Advances in High Energy Laser Diagnostics (HELD) for the Measurement of Particulate Matter

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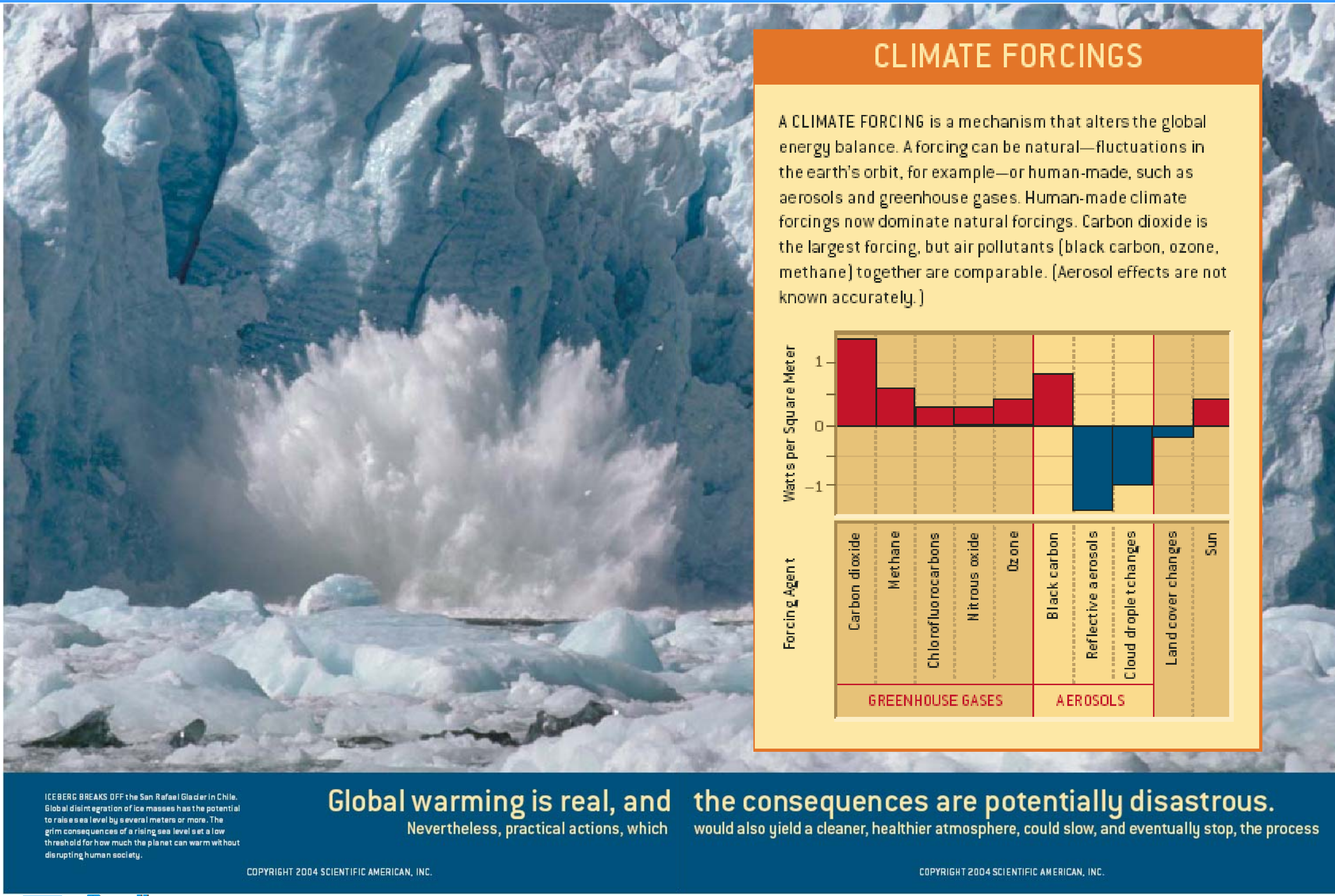
Outline

- Introduction
- High Energy Laser Diagnostics
  - LII
    - Laser-induced incandescence
  - ELS
    - Elastic light scattering
  - LIDELS
    - Laser-induced desorption with elastic light scattering
  - LIBS
    - Laser-induced breakdown spectroscopy
- Summary



CLIMATE FORCINGS

A CLIMATE FORCING is a mechanism that alters the global energy balance. A forcing can be natural—fluctuations in the earth's orbit, for example—or human-made, such as aerosols and greenhouse gases. Human-made climate forcings now dominate natural forcings. Carbon dioxide is the largest forcing, but air pollutants (black carbon, ozone, methane) together are comparable. (Aerosol effects are not known accurately.)

ICEBERG BREAKS OFF the San Rafael Glacier in Chile. Global disintegration of ice masses has the potential to raise sea level by several meters or more. The grim consequences of a rising sea level set a low threshold for how much the planet can warm without disrupting human society.

Global warming is real, and the consequences are potentially disastrous. Nevertheless, practical actions, which would also yield a cleaner, healthier atmosphere, could slow, and eventually stop, the process

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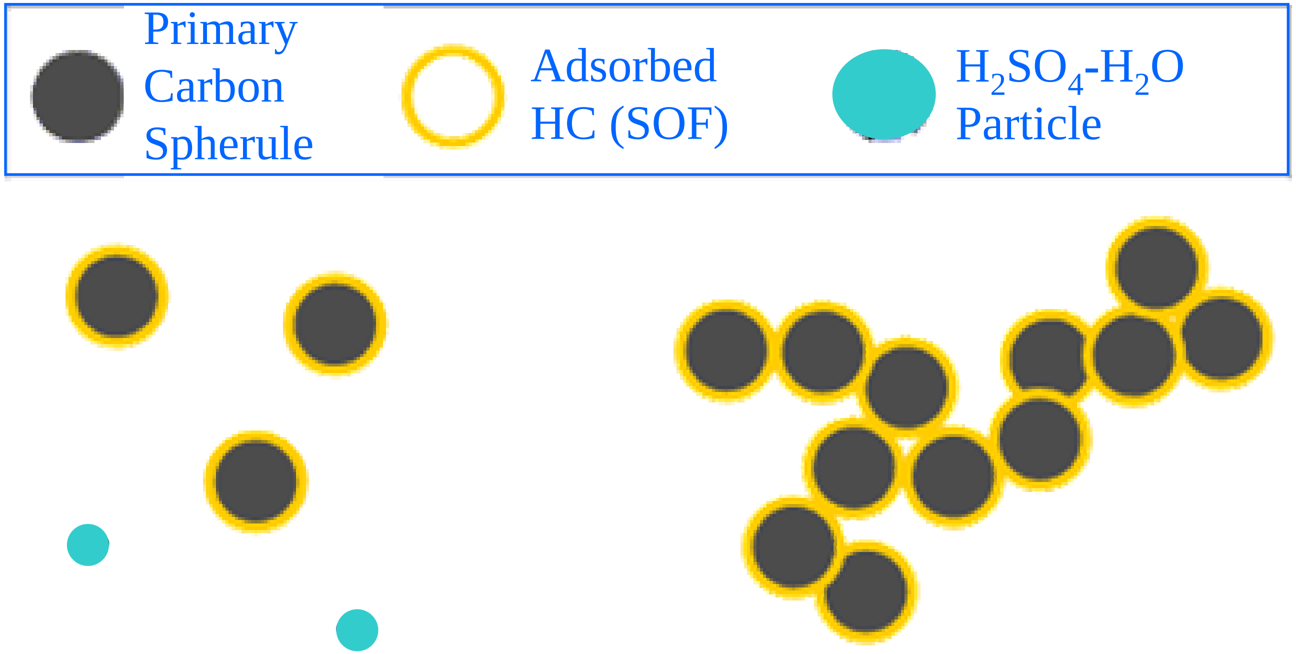
Particulate Matter Structure



Nuclei Mode
7-40 nm

Accumulation Mode
40-1000 nm

(from Dieselnet.com)

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High-Energy Laser Diagnostics (HELD)

- real-time PM measurement techniques are needed to investigate transients
- sensitive PM measurement techniques are needed to study cleaner vehicles
- *in situ* PM measurement techniques are needed to characterize engine-out/aftertreatment-in conditions
- *in situ* PM measurement techniques also avoid issues with sampling artifacts (dilution not required)
- optical diagnostic techniques, performed with pulsed lasers, can address these issues

HELD Advantages

- performed *in situ* or with extractive sampling
- performed without or with dilution
- large measurement range
- high sensitivity for very low concentrations
- high precision and repeatability
- high speed data acquisition and analysis
- real-time results
- applicable for steady and transient measurements
- spatially and temporally resolved
- suitable for engine and emissions control research, vehicle compliance enforcement, continuous emissions monitoring, etc.

HELD Techniques

Diagnostic	Property Measured
laser-induced incandescence (LII)	soot volume fraction, surface area, and primary particle diameter
laser-induced incandescence + elastic light scattering (LII + ELS)	aggregate size, number, and structure
laser-induced desorption + elastic light scattering (LIDELS)	volatile volume fraction
laser-induced breakdown spectroscopy (LIBS)	metallic ash (species and concentration)

Measurement of Diesel Particulate Matter

- laser-induced incandescence (LII) measures the elemental carbon (EC) component (soot) of diesel particulate matter (PM) emissions
- key advantages of LII:
  - —able to make real-time measurement in raw exhaust (i.e., without dilution)
    - assess particulate trap efficiency with before and after measurements
  - —high sensitivity (detection to better than 5 ppt)
  - —excellent transient response
  - —wide measurement range
  - —virtually no maintenance (24/7 hands-off operation)

Auto-Compensating LII (AC-LII)

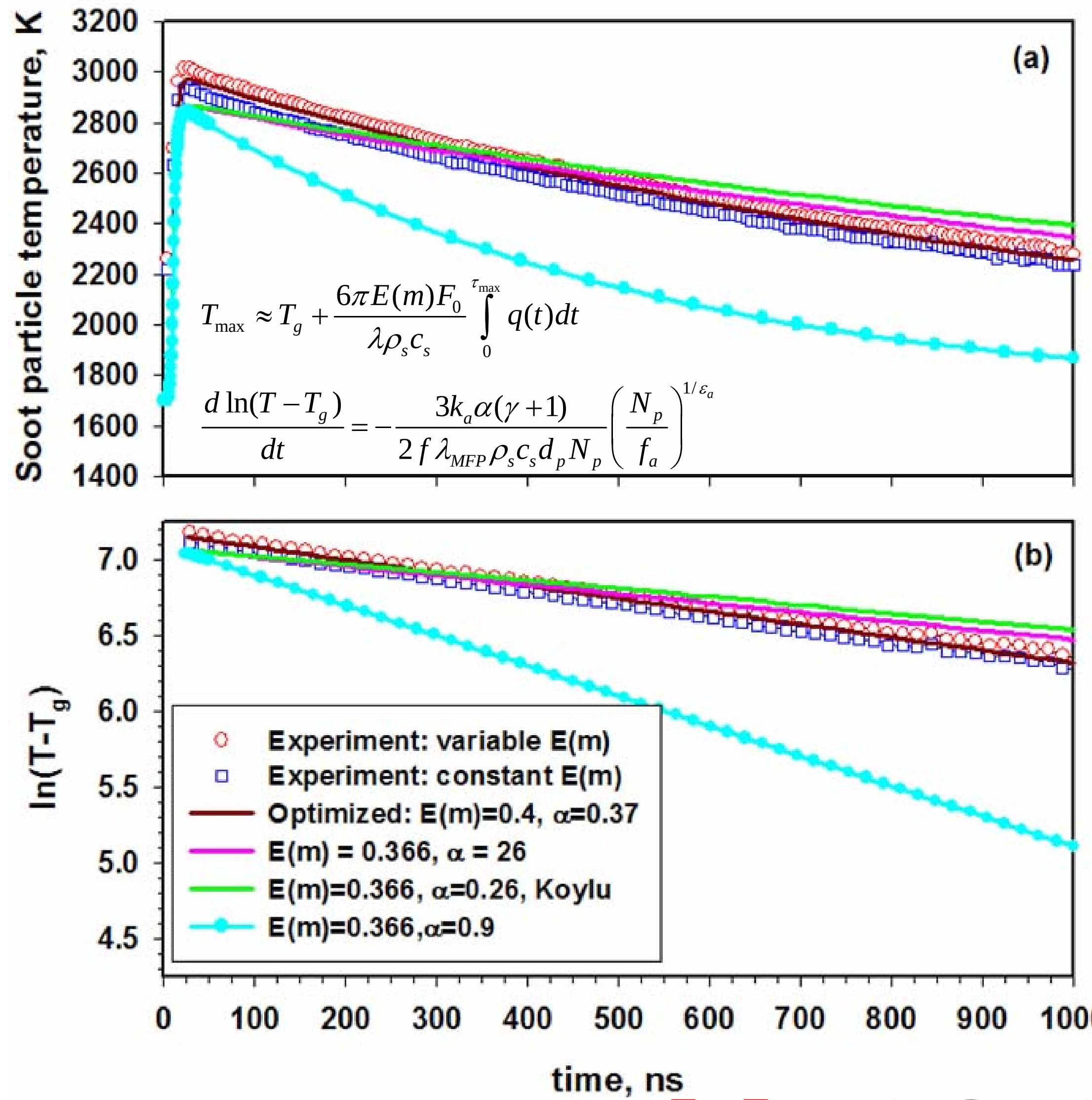
- two-color pyrometry coupled with LII to determine the time-resolved particle temperature
  - —permits use of low-fluence
  - —particles are kept below the sublimation temperature
- this new technique *automatically compensates* for any changes in the experimental conditions
  - —fluctuations in local ambient temperature
  - —variation in laser fluence
  - —laser beam attenuation by the particulate matter
  - —desorption of condensed volatile material

Accuracy of LII Measurements

- calibration source
  - radiance or irradiance calibration
- optics
  - absolute neutral density filter transmission
  - relative dichroic mirror reflectivity and interference filter transmission
- electronics
  - relative photodetector sensitivity
  - photodetector gain
  - amplifier gain
- dimensions of probe volume
- laser spatial fluence profile
- optical and other properties of soot at high temperatures

Determination of E(m) and α

- peak soot temperature dependent on the absorption function, *E(m)*
- temperature decay rate dependent on the thermal accommodation coefficient, α,
- combined approach of theoretical analysis and experimental particle temperature measurement applied to determine *E(m)* and α



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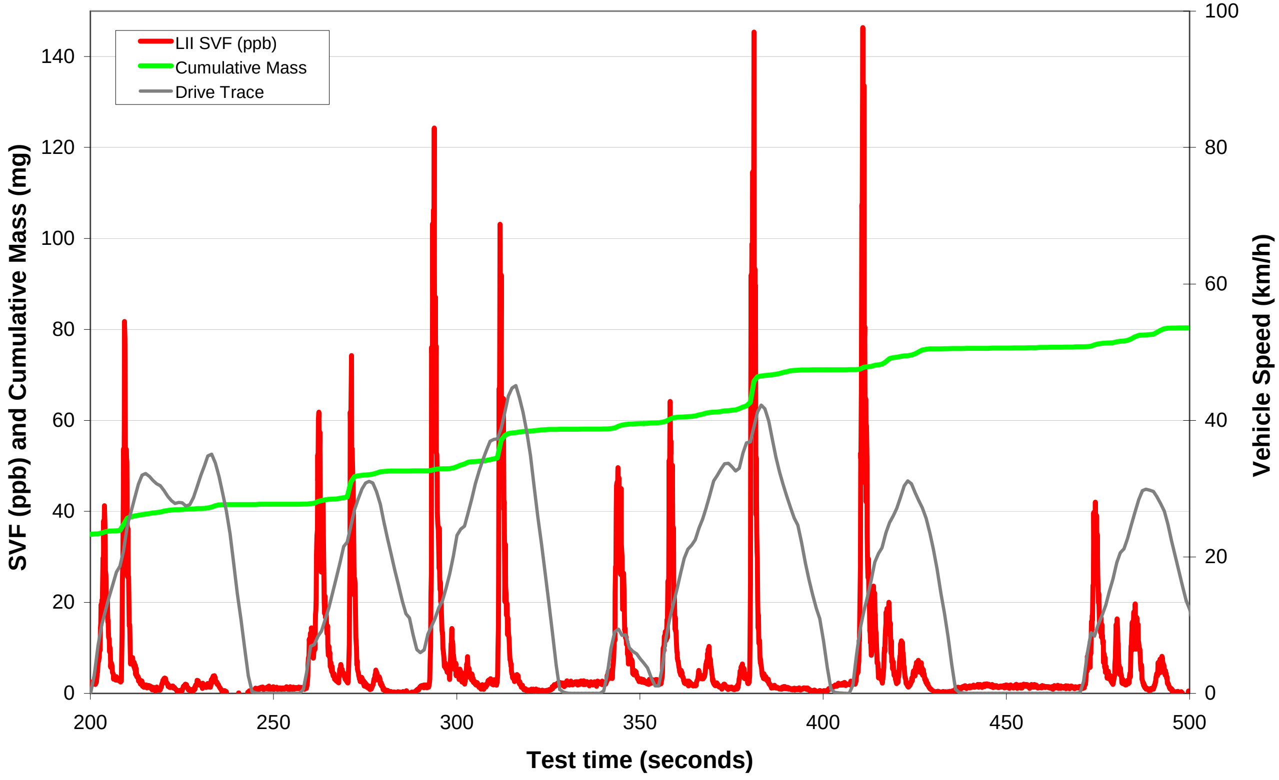
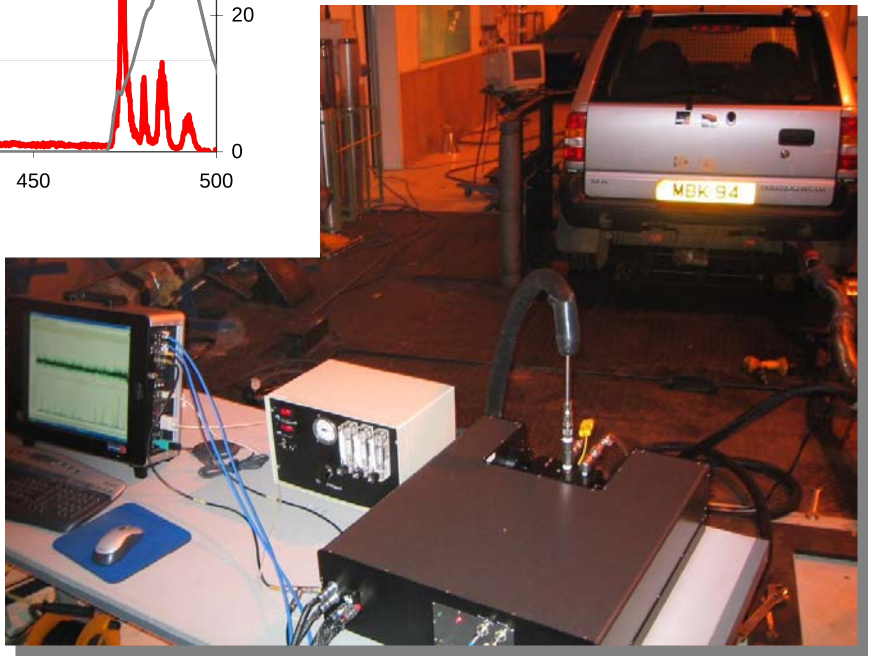
AC-LII in Use: Chassis Dynamometer



- cumulative mass determined from transient LII and exhaust flow measurements
- levels from 0.050 – 1500 ppb



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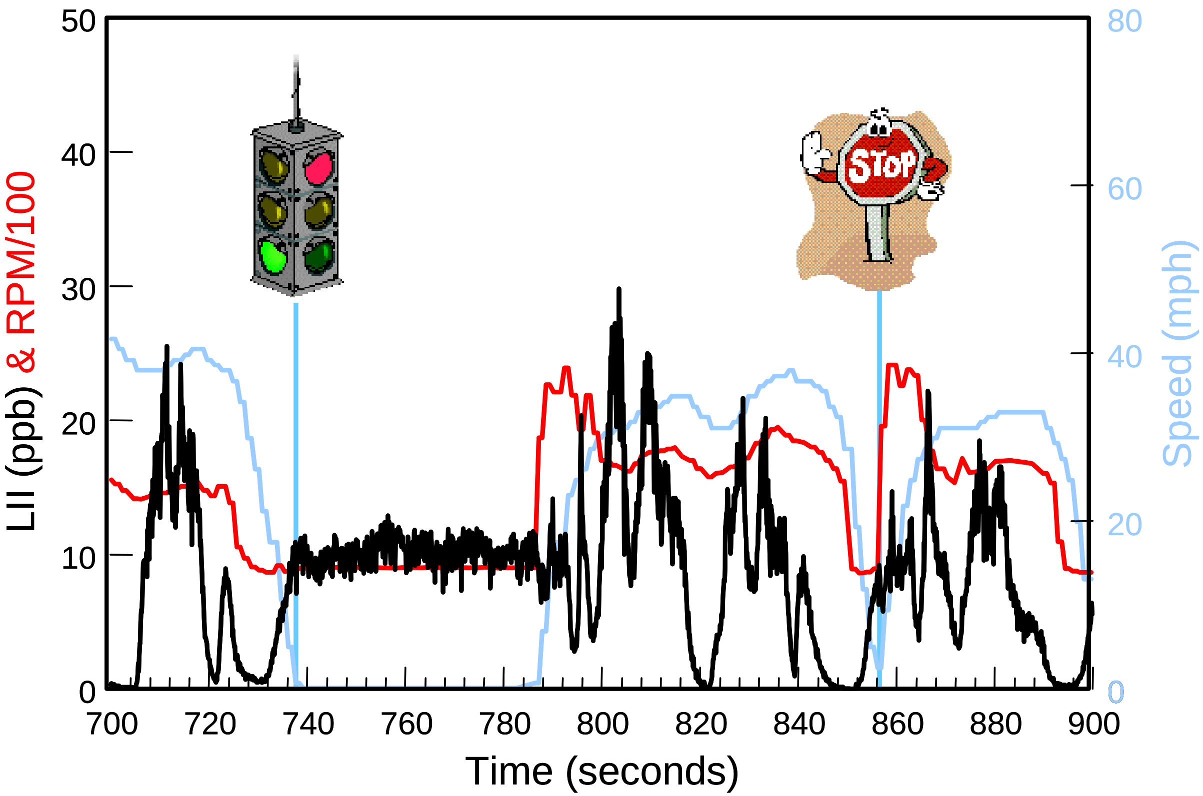
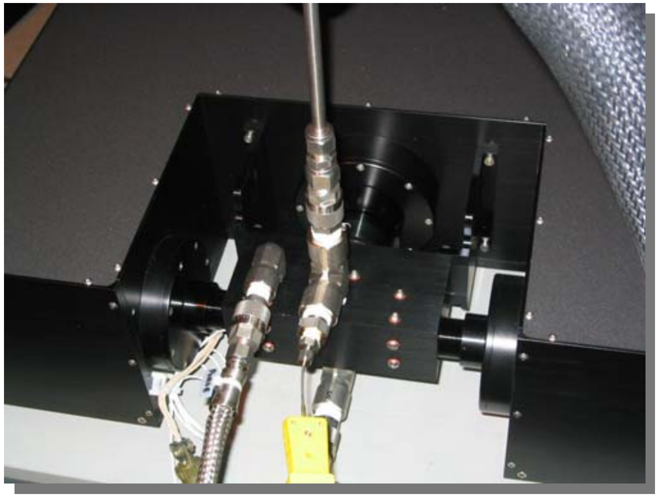
On-Vehicle AC-LII: Transient Measurements





13th CRC On-Road Vehicle Emissions Workshop, San Diego, April 7-9, 2003

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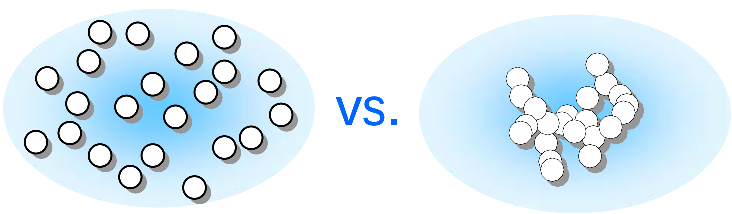
Determination of Soot Morphology

- it is clear that distribution parameters must be included to deconvolve information contained in time-resolved low fluence LII signals
- with many assumptions, this can be achieved by combined LII and elastic light scattering
- cannot use Rayleigh scattering theory, as the aggregates are far from spherical
- use RDG-PFA theory to interpret elastic scattered light, assume fractal parameters and a log normal distribution of N_p, the number of primary particles per aggregate
- combine with LII and iterate to determine N_g and d_p (assuming monodisperse primary particles)

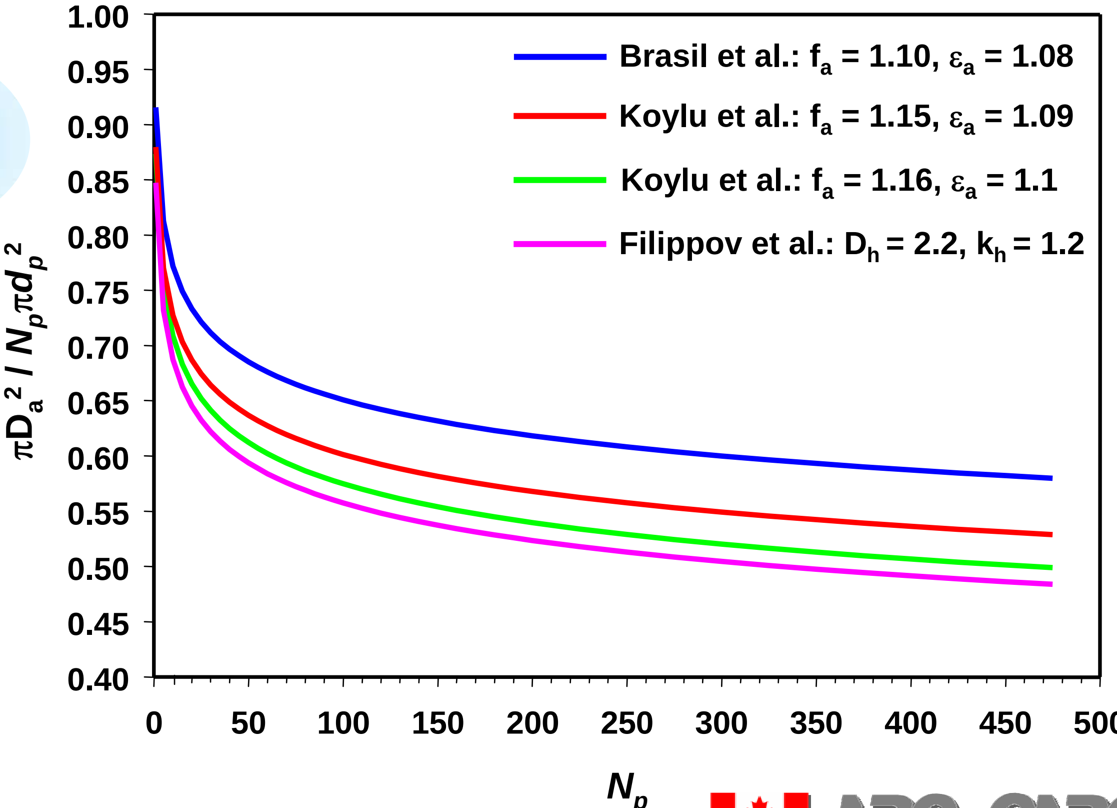
Consider Aggregate Effect on Cooling

- ratio of heat transfer areas of aggregated particles to non-aggregated particles
  - —shielding effect does not vary much with N_p for $N_p > 50$



$$D_a = \left(\frac{N_p}{f_a} \right)^{1/2\varepsilon_a} d_p$$

$$D_a = d_p (N_p / k_h)^{1/D_h}$$



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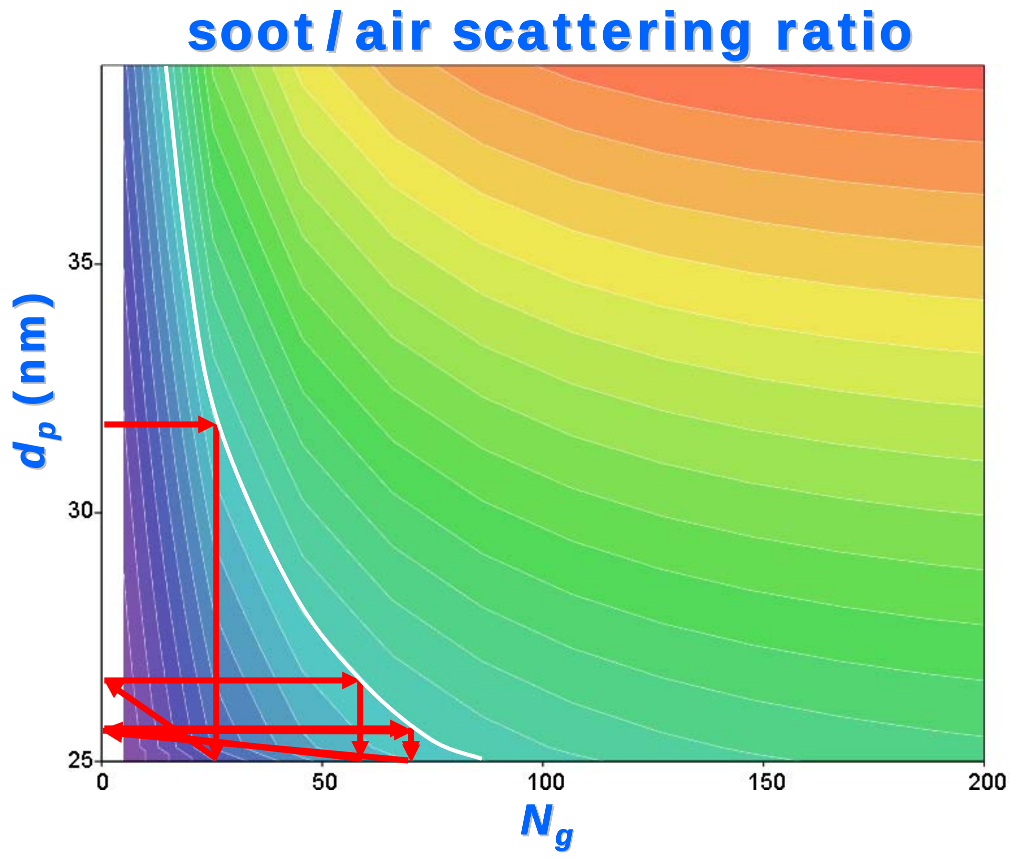
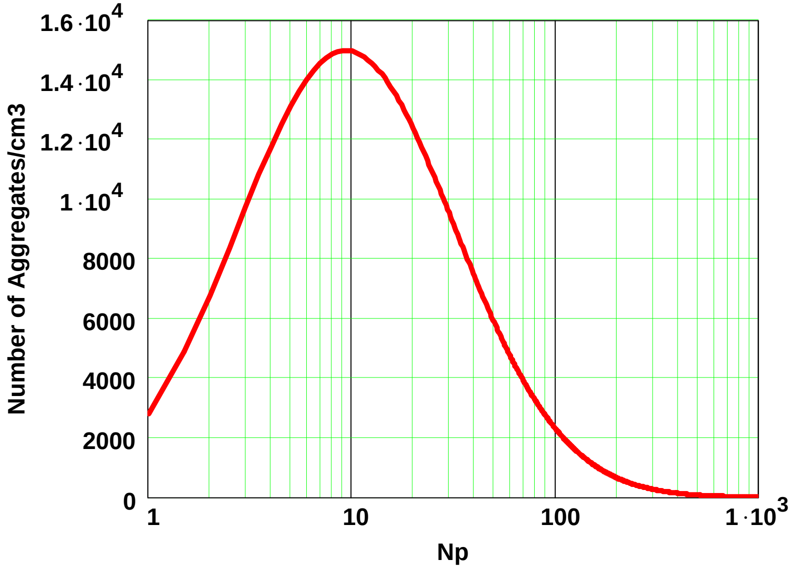
Apply LII/ELS to a DISI Vehicle

- results from direct injection spark-ignition (DISI) vehicle
  - —“steady-state”
  - —3 hp load
  - —1700 rpm
  - —gear selection held in D2
- experiment identical to AC-LII, except a third detector is added, using the same collection optics as for LII

Combined L I I and ELS





- result of LII and ELS:
  - SVF = 0.72 ppb
  - d_p = 25.7 nm
  - N_g = 42
  - N_{ag} = 9.1 x 10^5/cm^3

- improved description of soot morphology, and improved measure of primary particle diameter and surface area

Comparison of LII-ELS to SMPS

- 98 SMPS experiments, acquired in 11 sets on 9 different days over a one-month period
- variation in SMPS from experiment to experiment had a standard deviation of ~35% within a specific set, and a standard deviation of ~25% from the mean of one set to another
- calculate mobility diameter equivalent for LII-ELS results using:

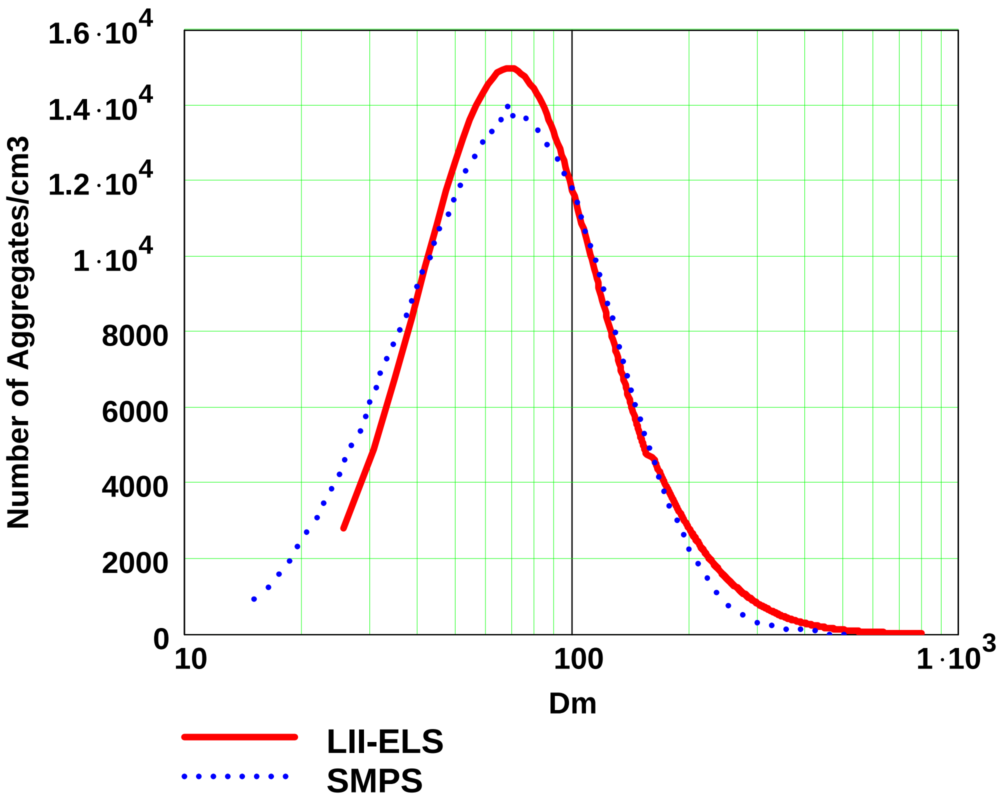
$$D_m = d_p N_p^{0.44}; \qquad N_p \leq 60$$

$$D_m = 0.7 d_p \left(\frac{N_p}{k_0} \right)^{\frac{1}{d_f}}; \qquad N_p > 60$$

SAE 2002-01-2715
(from Sorensen et al.)

Comparison of LII-ELS to SMPS

- assume σ_{2g} = 3.4
- reasonable agreement for shape of distribution
- however, SMPS is *not* corrected for dilution

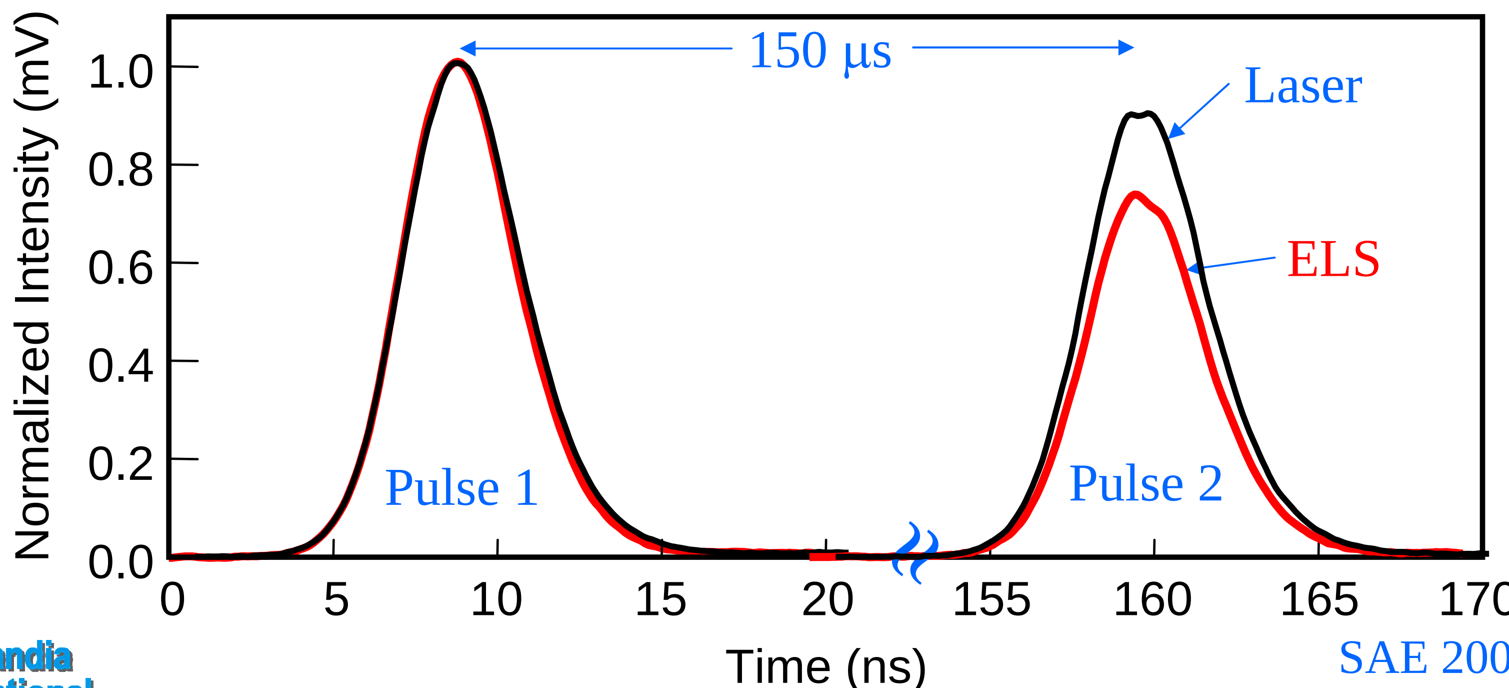


LIDELS

Laser-induced desorption (LID) with elastic laser scattering (ELS)

Double-pulse laser, measure ELS

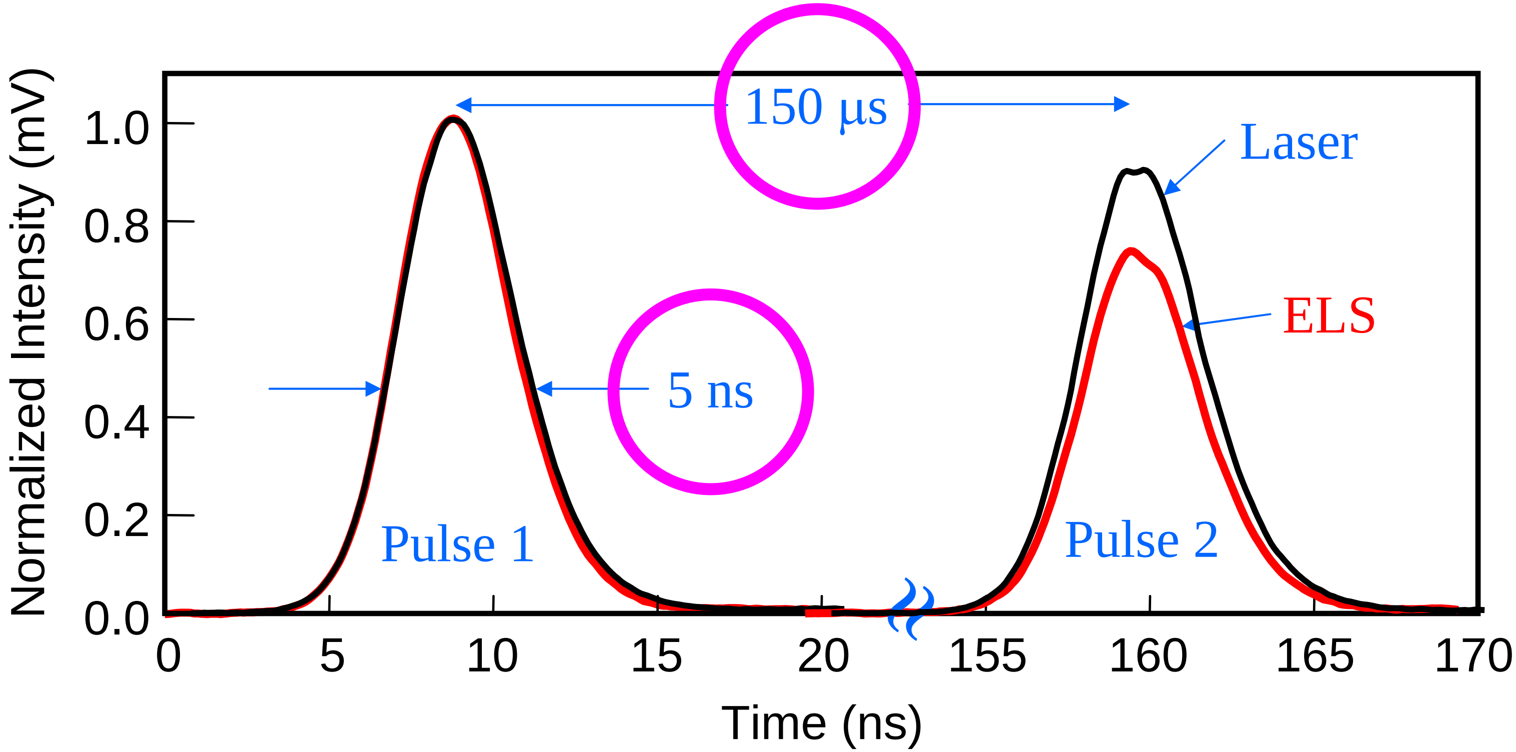
$$\left[\frac{A_{ELS-2}/A_{YAG-2}}{A_{ELS-1}/A_{YAG-1}}\right]^{0.5} = \text{solid volume fraction}$$



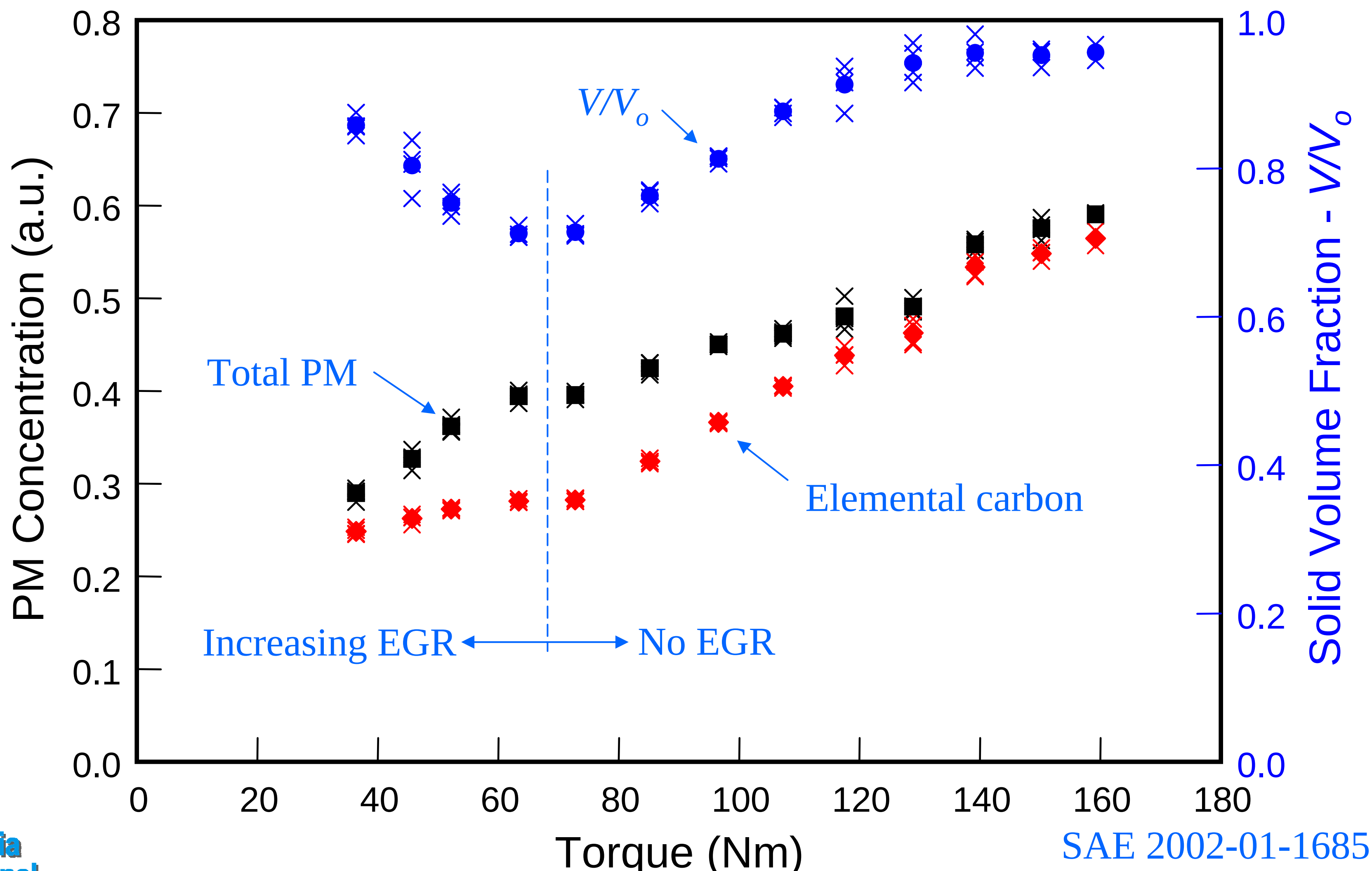
SAE 2002-01-1685

Simultaneous LIDELS Not Possible With One Laser

Data acquisition is by fast digital oscilloscope
150 μs at 5 giga samples/s = 750,000 points
∴ Sequential measurements are the only option



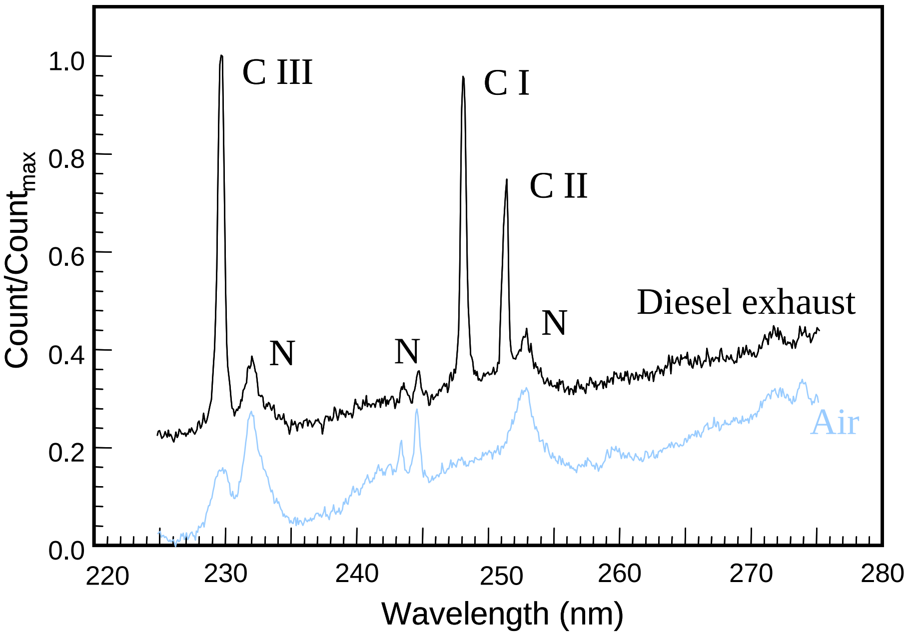
Sequential LIDELS For Load Sweep



SAE 2002-01-1685

Laser-Induced Breakdown Spectroscopy (LIBS)

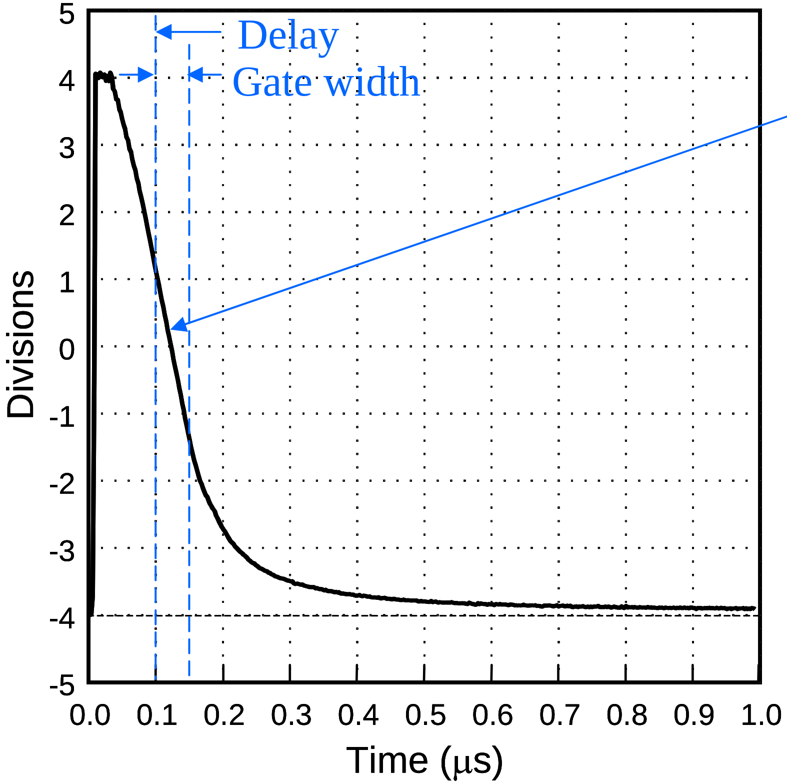
- focused laser beam creates a plasma that emits light characteristic of neutral and ionized atoms
- spectrometer output is imaged onto an intensified CCD camera



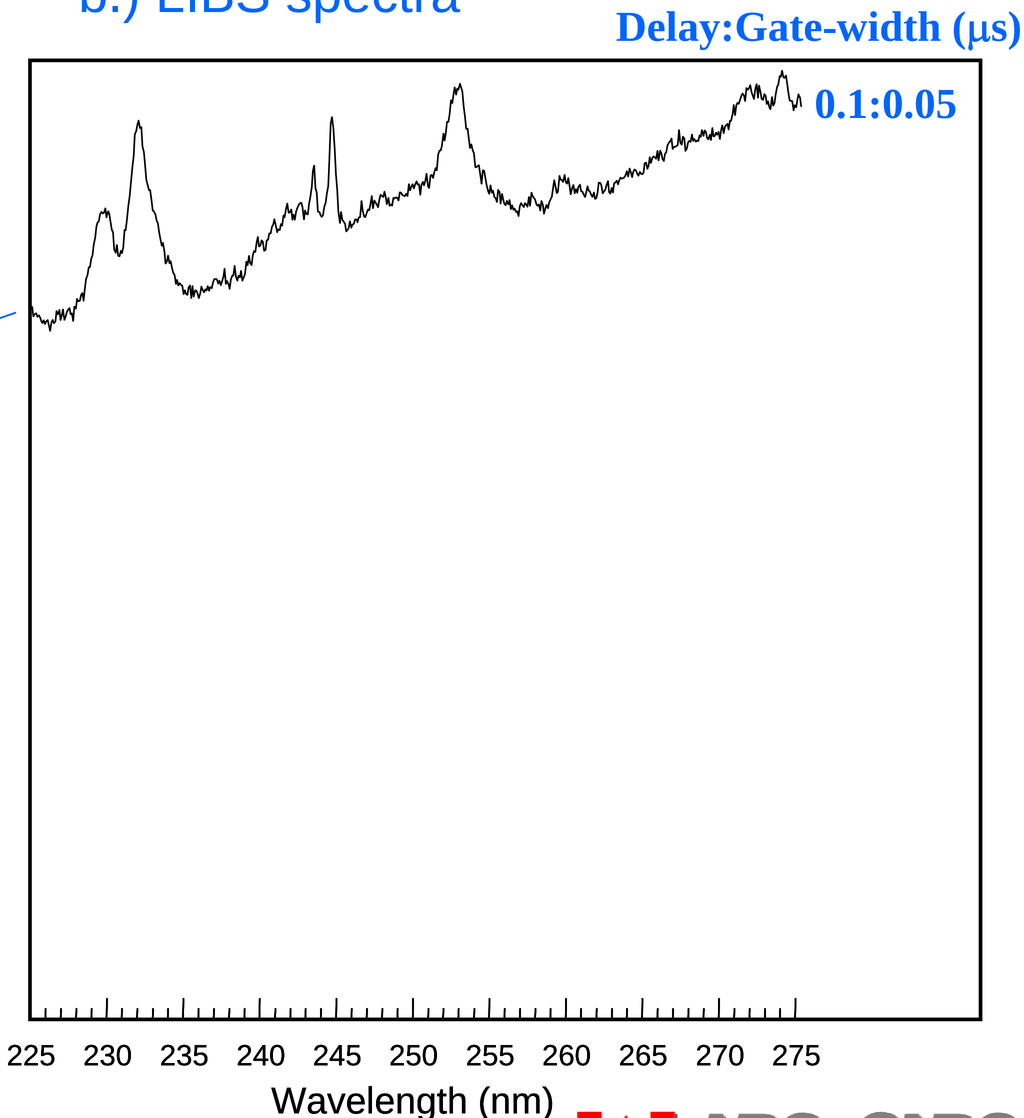
- line position provides species identification
- line intensity provides species concentration
- components from oil additives, engine wear, and corrosion of the exhaust system

Time-Resolved LIBS (TRELIBS) of Air

a.) Broadband emission from laser-induced spark



b.) LIBS spectra



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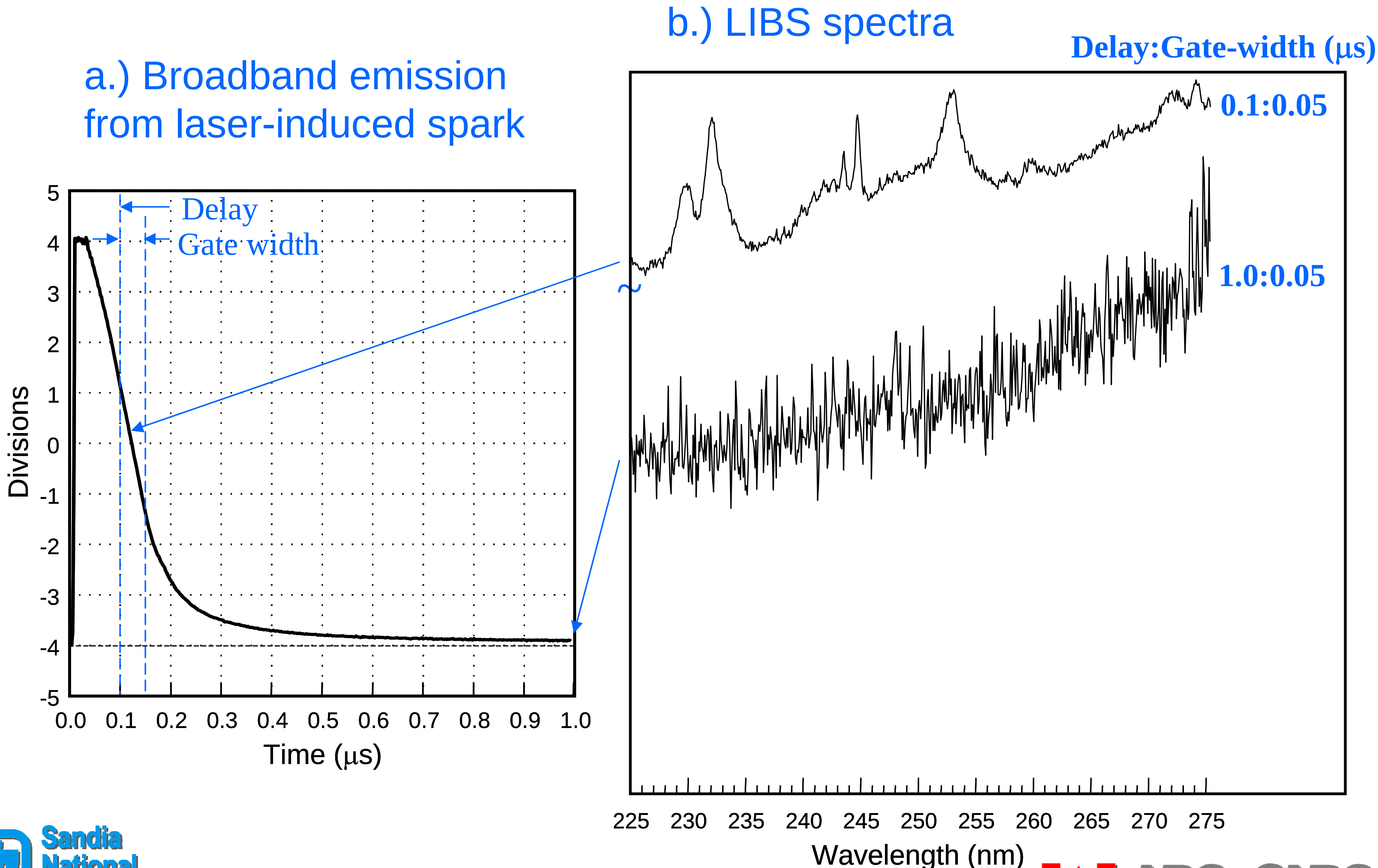

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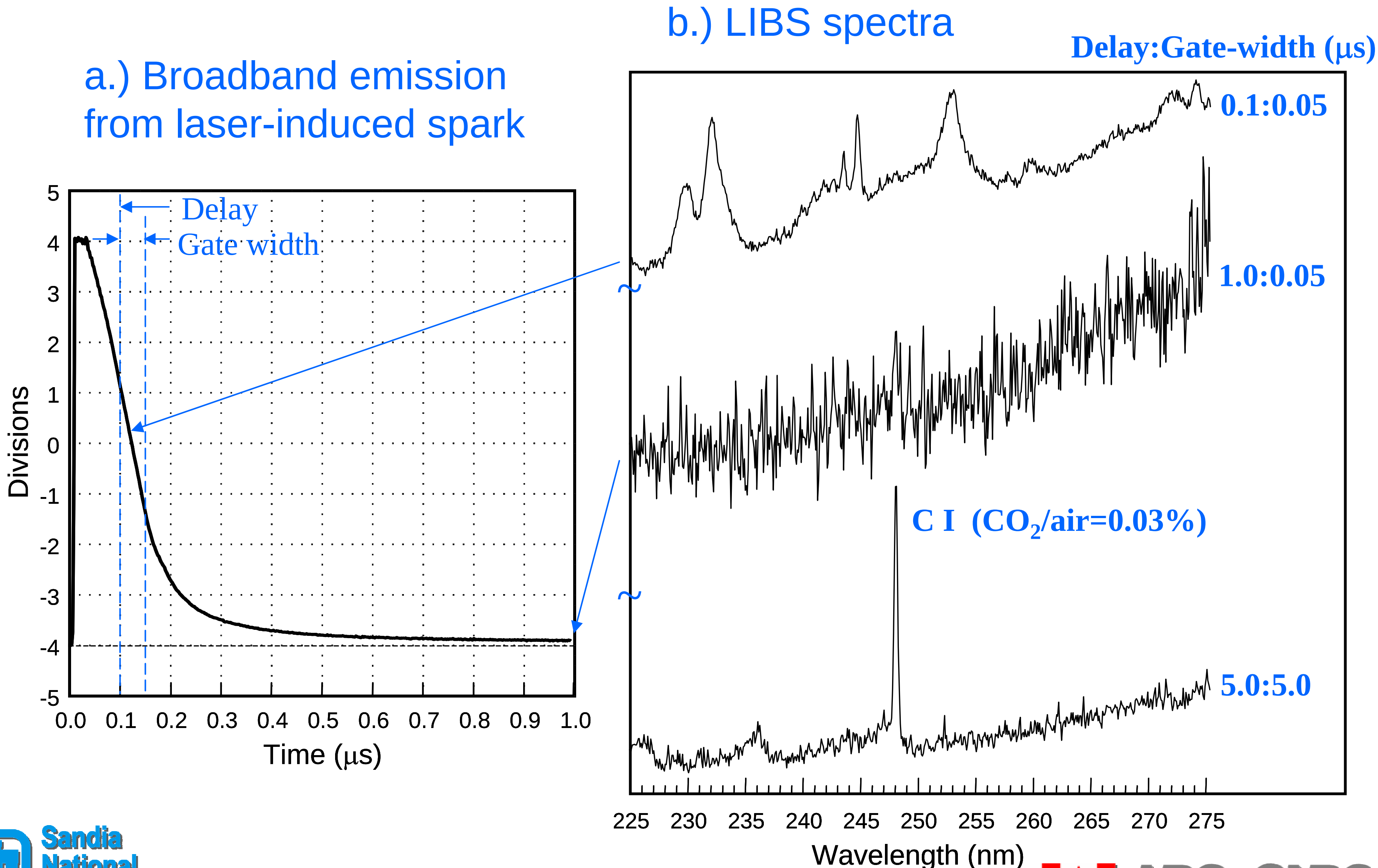
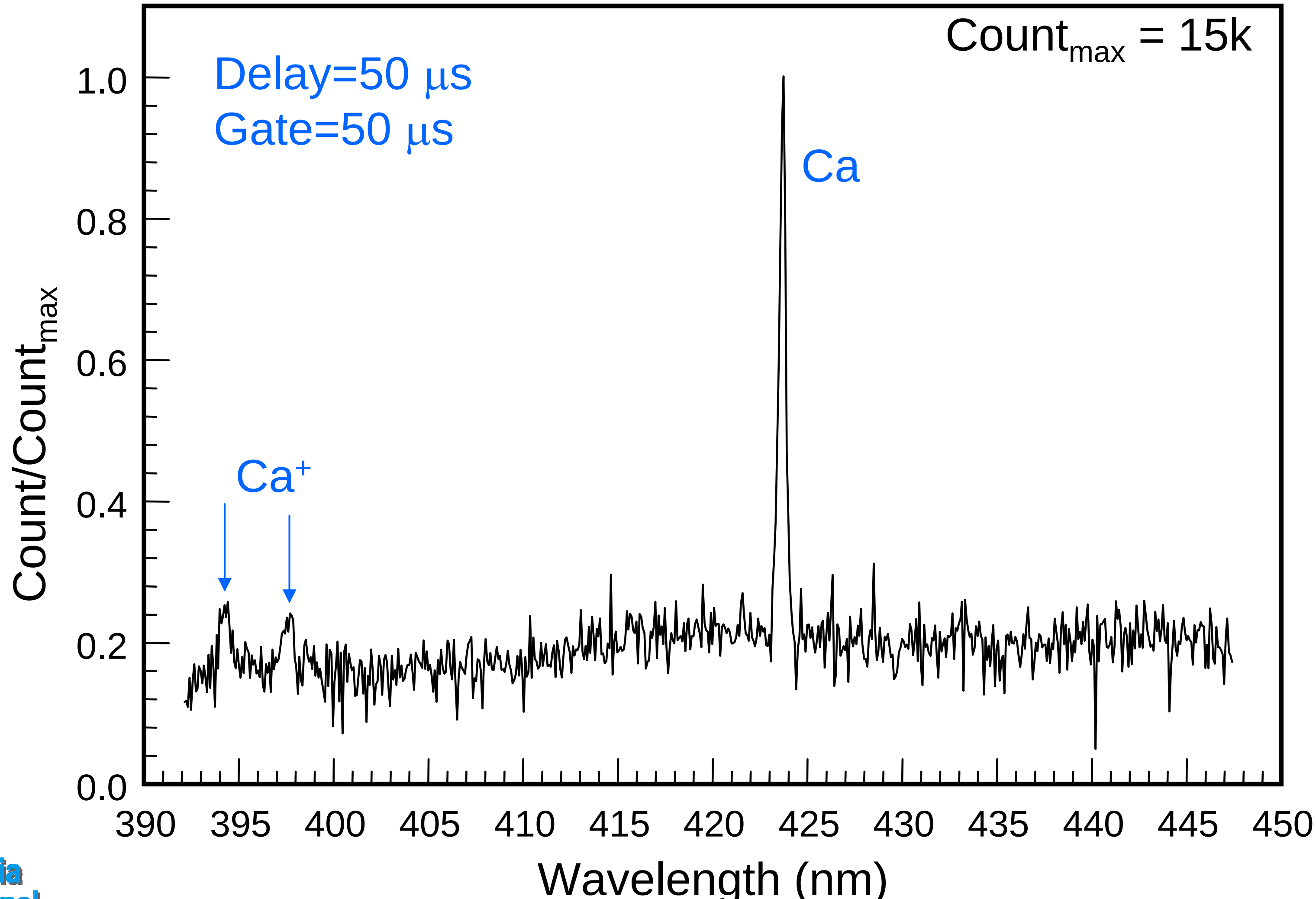
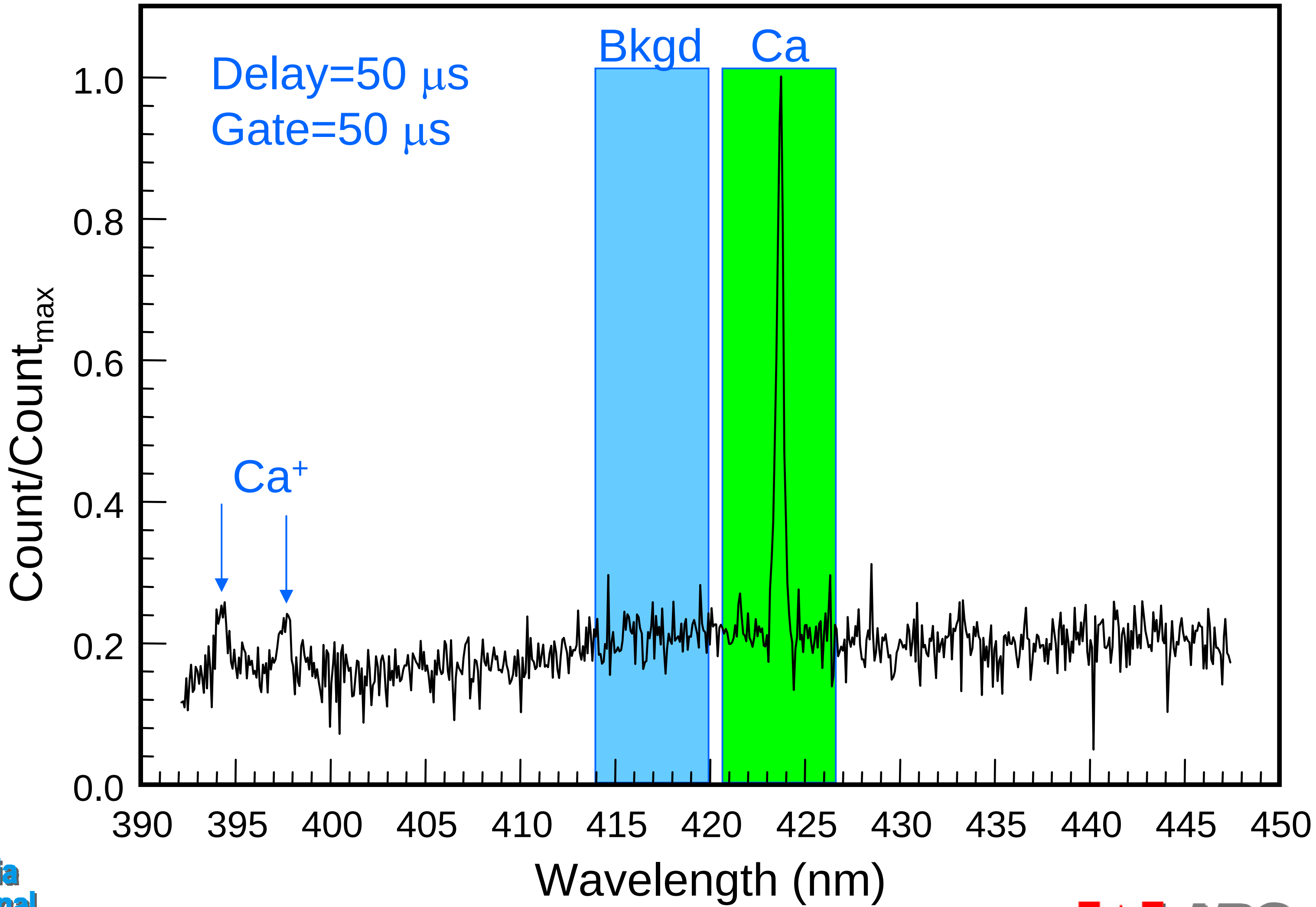


TRELIBS of Ca from Ash in Diesel Exhaust



Real-Time LIBS via Bandpass Filters



Current Status of HELD

- turn-key commercial AC-LII instruments are in use
- anticipate interest in LII/ELS using Rayleigh-Debye-Gans polydisperse fractal aggregate theory (RDG/PFA) for aggregate morphology
- LIDELS needs further evaluation and substantial development to prove viability as a quantitative technique, but there is strong industry interest
- LIBS has yet to generate much interest (and would require substantial development)

Acknowledgements

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... and don't forget to visit Poster 29 for examples of AC-LII applications