Light scattering patterns from single urban aerosol particles at Adelphi, Maryland; a classification relating to particle morphologies

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Techniques successfully used for analysis of atmospheric aerosol composition

- **Infrared absorption spectroscopic analysis:** Hoidale, G.B., and Blanco, A. (1969) Pure Appl. Geophys., vol 74, pp 151-164.
- **Temperature fractionation of aerosol particles:** Rosen, J.M. (1971) *J. Appl. Meteor., 10*, 1044-1046; Pinnick, R. G. et al (1987) *J. Atmos. Sci*., *44*(3), 562-576.
- **Temperature and humidity controlled nephelometry:** Larson, T. V. et al (1982) *Atmos. Environ., 16*, 1587-1590.
- **Pyrolysis and oxidation of carbon:** Rosen, H., et al, Appl. Opt.(1978) vol 17, pp 3859-3861; Dasch, J.M., and S.H. Cadle (1989) *Aerosol Sci. & Technol*., *10*, 236-248.
- **X-ray dispersive analyses:** Sheridan, P. J. et al (1993), *Atmos. Environ*., *27A*, 1169-1183.
- **Gas chromatography/mass spectrometry:** Rogge, W. F. et al (1993) *Atmospheric Environment, 27A*, 1309-1330.
- **Ion-exchange chromatography:** Decesari, S. et al (2002) *Atmospheric Environment*, *36*, 1827-1832.
- **Pyrolysis gas chromatography/mass spectrometry:** Gelencser, A. et al (2002) *Journal of Geophysical Research*, *107*(ACH2), 1-5.
- **X-ray fluorescence:** Chen, L. et al (2002) *Atmospheric Environment, 36*, 4541-4554.
- **Thermal-optical techniques:** Lim, H.-J., and B. J. Turpin (2002) *Environ. Sci. Technol*., *2002*(36), 4489-4496.
- **Laser-ablation mass spectrometry:** Prather, K.A. et al (1994) *Analytical Chem*., *66*, 1403-1407; Murphy, D. M. et al (2003) *Aerosol Science and Technology*, *37*, 382-391.
- **Laser-induced fluorescence:** Pinnick, R. G. et al (1995) *Aerosol. Sci. Tech*.23: 653-664; Chang, R.K. et al, US patent 6,947,134 B2, Sep. 20, 2005.
- **Laser-induced breakdown spectroscopy:** Hahn, D. W. (1998) *Applied Physics Letters, 72,* 2960-2962.
- **Size exclusion chromatography/capillary electrophoresis:** Krivacsy, Z.et al (2000) *Atmospheric Environment*, *34*, 4273-4281.
- **Epifluorescence microscopy/staining:** Bauer, H.et al (2002) *Atmospheric Research*, *64*, 109-119.

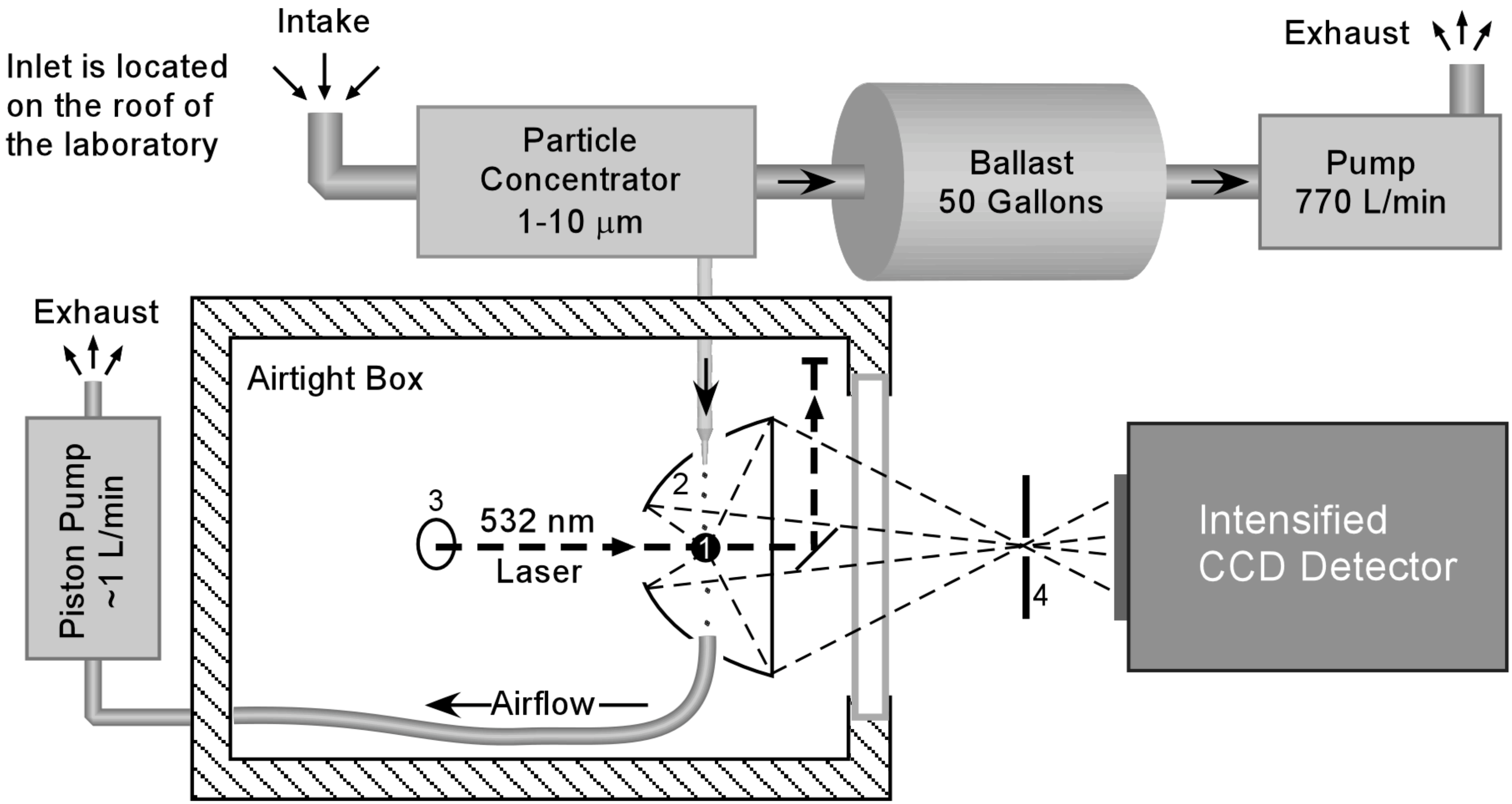
Definitive characterization of atmospheric aerosol morphologies (shape, internal structure and refractive index distribution) remains elusive

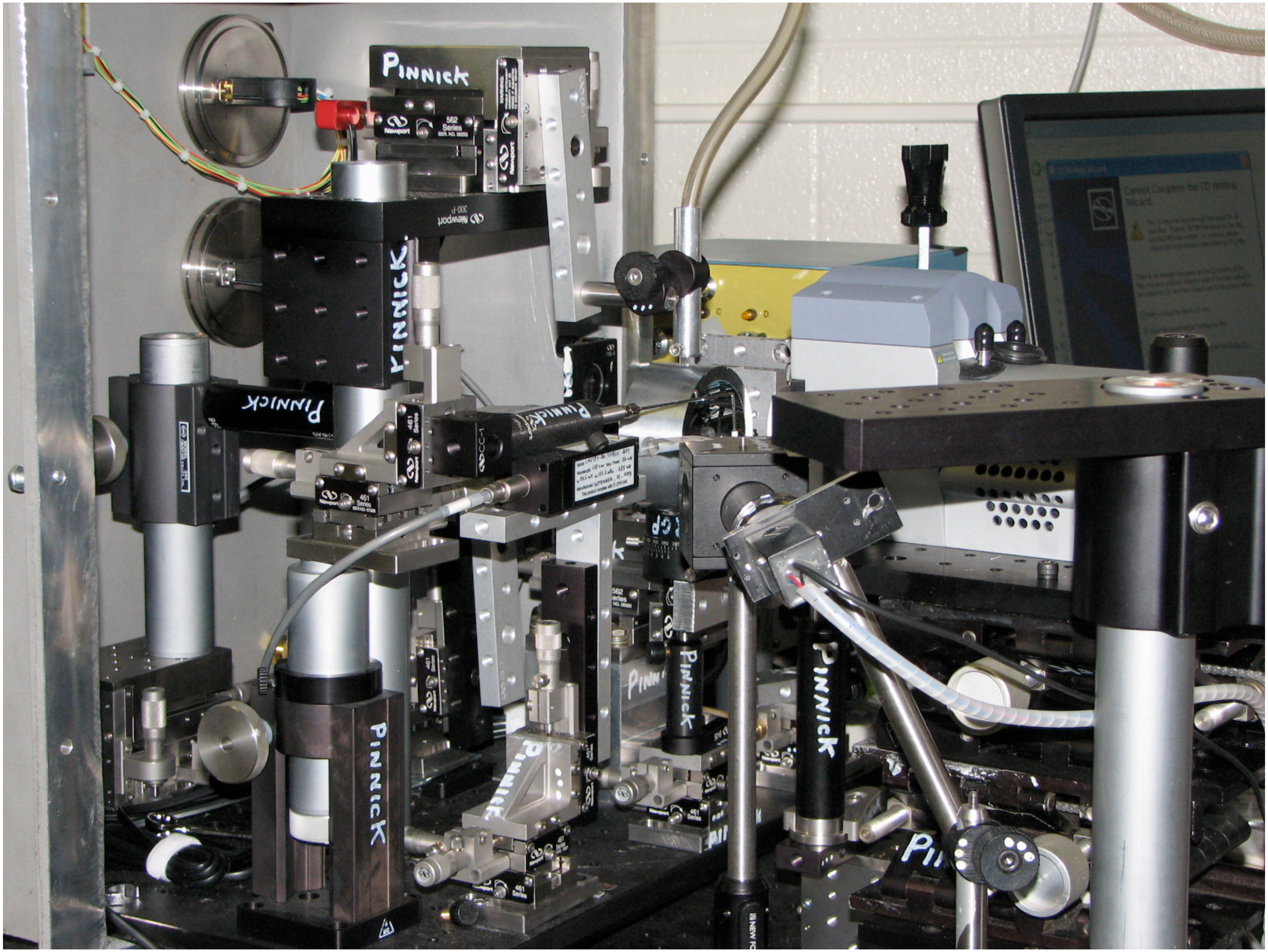
- Transmission electron microscopy (TEM) and scanning electron microscopy (SEM) not suitable for volatile particles
- SEM not sensitive to internal structure and variations in composition
- Single-particle angular elastic scattering techniques are sensitive, information-rich, and non-intrusive, but they are indirect, and inversion methods to determine morphology of non-spherical and agglomerate particles from scattering patterns is technically challenging

Previous attempts to characterize particles using angular elastic scattering

- **Gucker, F. T., J. Tuma, H. M. Lin, C. M. Huang, S. C. Ems, and T. R. Marshall (1973), Rapid Measurement of Light-Scattering Diagrams from Single Aerosol Particles in an Aerosol Stream and Determination of the Latex Particle Size, *Aerosol Science*, *4*, 389-404.**
- **Marshall, T. R., C. S. Parmenter, and M. Seaver (1976), Characterization of Polymer Latex Aerosols by Rapid Measurement of 360 degree Light-Scattering Patterns from Individual Particles, *Journal of Colloid and Interface Science*, *55*(3), 624-636.**
- **Bartholdi, M., G. C. Salzman, R. D. Hiebert, and M. Kerker (1980), Differential light scattering photometer for rapid analysis of single particles in flow, *Appl. Opt.*, *19*(10), 1573-1581.**
- **Wyatt, P.J., K.L. Schehrer, S.D. Phillips, C. Jackson, Y.J. Chang, R.G. Parker, D.T. Phillips, and J.R. Bottiger (1988), Aerosol-Particle Analyzer, *Applied Optics*, *27*(2), 217-221.**
- **Dick, W. D., P. J. Ziemann, P. F. Huang, and P. H. McMurry (1998), Optical shape fraction measurements of submicrometre laboratory and atmospheric aerosols, *Measurement Science & Technology*, *9*(2), 183-196.**
- **Kaye, P. H., E. Hirst, J. M. Clark, and F. Micheli (1992), Airborne Particle-Shape and Size Classification from Spatial Light-Scattering Profiles, *Journal of Aerosol Science*, *23*(6), 597-611.**
- **Pan, Y. L., K. B. Aptowicz, R. K. Chang, M. Hart, and J. D. Eversole (2003), Characterizing and monitoring respiratory aerosols by light scattering, *Optics Letters*, *28*(8), 589-591.**

(a) Cross-Sectional View

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Newport
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461 Series
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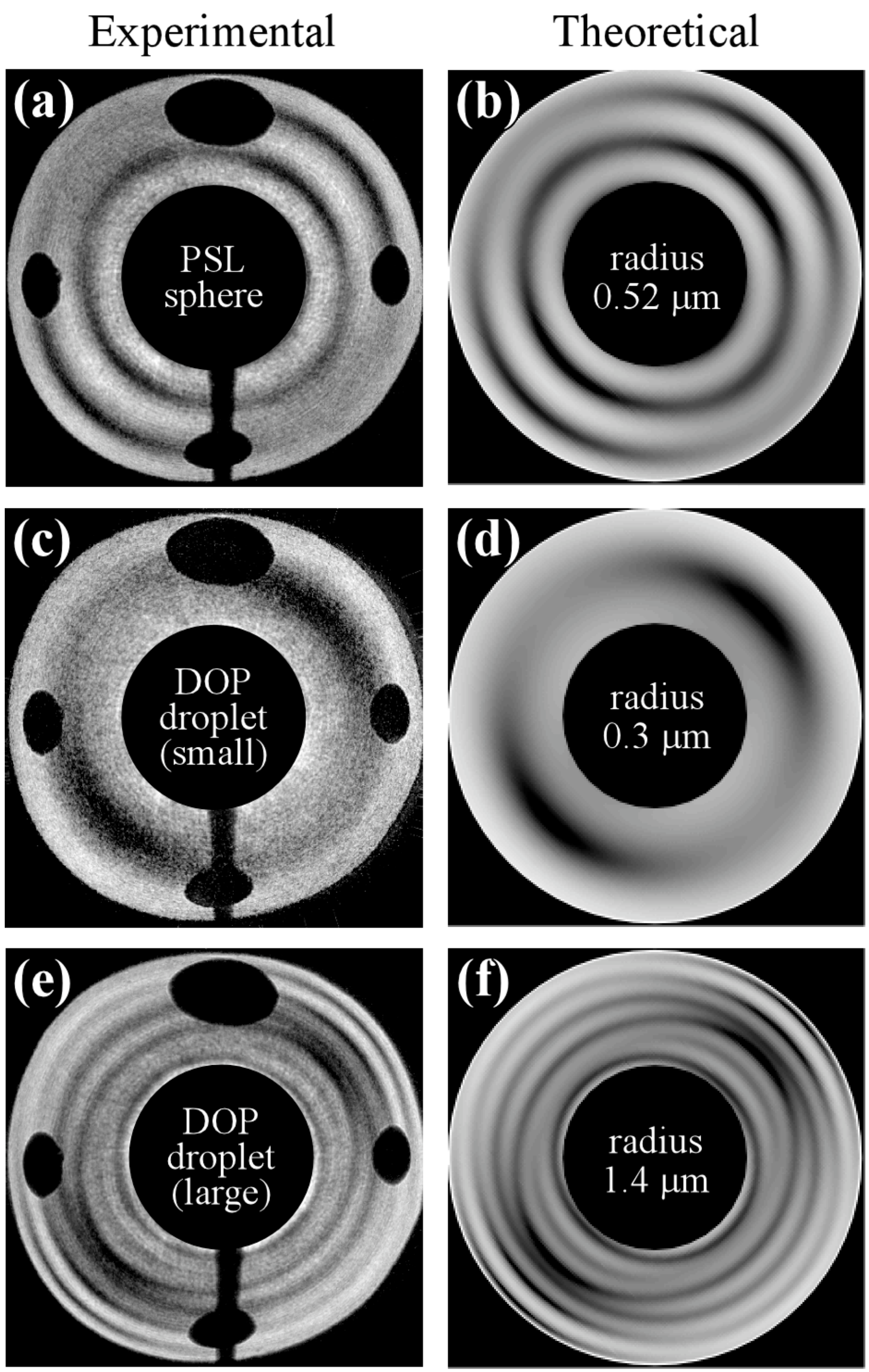
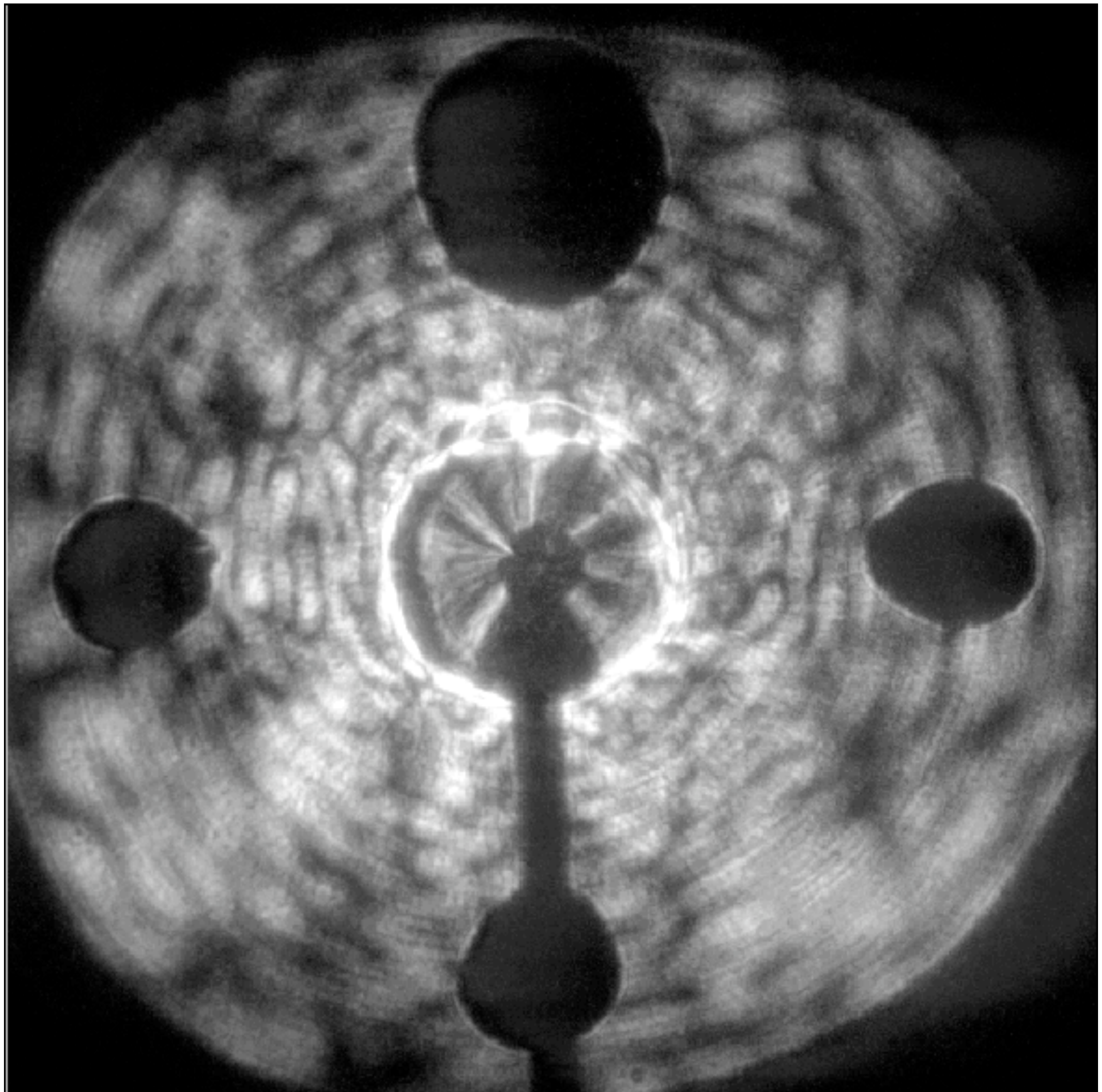


TAOS patterns of calibration particles compared with Lorenz-Mie calculations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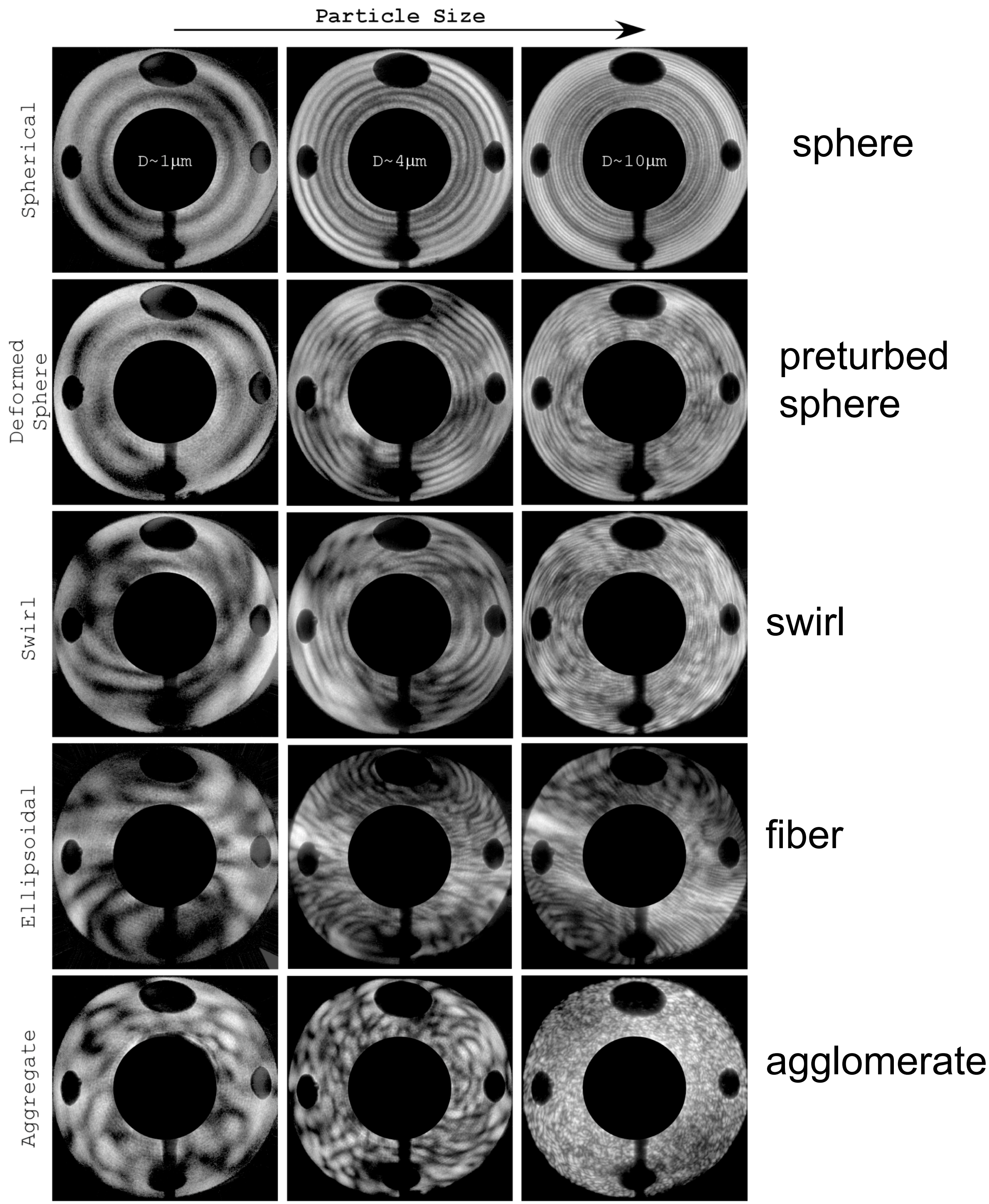
Scattering pattern classes



Possible aerosol particles corresponding to angular scattering pattern classes

- Sphere- organic carbon particles composed of single-ring, double-ring, and polycyclic aromatic hydrocarbons, organic polymers, humic acids, fulvic acids, humic-like substances; mixtures of acid sulfates/nitrates/sea salt with water; amorphous carbonaceous tar balls
- Preturbed sphere- multi-component particles (particularly particles with inclusions) such as organic carbon with sulfates; organic carbon with nitrates, organic carbon with crustal material, organic carbon with black carbon, acid sulfate particles with black carbon inclusions, metal inclusions in sulfuric acid , silicate fly ash, sulfuric acid mixed with crustal material, nearly spherical pollens or spores
- Swirl- mixtures suggested for the perturbed sphere class but with more non-homogeneity; crystalline-like leaf surface waxes dislodged by the wind; multiplet particles; silica shards produced by the combustion of coal
- Fiber- ammonium nitrate crystals; doublet particles of the same or different composition but with similar size
- Agglomerate- aggregate soot particles from oil-fired or coal-fired power plants; mineral dust of soil origin; agglomerates of particles of the same or different composition that may include sulfates, nitrates, quartz, clay minerals, organic carbon and black carbon; diesel engine emissions; cellular particles with complex morphology that are injected directly into the atmosphere including skin fragments, plant fragments, pollen, spores, bacteria, algae, and fungi

	Total Particles analyzed	Spheres (%)	Perturbed Spheres (%)	Swirls (%)	Fiber (%)	Aggregate (%)
1 micron	523	42	23	22	5	9
1.5 micron	505	21	24	26	4	25
3 micron	527	6	13	32	6	43
5 micron	510	<0.1	5	18	6	71

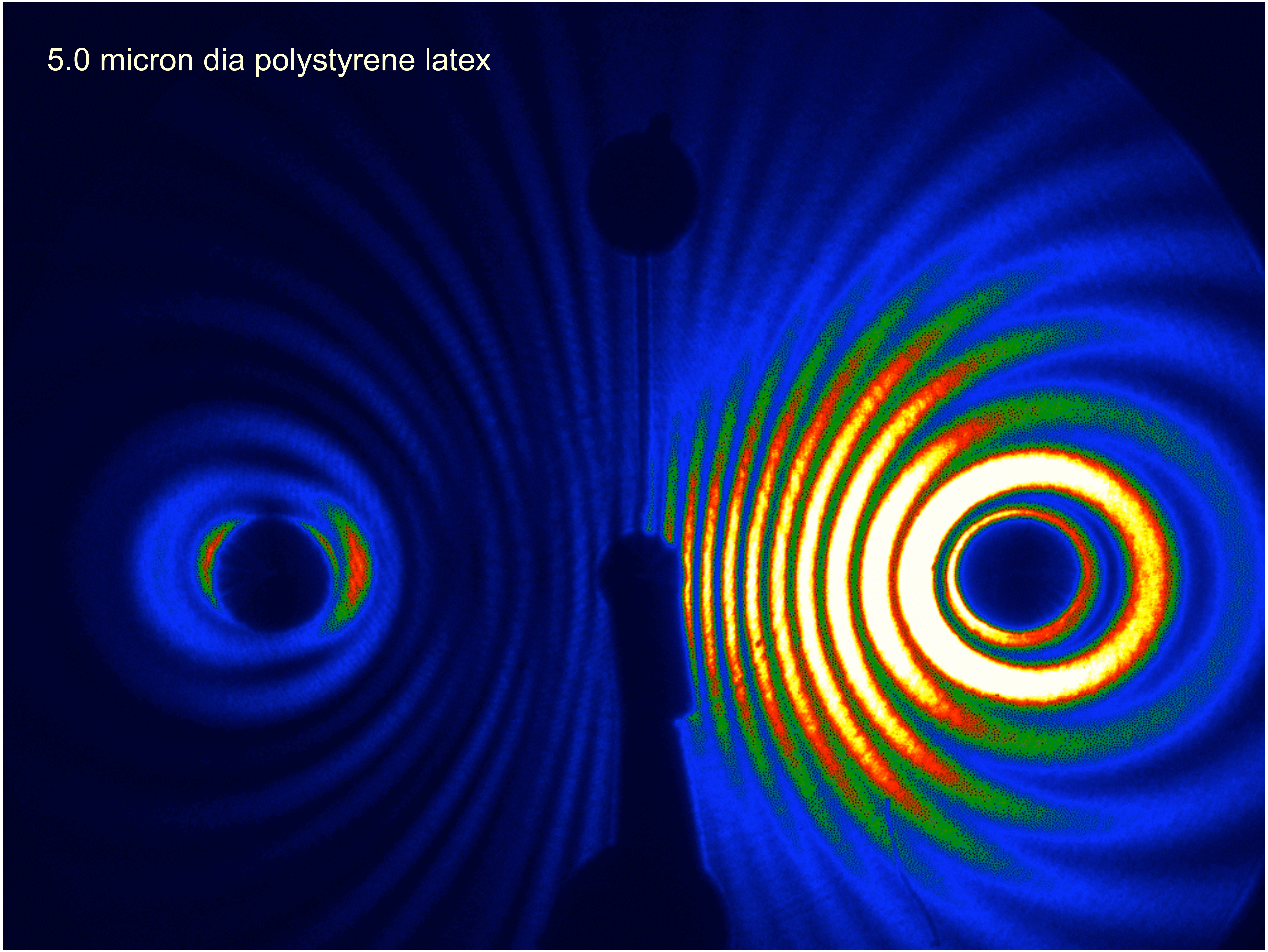
Probably formed mostly through gas-to-particle reactions

Probably formed mostly by direct injection into atmosphere

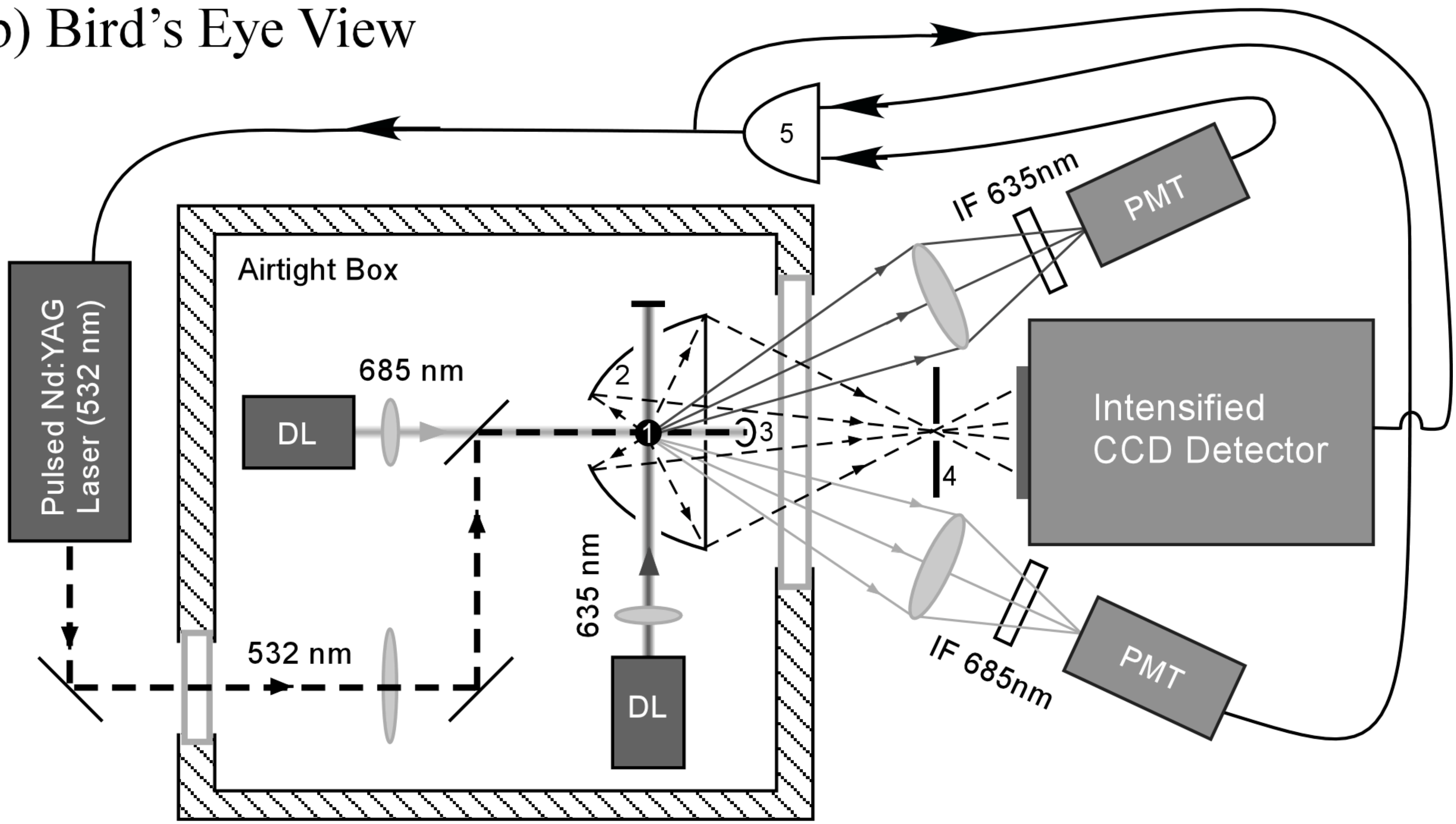
Frequency-of-occurrence of ambient aerosol scattering pattern class types for several particle sizes. Patterns collected at Adelphi, Maryland Oct 6-7, 2004

Future experiments

- Use more rigorous methods to classify angular scattering patterns.
- Change orientation of ellipsoidal mirror so that both forward and backward scattering patterns can be measured.
- Make improvements in the system (mainly improve the quality of the ellipsoidal mirror) so that refractive index and diameter of spherical particles can be inverted from the scattering data.


5.0 micron dia polystyrene latex

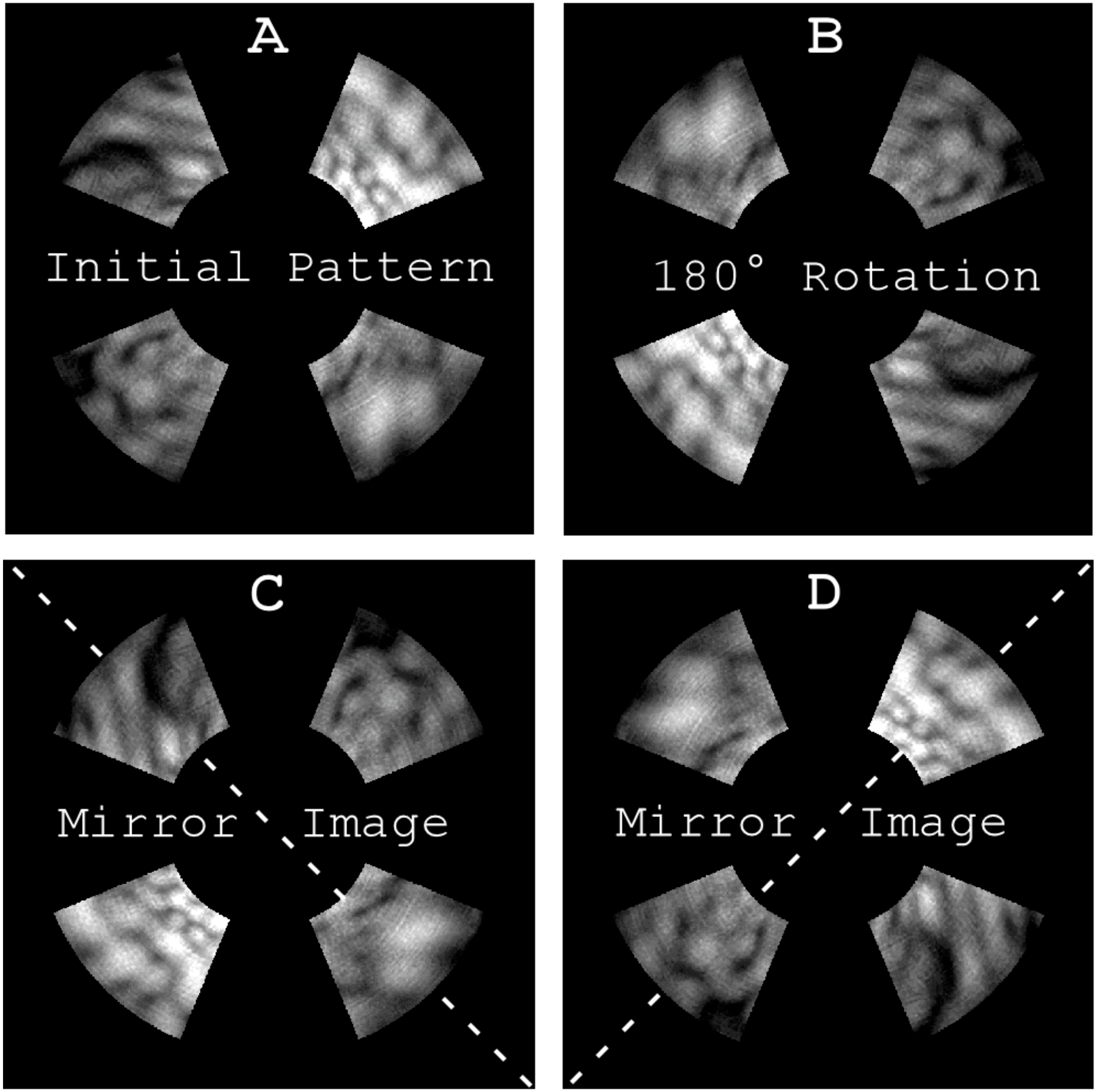
(b) Bird's Eye View



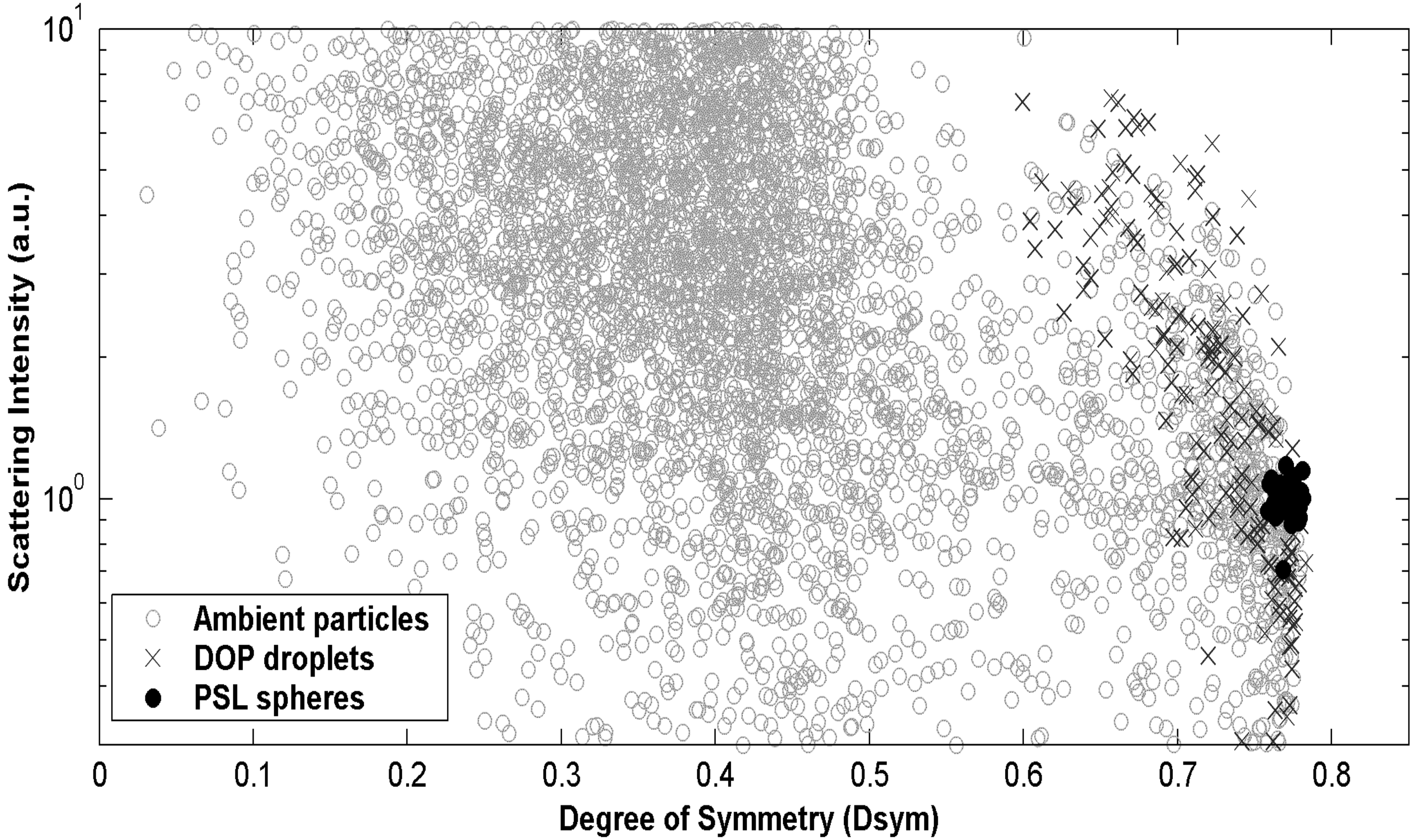
1. Trigger and scattering volume
2. Ellipsoidal reflector
3. 45 degree mirror
4. Iris
5. AND gate (pre-amplifiers not shown)

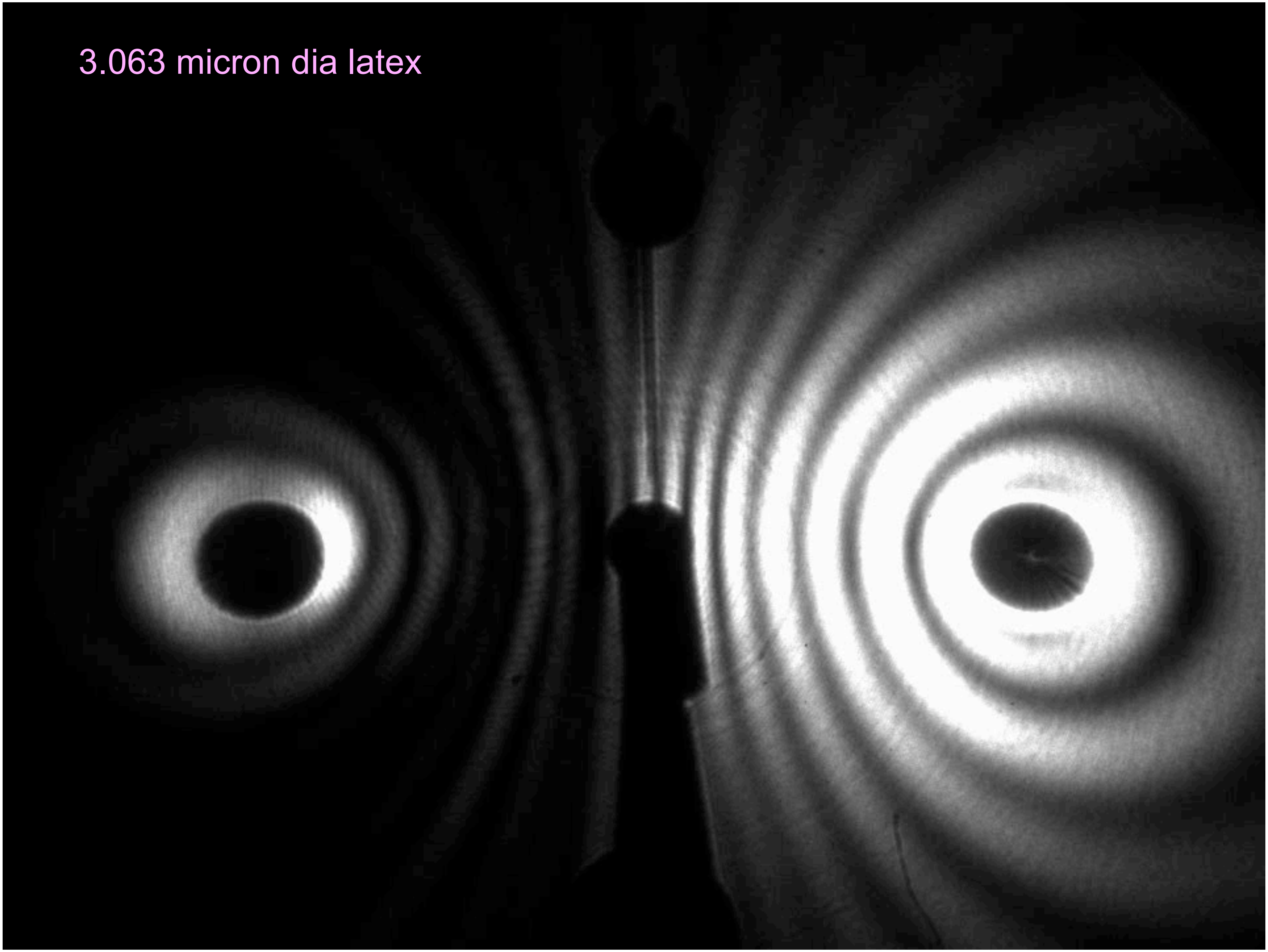
DL: Diode Laser (CW mode)
PMT: Photo-multiplier Tube
IF: Interference Filter

$$Dsym = \frac{1}{3}\sum_{i=1}^{3}\left(1 - \frac{\left(\sum_{pixel\ subset}(A - B_i)^2\right)^{1/2}}{2 \times RMS(A)}\right)$$


A
Initial Pattern
B
180° Rotation
C
Mirror Image
D
Mirror Image

Degree of Symmetry (Dsym) versus integrated scattering intensity, which roughly correlates to particle size, for calibration and ambient particles




3.063 micron dia latex