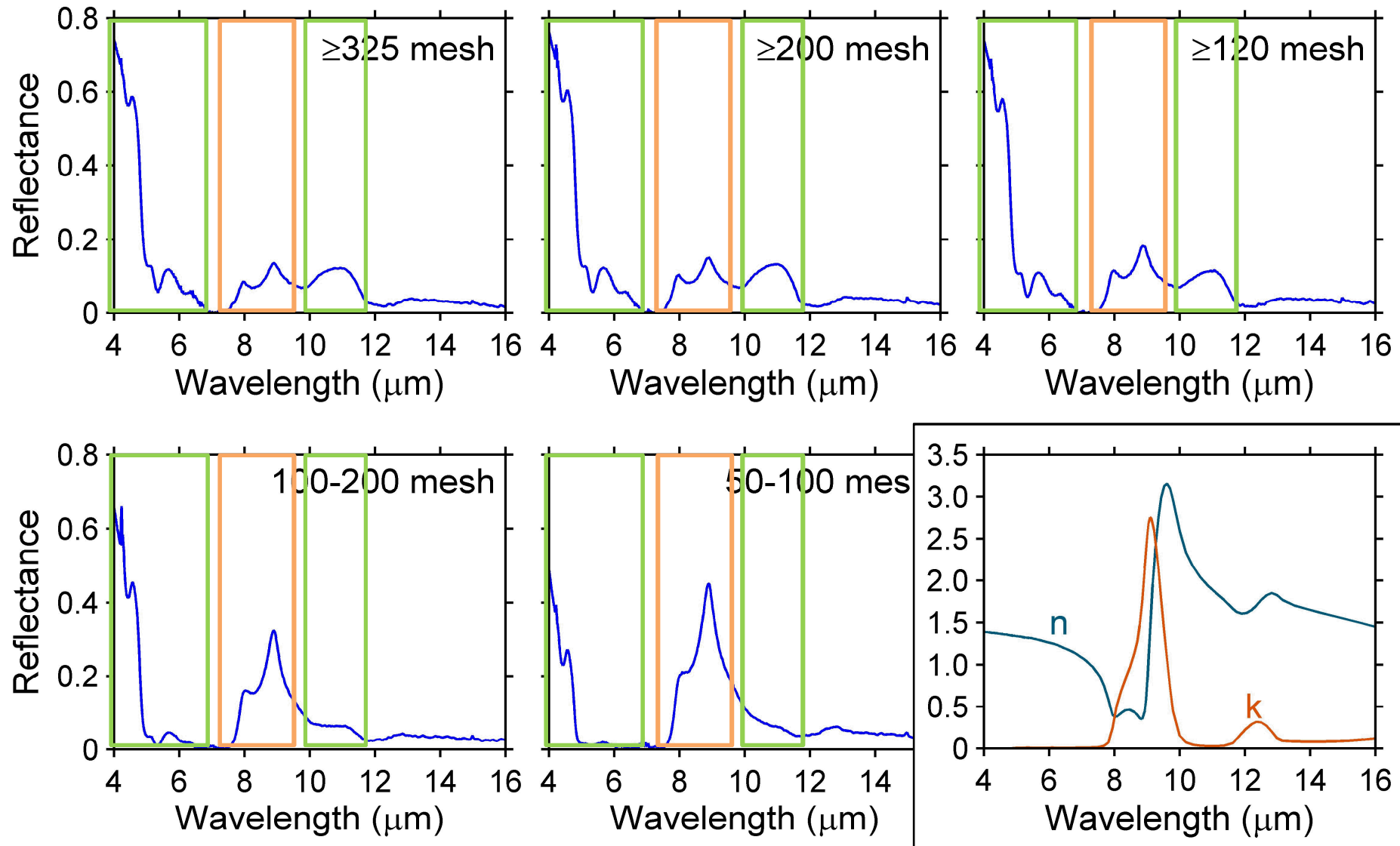
November 17th, 2017

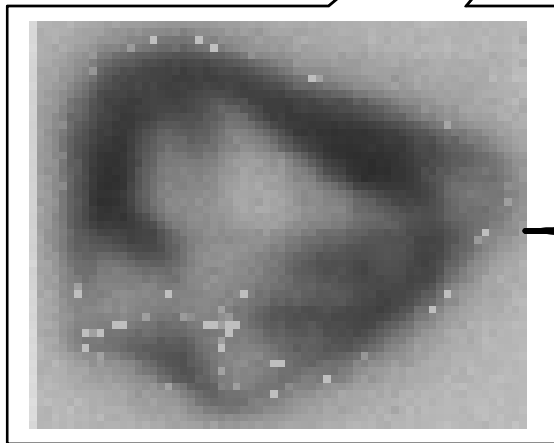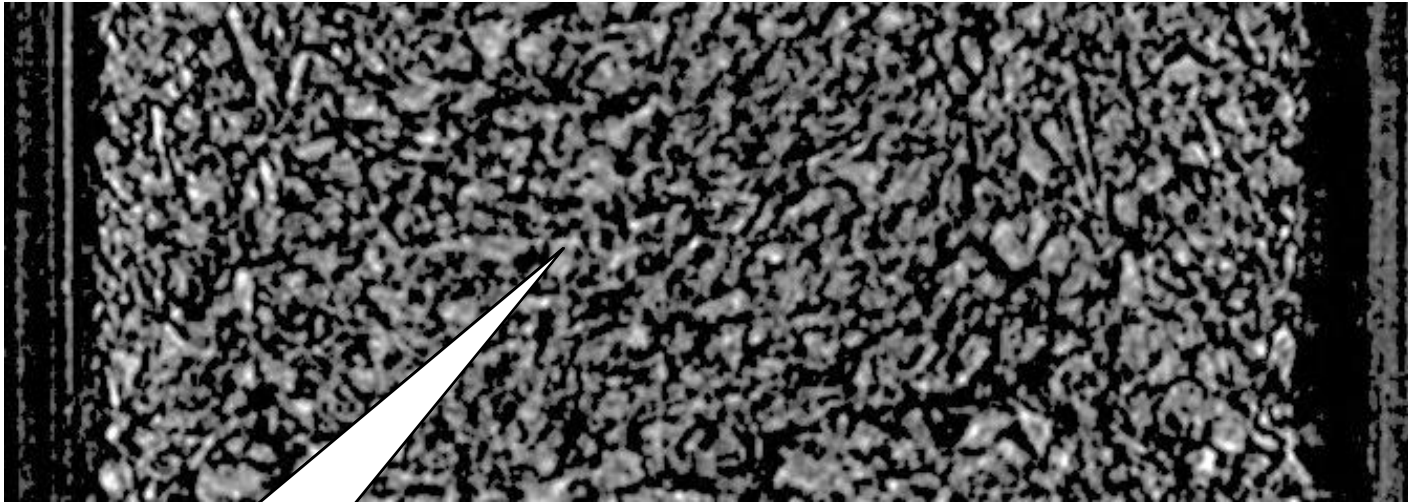# Silica Powders: A Visible Example of Morphological Dependence

Transitioning from the finer to the coarser powders:

- Less scattering per unit volume

- Appear less bright

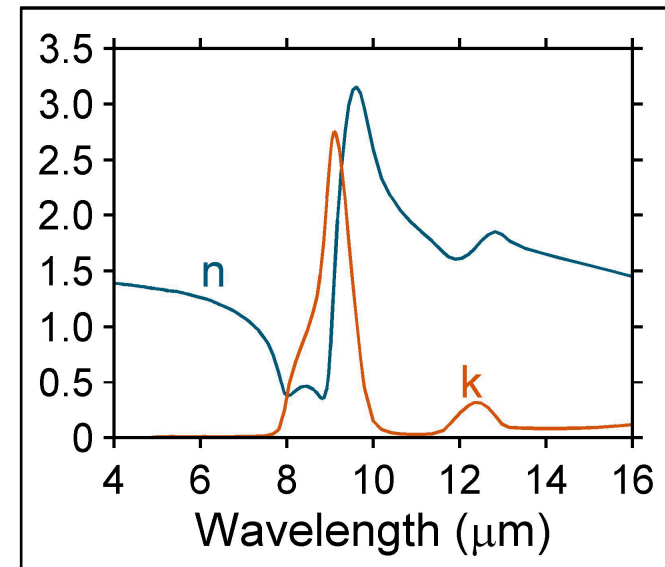# Silica Powders: An Infrared Example of the Morphological Dependence

Silica Powders: An Infrared Example of the Morphological Dependence

$$\nabla \cdot \mathbf{D}(\mathbf{r}) = \rho(\mathbf{r})$$
$$\nabla \times \mathbf{E}(\mathbf{r}) = i\omega\mu\mathbf{H}(\mathbf{r})$$
$$\nabla \cdot [\mu\mathbf{H}(\mathbf{r})] = 0$$
$$\nabla \times \mathbf{H}(\mathbf{r}) = \mathbf{J}(\mathbf{r}) - i\omega\mathbf{D}(\mathbf{r})$$
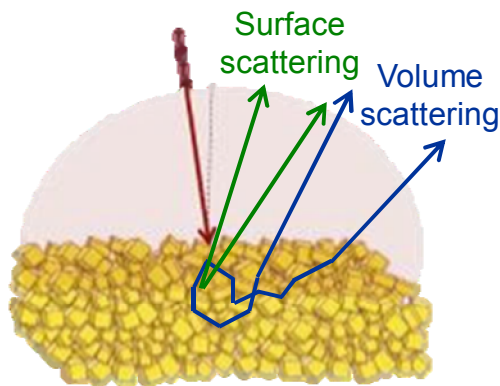$$= -i\omega\varepsilon\mathbf{E}(\mathbf{r})$$

$$n + ik = c(\varepsilon\mu)^{1/2}$$
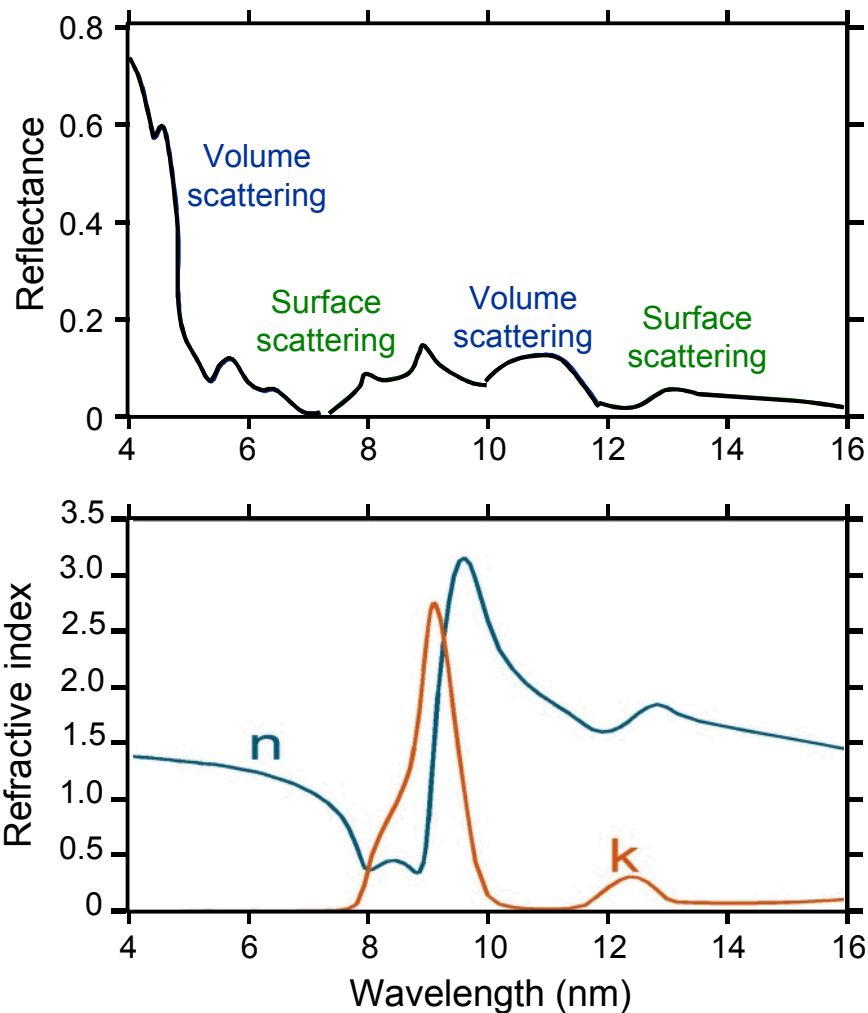
Introduce phenomenological "surface scattering" vs. "volume scattering"
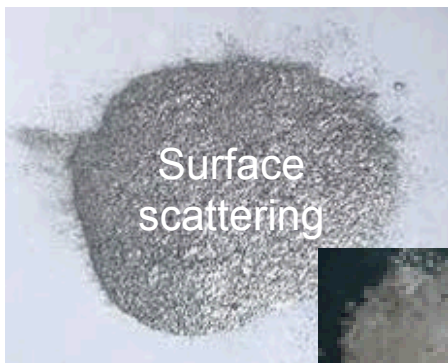


Surface scattering

Volume scattering

Surface scattering from particles: High k

Volume scattering from particles: Low k

Surface scattering

Volume scattering

Volume scattering

Surface scattering

Volume scattering

Surface scattering

Reflectance

n

k

Refractive index

Wavelength (nm)

- **Forward/inverse radiative transfer (RT) model for an optically thick deposit**
  - Optimized, the model should demonstrate agreement with reflectance spectra…
  - …while the extracted parameters should agree with independent measurements

- **Forward electromagnetic scattering model for sparse particulate deposits**

- **Our model-system measurements that have enabled both efforts**

Mesh	Size (µm)
325	44
200	74
120	125
100	149
50	297
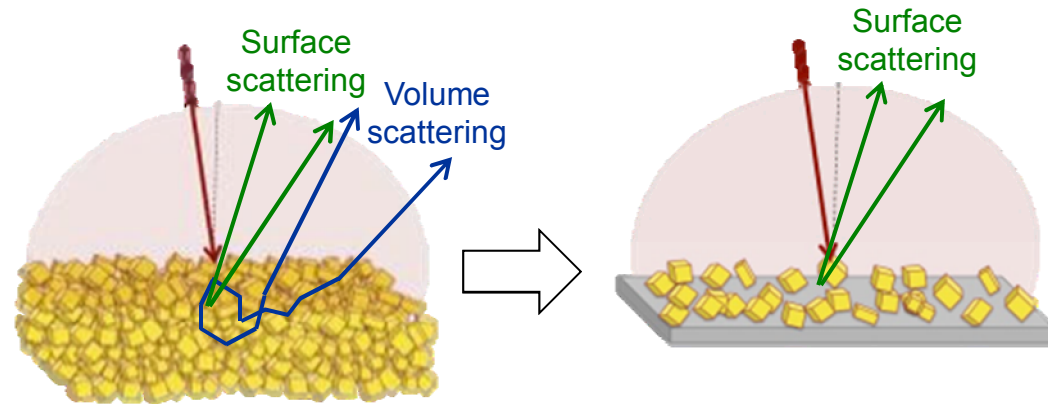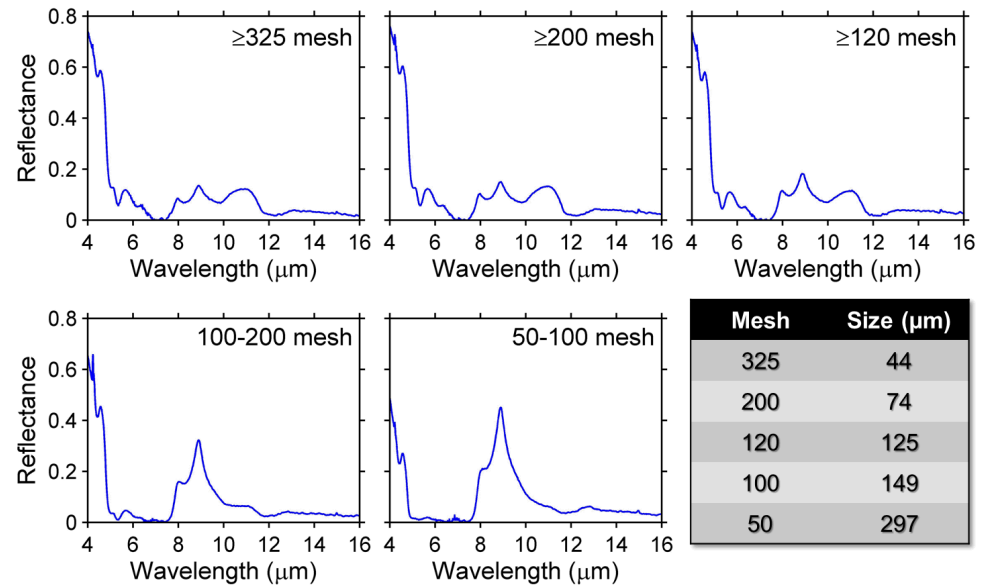
Forward/Inverse Radiative Transfer (RT) Model for an Optically Thick Deposit

Work funded under the HARD Solids Venture by the DOE Office of Defense Nuclear Nonproliferation R&D (DNN R&D)

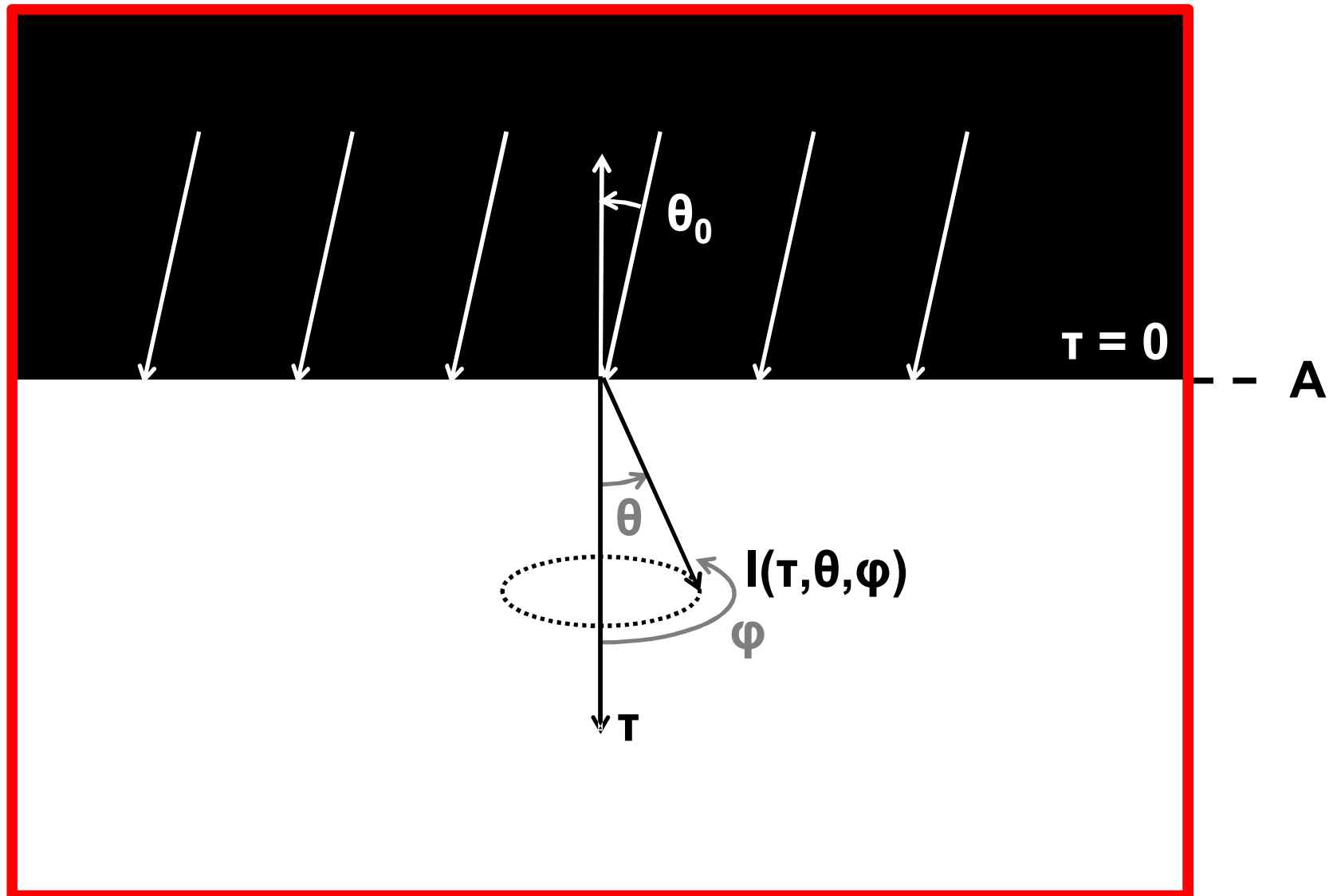# Plane Albedo (Directional, Hemispherical Reflectance) Measurement
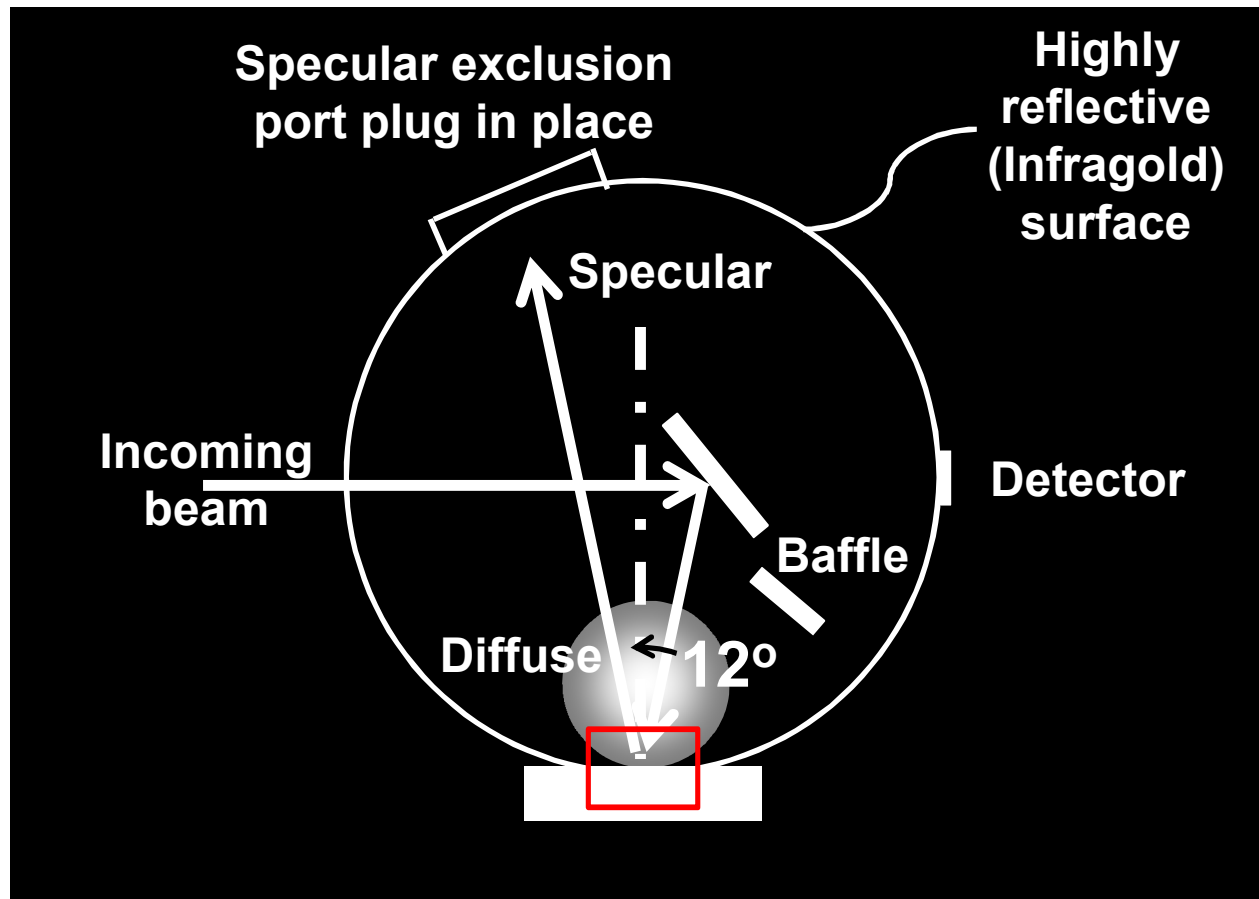
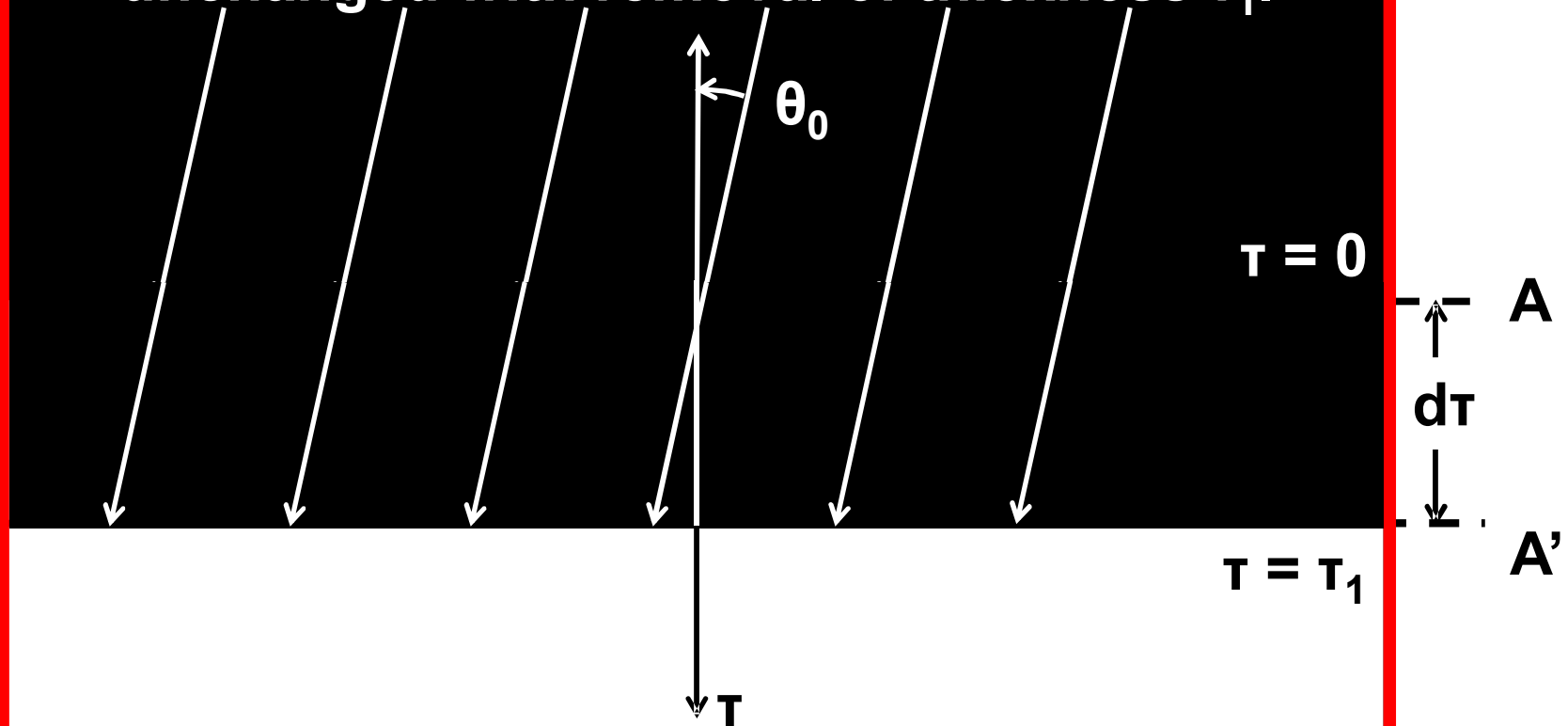Measuring plane albedo with 12° incidence angle

Invariant Imbedding Solution

τ = 0

– – A

Invariant Imbedding Solution

Invariance relationship: Reflectance is unchanged with removal of thickness τ_1.

θ_0

$\tau = 0$

A

$d\tau$

A'

$\tau = \tau_1$

τ

Solve for reflectance R as a Fourier series in azimuth

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Ambartsumian Nonlinear Integral Equation Solves for Fourier Coefficients of Reflectance R^m

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

$$(\mu + \mu_0) R^m(\mu, \mu_0) = \frac{\varpi}{4} P^m(-\mu, \mu_0)$$

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

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

$$+ \varpi \mu \mu_0 \int_0^1 \int_0^1 R^m(\mu, \mu') P^m(-\mu', \mu'')$$

$$\cdot R^m(\mu'', \mu_0) \, d\mu' \, d\mu''$$

$\left| \cos(\theta_{\text{observation}}) \right|$ $\left| \cos(\theta_{\text{incident}}) \right|$

Fourier coefficients of particle scattering function

Single scattering albedo

$$\boldsymbol{P^m, \varpi} \longrightarrow \boldsymbol{R^m}$$

Solution Provided by Michael Mishchenko (NASA GISS)

Bidirectional reflectance of flat, optically thick particulate layers: an efficient radiative transfer solution and applications to snow and soil surfaces

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[a]NASA Goddard Institute for Space Studies, 2880 Broadway, New York, NY 10025, USA
[b]Laboratory for Radiative Transfer Theory, The Main Astronomical Observatory, Ukrainian National Academy of Sciences, Golosiiv, 252650 Kyiv-22, Ukraine
[c]Science Systems and Applications, Incorporated, 2880 Broadway, New York, NY 10025, USA

Abstract

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. © 1999 Elsevier Science Ltd. All rights reserved.

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Electromagnetic Scattering by Particles and Surfaces

FORTRAN Codes for the Computation of the Bidirectional Reflection Function for Flat Particulate Layers and Rough Surfaces

By *Michael I. Mishchenko* and *Nadia T. Zakharova*

This webpage provides access to two collections of FORTRAN codes.

The first one can be used to compute the (scalar) bidirectional reflectance of a semi-infinite homogeneous slab composed of arbitrarily shaped, randomly oriented particles based on a rigorous numerical solution of the radiative transfer equation.

The second one can be used to compute the Stokes reflection matrix of a rough interface separating two homogeneous half-spaces with different refractive indices (e.g., a rough ocean surface).

Particulate Semi-Infinite Layers
The code brf.f solves the Ambartsumian's nonlinear integral equation for the reflection function using a simple iterative method. Since this technique bypasses the computation of the internal field, it is by far the fastest and most accurate numerical approach available.

The codes are ideally suitable to computing the BRF for flat snow, soil, and powder surfaces and optically thick clouds and may find applications in geophysics, physics, biophysics, and industrial research.

A detailed user manual to the codes has been published: M. I. Mishchenko, J. M. Dlugach, E. G. Yanovitskij, and N. T. Zakharova, Bidirectional reflectance of flat, optically thick particulate layers: An efficient radiative transfer solution and applications to snow and soil surfaces, *J. Quant. Spectrosc. Radiat. Transfer*, **63**, 409–432 (1999). A hardcopy reprint of this paper is available from Michael Mishchenko upon request. Please 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.
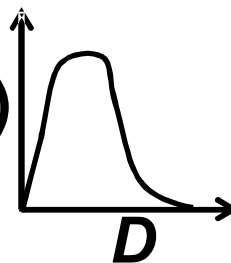
Benchmark results
The following output files were computed by the codes in their current settings and may provide a useful test of the performance of the codes on different computers:
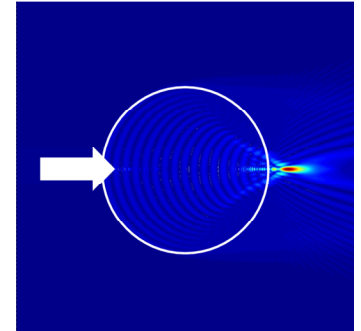
- spher.print

- spher.write

- refl.print

- interp.write

The file refl.write is not given here because of its large size.
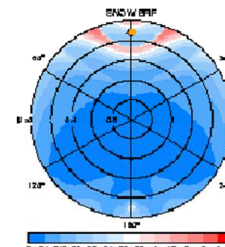
Step #1: n,k ➕ $N(D)$ ➡ $P^m, \tilde{\omega}$
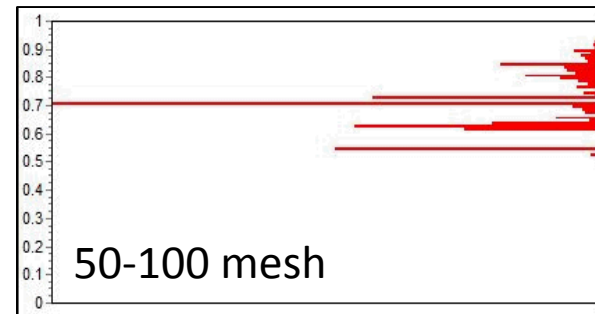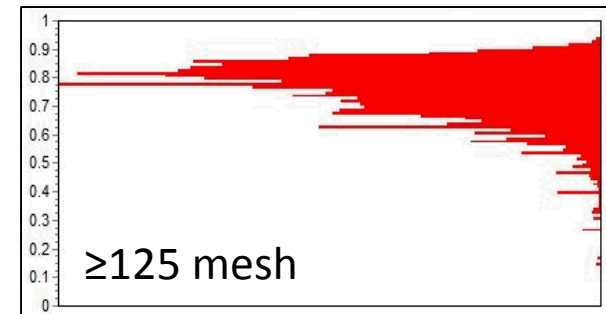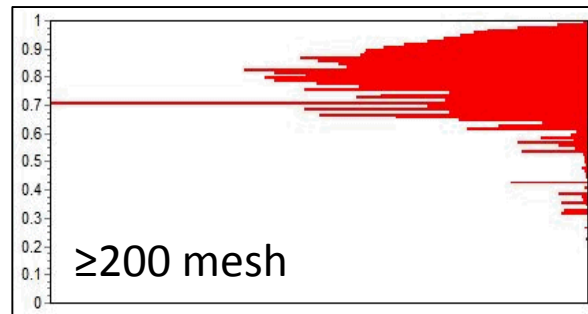
Step #2: $P^m, \tilde{\omega}$ ➡ R^m

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

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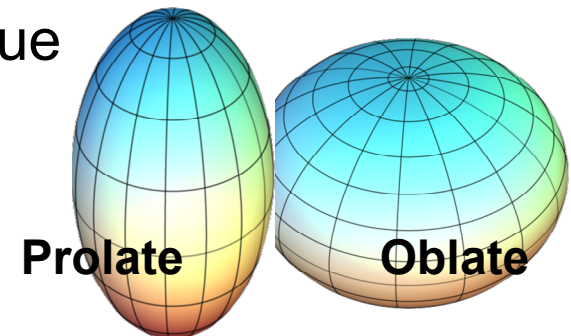
$$+ \varpi \mu \mu_0 \int_0^1 \int_0^1 R^m(\mu, \mu') P^m(-\mu', \mu'') \cdot R^m(\mu'', \mu_0) \, d\mu' \, d\mu''$$

Step #3:
$$\begin{cases} R(\mu, \mu_0, \varphi) = R^0(\mu, \mu_0) + 2 \sum_{m=1}^{m_{\max}} R^m(\mu, \mu_0) \cos(m\varphi) \\ \text{Plane albedo } (\mu_0) = 2 \int_0^1 R^0(\mu, \mu_0) \mu \, d\mu \end{cases}$$

Our Canonical Geometry: Spheroids

≥325 mesh

≥200 mesh

≥125 mesh

100-200 mesh

50-100 mesh

- This range of shapes and aspect ratios (ARs) is greatly simplified

- Shape = spheroid, AR = one characteristic value

- 2 shape bins: one bin each per prolate, oblate

Prolate   **Oblate**

Particle Size Distribution (PSD)

- Approximate with skewed *area* log-normal distribution

- Expression with 3 parameters: r_g, σ_g, γ
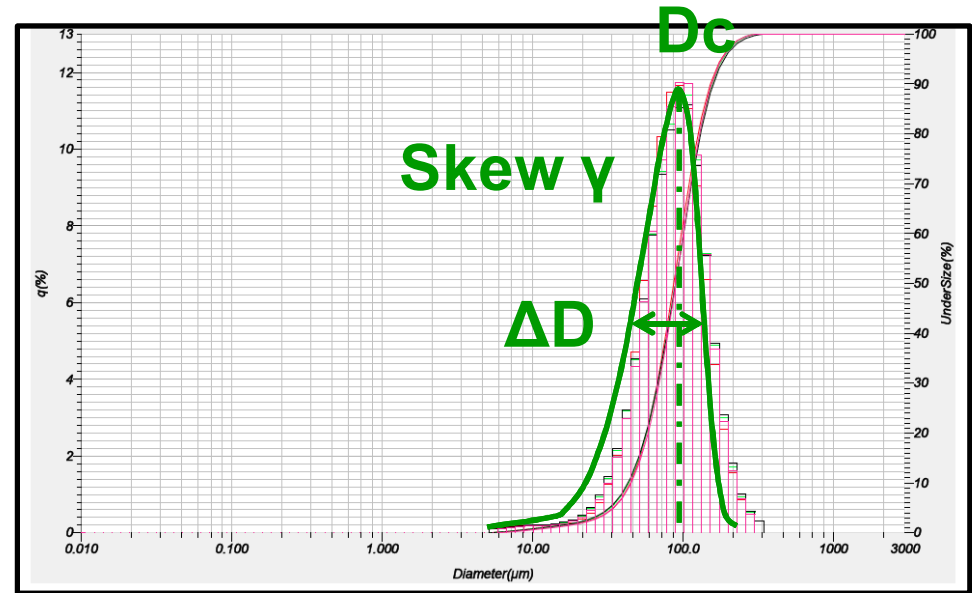
$$n(r) = \text{constant} \times r^{-3} \exp\left[-\frac{(\ln r - \ln r_g)^2}{2\ln^2 \sigma_g}\right] \times \left[1 + \text{erf}\left(\frac{\alpha(\ln r - \ln r_g)^2}{2\sqrt{2}\ln^2 \sigma_g}\right)\right]$$
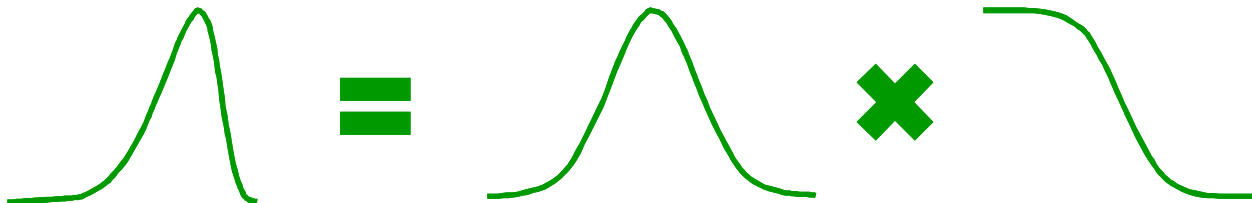
"Patching" the Scattering Properties

- 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

$$p(\theta) = \frac{4\pi}{C_{sca}} \frac{dC_{sca}}{d\Omega} S(\theta)$$
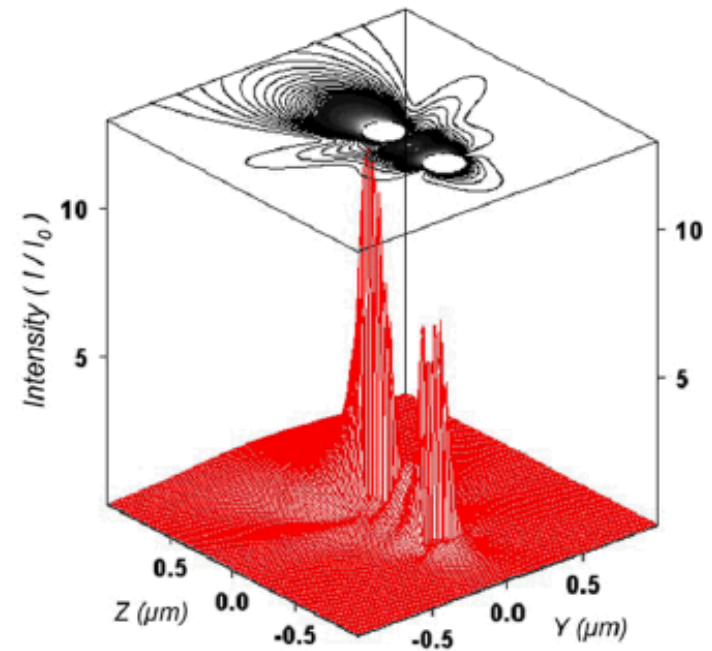
J.-C. Auger and B. Stout, "Local field intensity in aggregates illuminated by diffuse light: T matrix approach," Appl. Opt. **47**, 2897-2905 (2008).

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"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
  - $S(\theta) = F(f_{SSF}, u)$

  - $u = \dfrac{8\pi r_0}{\lambda} \sin\left(\dfrac{\theta}{2}\right)$

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

    Scattering angle

    Size parameter

$$p(\theta) = \frac{4\pi}{C_{\text{sca}}} \frac{dC_{\text{sca}}}{d\Omega} S(\theta)$$
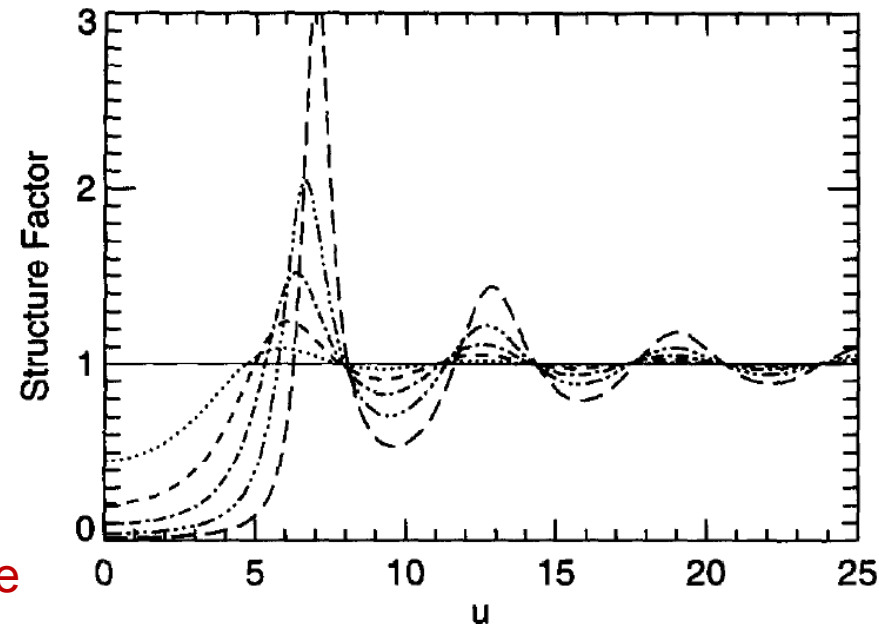
M. I. Mishchenko, "Asymmetry parameters of the phase function for densely packed scattering grains," JQSRT **52**, 95-110 (1994).

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1st-Surface Reflectance and Surface Roughness

$$F_{\text{Fresnel}} \times R_{\text{Fresnel}}$$

Fractional area $F_{Fresnel}$
of refractive index n+ik

$$\text{medium} \qquad \times R_{\text{Fresnel}}$$

Medium of particles of
refractive index n+ik

With surface roughness
characterized by θ_{eff}
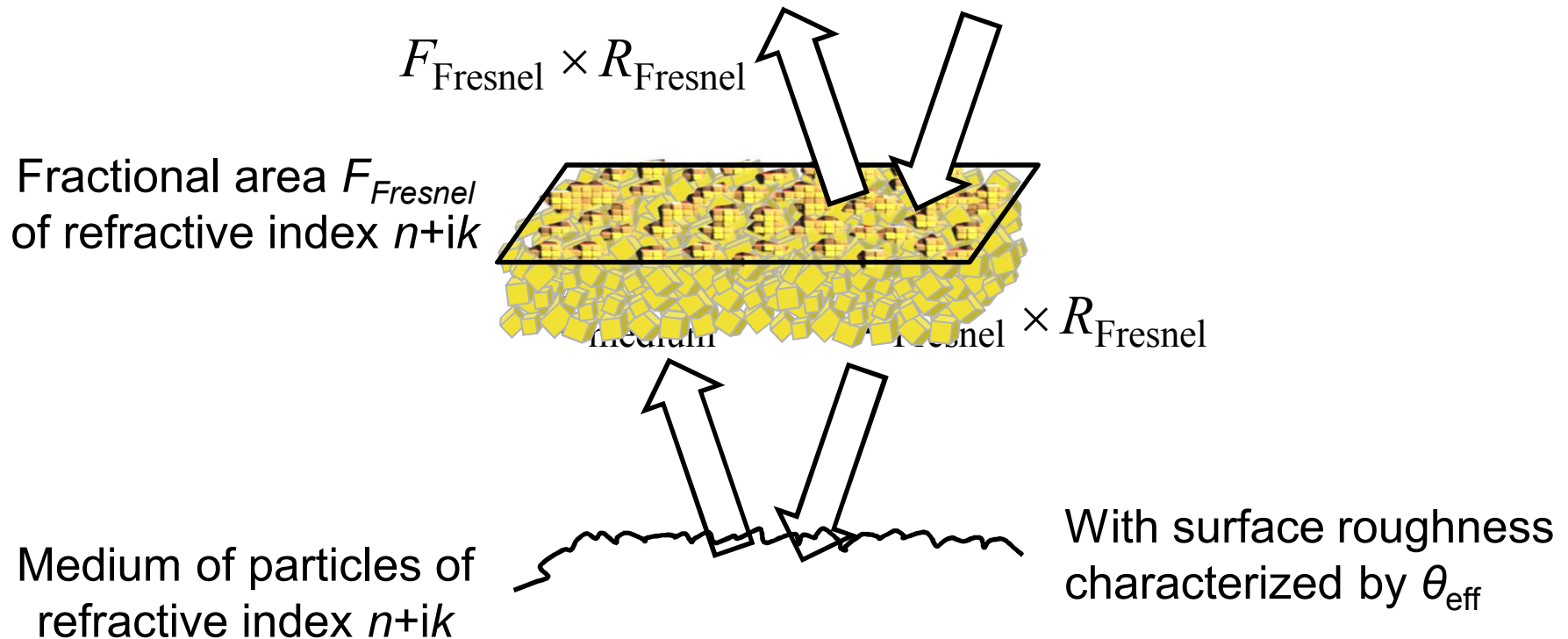
$$R = F_{\text{Fresnel}} \times R_{\text{Fresnel}} + \left(1 - F_{\text{Fresnel}} \times R_{\text{Fresnel}}\right) \times R_{\text{medium}}$$

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Full Characterization of Material Systems

Unpolarized hemispheric total reflection measurements

Polarized directional measure-ments

Optical scattering from particle ensembles

Microscopy for single-particle imaging

Spectral data

≥325 mesh

≥200 mesh

≥120 mesh

100-200 mesh

50-100 mesh
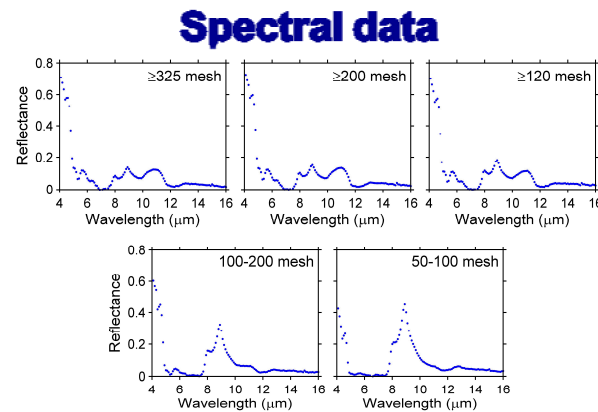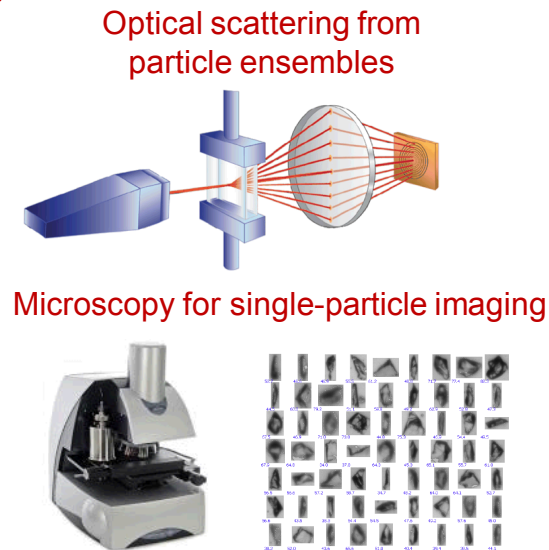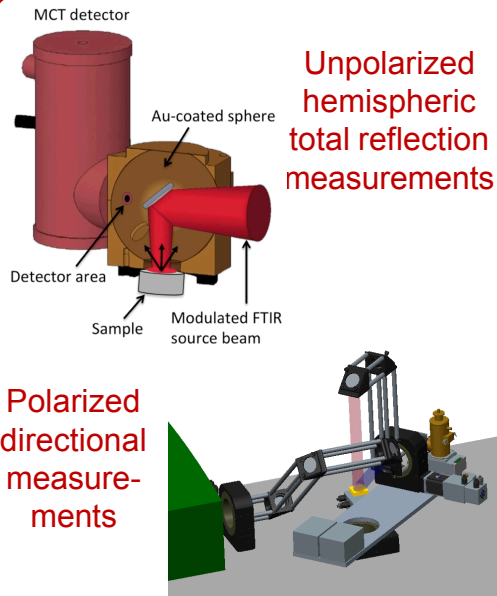
Individual particle morphology

Particle size distribution

Optical diffraction

Image analysis

Mass

Particle aspect ratio distribution

AR = W/L

Particle shape distributions
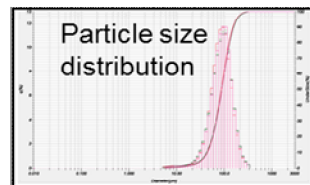
Aggregate morphology

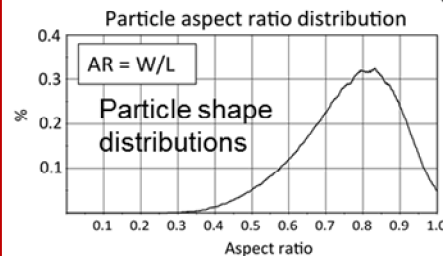3D Material distribution

XRT

Model system

Packing density

Frequency (wavenumbers)

Pluviated and Packed Powder
1.5 micron Alumina
Diffuse Reflectance

High density (1.54 g/cc)
Low density (0.50 g/cc)
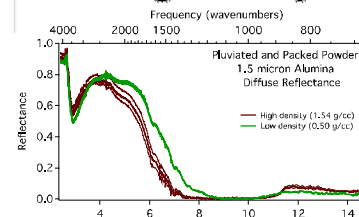
X-ray tomography for full structure of particulate solid
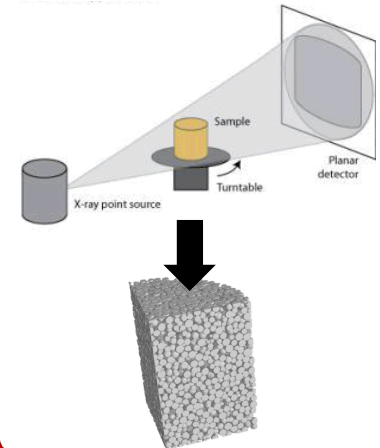
Model Assessment

Particle aspect ratio

Particle size distribution: Mean, width, and skew

Packing density

Surface roughness

1st-surface reflection

~7 Parameters

Complex refractive index

DAKOTA

$$\nabla \cdot \mathbf{D}(\mathbf{r}) = \rho(\mathbf{r})$$

$$\nabla \times \mathbf{E}(\mathbf{r}) = i\omega\mu\,\mathbf{H}(\mathbf{r})$$

$$\nabla \cdot [\mu\,\mathbf{H}(\mathbf{r})] = 0$$
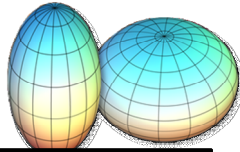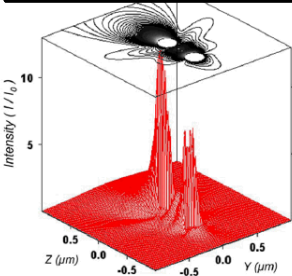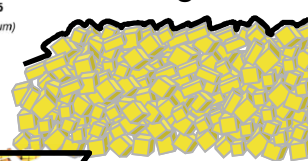
$$\nabla \times \mathbf{H}(\mathbf{r}) = \mathbf{J}(\mathbf{r}) - i\omega\mathbf{D}(\mathbf{r}) = -i\omega\varepsilon\,\mathbf{E}(\mathbf{r})$$

$$(\mu + \mu_0)R^m(\mu, \mu_0)$$

$$= \frac{\varpi}{4}P^m(-\mu, \mu_0)$$

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

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

$$+ \varpi\mu\mu_0 \int_0^1 \int_0^1 R^m(\mu, \mu')\,P^m(-\mu', \mu'')$$

$$\cdot R^m(\mu'', \mu_0)\,d\mu'\,d\mu''$$

Physics-Based Model
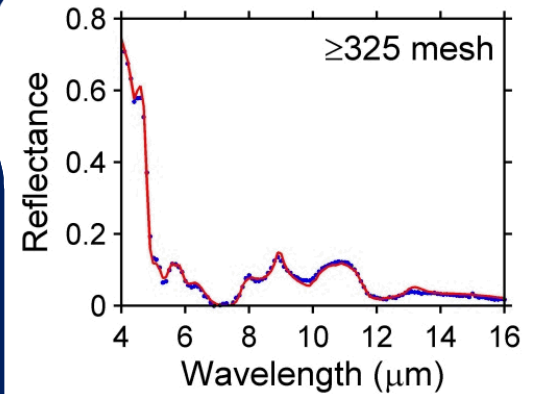
Compare to measured reflectance spectrum

≥325 mesh
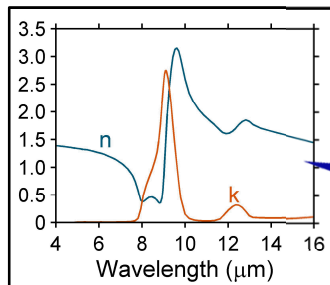
Measured and Modeled Reflectance Spectra of Silica Powders

Reflectance plots:
(a) ≤45 μm
(b) ≤75 μm
(c) ≤125 μm
(d) 75-150 μm
(e) 150-300 μm
Wavelength (μm)
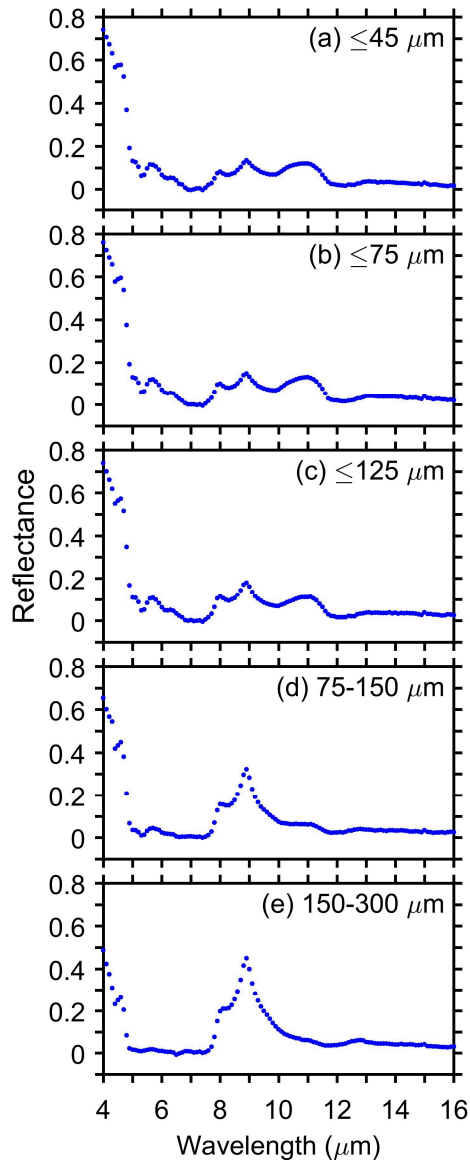
Nice agreement, but…

"Give me seven free parameters and I can fit an elephant"
– K. Lumme and A. Penttilä, JQSRT **112**, 1658-1670 (2011).

Are the fits extracting physical parameters?

Or are we just shape-fitting?

Particle size distribution: Mean, width, and skew
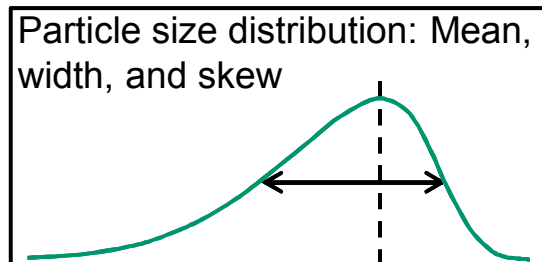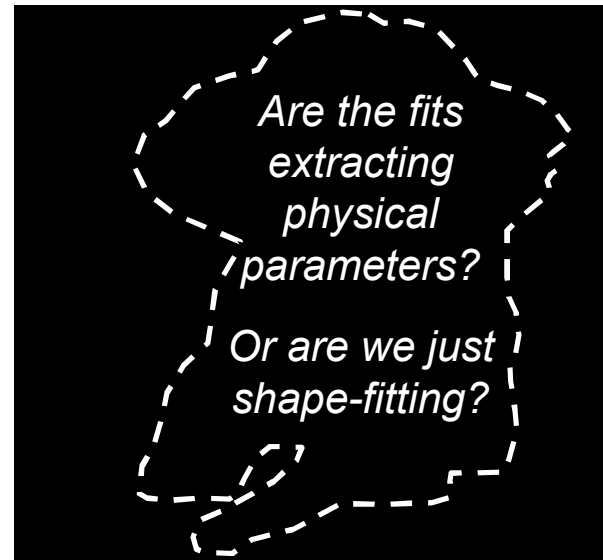
vs
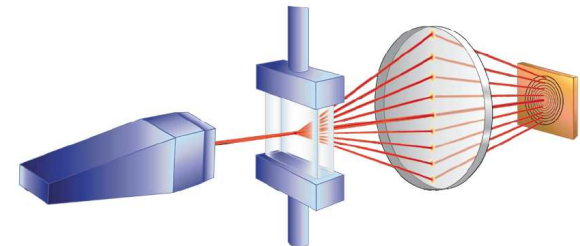
Measured and Modeled Reflectance Spectra and PSDs of Silica Powders
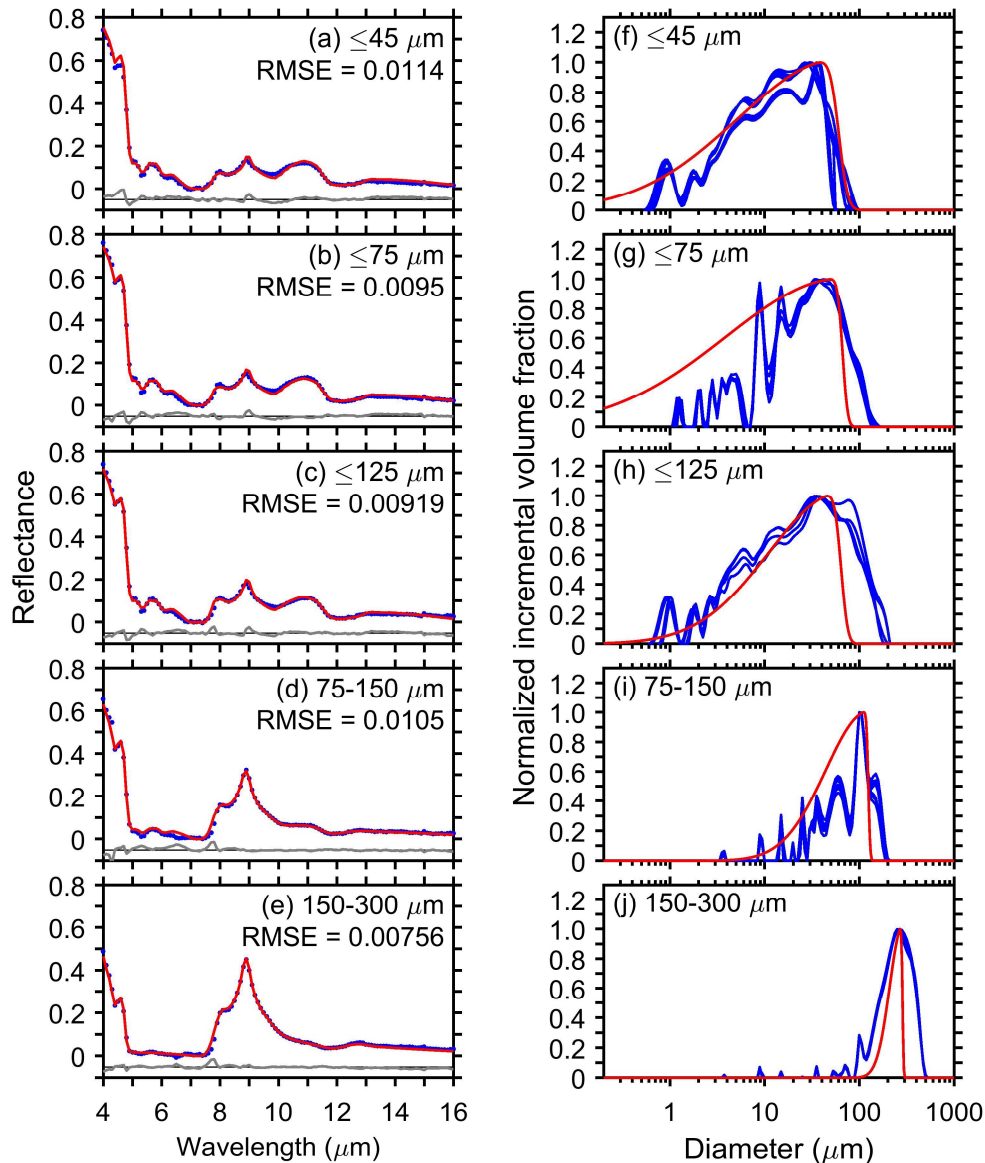
- PSDs derived via optimizing reflectance model to measured spectra demonstrate general agreement with PSDs measured via laser diffraction

- Exact level of agreement confounded by broad distributions provided by manufacturer

- Repeat process on samples sieved in-house

Measured and Modeled Reflectance Spectra and PSDs of Silica Powders

(a) 74-88 μm
RMSE = 0.0116

(b) 105-125 μm
RMSE = 0.00866

(c) 149-177 μm
RMSE = 0.0081

(d) 210-250 μm
RMSE = 0.00947

(e) 297-360 μm
RMSE = 0.00832

Reflectance

Wavelength (μm)

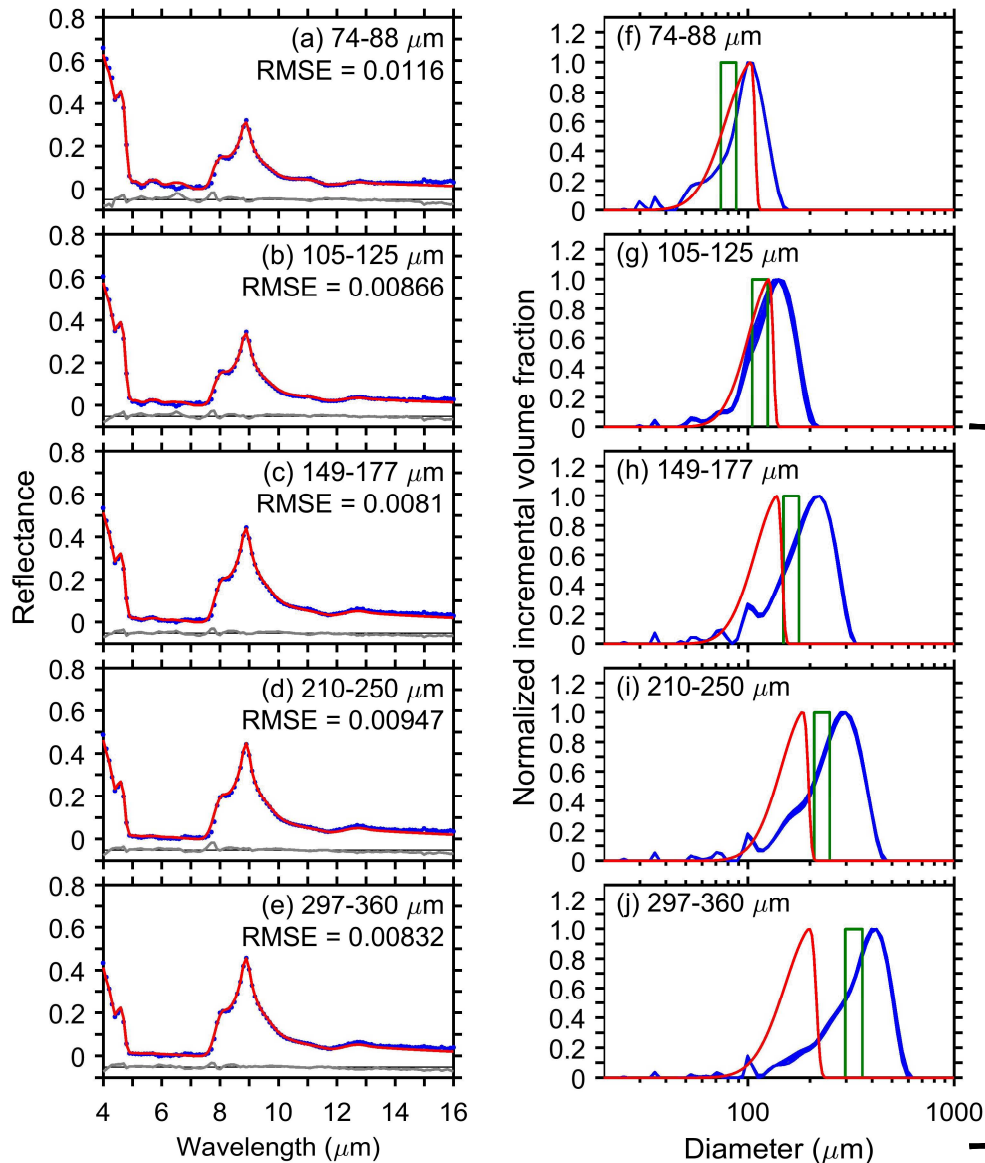(f) 74-88 μm

(g) 105-125 μm

(h) 149-177 μm

(i) 210-250 μm

(j) 297-360 μm

Normalized incremental volume fraction

Diameter (μm)

- Departure from measured PSDs is evident for larger particles
  - Model-derived PSDs are skewed to smaller particle sizes

- Potential cause: The reflectance of larger particles is spectrally similar to the $R_{Fresnel}$ of the bulk material
  - Optimization is likely attributing reflectance contribution of larger particles to 1st-surface reflectance term

VS
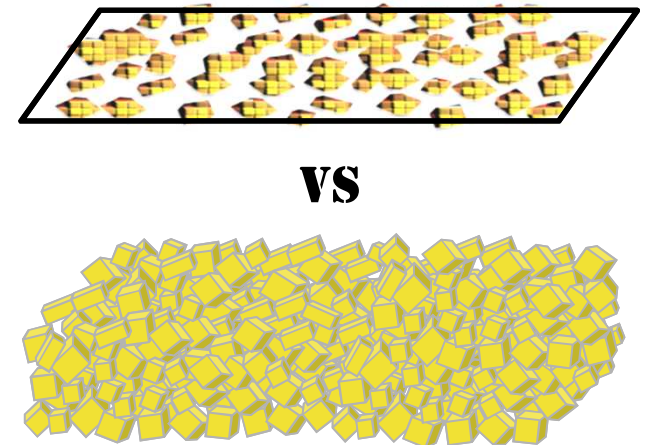
G. Ito, J. A. Arnold, and T. D. Glotch, "T-matrix and radiative transfer hybrid models for densely packed particulates at mid-infrared wavelengths," J. Geophy. Res. Planets **122**, 822-838 (2017).
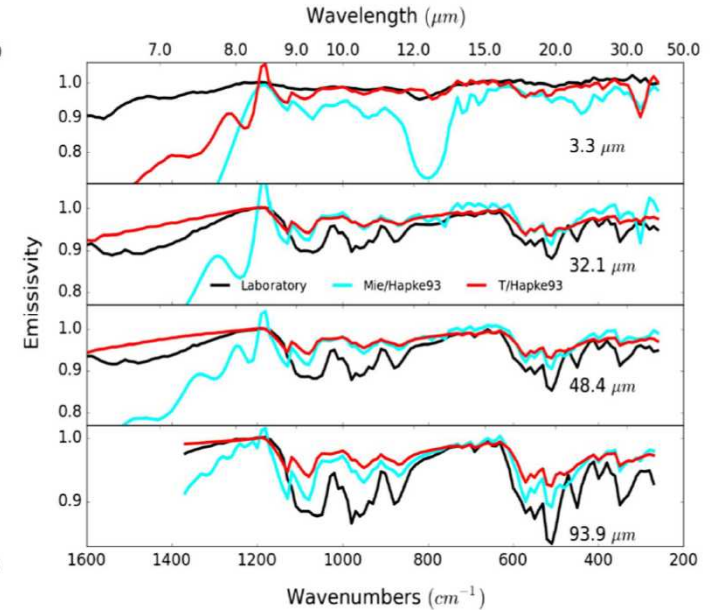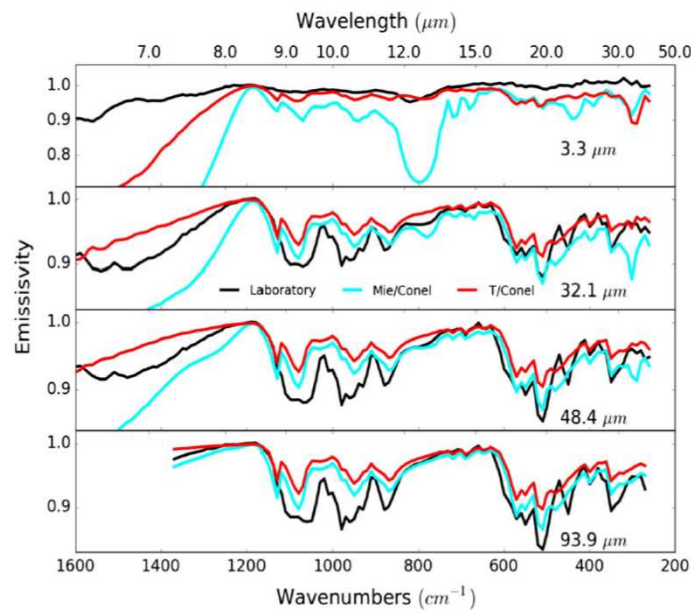
B. Ramezan pour and D. W. Mackowski, "Radiative transfer equation and direct simulation prediction of reflection and absorption by particle deposits," JQSRT **189**, 361-368 (2017).
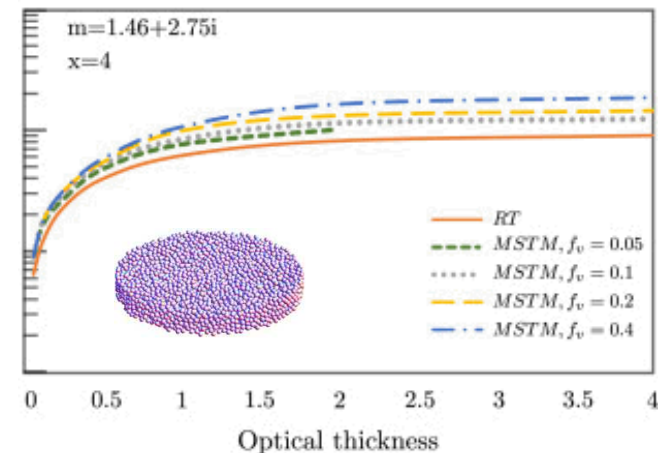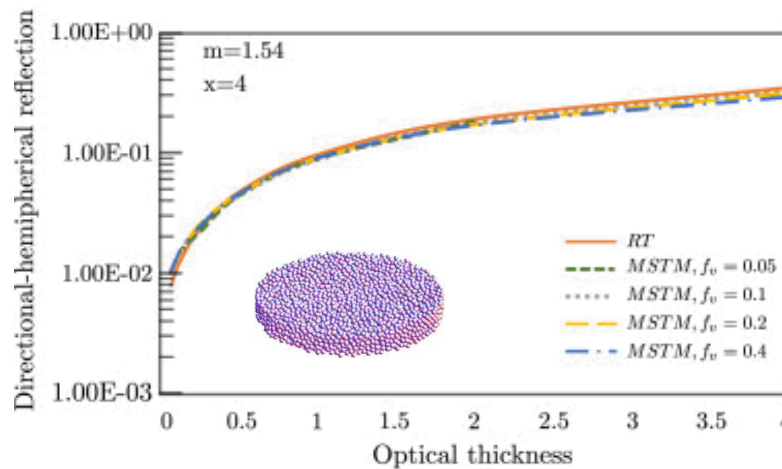
Forward electromagnetic scattering model for sparse particulate deposits

Work funded by the IARPA's SILMARILS Program

Recall phenomenological "surface scattering" vs. "volume scattering"
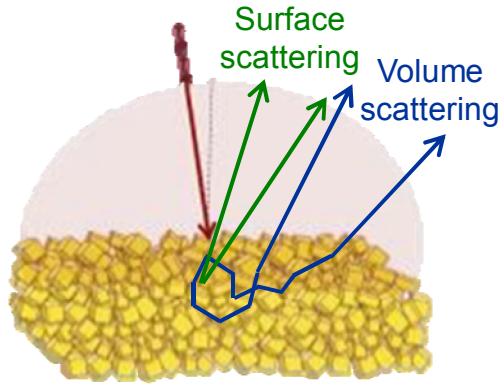
Surface scattering from particles: High k

Volume scattering from particles: Low k
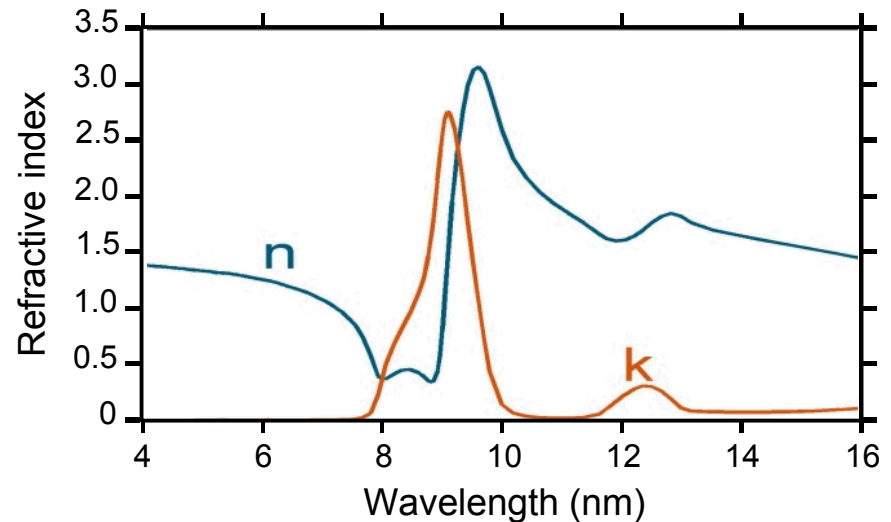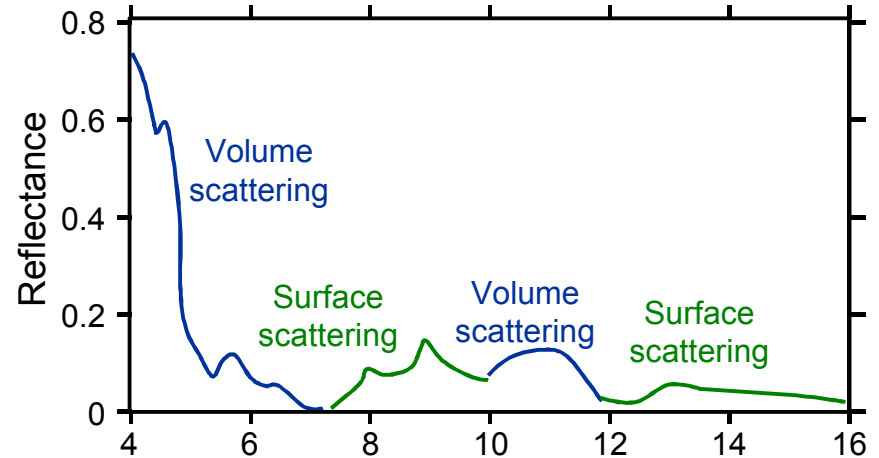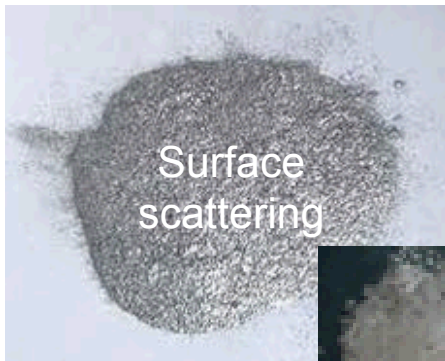
Now consider *low particulate loadings* (vs. optically thick deposits)
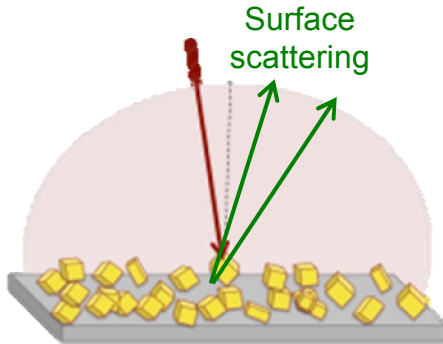
Surface scattering from particles: High k

Volume scattering from particles: Low k

1) *Volume*-scattering features will effectively disappear.

2) *Surface* scattering remains, but (for small particles) the substrate will be its primary source.

3) For small particles, the spectrum will appear similar to the substrate reflectance, but attenuated by particle absorption (defined by *k*).
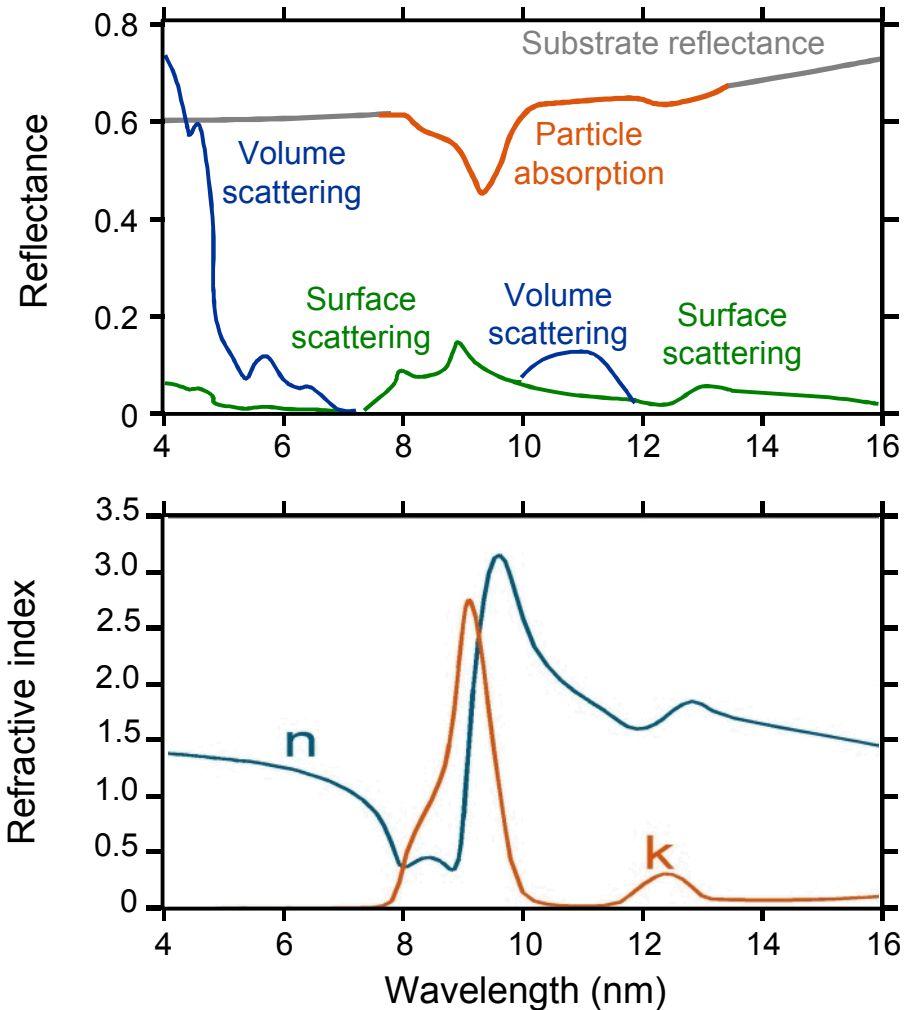
EM Scattering by Surface-Bound Particle

Surface-bound sphere

$D = \lambda$, $n = n_2$

Angular distribution of reflected light impacted by both particle and surface

$n_1 = 1$

$n_2 = 3$

Multiple particle/ surface interactions impact the reflectance signature

Figures from W. Sun et al., "Finite-difference time-domain solution of light scattering by arbitrarily shaped particle and surfaces," Chapter 3 of *Light Scattering Reviews 6: Light Scattering and Remote Sensing of Atmosphere and Surface*, A. A. Kokhanovsky (ed.), Springer-Verlag, Berlin (2012).
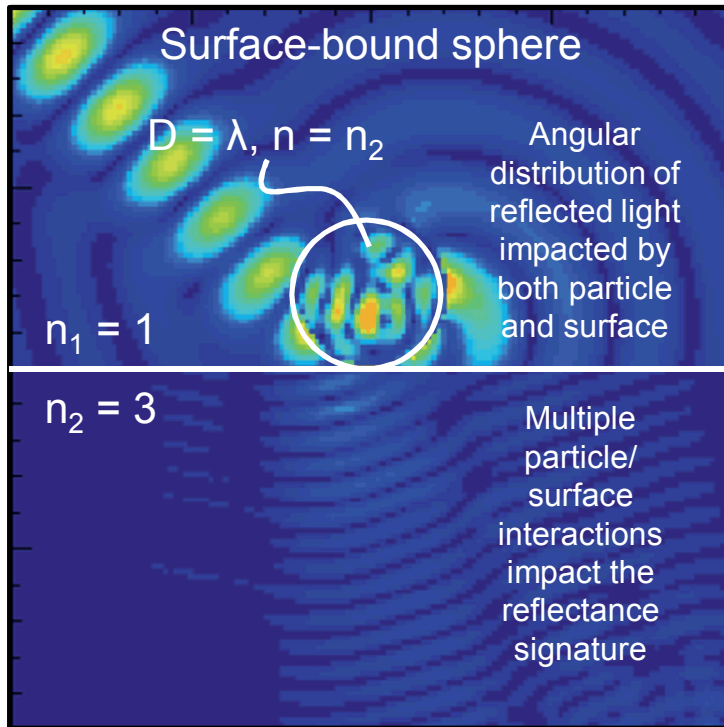
T-matrix solution:

1) Calculate T matrix of particle

2) Calculate reflection matrix **A** of surface

3) Solve for f_{mn} and g_{mn} in

$$\left(I - \left[T_{mn,mn_1}\right]\left[A_{mn_1 n}\right]\right)\begin{bmatrix} f_{mn} \\ g_{mn} \end{bmatrix} = \left[T_{mn,mn_1}\right]\begin{bmatrix} a_{mn_1} \\ b_{mn_1} \end{bmatrix}$$

where a_{mn_1} and b_{mn_1} are expansion coefficients of incident/reflecting wave

4) Use f_{mn} and g_{mn} to compute scattering and extinction for surface-bound particle
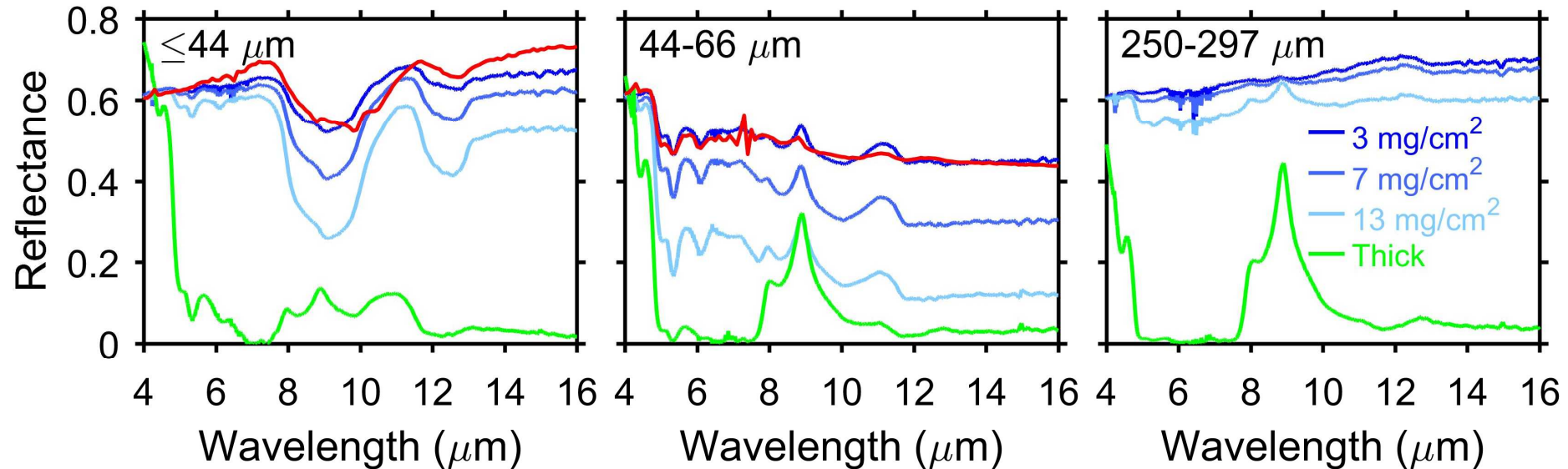
- 3 different particle sizes (≤44 µm, 44-66 µm, and 250-297 µm)
- 3 deposits each (with optically thick spectrum for reference)
- Transition between thin and thick spectra depends on particle size

→ *Detection algorithms assuming linear scaling of signature with concentration will be challenged by the physics of light scattering and extinction.*

Related Recent Work

Q. Lin, Y. Xuan, and Y. Han, "Prediction of the radiative properties of surfaces covered with particulate deposits," JQSRT **195**, 112-122 (2017).
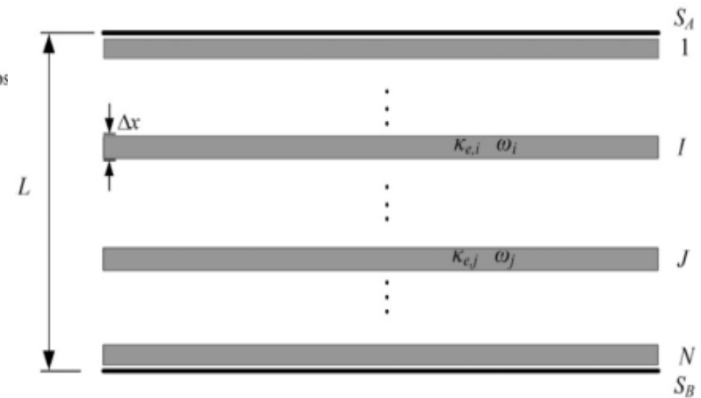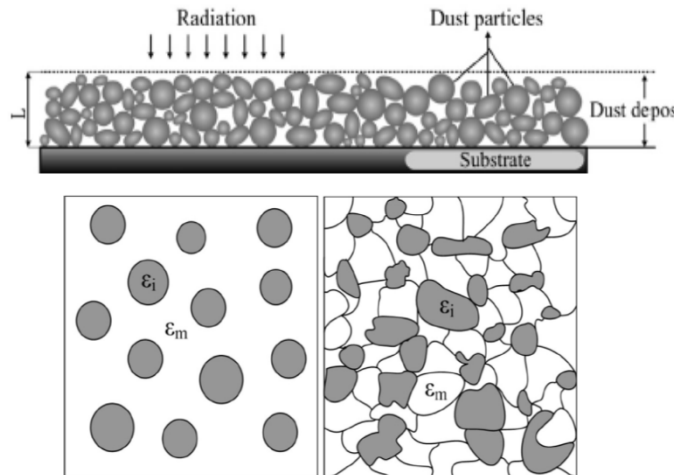
A. Egel, Y. Eremin, T. Wriedt, D. Theobald, U. Lemmer, and G. Gomard, "Extending the applicability of the T-matrix method to light scattering by flat particles on a substrate via truncation of sommerfield integrals," JQSRT **202**, 279-285 (2017).
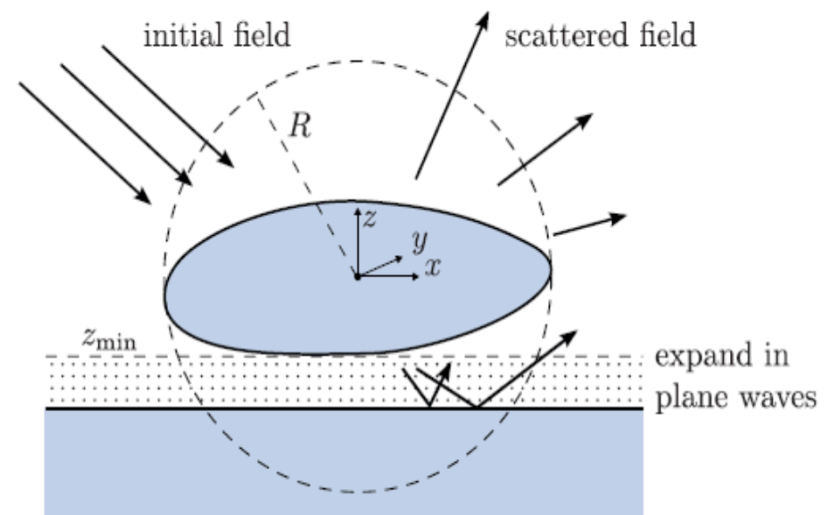
F. Rivera-Hernandez, J. L. Bandfield, S. W. Ruff, and M. J. Wolff, "Characterizing the thermal infrared spectral effects of optically thin surface dust: Implications for remote-sensing and in situ measurements of the martian surface," Icarus **262**, 173-186 (2015).
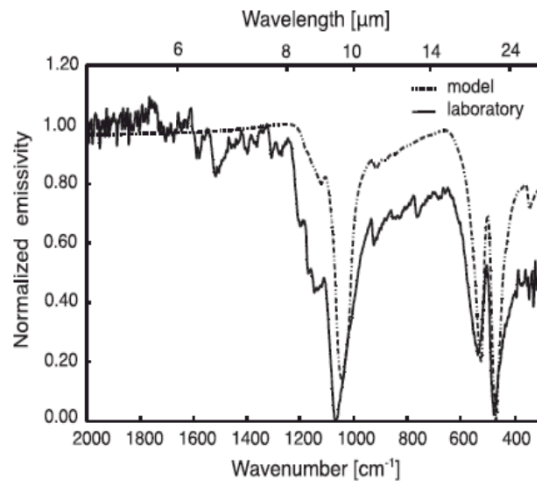
Summary and Next Steps

- Demonstrated numerically invertible physics-based model for the reflectance spectrum of an *optically thick particulate medium*
  - Requires as input both fundamental optical properties (n,k) and morphological parameters (PSD, packing density, surface roughness)
  - Optimized, the model spectra demonstrate agreement with measured spectra
  - Model-extracted PSDs agree with laser-diffraction measurements for smaller particles
  - Next steps: Consideration of packing density and surface roughness

- Demonstrated impact of particle size and deposited mass upon reflectance spectrum of a *sparse particulate deposit*
  - Next steps: Numerical inversion & consideration of directional effects

Acknowledgments

- Michael Mishchenko (NASA GISS) – Modeling advice and assistance

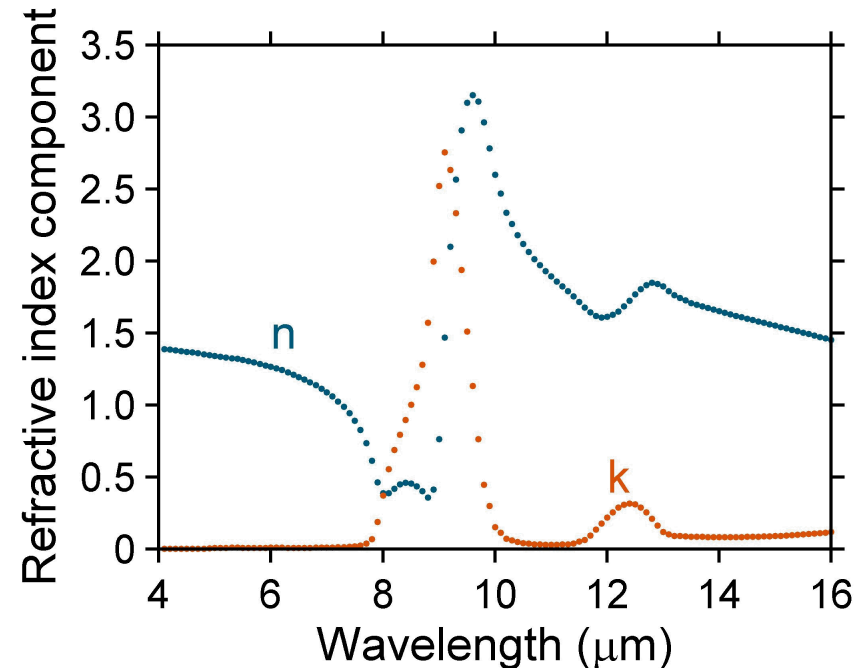- Patty Hough (Sandia National Laboratories, CA) – Model inversion via Dakota, parallel computing

Thanks! Questions?

Extra Viewgraphs

Optimization via "nl2sol" in DAKOTA

- 121 λs: 4-16 μm @ 0.1 μm resolution

- The 121 λ-dependent calculations are divided among 64 processors (4 nodes, 16 cores/node)

- ~10 min/spectrum, 100+ such calculations required for convergence

- 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
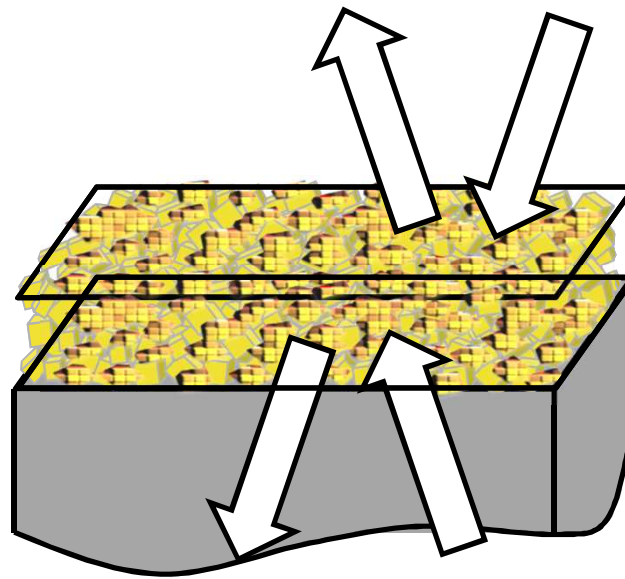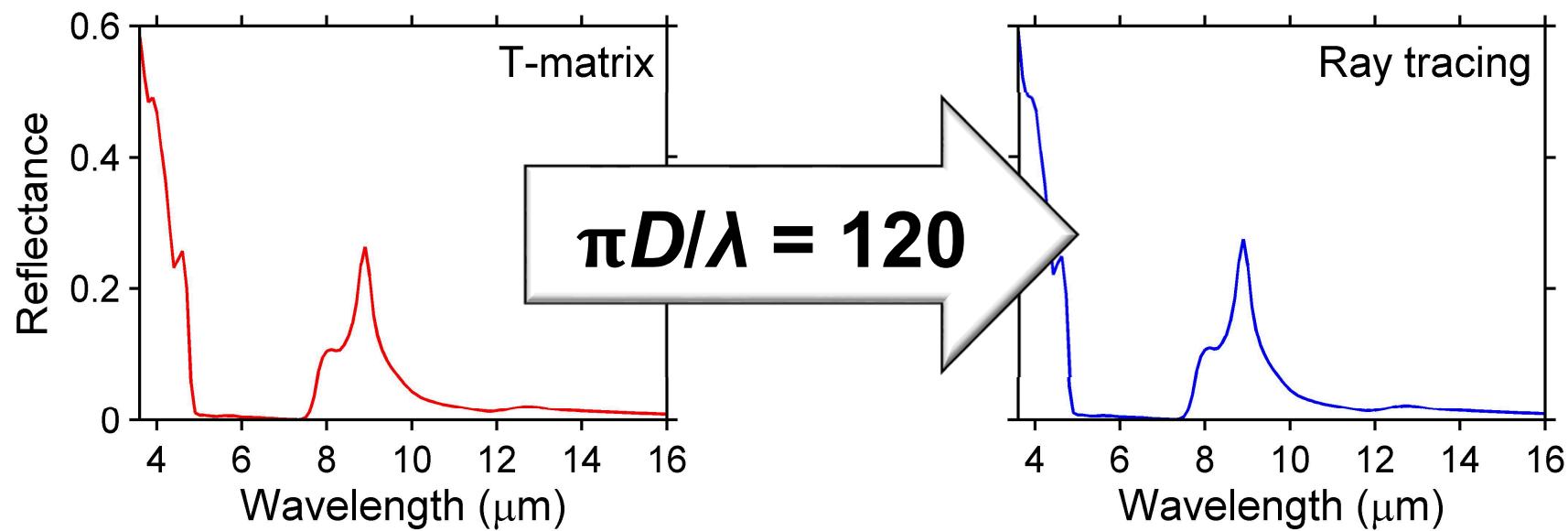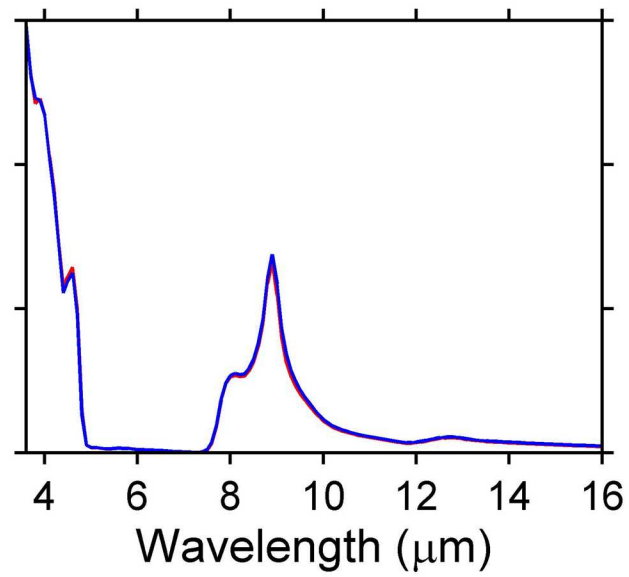
Should $R_{substrate}$ be modified to account for surface reflection from deposit?

$R_{substrate}$

T-matrix vs. Ray tracing

$\pi D/\lambda = 120$

Similar Approximations Made by Others

- J. L. Bandfield, P. O. Hayne, J.-P. Williams, B. T. Greenhagen, and D. A. Paige, "Lunar surface roughness derived from LRO Diviner Radiometer observations," Icarus **248**, 357-372 (2015).

- P. Helfenstein and M. K. Shepard, "Submillimeter-scale topography of the lunar regolith," Icarus **141**, 107-131 (1999).

- M. K. Shepard, R. A. Brackett, and R. E. Arvidson, "Self-affine (fractal) topography: Surface parameterization and radar scattering," J. Geophys. Res. **100**, 11709-11718 (1995).

- B. Hapke, "Bidirectional reflectance spectroscopy. 3. Correction for Macroscopic Roughness," Icarus **59**, 41-59 (1984).

But is this approximation sufficient?