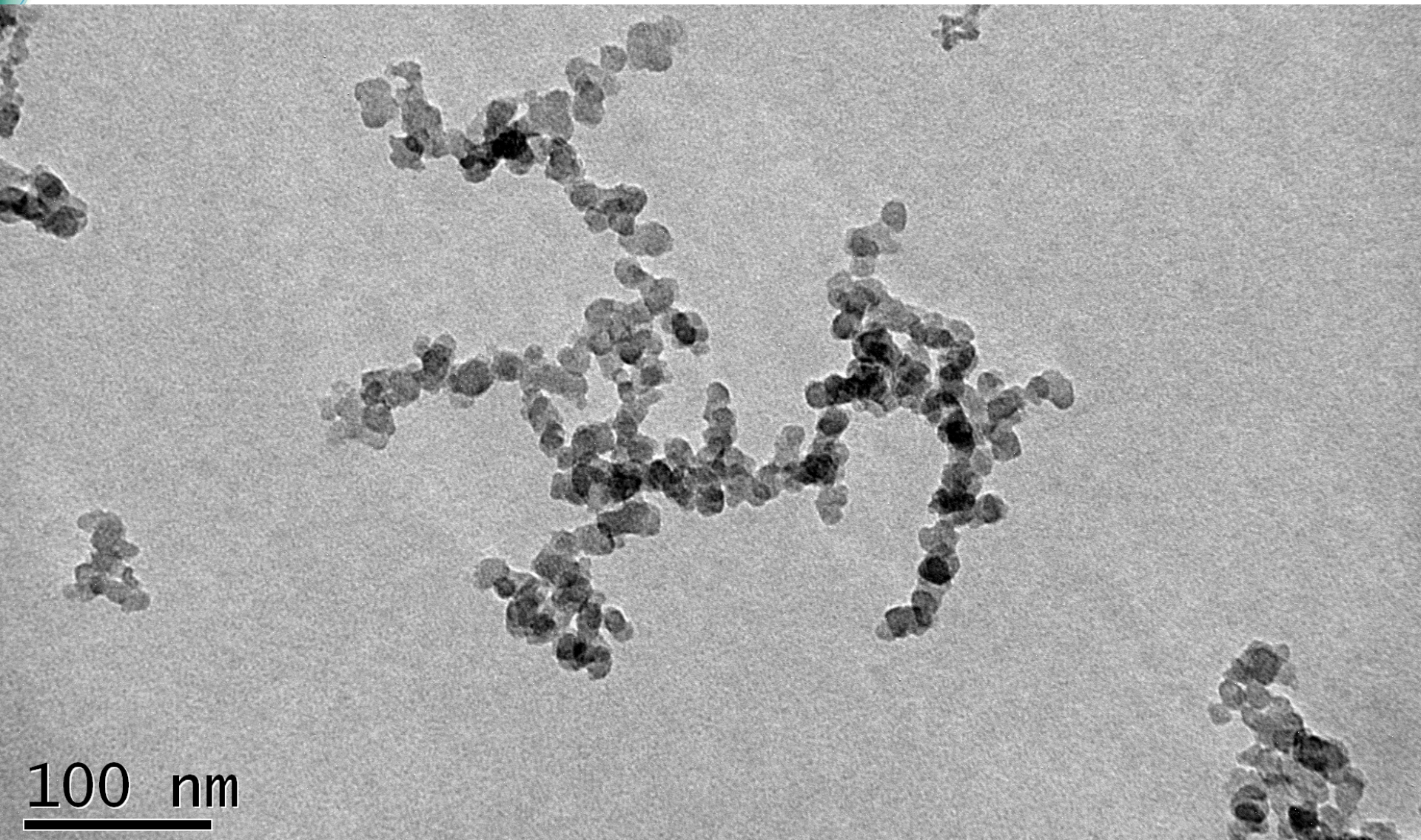
Effects of Soot Composition and Morphology on Optical Diagnostics:

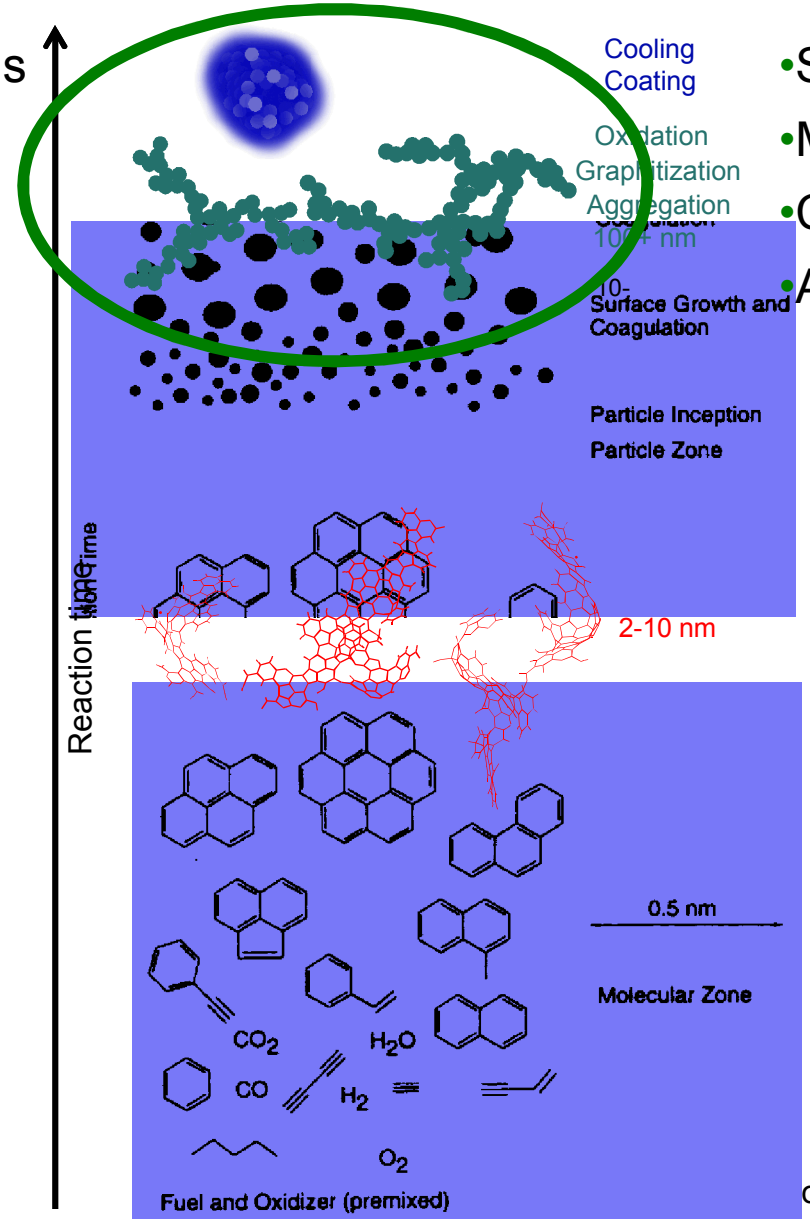
Experimental Artifacts or Useful Tools?

SAND2014-4707C



Soot formation and evolution

- What are the mechanisms and rates for growth and oxidation?
- How do particles physically and chemically evolve?



- Size
- Morphology
- Composition
- Abundance

Violi, *personal communication*

ot Formation in Combustion" 1994

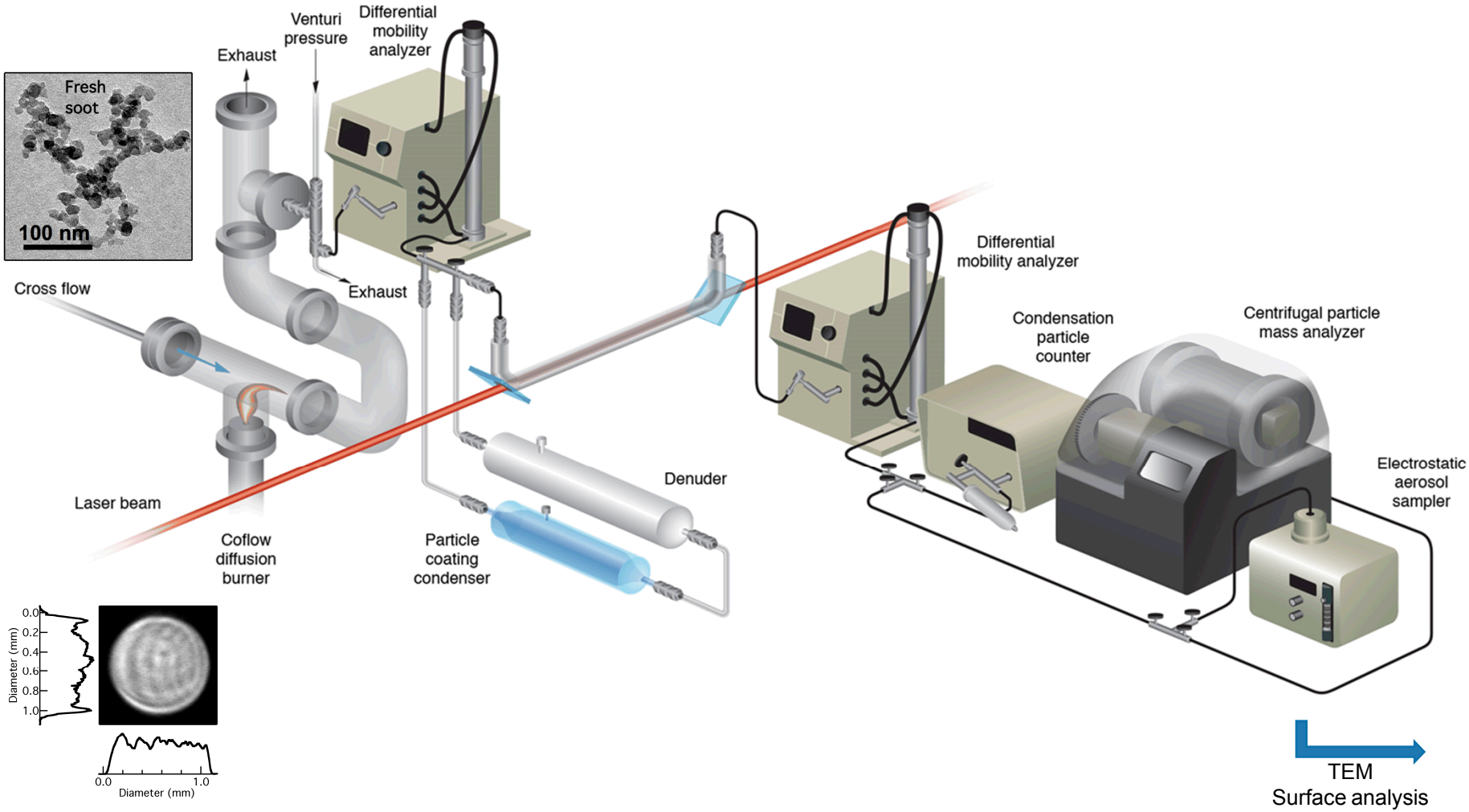
Why do we care about soot coatings?

- Particles in combustors and exhaust streams often coated with
  - Unburned fuel, oil, ash, combustion by-products

- Coatings can change particle
  - Optical properties, morphology, heat capacity, chemical reactivity
  - Global-warming potential
  - Contribution to air pollution
  - Impact on human health

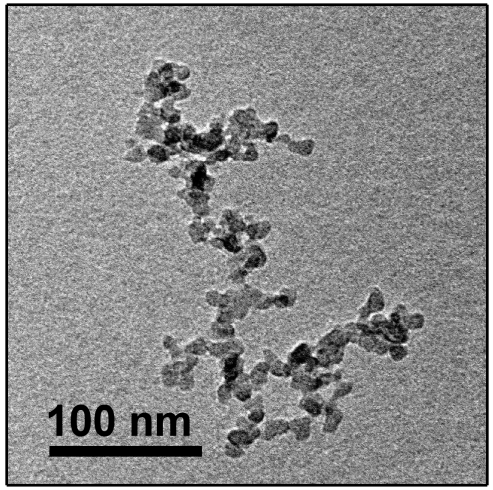
- Coatings can affect
  - After-treatment systems
  - Emissions
  - Optical diagnostics

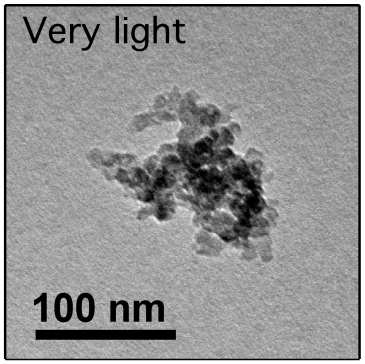
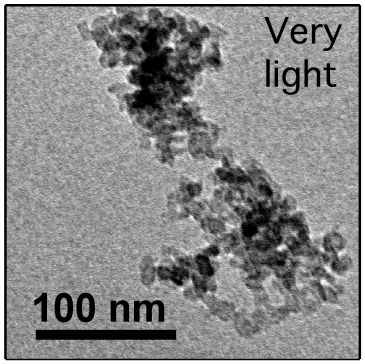
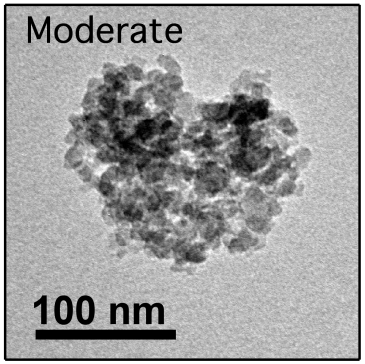
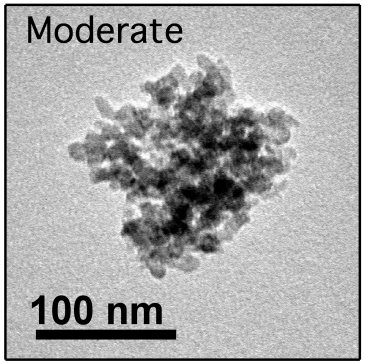
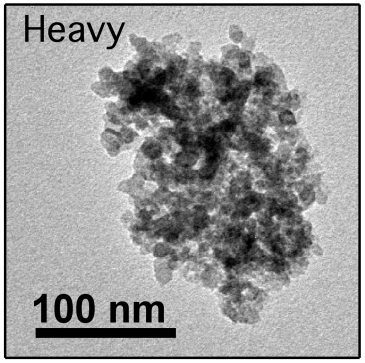
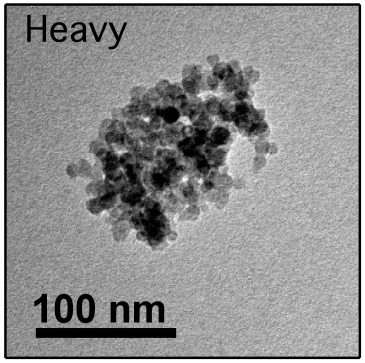
Effects of coatings on soot diagnostics



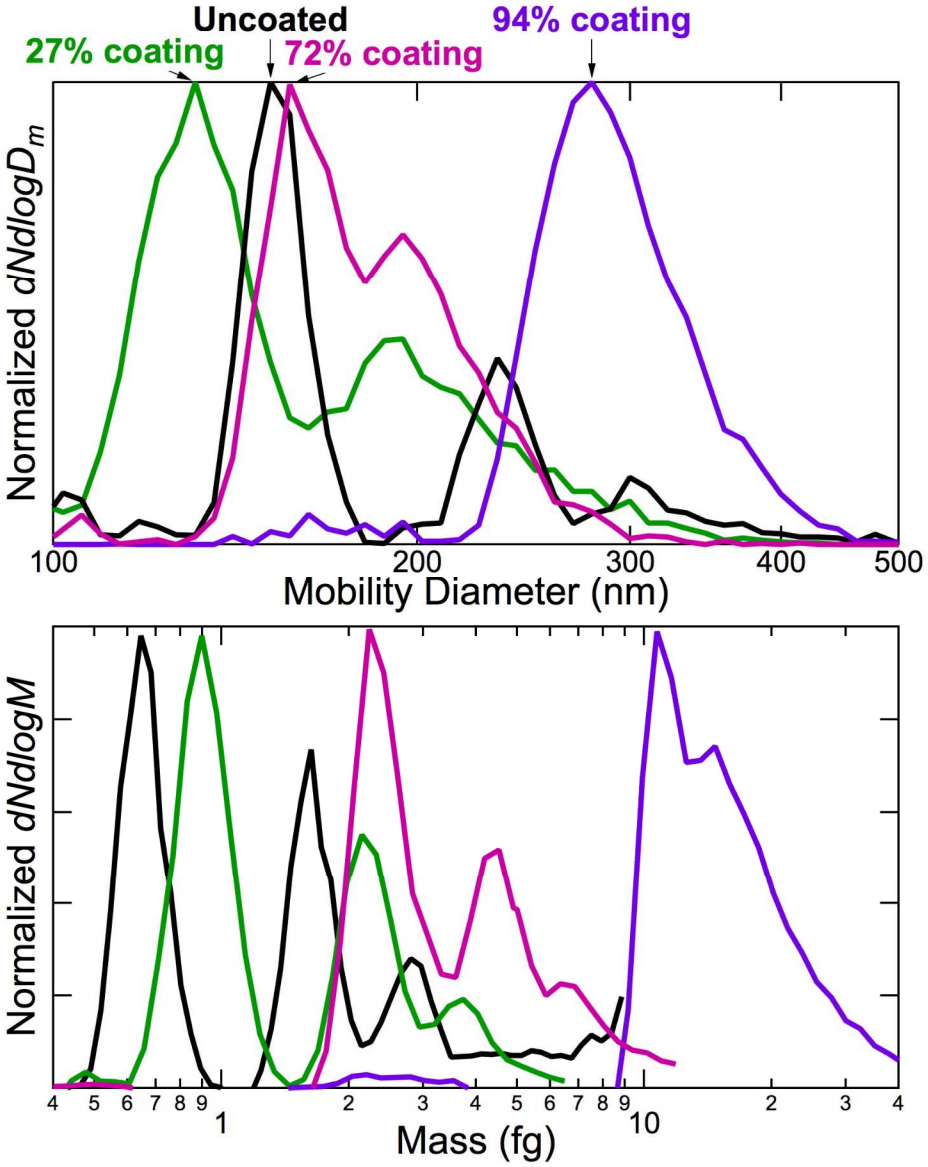
Effects of coatings on soot morphology

Flame-generated soot

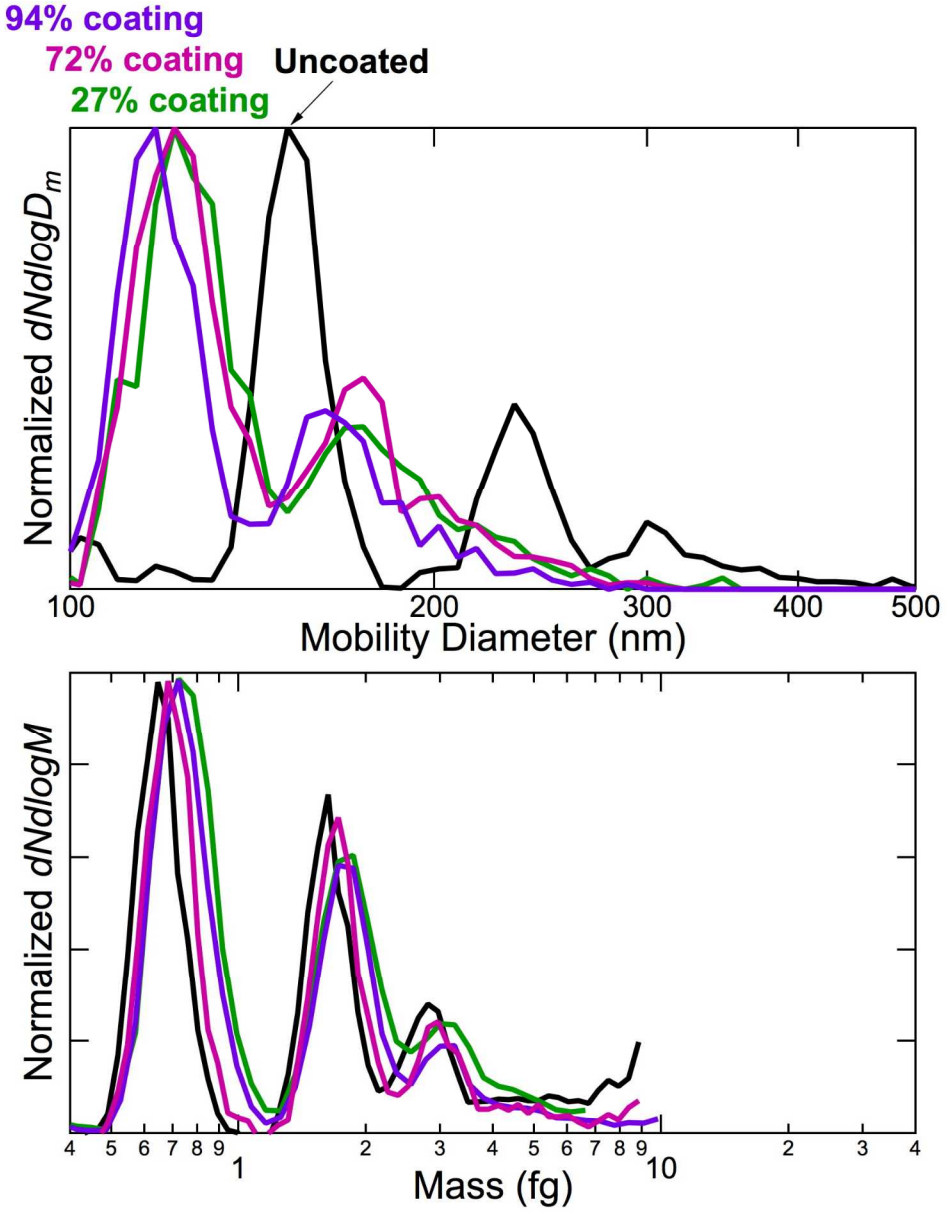


Coated w/ oleic acid	Coated, denuded	
		5% by mass
		72% by mass
		94% by mass

Effects of coatings on soot morphology

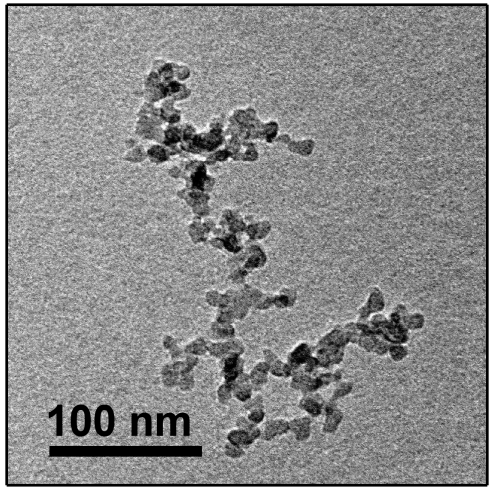


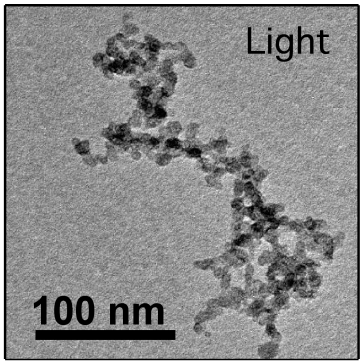
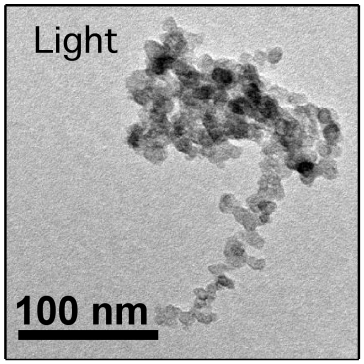
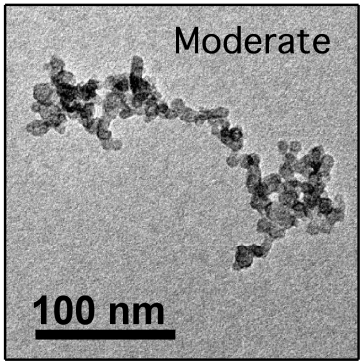
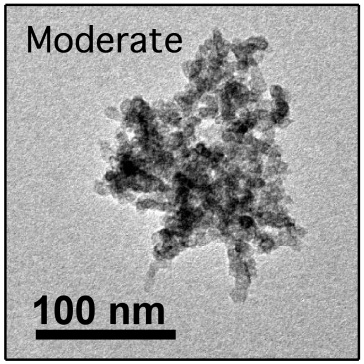
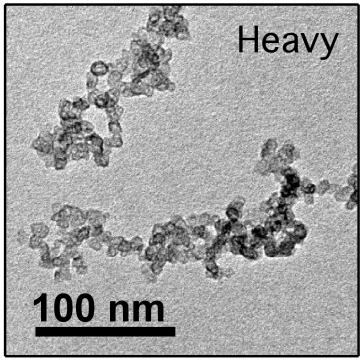
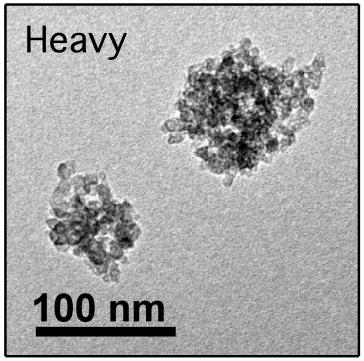
Effects of coatings on soot morphology



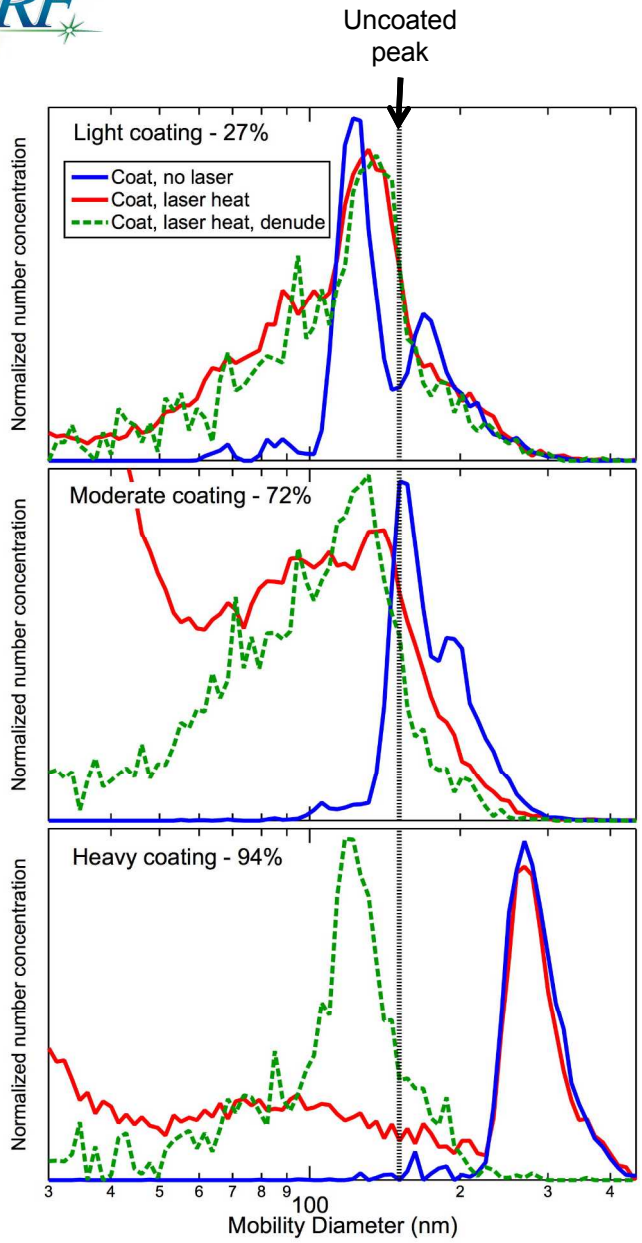
Effects of laser heating on morphology

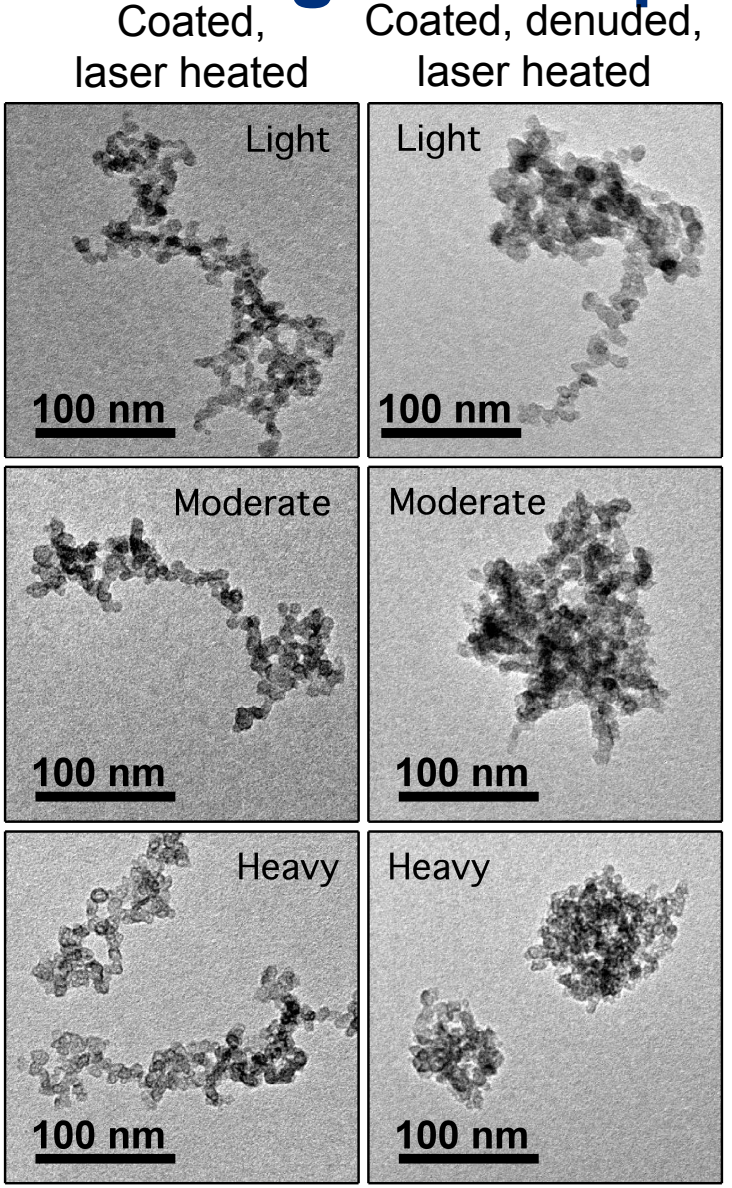
Flame-generated soot



Coated, laser heated	Coated, denuded, laser heated	
		27% by mass
		72% by mass
		94% by mass

Effects of laser heating on morphology





27% by mass

72% by mass

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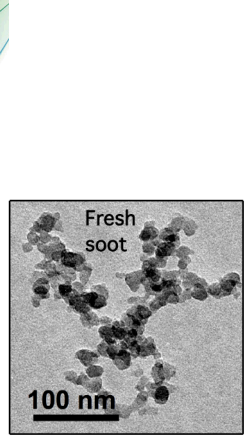
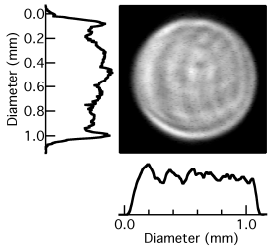
Effects of coatings on soot diagnostics

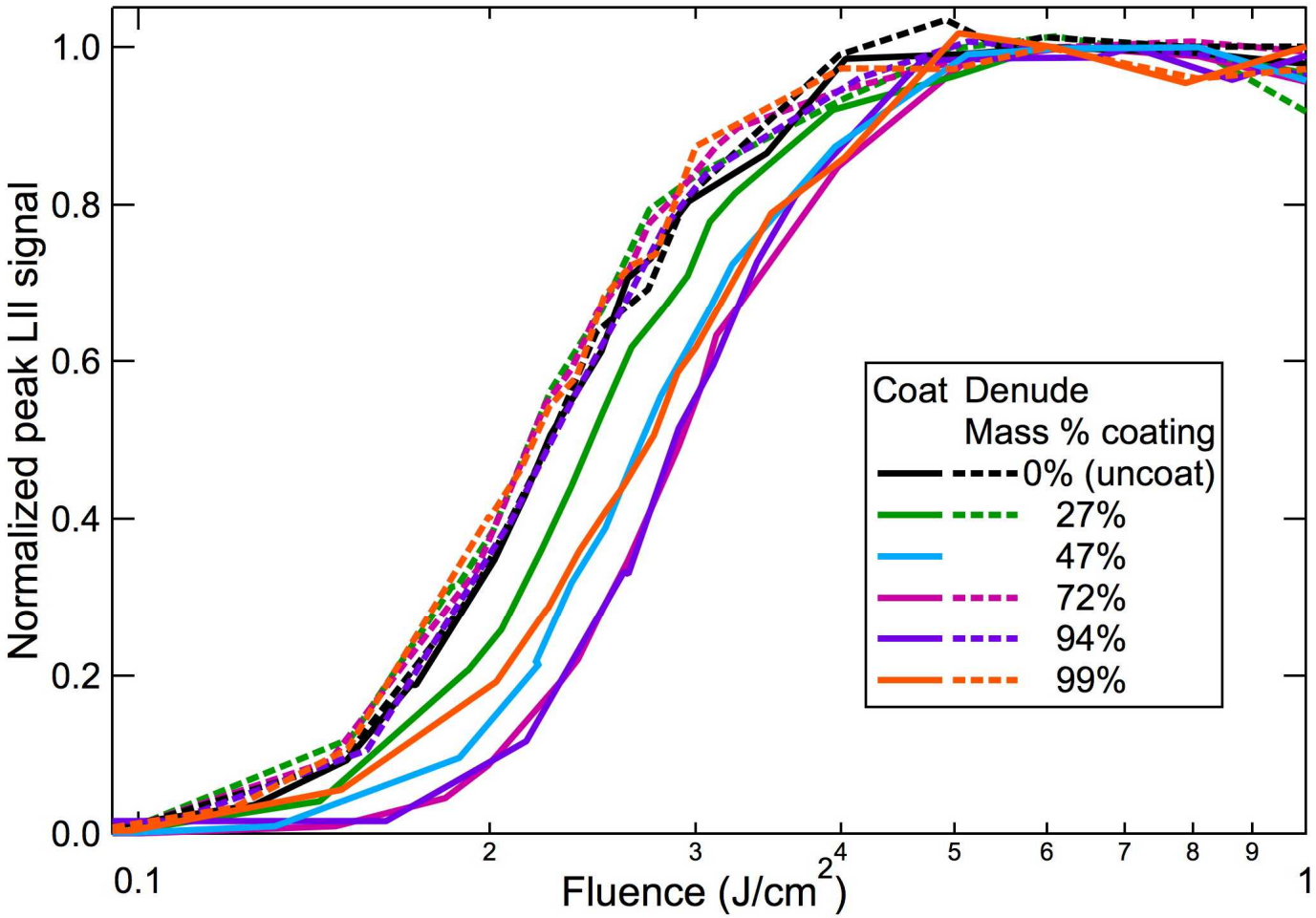


Nd:YAG laser
3-mm aperture
Thin-film polarizers
Half-wave plate
152.5-cm lens
50-cm lens
PMT
1-Ghz scope
Optical detection system
Relative humidity sensor
Venturi pressure
Exhaust
Differential mobility analyzer
Exhaust
Cross flow
Differential mobility analyzer
Condensation particle counter
Centrifugal particle mass analyzer
Electrostatic aerosol sampler
Denuder
Laser beam
Coflow diffusion burner
Particle coating condenser



TEM
Surface analysis

Effects of coatings on LII signal



Effects of coatings on LII signal

- Assuming absorptive heating dominates energy change (i.e., low fluence and short times), energy balance eqn is

$$\left[Mc_s(T) + M_{coat}c_{coat}(T)\right]\frac{dT}{dt} = \dot{Q}_{abs} - \dot{Q}_{coat}$$

- Integrating and rearranging gives:

Amount of coating evaporated

Absorption cross section of core particle

$$T_{\max} = \frac{\sigma_{abs}F_L}{Mc_s} - \frac{M_{vap}}{Mc_s}\left[\frac{\Delta H_{coat}}{W_{coat}} + c_{coat}\left(T_{boil} - T_0\right)\right] + T_0$$

Coating properties

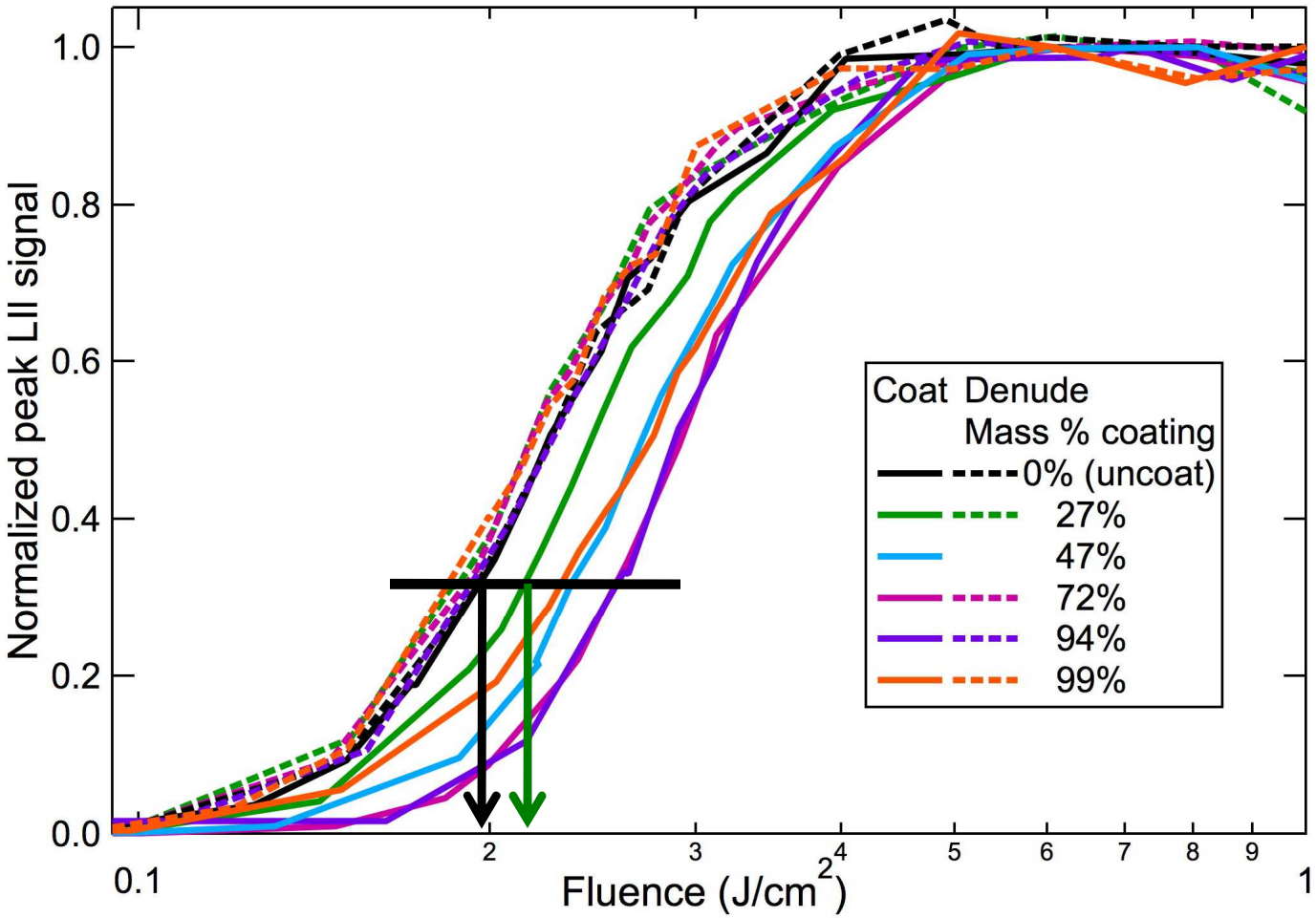
- When S_{max}(LII) is equal for coated and uncoated particles, T_{max} is also equal, and

$$F_L(\text{Coated}) = \frac{\sigma_{uncoat}}{\sigma_{coat}}F_L(\text{Uncoated}) + \frac{M_{vap}}{\sigma_{coat}}\left[\frac{\Delta H_{coat}}{W_{coat}} + c_{coat}\left(T_{boil} - T_0\right)\right]$$

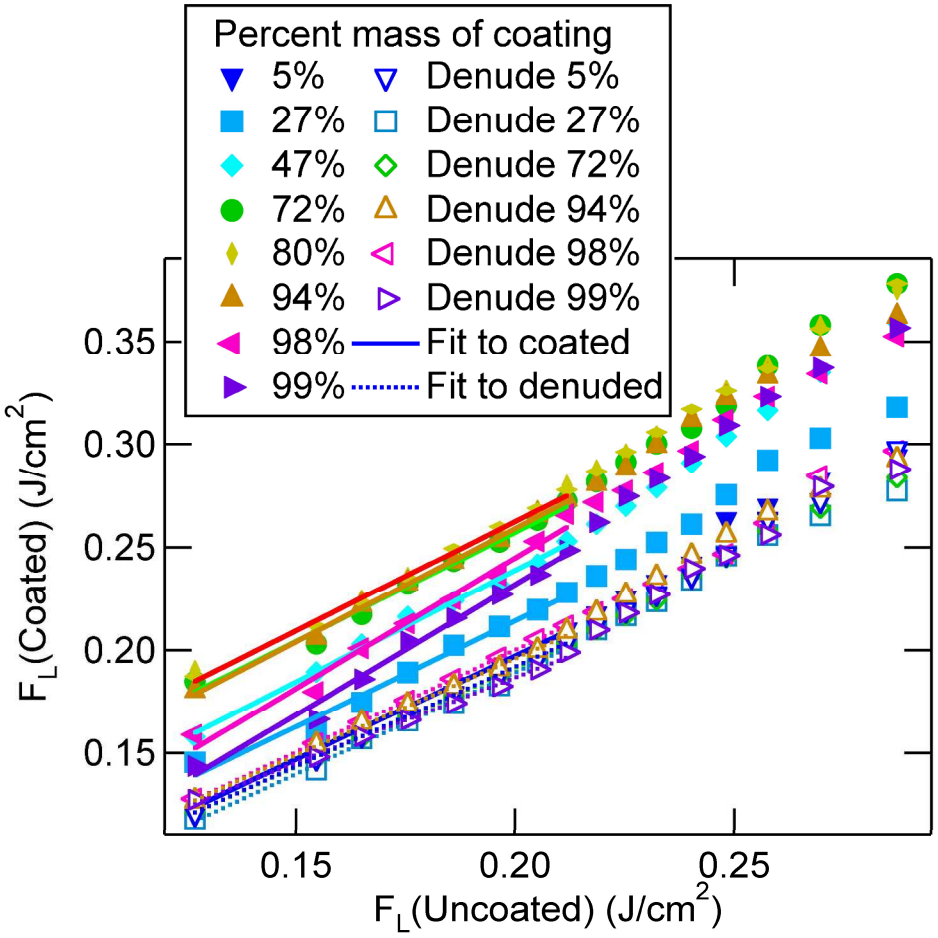
Slope^{-1} = Coating enhancement

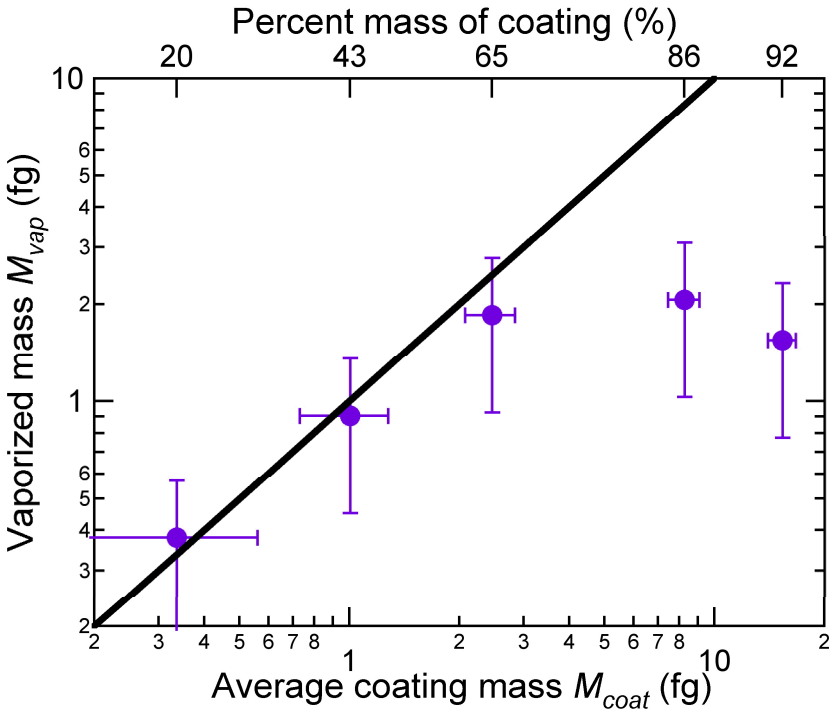
Intercept = Info about evaporated coating

Effects of coatings on LII signal



Effects of coatings on LII signal





$$\frac{1}{\text{Slope}} = \frac{\sigma_{coat}}{\sigma_{uncoat}}$$

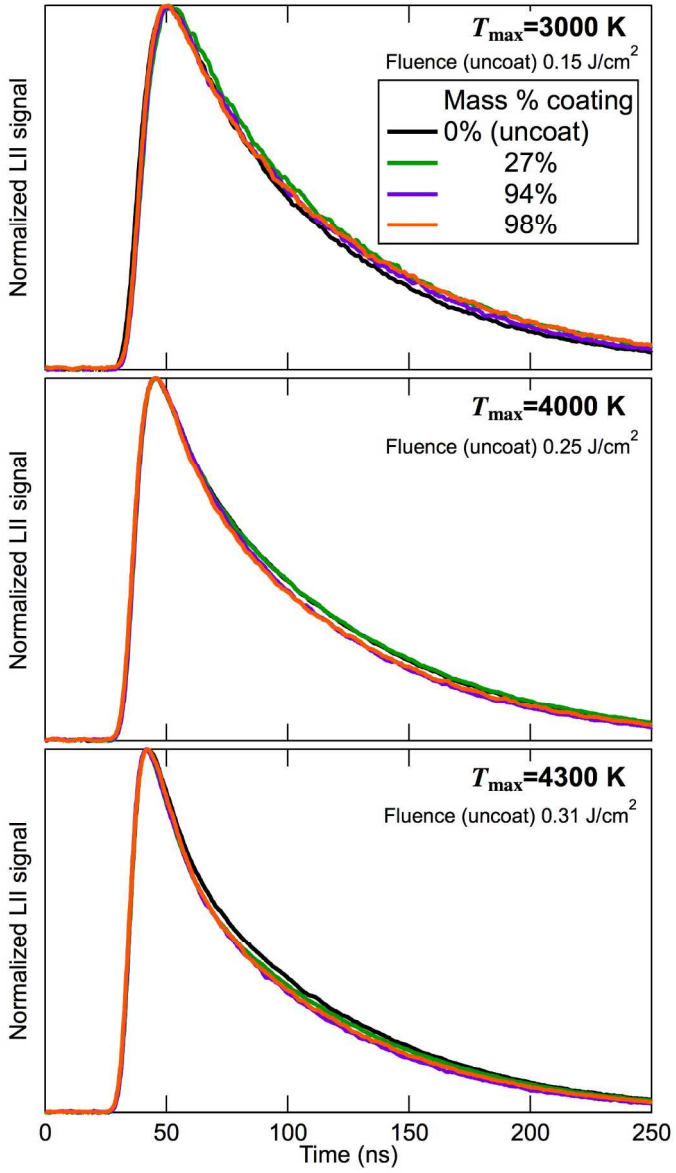
Slope^{-1} ≤ 1

$$M_{vap} = \frac{\text{Intercept}}{\text{Slope}} \frac{\sigma_{uncoat}}{\Gamma}$$

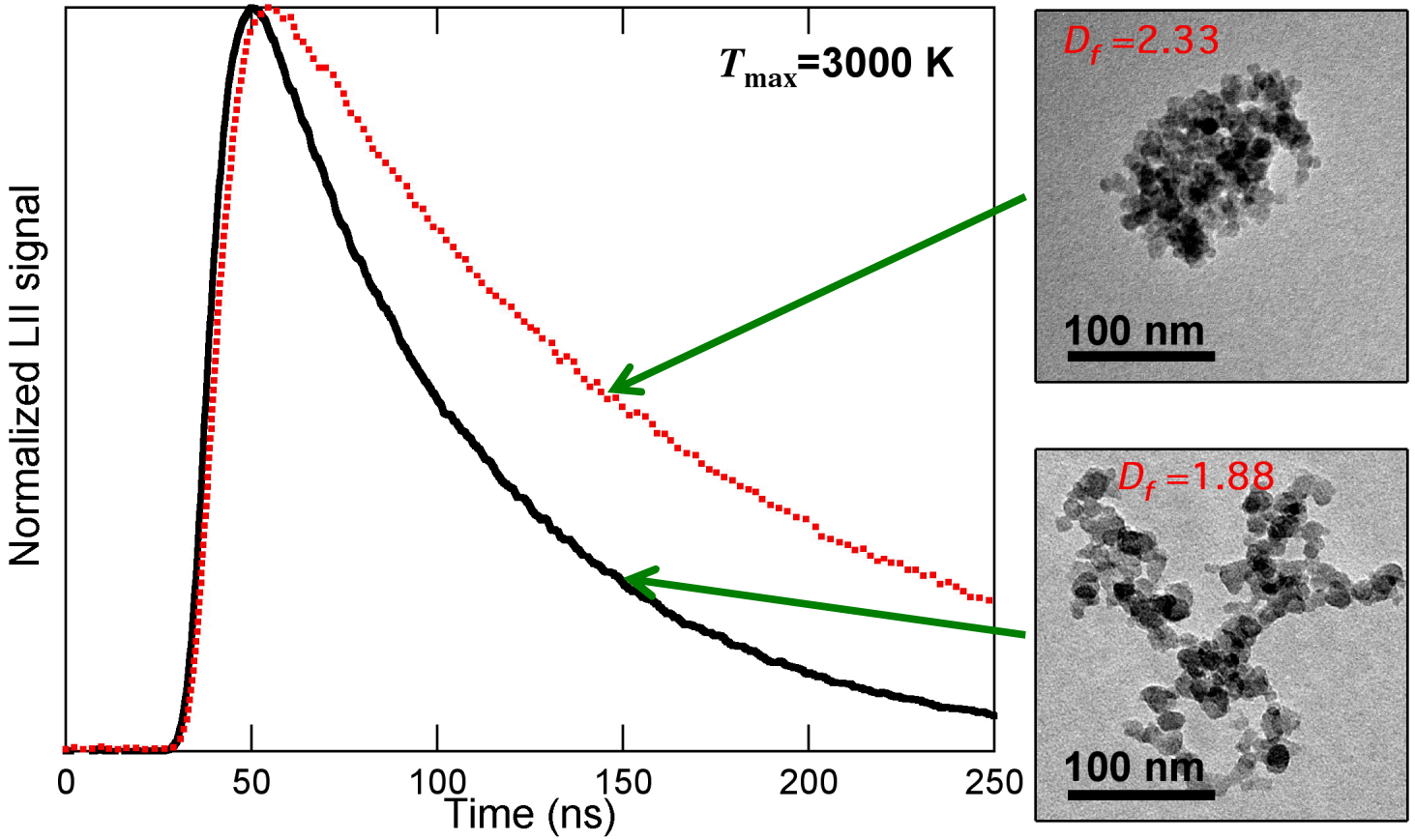
$$\sigma_{uncoat} = N \frac{\pi^2 d_p^3 E(m)}{\lambda_L}$$

$$\Gamma = \left[\frac{\Delta H_{coat}}{W_{coat}} + c_{coat} \left(T_{boil} - T_0 \right) \right]$$

Effects of coatings on LII signal



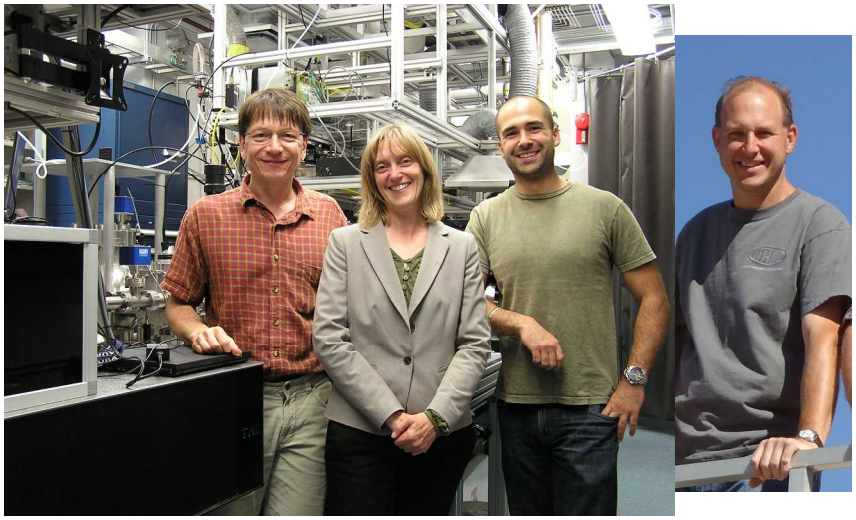
Effects of morphology on LII signal



Summary of results

- Coatings
  - Lead to particle collapse
  - Significantly reduce LII signal at some fluences
- Particle morphology
  - Has significant influence on LII signal decay rates
  - Complicates primary-particle sizing
- LII sensitivity to coatings and morphology may be useful
  - Fluence shift with coatings could be used to characterize composition
  - Coupled ELS and LII could be used to characterize morphology and size

Acknowledgements



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Paul Schrader
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Research and Development
DOE Office of Basic Energy Sciences

THANK YOU


CRF