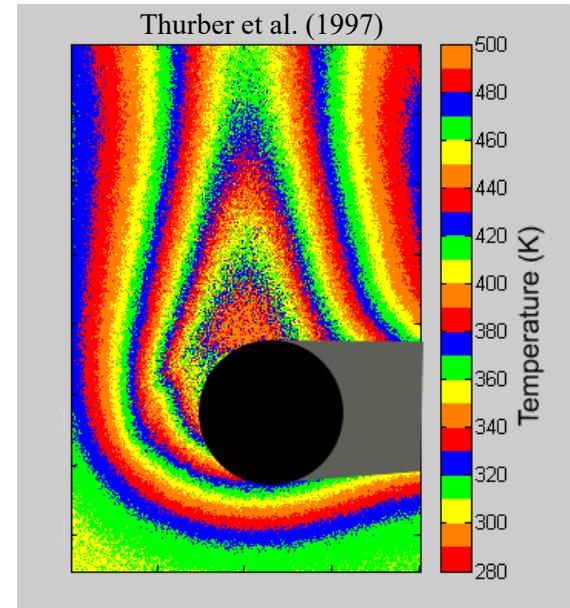


# Quantitative Laser Diagnostics for Combustion Chemistry and Propulsion

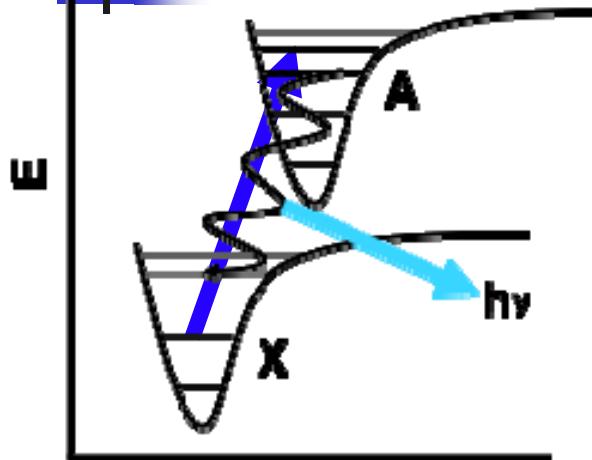
## Lecture 15: Application Examples of LIF, PLIF Large Molecules (Tracers)

1. Introduction to flow tracer PLIF
2. Acetone PLIF to image fuel mixing
3. 3-pentanone PLIF as a flow tracer
4. 3-pentanone PLIF in IC-engines
5. CW PLIF for high-frame-rate imaging
6. Toluene PLIF for temperature (1 camera)
7. Toluene PLIF for temperature (2 camera)
8. The future



Temperature image; single-shot PLIF of acetone in flow around heated rod

# 1. Introduction to Flow Tracer PLIF



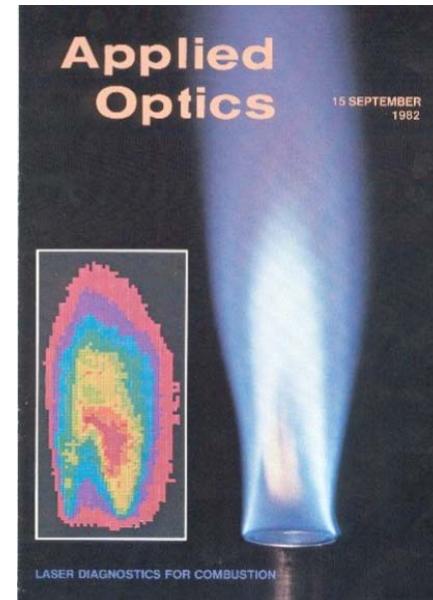
## Planar Laser-Induced Fluorescence

$$S_f = \frac{E}{h\nu} n \sigma(\lambda, T) \phi(\lambda, T, p_i) \eta$$

Diagram illustrating the Planar Laser-Induced Fluorescence (PLIF) signal equation:

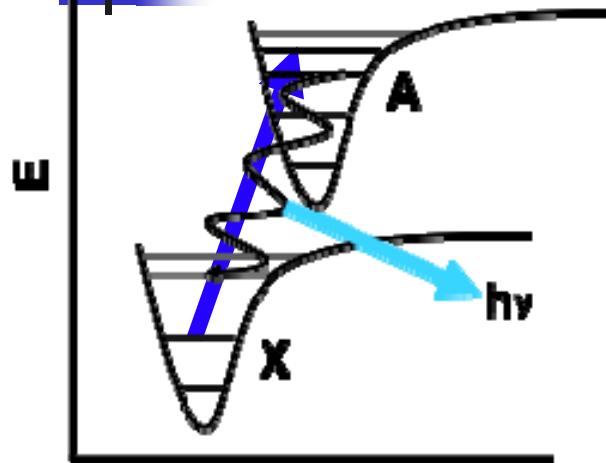
- LIF Signal**: The output signal  $S_f$ .
- Incident Photons**: Represented by an orange arrow pointing to the absorption term.
- Photons Absorbed**: Represented by a red arrow pointing to the absorption term.
- Fluorescence Quantum Yield**: Represented by a green arrow pointing to the quantum yield term.
- Collection Efficiency**: Represented by a purple arrow pointing to the collection efficiency term.

- History of PLIF begins at Stanford  
1982 - Kychakoff, Howe, Hanson,  
and McDaniel using OH
- Now we examine PLIF from tracers,  
mostly ketones, to control  $\phi$



# 1. Introduction to Flow Tracer PLIF

## LIF of tracer molecules added to flow

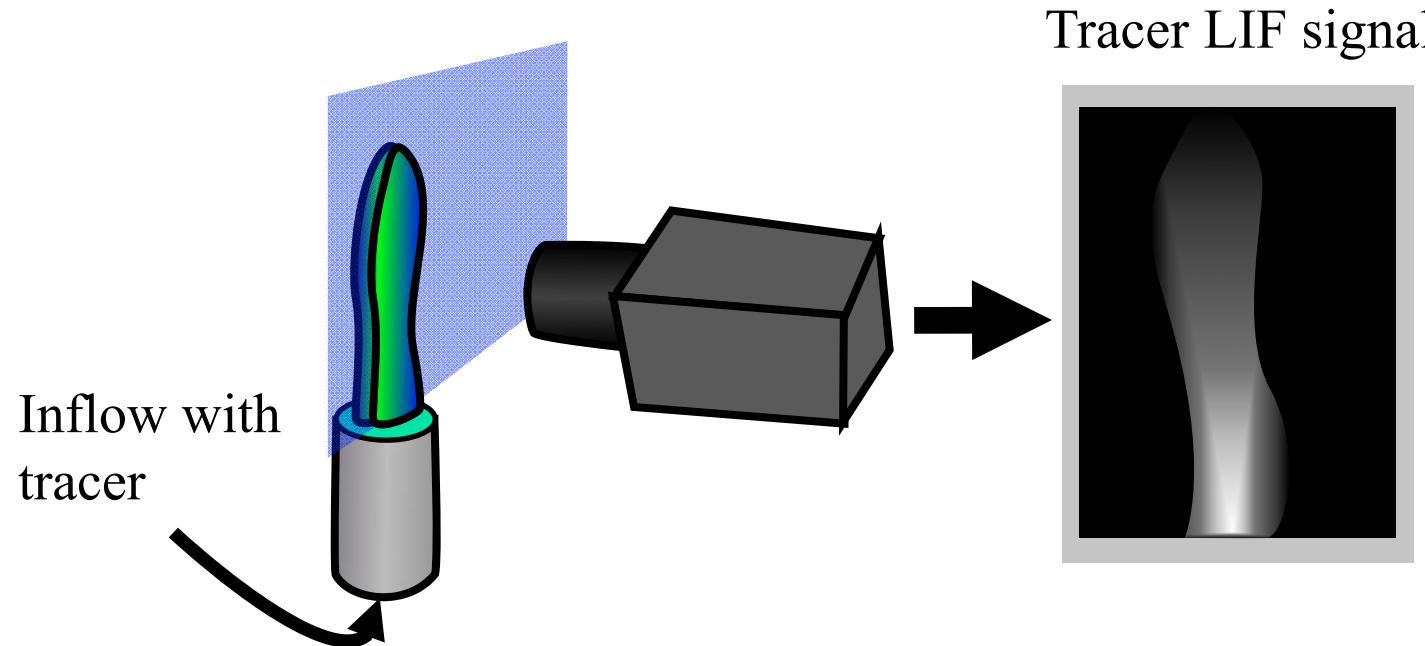


### Planar Laser-Induced Fluorescence

$$S_f = \frac{E}{h\nu} n \sigma(\lambda, T) \phi(\lambda, T, p_i) \eta$$

Diagram illustrating the Planar Laser-Induced Fluorescence (PLIF) signal generation process:

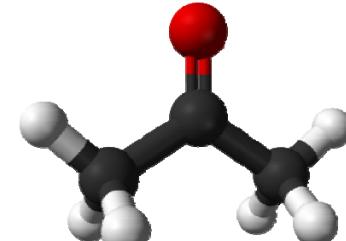
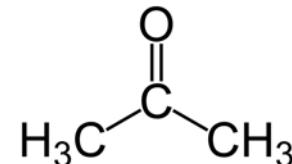
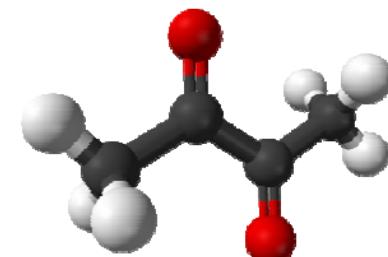
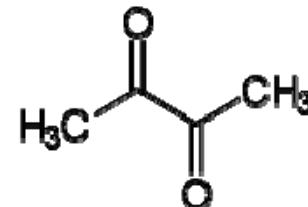
- LIF Signal**: The final output of the fluorescence signal.
- Incident Photons**: Represented by an orange arrow pointing to the absorption term.
- Photons Absorbed**: Represented by a red arrow pointing to the absorption term.
- Fluorescence Quantum Yield**: Represented by a green arrow pointing to the quantum yield term.
- Collection Efficiency**: Represented by a purple arrow pointing to the collection efficiency term.



# 1. Introduction to Flow Tracer PLIF

## Biacetyl and acetone first tracers used

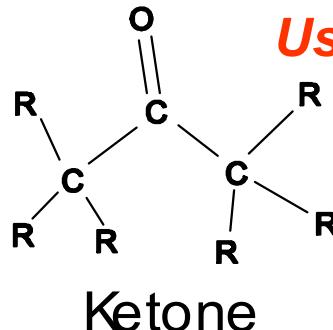
- Background: Addition of a fluorescent tracer enabling LIF and PLIF in air (neither O<sub>2</sub> nor N<sub>2</sub> are readily accessible)
  - 1980's at Stanford HTGL  
Biacetyl ((CH<sub>3</sub>CO)<sub>2</sub>), but had two problems:
    - 1) smelly and hard to handle
    - 2) highly quenched by O<sub>2</sub>
  - Early 1990's by Lozano at Stanford  
Acetone (CH<sub>3</sub>COCH<sub>3</sub>), has become a popular laser diagnostic in laboratories worldwide



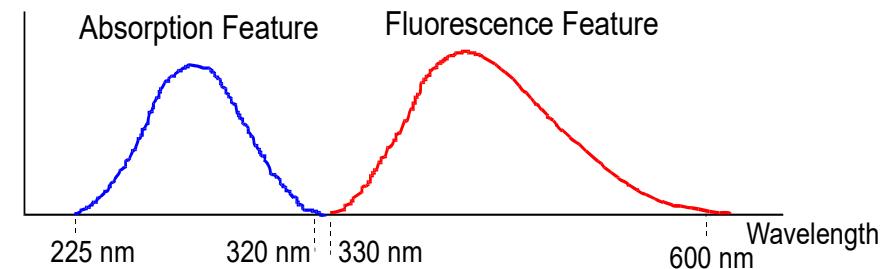
# 1. Introduction to Flow Tracer PLIF

## Ketones are good flow tracers

- How can PLIF be used to study mixing?

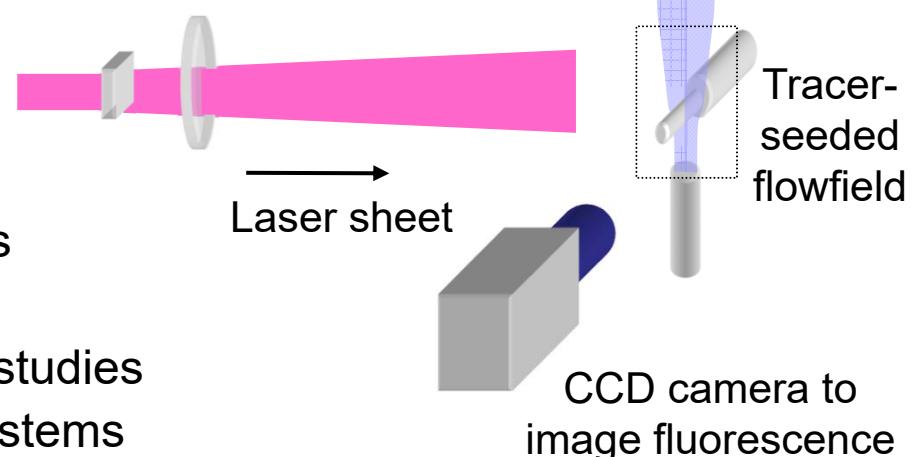


**Use Ketones as Tracers: Acetone and 3-Pentanone**



### ■ Advantages of Ketones as Tracers

- + Strong signals
  - Accessible absorption feature
  - Non-resonant fluorescence
- + Resistant to bath gas effects
- + Similar to common hydrocarbon fuels



### ■ Applications

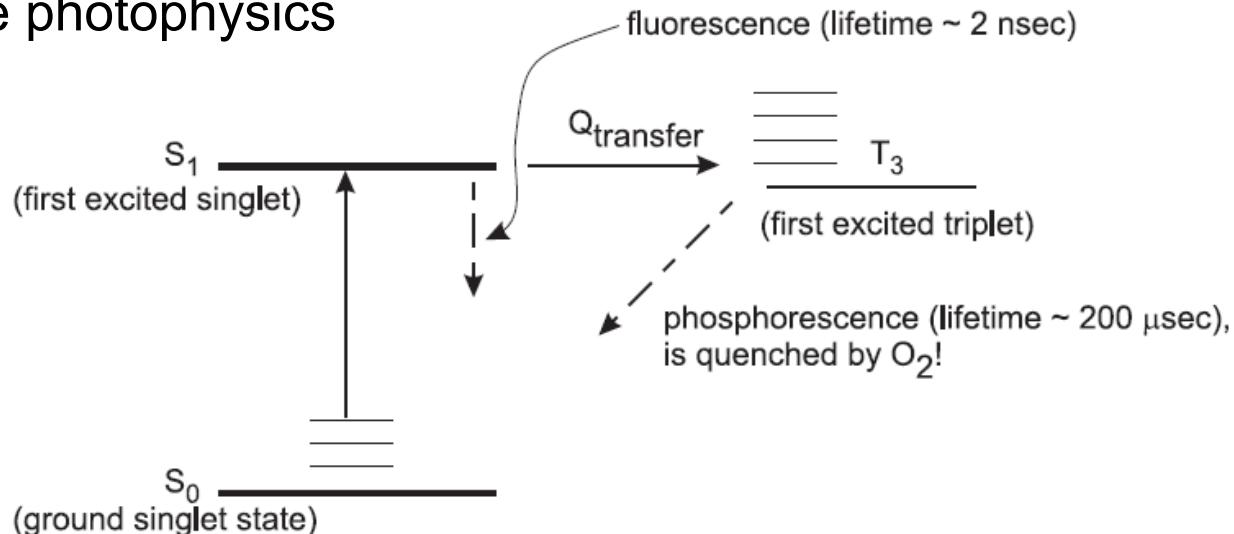
- Fundamental fluid/heat transfer studies
- Fuel/air mixing in combustion systems

→ *Application is straightforward in isothermal, isobaric flows , i.e., Signal is proportional to mixture fraction of tracer*

# 1. Introduction to Flow Tracer PLIF

## A brief overview of acetone LIF

- Acetone photophysics

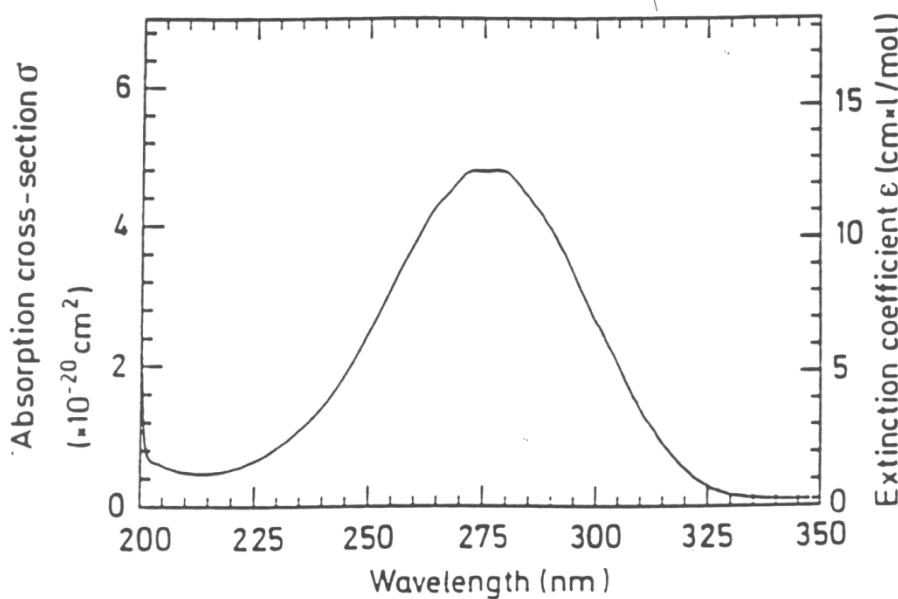


- Vapor pressure is 180Torr at room temperature
- Cheap, non-toxic, easy to handle
- Fluorescence lifetime is approximately 2ns and is independent of O<sub>2</sub>
- Constant fluorescence yield of about 0.2% at room temperature
- Each state is a manifold of vibrational levels
- “Intramolecular intersystem crossing” at rate Q<sub>t</sub> dominates, therefore the FY =  $A_{10}/(A_{10} + Q_t) \approx A_{10}/Q_t \approx 0.2\%$  and SF  $\propto n_{\text{acetone}} I_v$  (or E)!
- Absorption and fluorescence are broadband

## 2. Acetone PLIF to Image Fuel Mixing

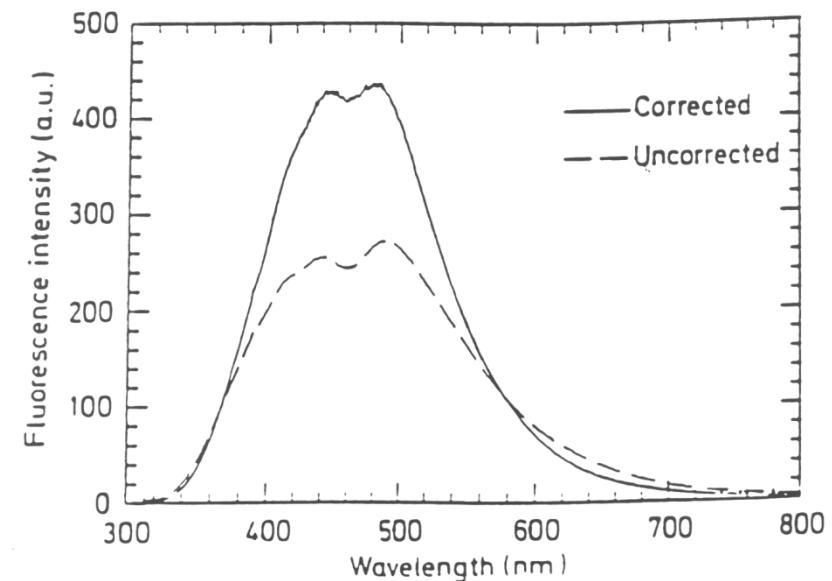
### Absorption and fluorescence wavelengths

- Acetone: A tracer for concentration measurements in gaseous flows by planar LIF (Lozano, Yip & Hanson, 1992)



Acetone absorption spectrum corresponding to excitation from the ground state to the first excited singlet

- Reveals range of suitable excitation wavelengths



Acetone fluorescence spectrum when excited at 308nm: the dashed line corresponds to the detected signal; the solid line is the same curve corrected for the detection system responsivity

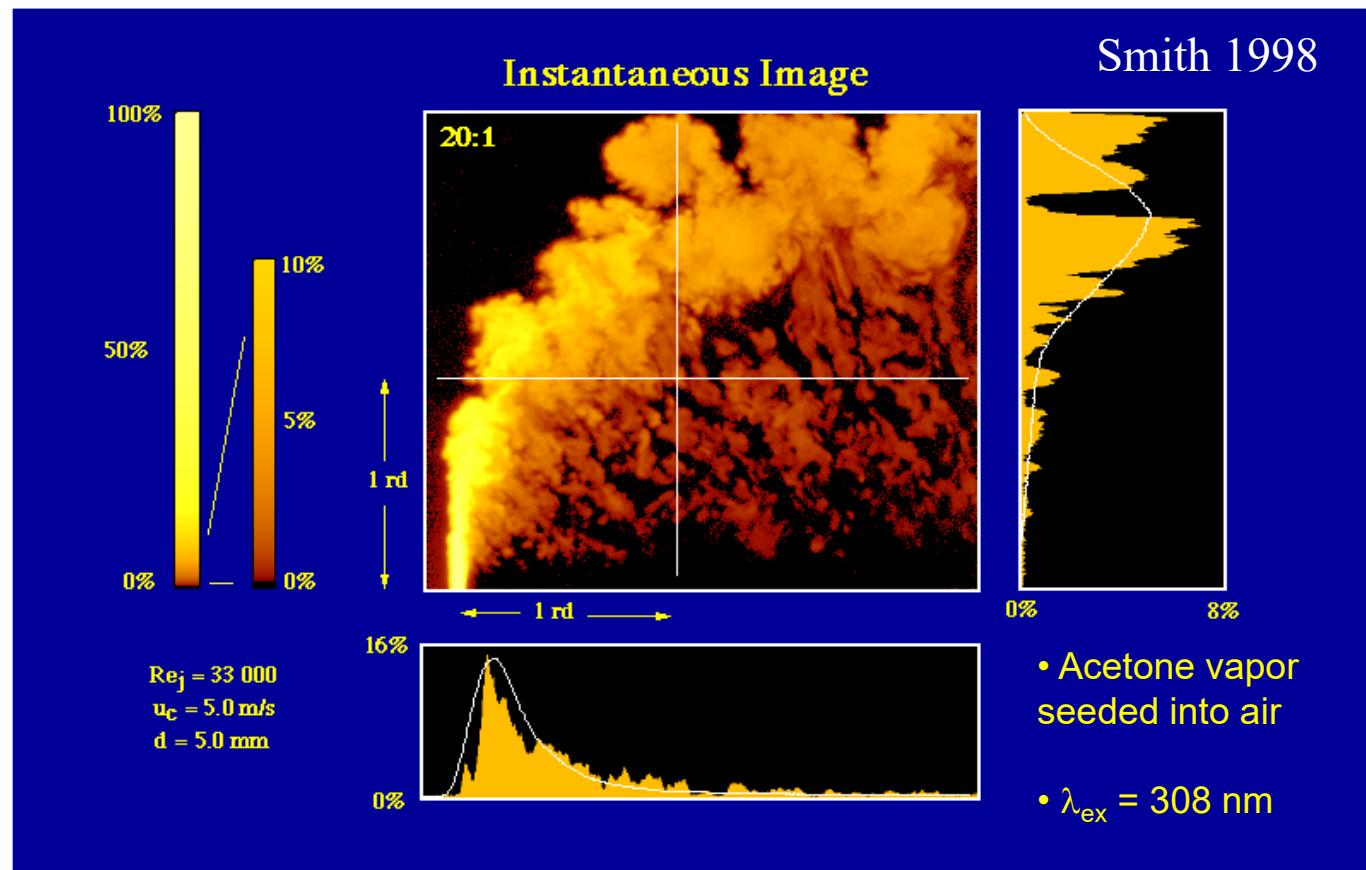
- Reveals separation of emission wavelength from excitation

## 2. Acetone PLIF to Image Fuel Mixing

- Example of acetone PLIF to investigate jet in crossflow: isothermal, isobaric mixing study

Side-view

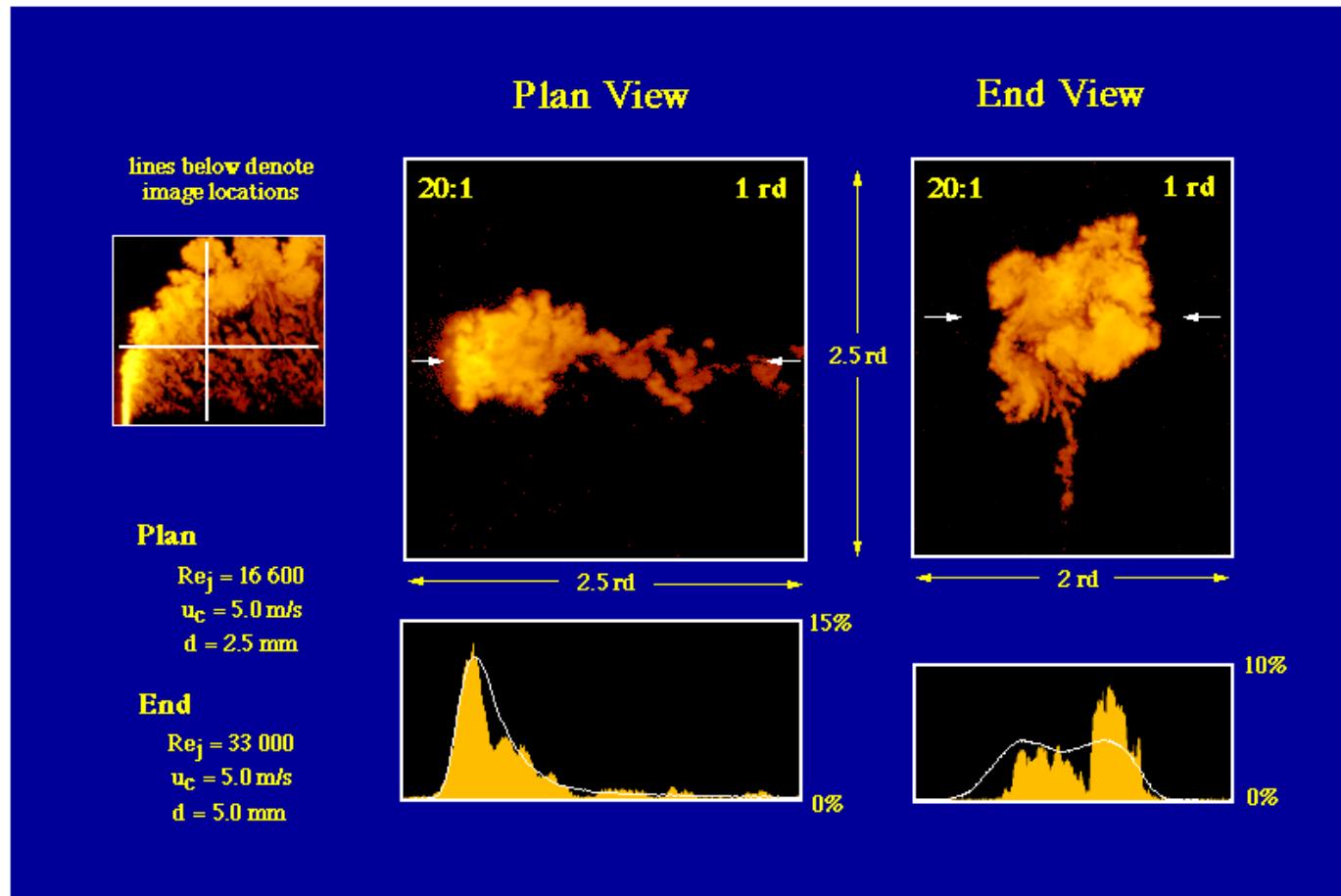
$$\begin{aligned}U_{\text{crossflow}} &= 5 \text{ m/s} \\d_{\text{jet}} &= 5 \text{ mm} \\U_{\text{jet}}/U_{\text{crossflow}} &= 20/1 = r \\1 \text{ rd} &= 10 \text{ cm}\end{aligned}$$



- For isothermal, isobaric case, fluorescence signal is proportional to tracer mole fraction

## 2. Acetone PLIF to Image Fuel Mixing

Example of acetone PLIF to investigate jet in crossflow:  
isothermal, isobaric mixing study



- Detail provided by PLIF provides important tests of model predictions

### 3. 3-pentanone PLIF as a Flow Tracer

#### Photophysics Database and Modeling: Fundamentals

- PLIF with ketone/aromatic tracers widely used, but not quantitative at high P & T

$$S_f = \frac{E/h\nu}{\text{signal}} \times \frac{L}{\text{number of photons}} \times \frac{n_{ab}}{\text{path length}} \times \underbrace{\sigma(\lambda, T) \times \phi(\lambda, T, P)}_{\text{Photophysical parameters}} \times \eta\Omega/4\pi$$

absorption cross-section      fluorescence yield (FQY)

- Measurements and modeling needed for  $s(l, T)$ ,  $f(l, T, P)$  at high P & T

$$\phi = \sum_{n=0}^{\infty} \frac{k_f}{k_f + k_{vib} + k_{nr}(E_n) + k_{O2}(E_n)} p(E_n)$$

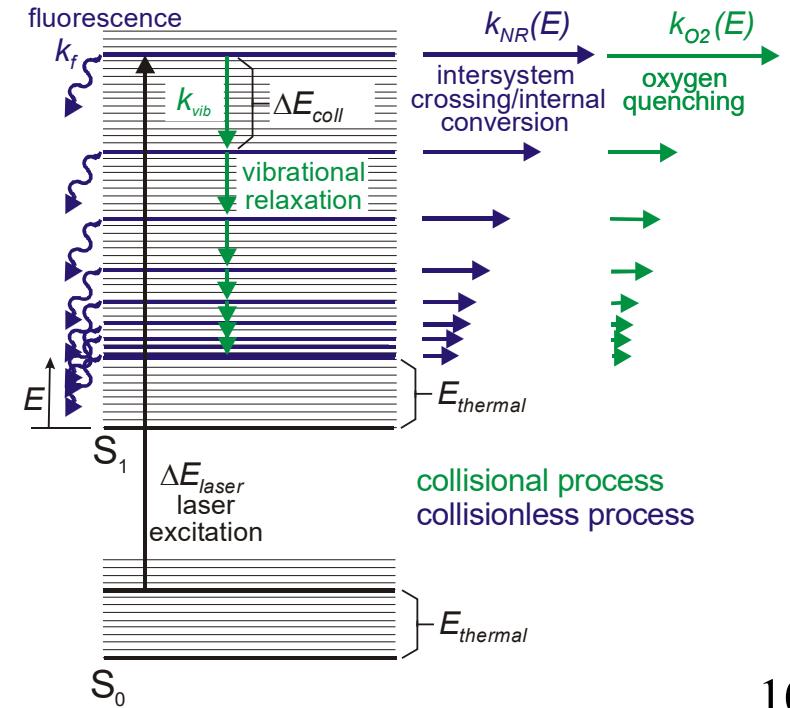
$k_{vib}$  = collisional rate ( $s^{-1}$ )

n = n<sup>th</sup> collisional time step

Energy at n<sup>th</sup> step:  $E_{n+1} = E_n - \Delta E_{coll} = E_n - \alpha(T)[E - E_{thermal}]$

- Model has four parameters,  $\alpha$ ,  $k_{nr}$ ,  $k_{O2}$ ,  $k_f$

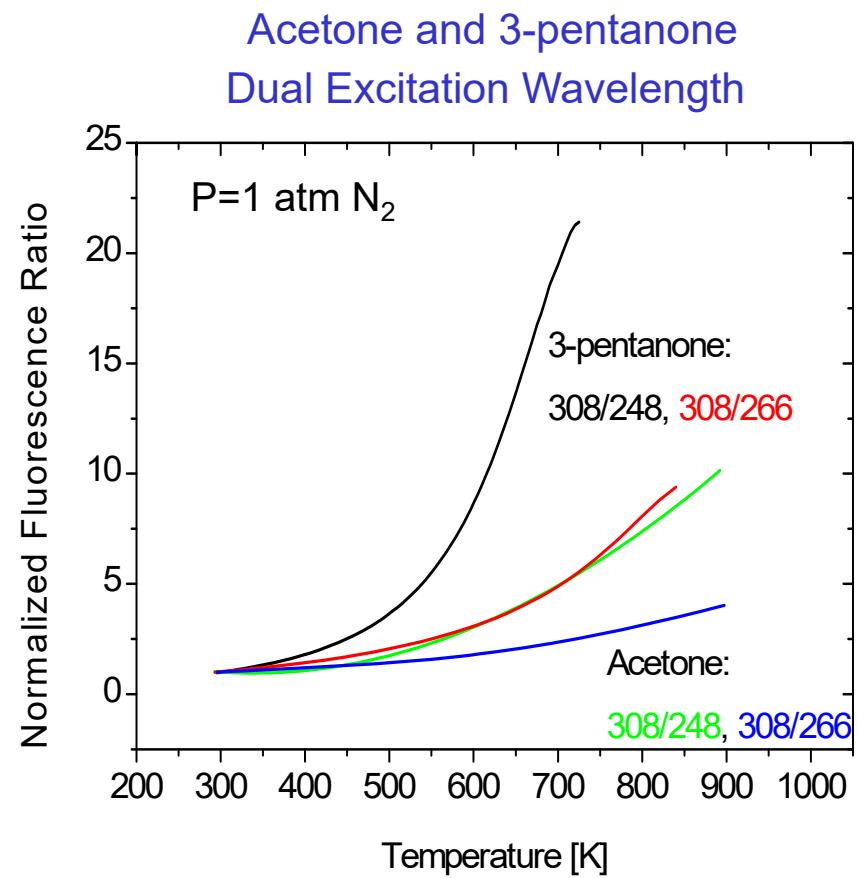
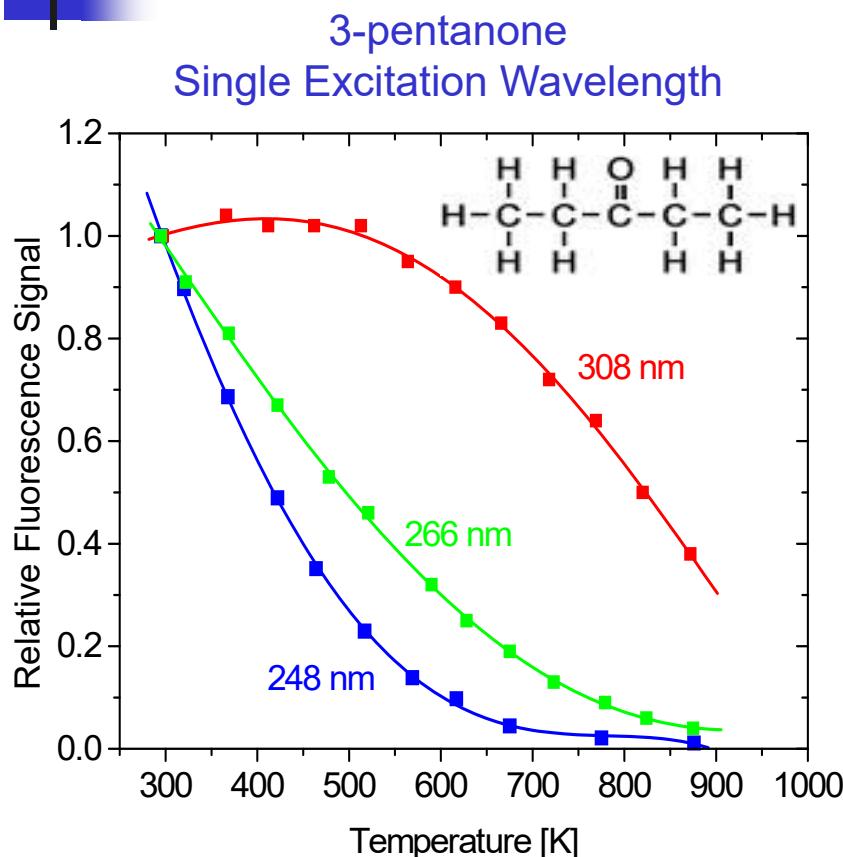
- $k_{nr}$  &  $k_{O2}$  measurable from lifetime data
  - $\alpha$ ,  $k_f$  from f data over range of P,T



### 3. 3-pentanone PLIF as a Flow Tracer

3-pentanone: Better match to physical properties of fuels

Photophysical behavior qualitatively similar to acetone but quantitatively different

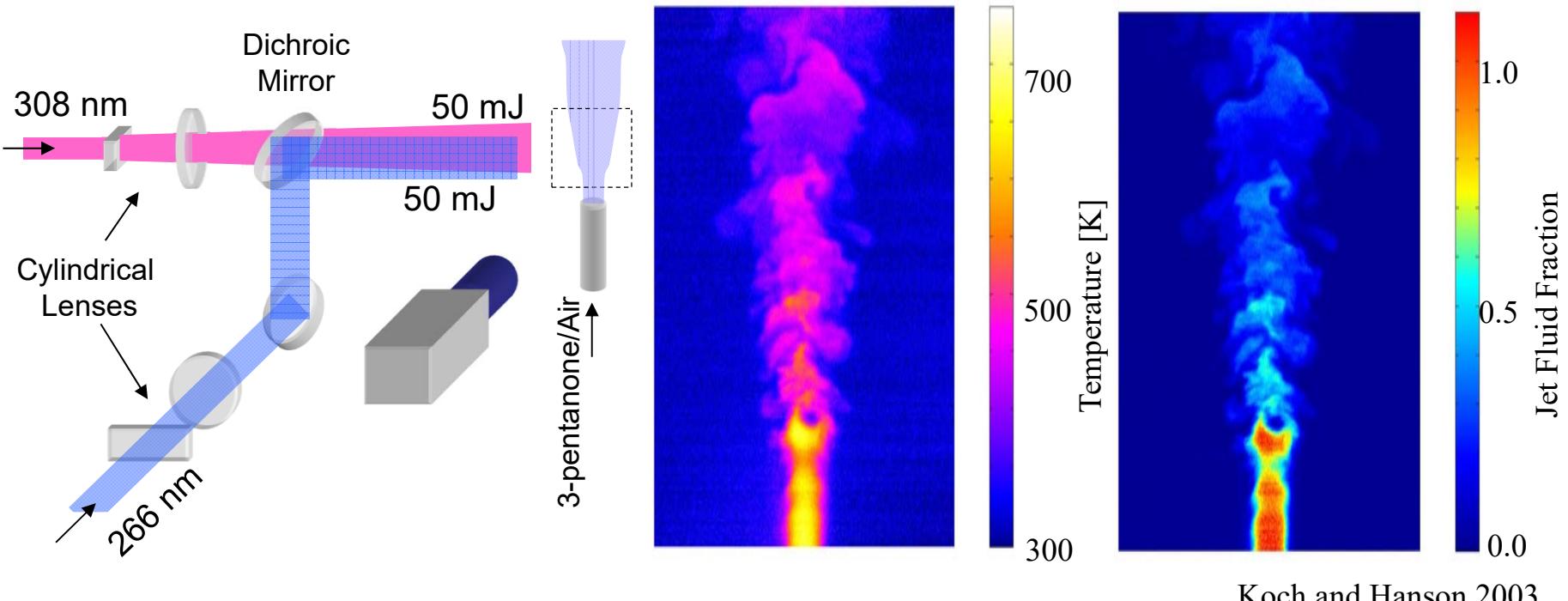


- 308 nm, T< 550K - ideal regime for straightforward concentration imaging
- 248 nm highly T-sensitive – good for temperature imaging

- Long/short wavelength ratios are most temperature sensitive
- 3-pentanone is more T-sensitive than acetone

### 3. 3-pentanone PLIF as a Flow Tracer Simultaneous Imaging of $\chi_i$ , $T$

- Strong T dependencies of 3-pentanone enable quantitative imaging



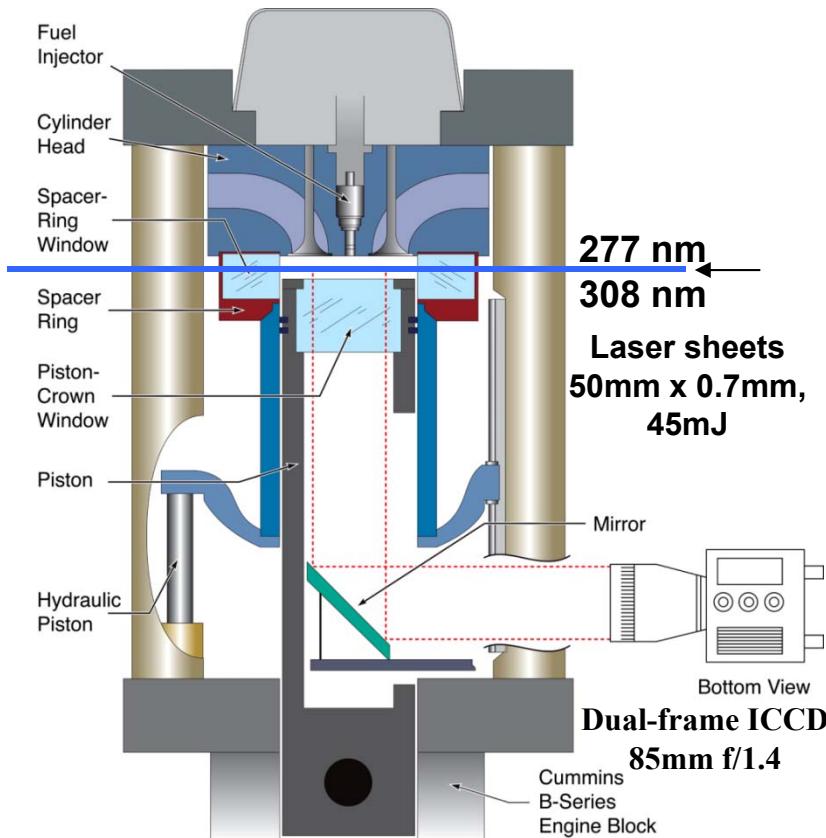
Koch and Hanson 2003

- Laser excitation at 308 and 266 nm
- Frame transfer CCD camera acquires 2 images within 2  $\mu$ s
- Instantaneous  $T$ ,  $\chi_i$  images from a heated 3-p/air jet after 308/266 nm excitation
- Flowfield contains 1-4% 3-pentanone (2.9% is stoichiometric)

# 4. 3-pentanone PLIF in IC-Engines

## Homogeneous Charge Compression Ignition (HCCI)

- Natural thermal stratification (TS) important for HCCI engine ignition
- Increase of TS could extend high-load operation
- PLIF of temperature and residuals an important tool to study TS



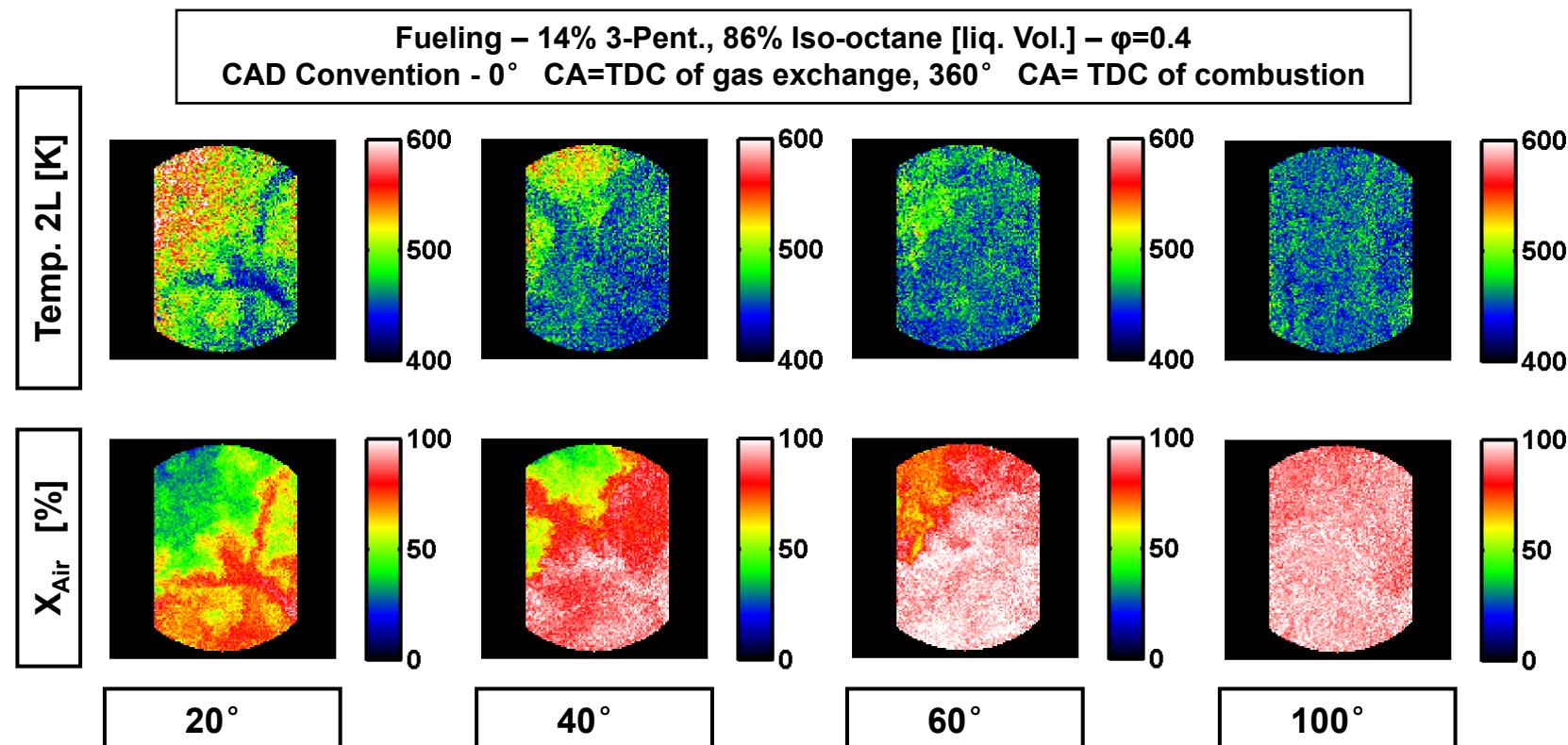
HCCI Engine Combustion Fundamental  
Laboratory, Sandia National Labs

- Tracer-based PLIF strategies used
  - Two-line (277nm/308nm ratio) – simultaneous T and  $X_i$
  - Single-line (277nm) – T (homogeneous composition)
  - Tracer selection: 3-Pentanone – good sensitivity, minimal effect of oxygen quenching
- Investigate the thermal stratification in HCCI engine: motored & fired operation
  - Early residual mixing
  - Thermal stratification evolution during compression

## 4. 3-pentanone PLIF in IC-Engines

Mixing of Residuals with Fresh Intake Can Produce Stratification

- Incomplete mixing of hot retained residuals (internal EGR) with fresh intake can result in TS
- Two-line PLIF of  $T$  and  $X_{\text{air}}$  to track evolution of intake-air/residual gas mixing

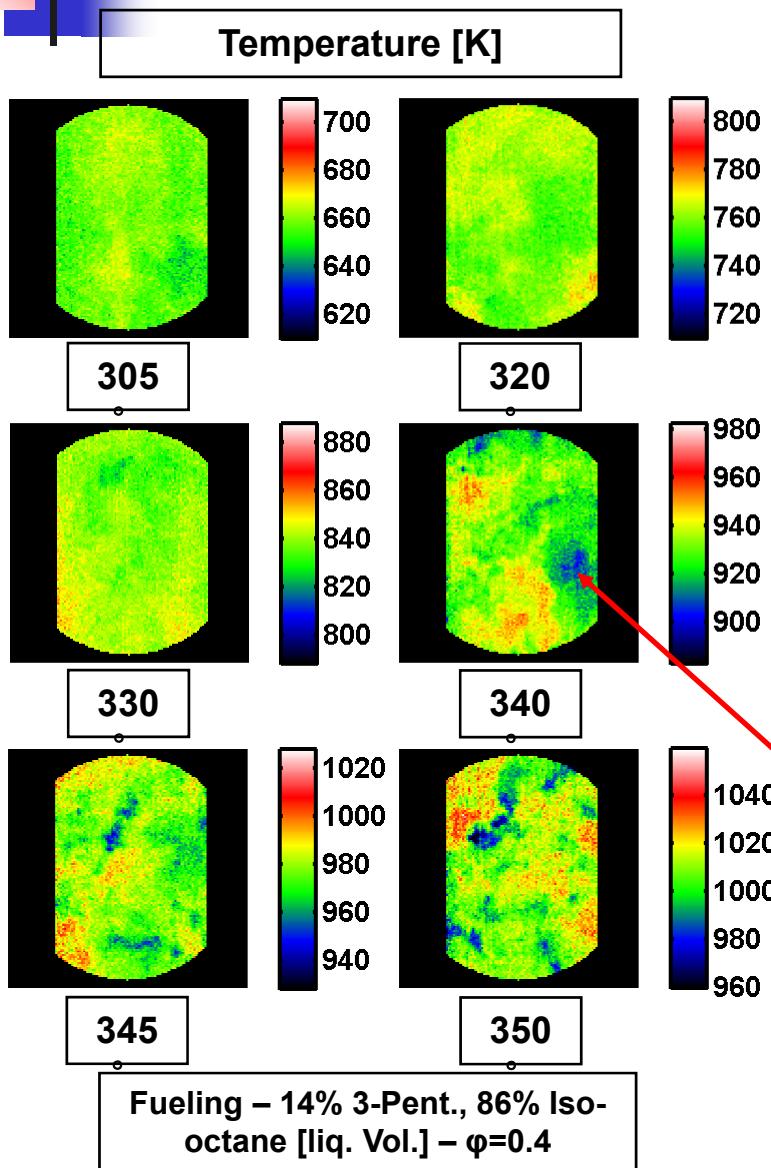


- Mixing complete after 100° of CA (bottom of stroke at 180°)
- Mixing of EGR and fresh intake does not increase the thermal stratification for these conditions

Snyder et al. 2011

## 4. 3-pentanone PLIF in IC-Engines

One-line PLIF in Engines:  
Natural Thermal Stratification (TS) Increase Near TDC



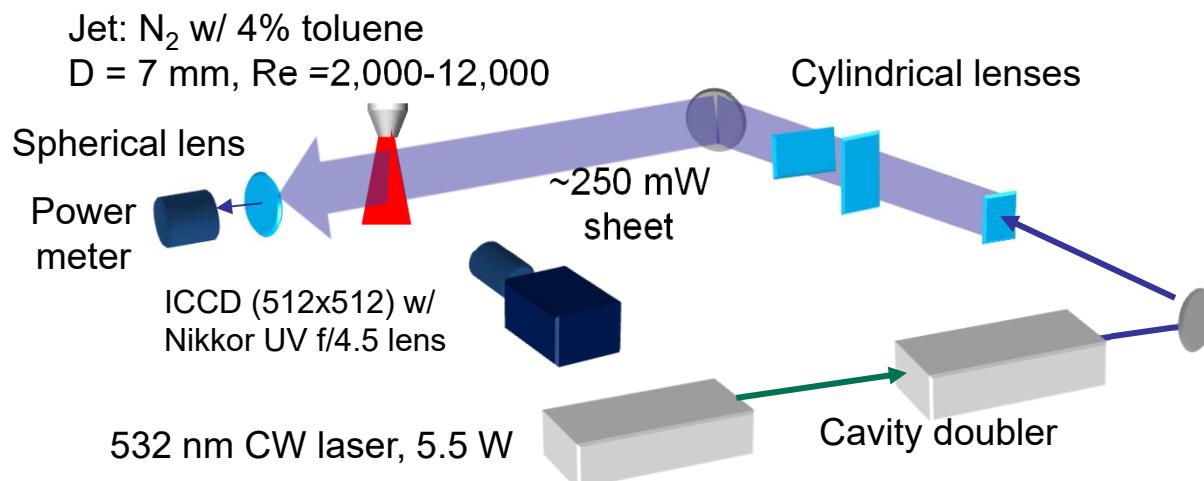
- Single-line (277nm) PLIF of temperature to monitor TS evolution during compression
- High sensitivity of measurement (4-5K) allows detection of small variations – important for HCCI
- Initial TS is small (305°CA) but drastically increases throughout remaining compression
  - TS near TDC results in sequential auto-ignition that reduces the rate of heat release – important for extension to high-load operation

Heat transfer with wall surfaces in boundary layer leads to pockets of cooler gas that convect into core cylinder region and affect auto-ignition

# 5. CW PLIF for High-Frame-Rate Imaging

New Diagnostic for Real-time Measurements of Turbulent Mixing

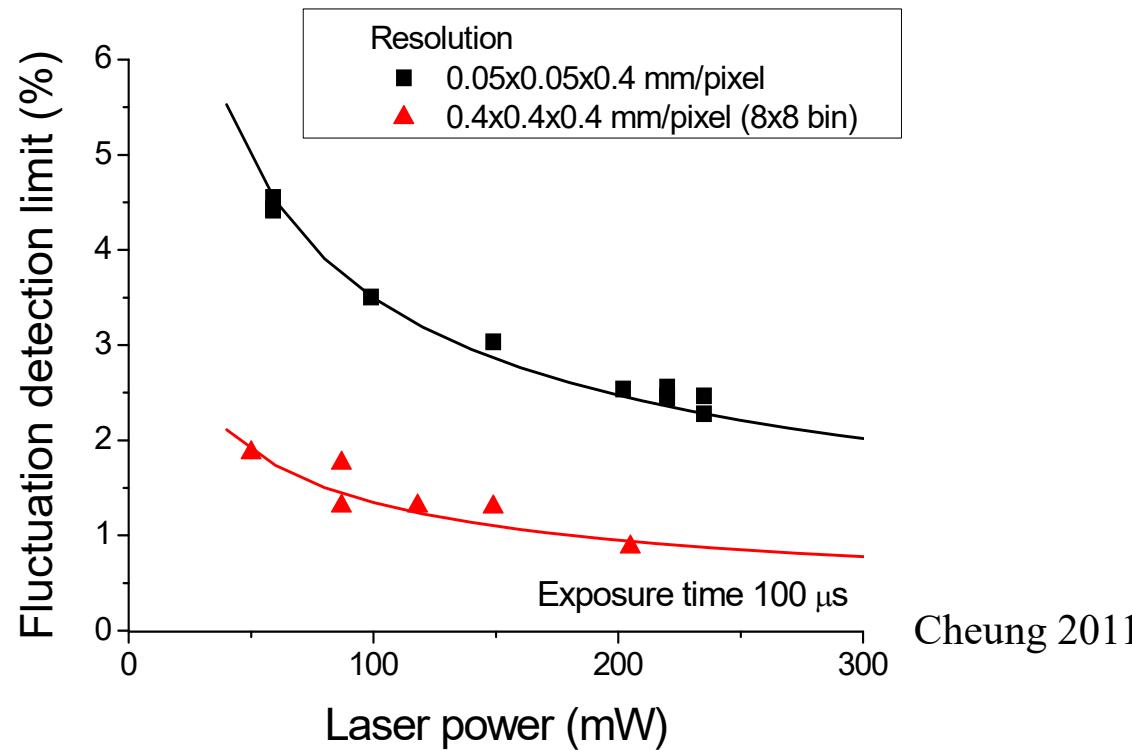
- **Problem: Use of pulsed lasers limits temporal resolution of tracer-based PLIF**
- Strategy: Use CW laser at 266 nm and toluene (**high  $\phi$** ) tracer for CW-LIF/PLIF
- CW-LIF/PLIF would offer useful diagnostic for turbulent/fluctuating flows
  - Turbulent mixing of injected fuel streams
  - Studies of jet noise
- Apply CW PLIF to a toluene-seeded, steady N<sub>2</sub> jet to study fluctuation detection limits
  - CW UV light @ 266nm from cavity doubling of commercial 532 nm lasers (5-20W)
  - LIF signal imaged with intensified CCD camera



## 5. CW PLIF for High-Frame-Rate Imaging

### CW PLIF Detection Limits for 266nm Excitation of Toluene

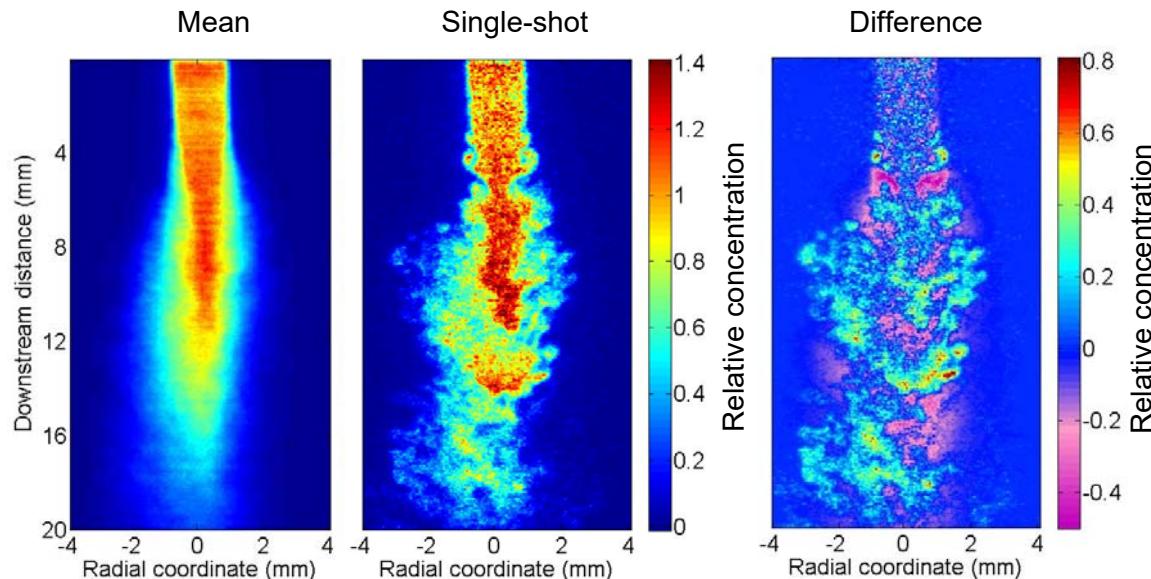
- What is the minimum detectable fluctuation in  $\chi_i$ ?



- Data consistent with shot-noise detection limit as  $\text{SNR} \propto (\text{signal})^{1/2}$
- Mixture fraction fluctuations  $< 1\%$  can be detected with 100  $\mu$ s resolution!
  - 10 kHz frame rate equivalent at each 0.4x0.4x0.4mm pixel

## 5. CW PLIF for High-Frame-Rate Imaging

CW PLIF Imaging of Mixing in Turbulent Jet: First Demonstrations



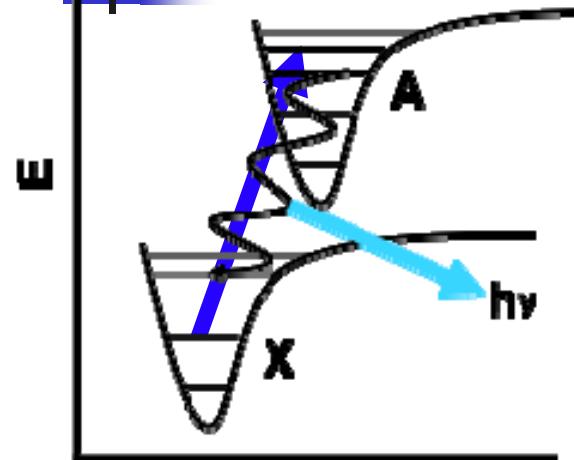
10  $\mu$ s exposure time  
(100 kHz frame rate equivalent)  
200 mW laser power  
0.05x0.05x0.4 mm/pixel

- N<sub>2</sub> jet seeded with 4% toluene (Re = 10700) in N<sub>2</sub> coflow
- Single-shot images of X<sub>mixture</sub> show time-varying features of mixing

### The Future:

- Improve laser intensity with higher power source, beam homogenizer
- Demonstrate high-speed imaging

## 6. Toluene PLIF for T Imaging: (1-camera)



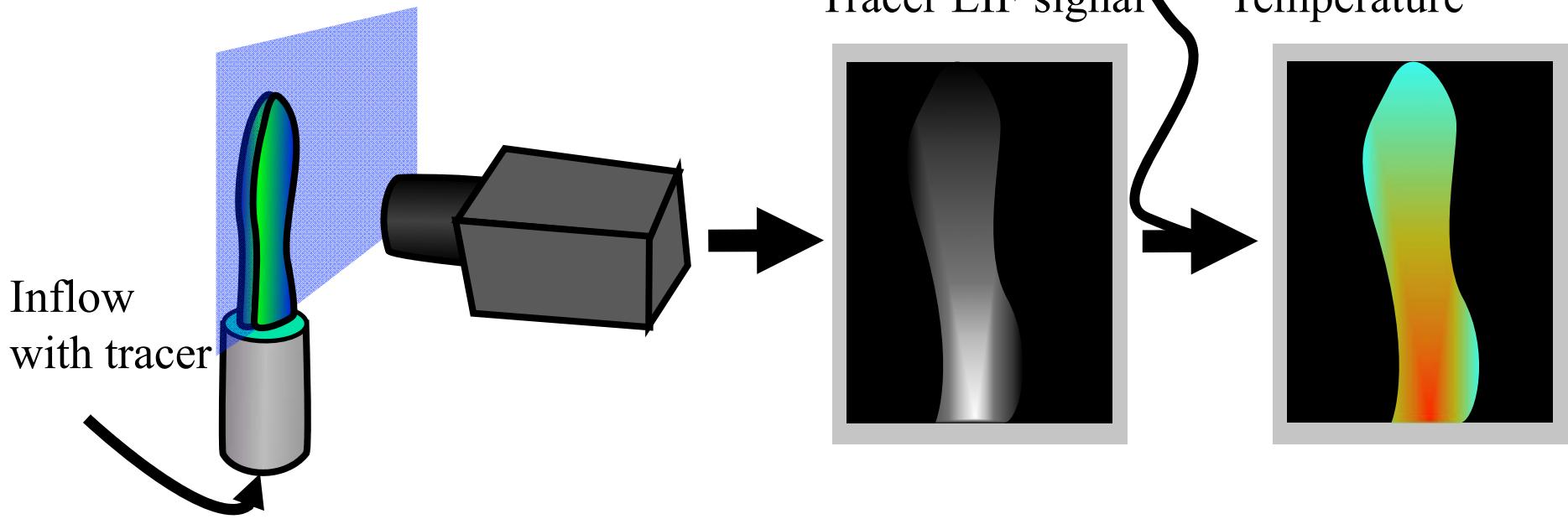
### Planar Laser-Induced Fluorescence

$$S_f = \frac{E}{h\nu} n \sigma(\lambda, T) \phi(\lambda, T, p_i) \eta$$

Diagram illustrating the Planar Laser-Induced Fluorescence (PLIF) signal generation process:

- LIF Signal**: The output signal from the fluorescence process.
- Incident Photons**: Represented by an orange arrow pointing to the absorption term.
- Photons Absorbed**: Represented by a red arrow pointing to the absorption term.
- Fluorescence Quantum Yield**: Represented by a green arrow pointing to the quantum yield term.
- Collection Efficiency**: Represented by a purple arrow pointing to the collection efficiency term.

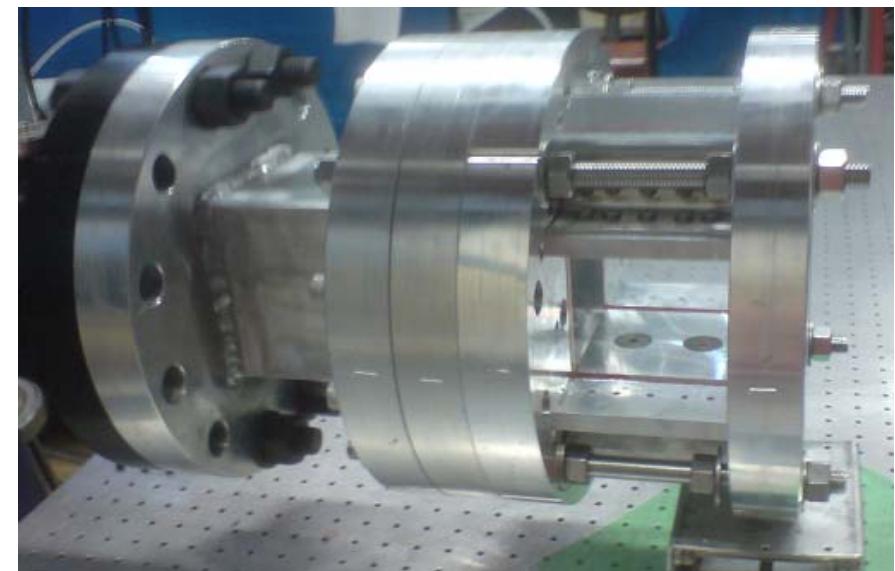
The equation shows the LIF Signal ( $S_f$ ) is proportional to the energy per photon ( $E/h\nu$ ), the number of molecules ( $n$ ), the cross-section for absorption ( $\sigma(\lambda, T)$ ), the fluorescence quantum yield ( $\phi(\lambda, T, p_i)$ ), and the collection efficiency ( $\eta$ ).



## 6. Toluene PLIF for T Imaging (1-camera)

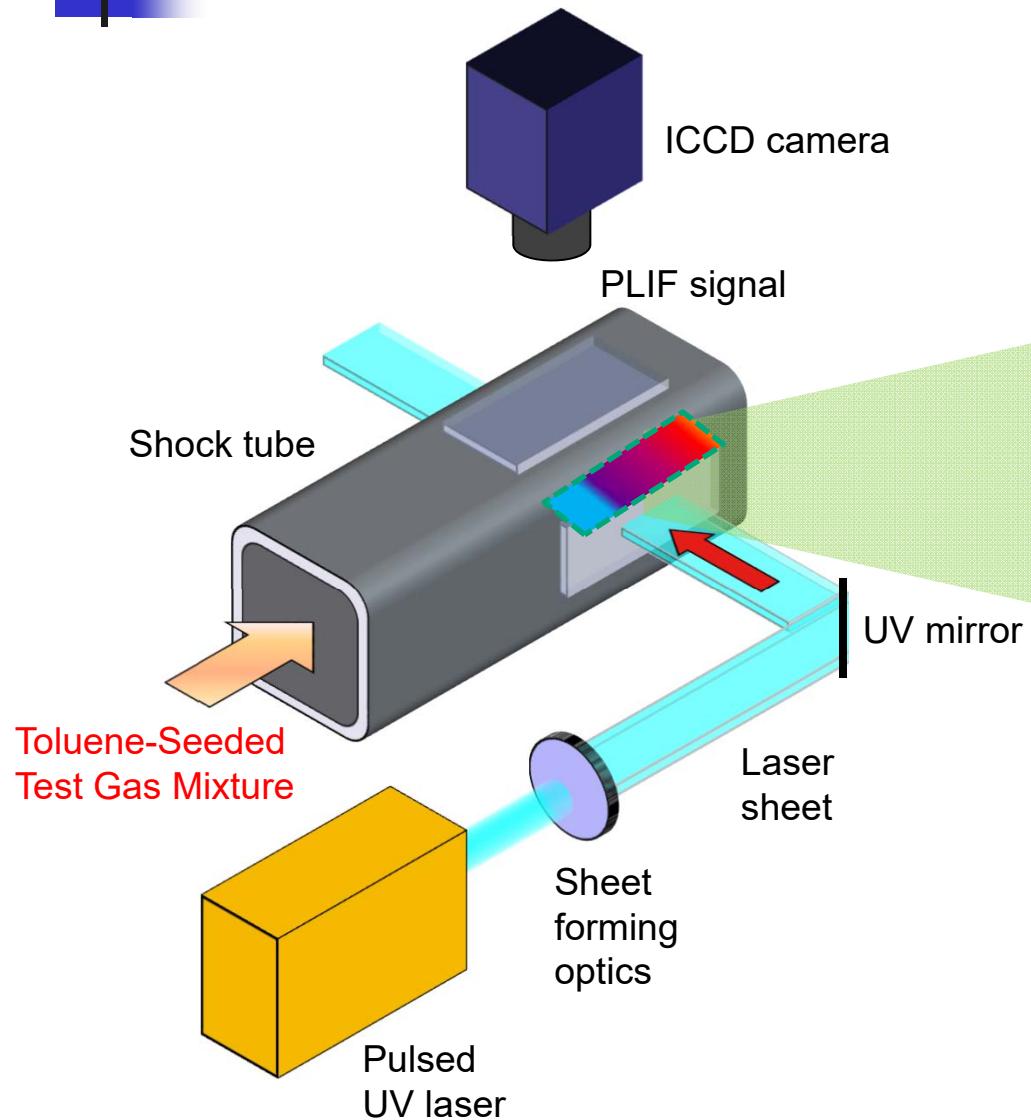
- Critical need for accurate targets to validate CFD simulations of shock/boundary layer interactions  
    ➔ **PLIF Imaging in a Shock Tube**
- Development of new high-sensitivity temperature measurement strategy for imaging  
    ➔ **Toluene-Based PLIF Temperature Measurement**
- Shock tube viewing section enables imaging of reflected shock wave/boundary layer interaction

Four-Window Square  
Endwall Viewing Section:  
Stanford Shock Tube

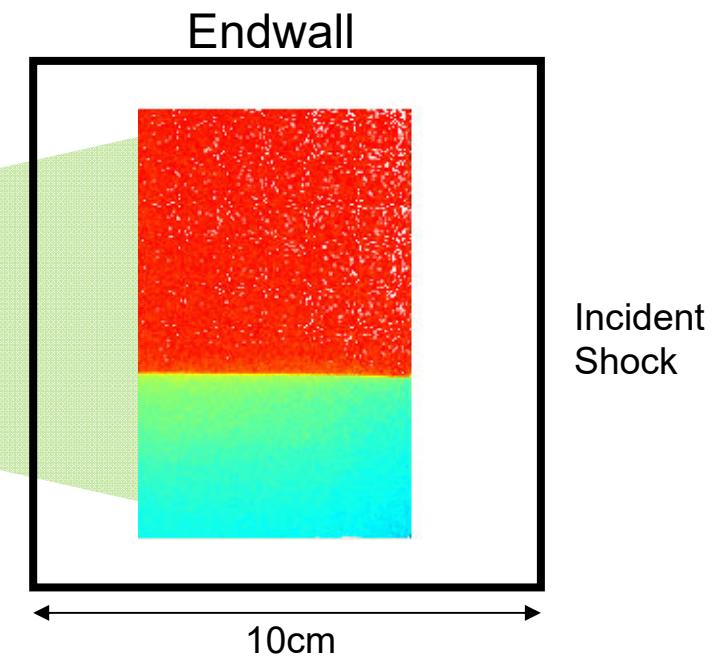


## 6. Toluene PLIF for T Imaging (1-camera)

PLIF of Incident Shock Wave Arrival



Camera PLIF Image

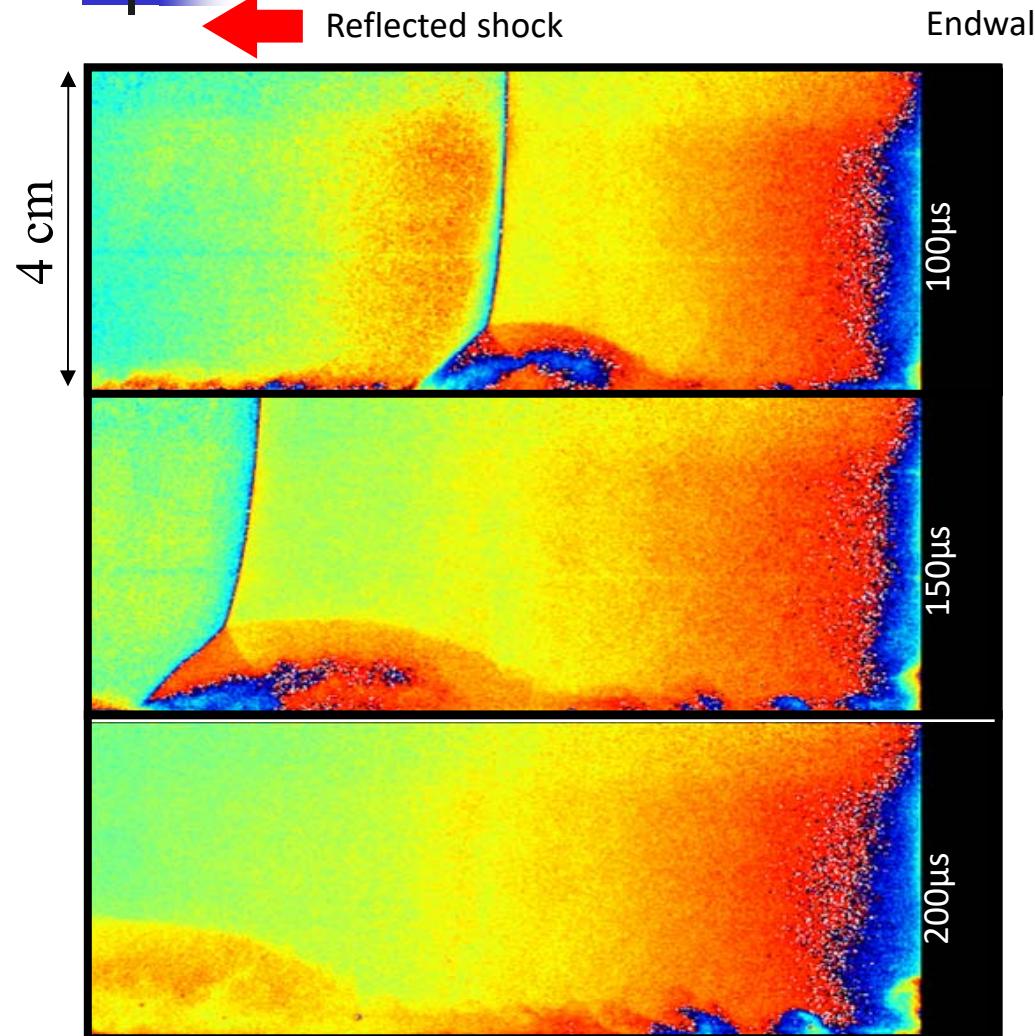


**Shock Flow Conditions:**

$T_1=296\text{K}$ ,  $T_2=438\text{K}$ ,  
 $P_1=44\text{torr}$ ,  $P_2=0.24\text{atm}$ ,  
 $X_f=7\%$ ,  $\text{Incident } V_s=605\text{m/s}$

## 6. Toluene PLIF for T Imaging (1-camera)

Shock Wave/BL Interaction: Reflected Shock  
Bifurcation Time Sequence



- First toluene PLIF images of shock bifurcation
  - Interaction of reflected shock wave with boundary layer
- Toluene provides sensitive data for **temperature** and **flow structure** for CFD validation
- Opportunity to apply to other shock/boundary layer flowfields

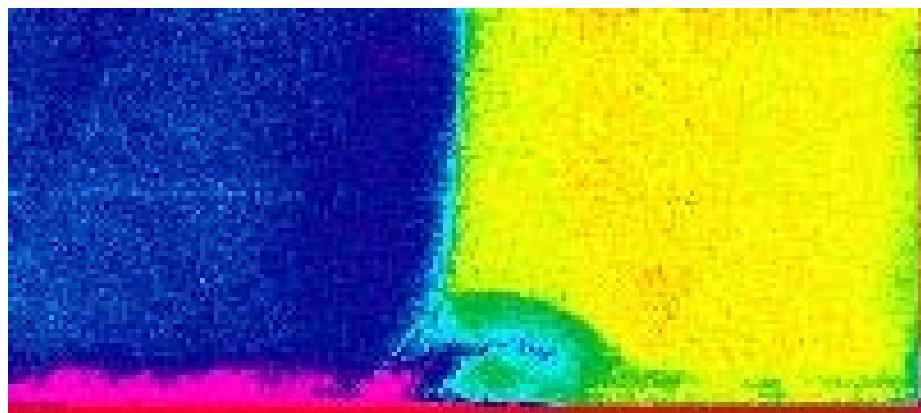
### Shock Flow Conditions:

$T_1=296K$ ,  $T_2=498K$ ,  $T_5=696K$   
 $P_1=32\text{ torr}$ ,  $P_2=0.25\text{ atm}$ ,  $P_5=1.05\text{ atm}$   
 $X_f=8\% \text{ Tol}/N_2$  Incident  $V_s=710\text{ m/s}$

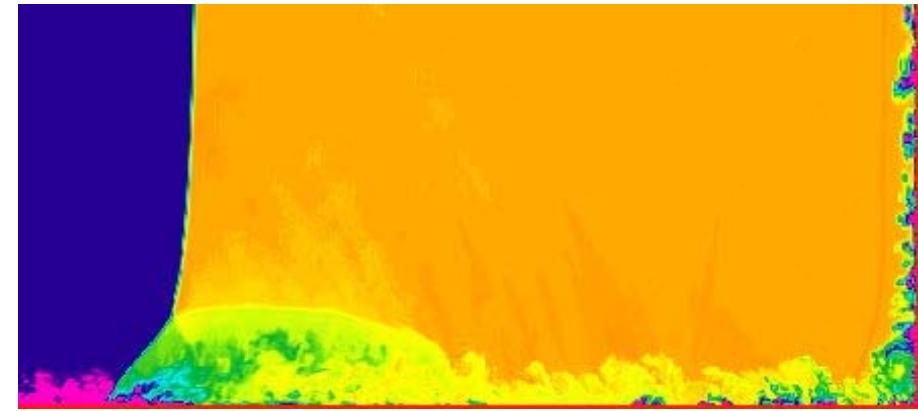
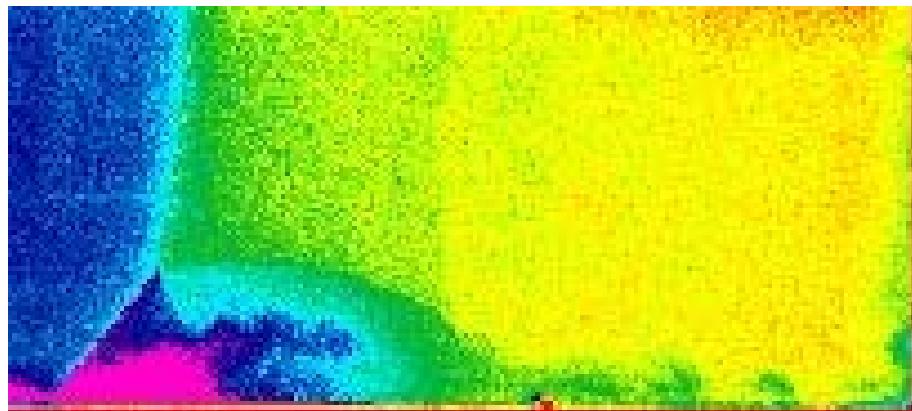
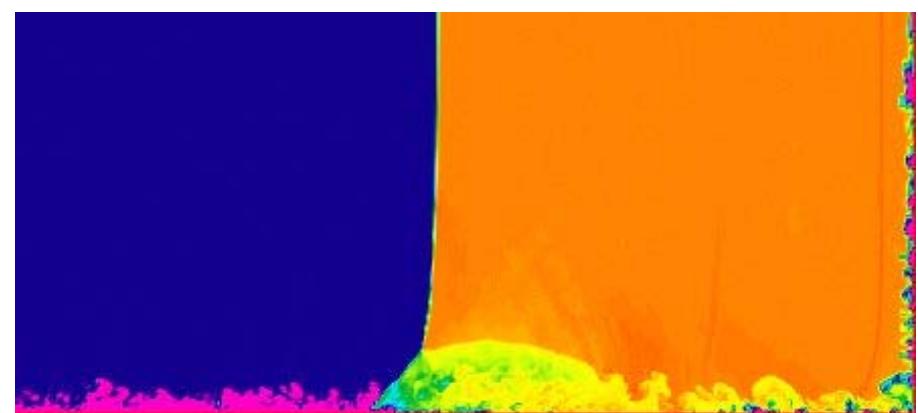
# 6. Toluene PLIF for T Imaging (1-camera)

PLIF Signal Comparison with CFD Simulation

Experiment



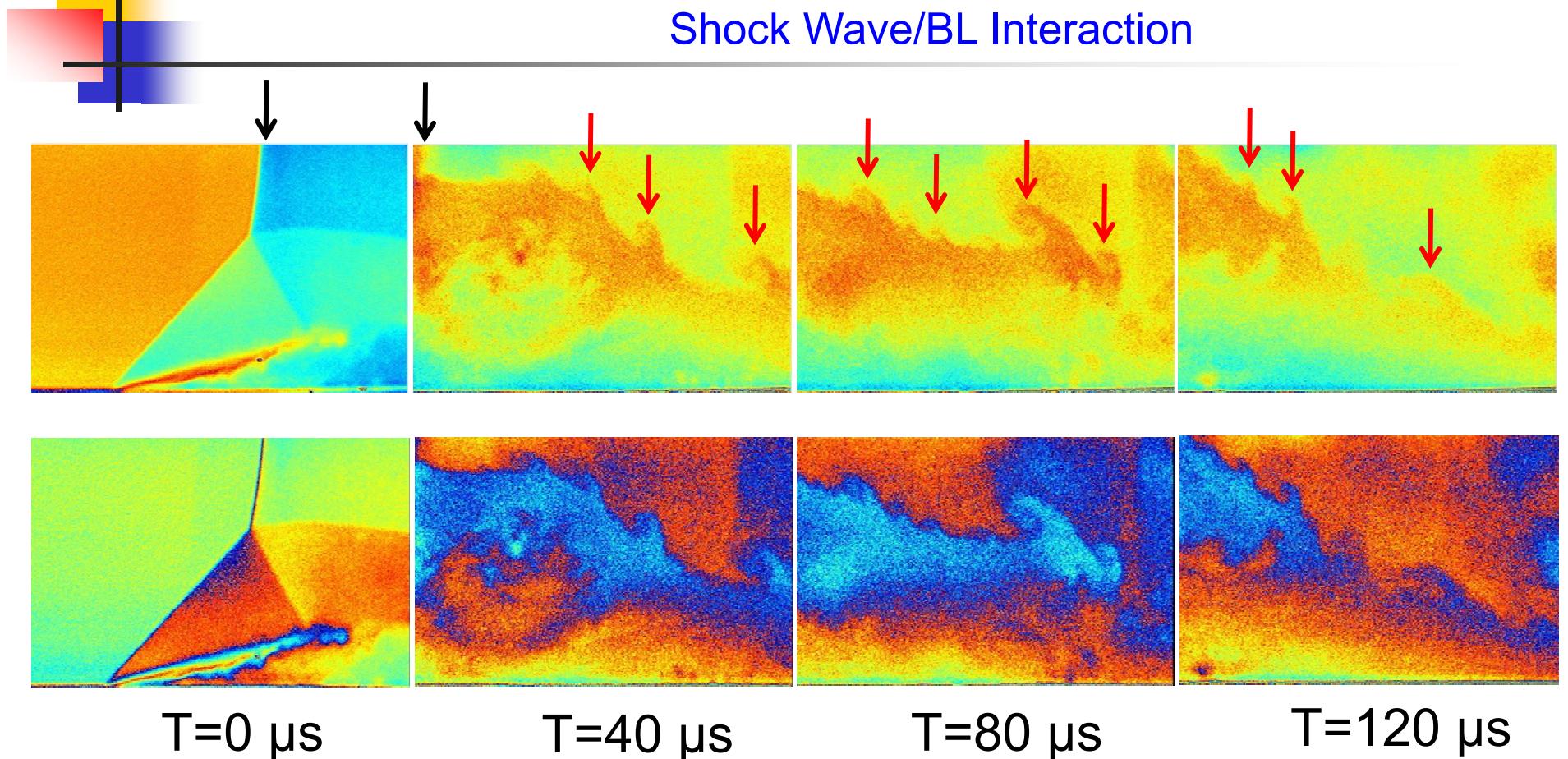
Simulation (Charles)



## Shock Flow Conditions:

$T_1=296\text{K}$ ,  $T_2=498\text{K}$ ,  $T_5=696\text{K}$   
 $P_1=32\text{torr}$ ,  $P_2=0.25\text{atm}$ ,  $P_5=1.05\text{atm}$     $X_f=8\%\text{Tol}/N_2$    Inc.  $V_s=710\text{m/s}$

# 6. Toluene PLIF for Temperature Imaging



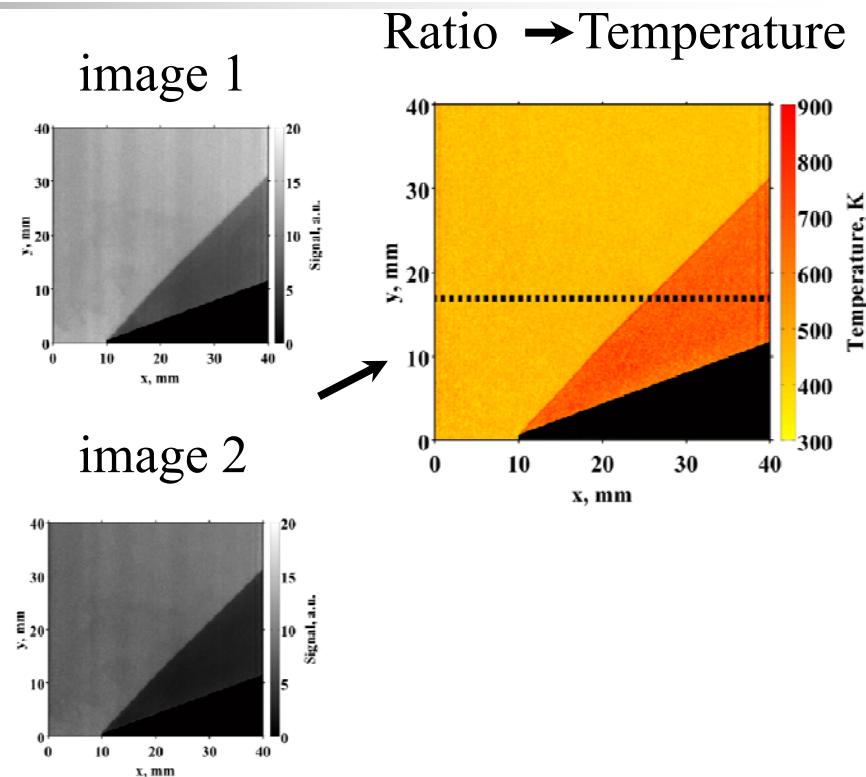
- Two different color renditions
- Structure formation revealed

## Shock data

$P_1 = 21 \text{ torr}$      $P_2 \approx 0.143 \text{ atm}$      $P_5 \approx 0.517 \text{ atm}$   
 $T_2 \approx 521\text{K}$      $T_5 \approx 772\text{K}$   
 $V_s \approx 737 \text{ m/s}$   
0.6%Toluene/ $\text{N}_2$

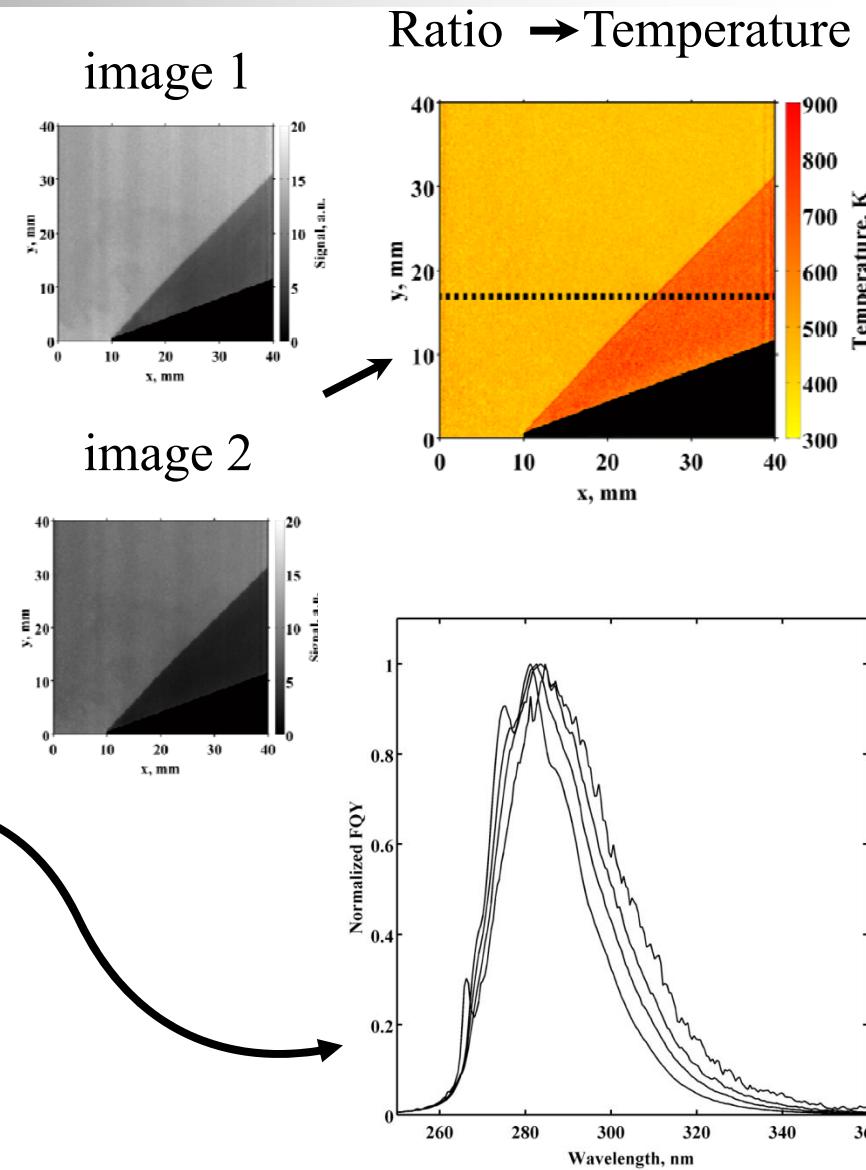
## 7. Toluene PLIF for T Imaging (2-camera)

- Two cameras simultaneously image different spectral regions of fluorescence spectrum
- Select target species or tracer based upon conditions and quantity of interest (e.g., T or  $x_{O_2}$ )
- e.g., toluene, 3-pentanone, acetone



## 7. Toluene PLIF for T Imaging (2-camera)

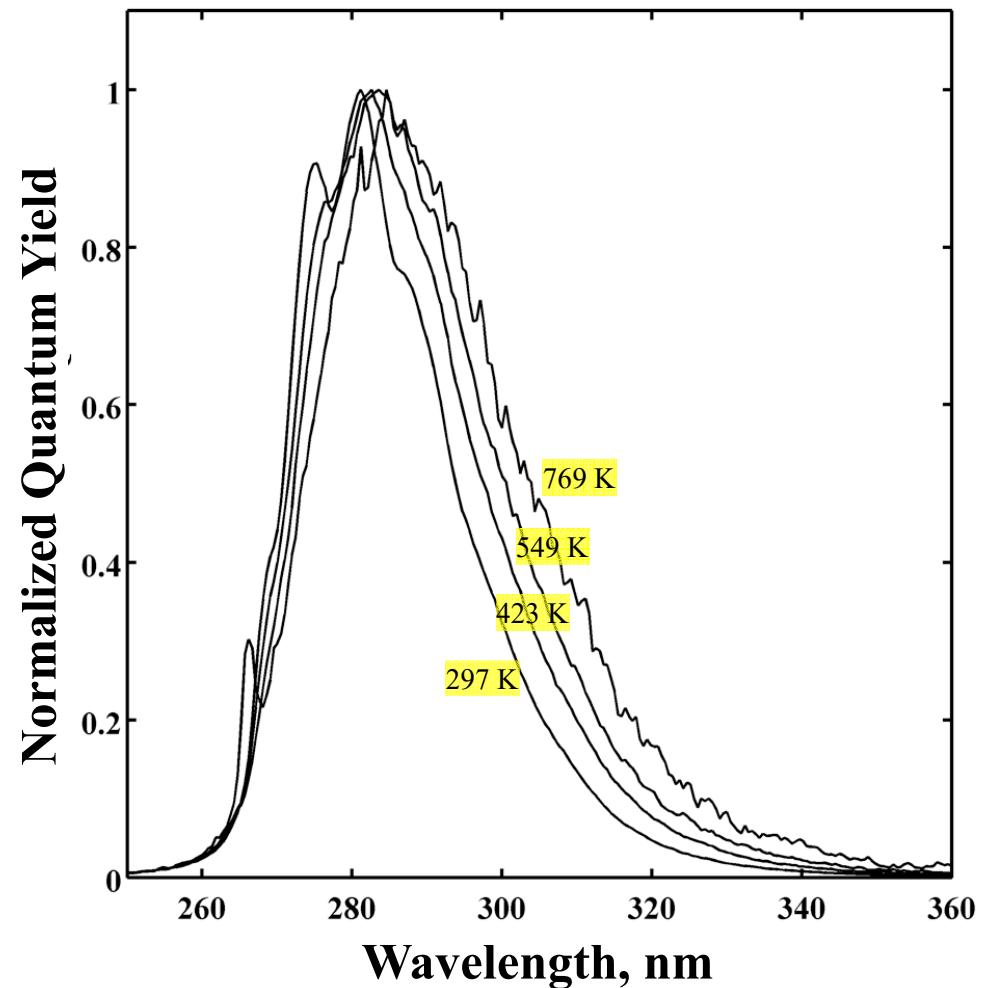
- Two cameras simultaneously image different spectral regions of fluorescence spectrum
- Select target species or tracer based upon conditions and quantity of interest (e.g., T or  $X_{O_2}$ )
- e.g., toluene, 3-pentanone, acetone
- With understanding of photophysics of fluorescing species, physical quantities (e.g., T) can be inferred from ratio of images
- Allows for measure of quantity independent of tracer number density



## 7. Toluene PLIF for T Imaging (2-camera)

- Toluene fluorescence is function of temperature

$$S_f = \frac{E}{h\nu} \frac{P_i}{RT} \sigma(T) \phi(T, \lambda) \eta$$



From Koban, Koch, Hanson, Schulz, 2005

27

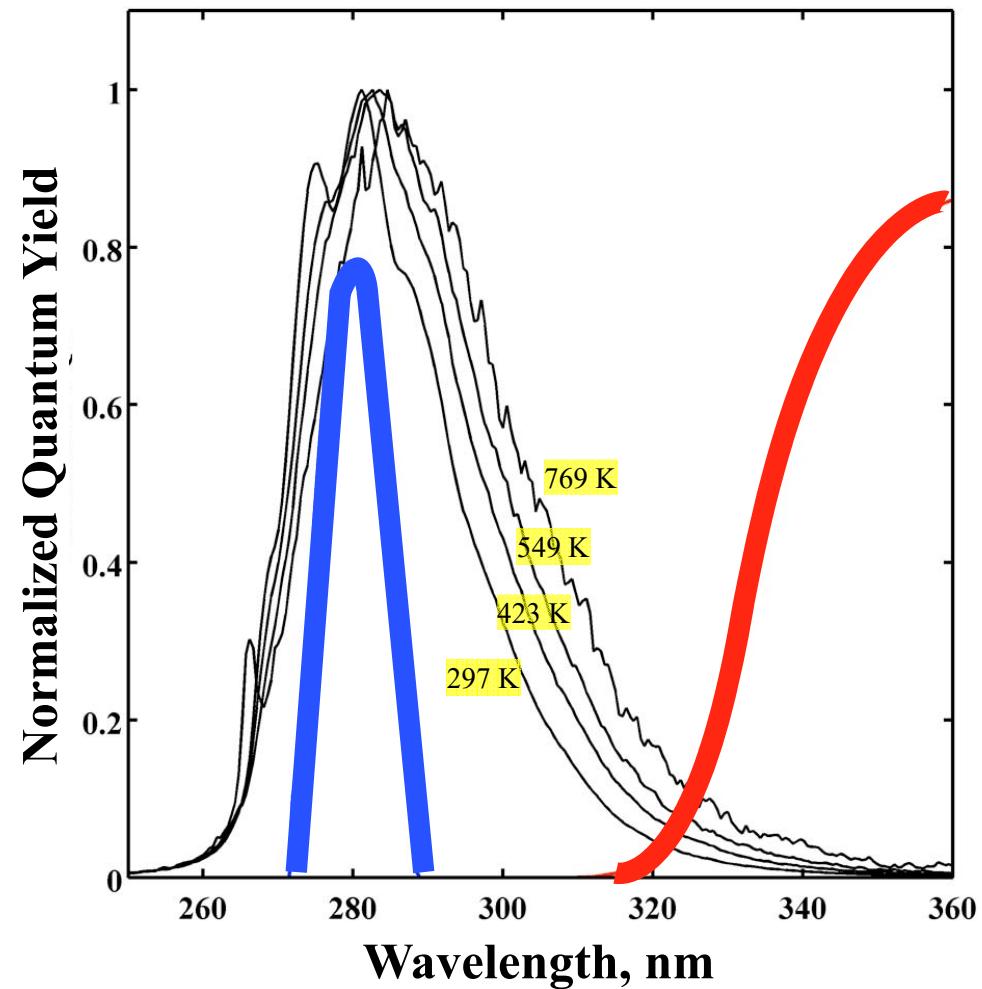
## 7. Toluene PLIF for T Imaging (2-camera)

- Toluene fluorescence is function of temperature

$$S_f = \frac{E}{h\nu} \frac{P_i}{RT} \sigma(T) \phi(T, \lambda) \eta$$

but depends on filter,  $F(\lambda)$ , thus

$$S_f = \frac{E}{h\nu} n \sigma(T) \eta \int \phi(\lambda, T) F(\lambda)$$



From Koban, Koch, Hanson, Schulz, 2005

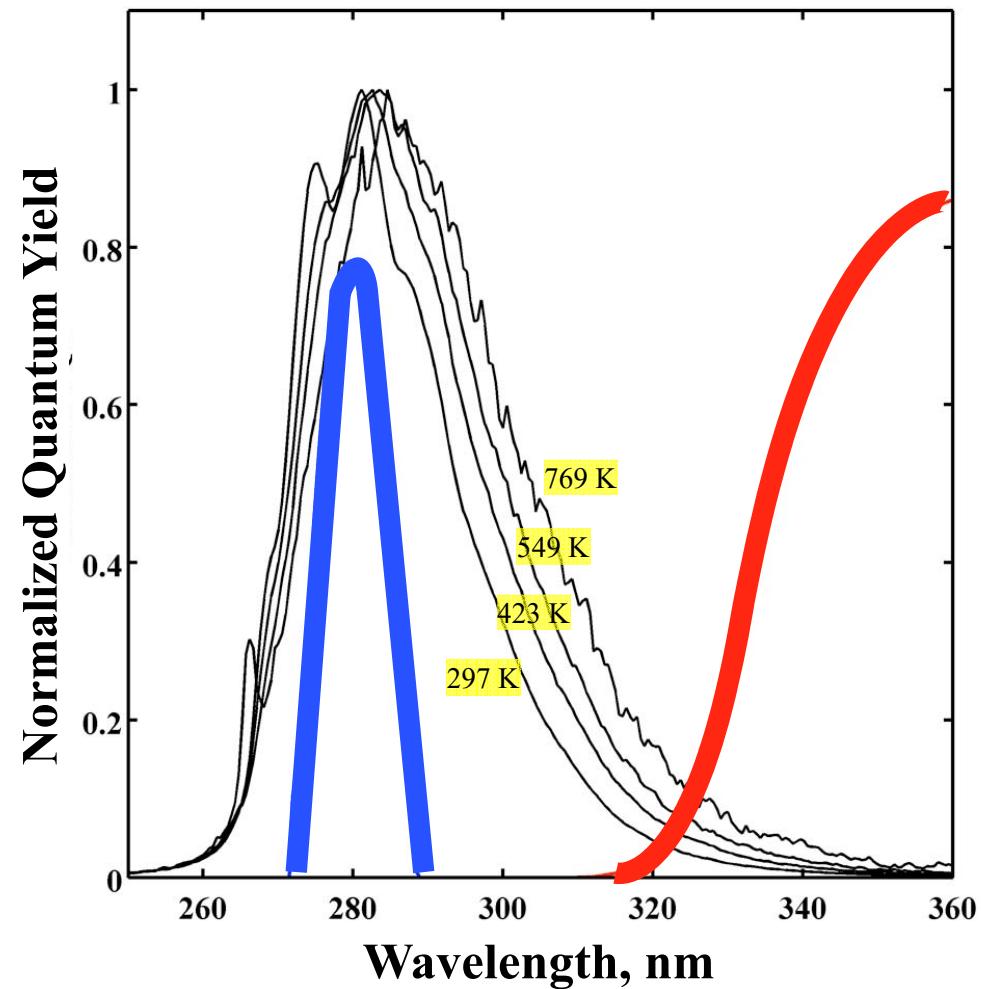
## 7. Toluene PLIF for T Imaging (2-camera)

- Toluene fluorescence is function of temperature

$$S_f = \frac{E}{h\nu} \frac{P_i}{RT} \sigma(T) \phi(T, \lambda) \eta$$

$$S_f = \frac{E}{h\nu} n \sigma(T) \eta \int \phi(\lambda, T) F(\lambda)$$

$$\frac{S_1}{S_2} = \frac{\int \phi(\lambda, T) \underline{F_1(\lambda)}}{\int \phi(\lambda, T) \underline{F_2(\lambda)}}$$



From Koban, Koch, Hanson, Schulz, 2005

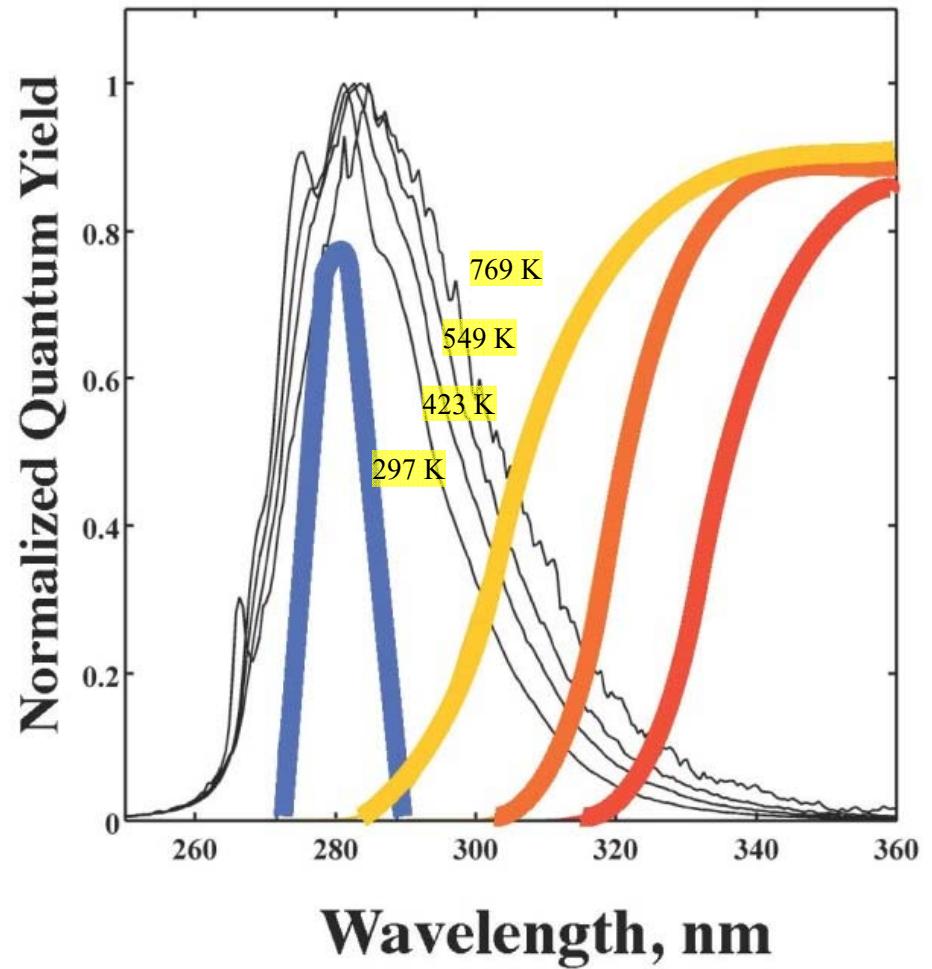
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$$S_f = \frac{E}{h\nu} n \sigma(T) \eta \int \phi(\lambda, T) F(\lambda)$$

$$\frac{S_1}{S_2} = \frac{\int \phi(\lambda, T) \underline{F_1(\lambda)}}{\int \phi(\lambda, T) \underline{F_2(\lambda)}}$$



- Three sets of filters examined

From Koban, Koch, Hanson, Schulz, 2005

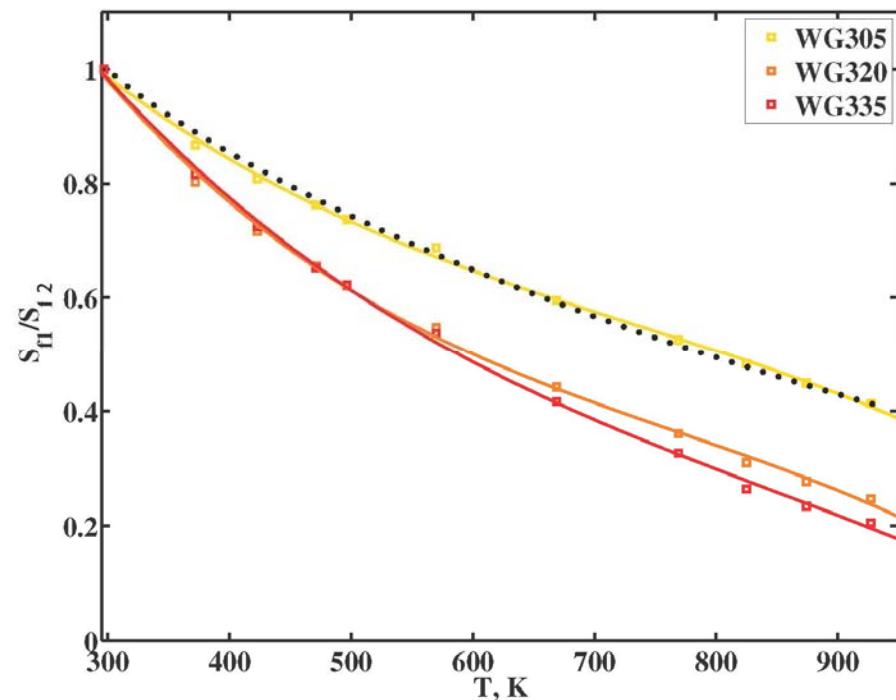
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$$S_f = \frac{E}{h\nu} n \sigma(T) \eta \int \phi(\lambda, T) F(\lambda)$$

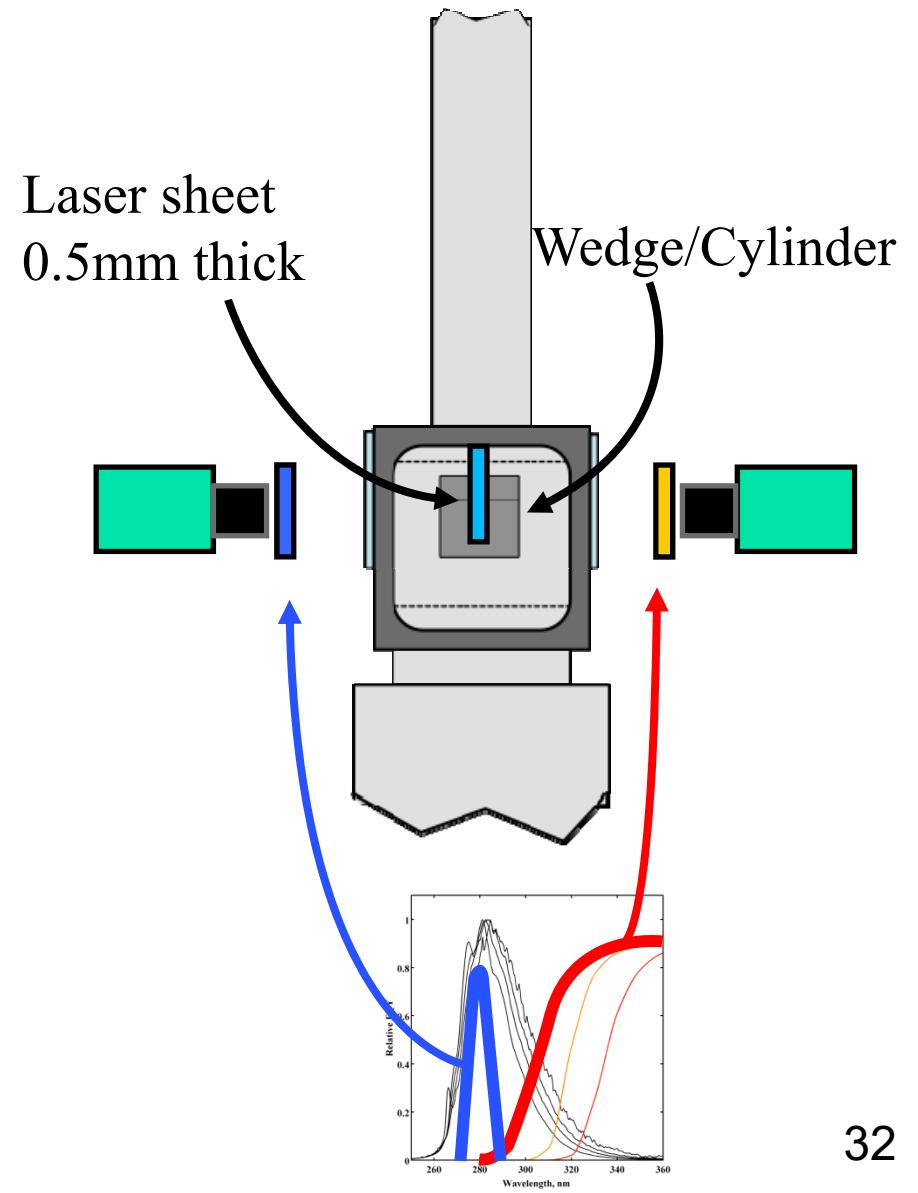
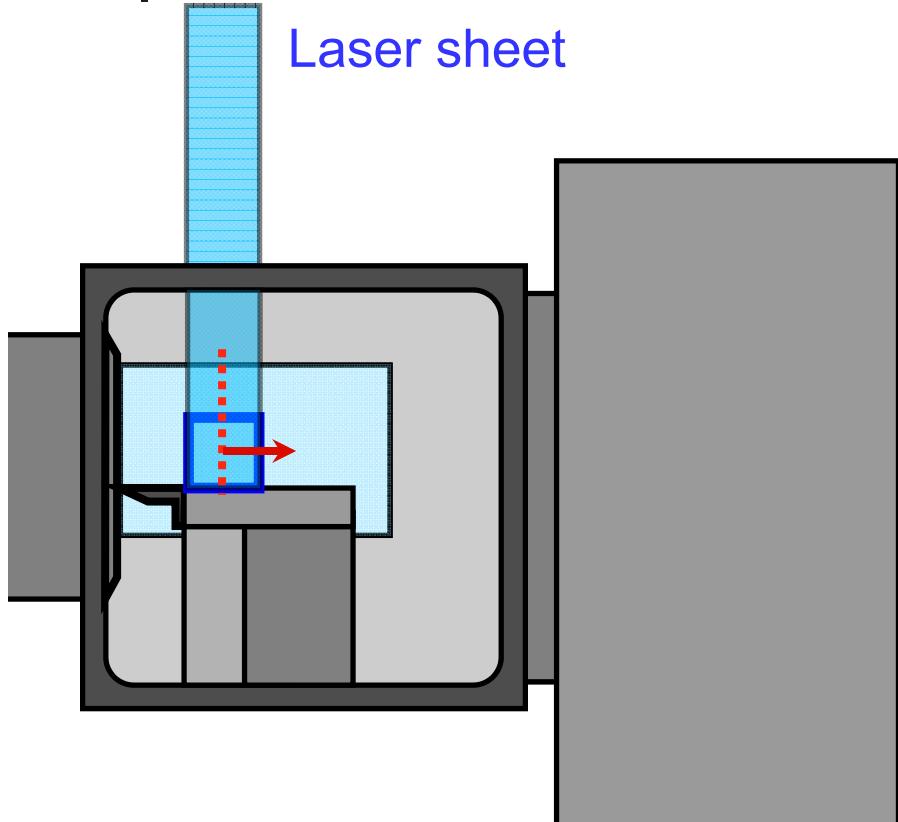
$$\frac{S_1}{S_2} = \frac{\int \phi(\lambda, T) \underline{F_1(\lambda)}}{\int \phi(\lambda, T) \underline{F_2(\lambda)}}$$



**Next: 3 demonstrations of 2-camera T imaging**

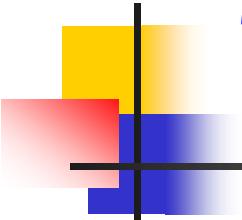
## 7. Toluene PLIF for T Imaging (2-camera)

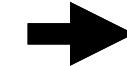
### Experimental setup

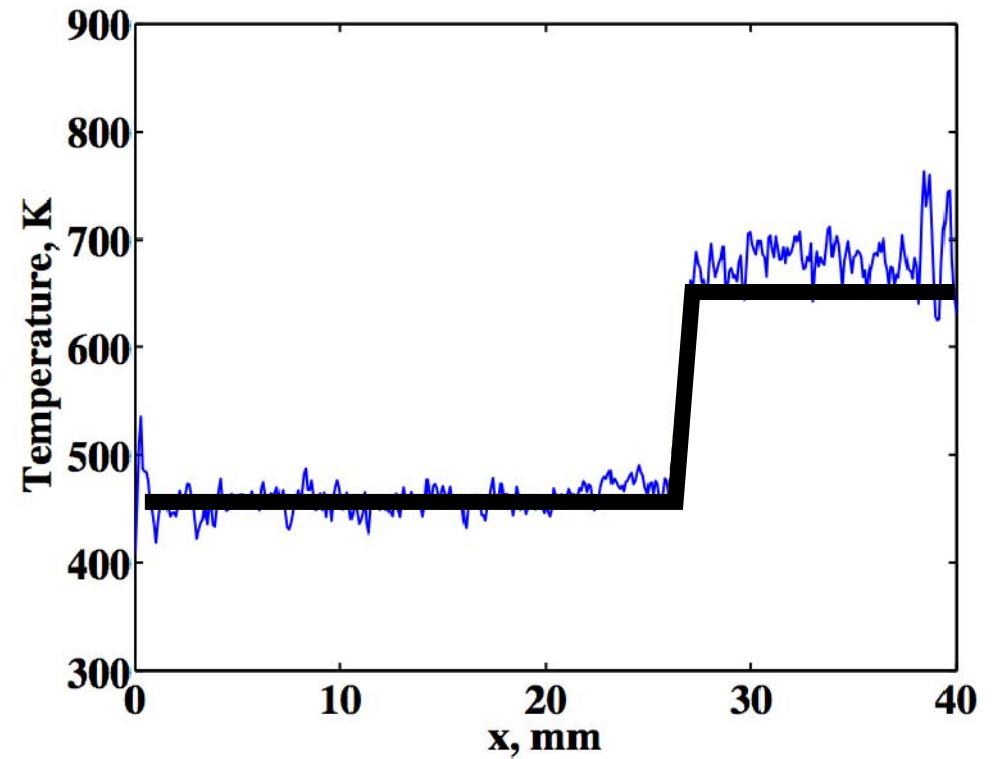
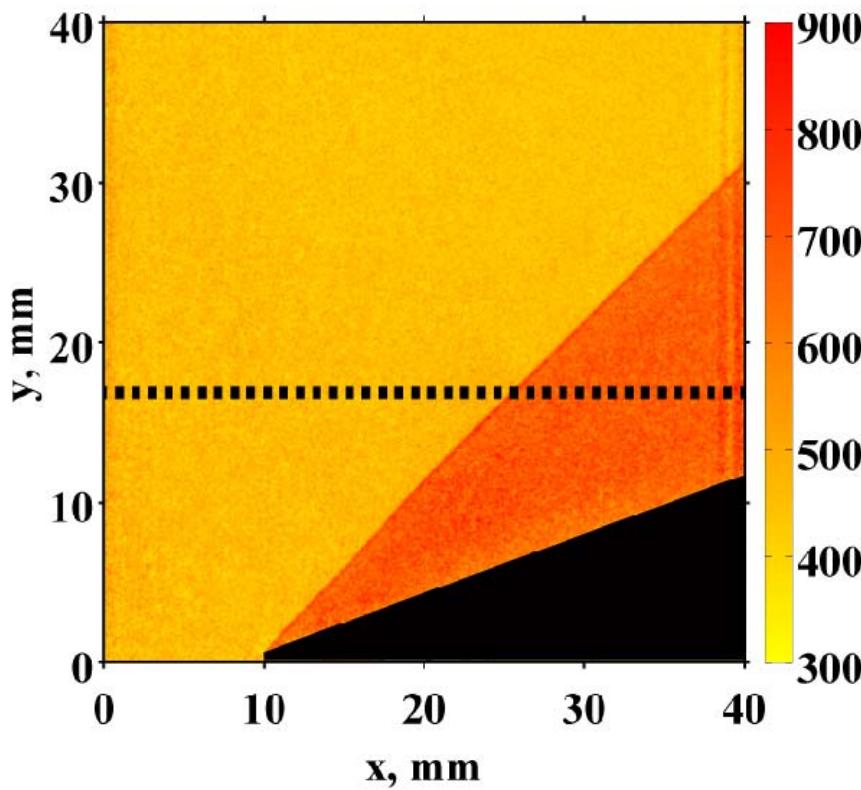


## 7. Toluene PLIF for T Imaging (2-camera)

Wedge Flow: Temperature Imaging

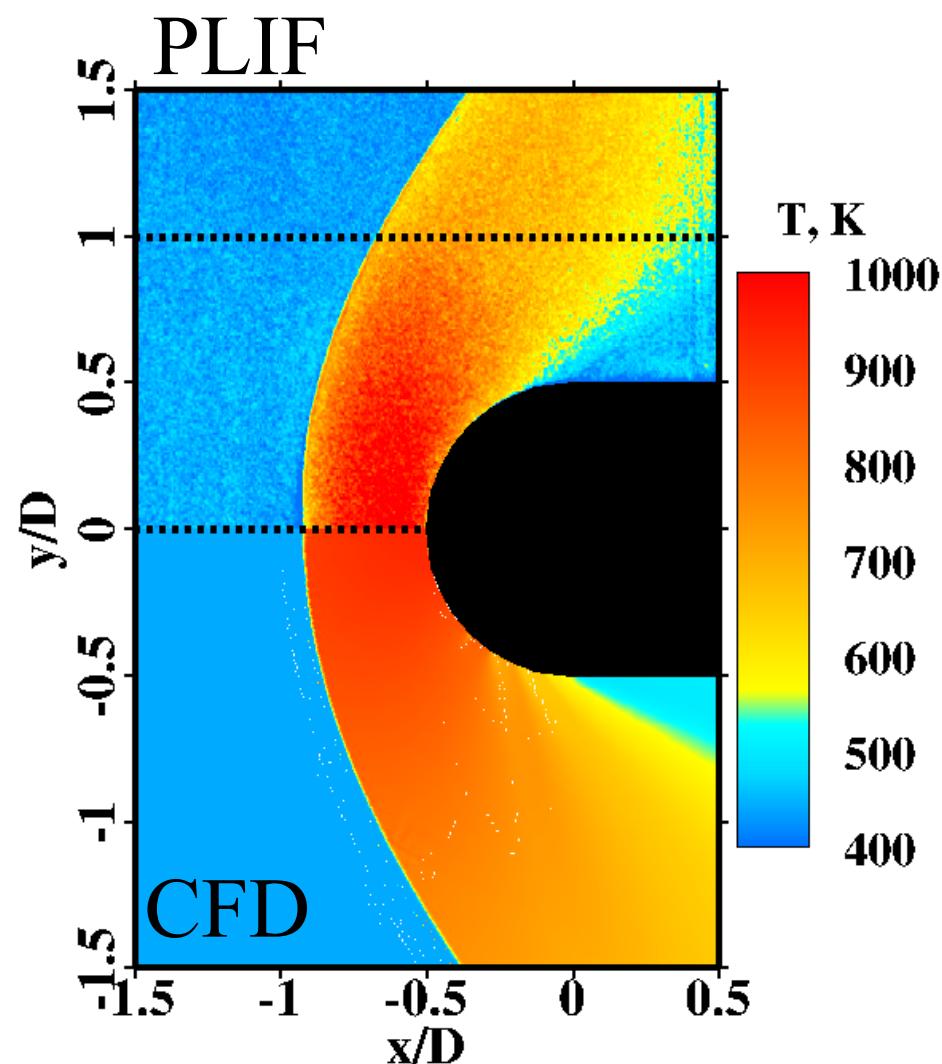


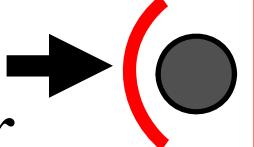
$M = 2.3$    
 $P = 1 \text{ bar}$  



## 7. Toluene PLIF for T Imaging (2-camera)

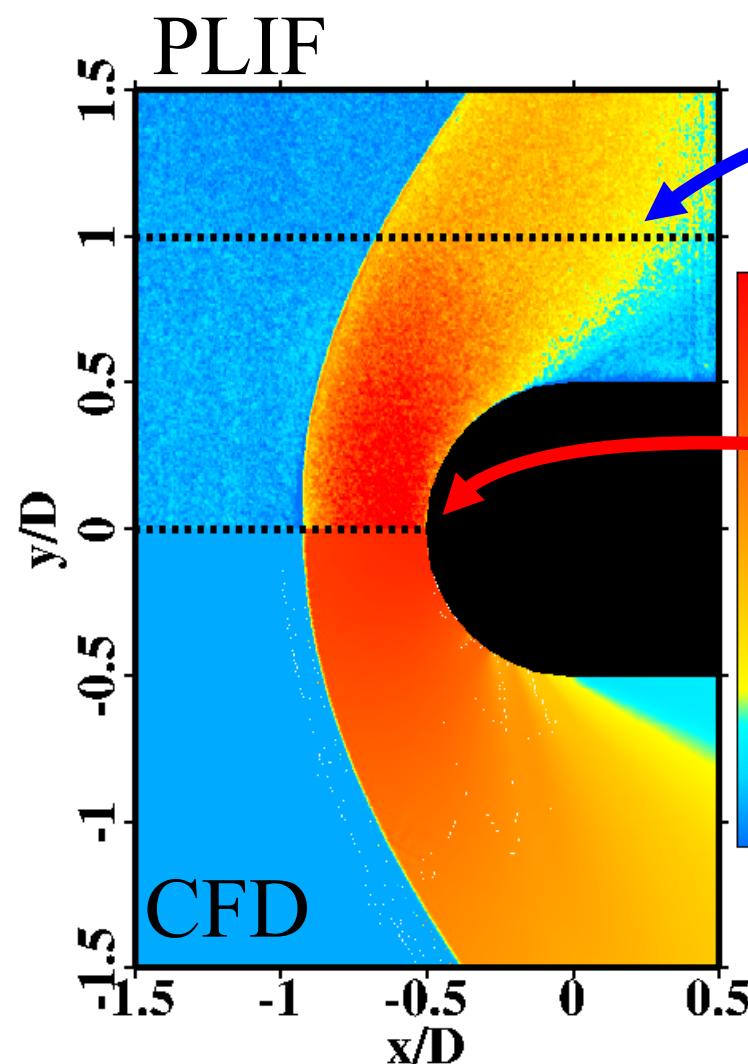
Cylinder Flow: Temperature Imaging



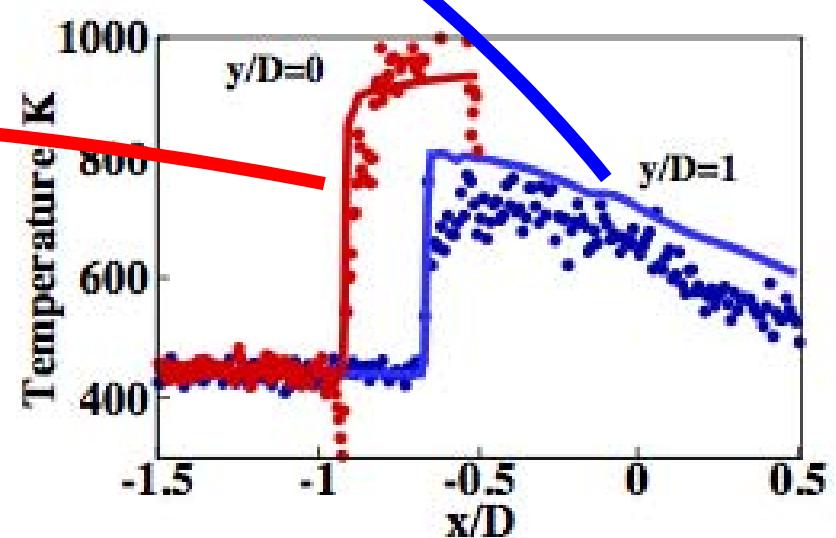
$M = 2.3 \rightarrow$    
 $P = 1 \text{ bar}$

## 7. Toluene PLIF for T Imaging (2-camera)

Cylinder Flow: Temperature Imaging



$M = 2.3$   $\rightarrow$

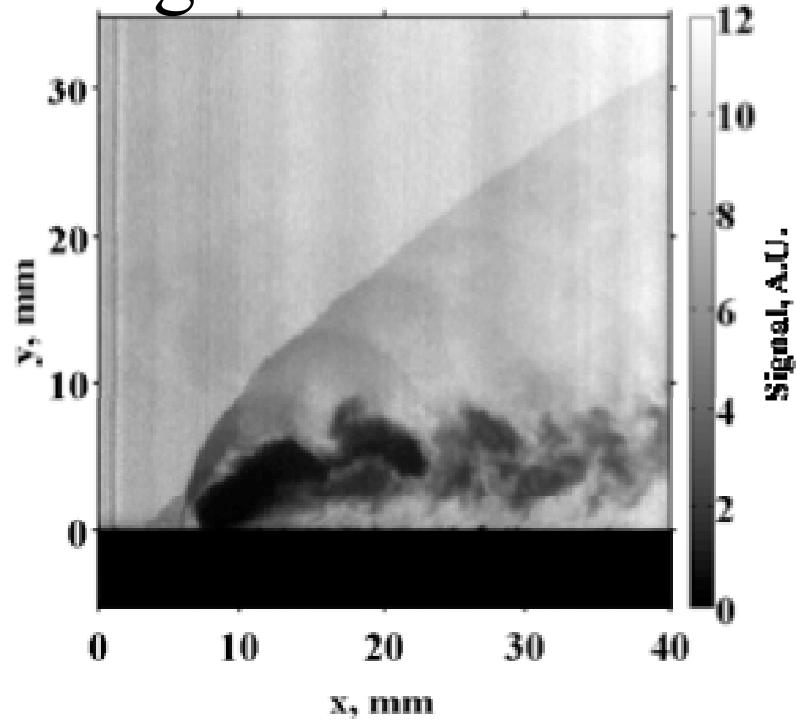


## 7. Toluene PLIF for T Imaging (2-camera)

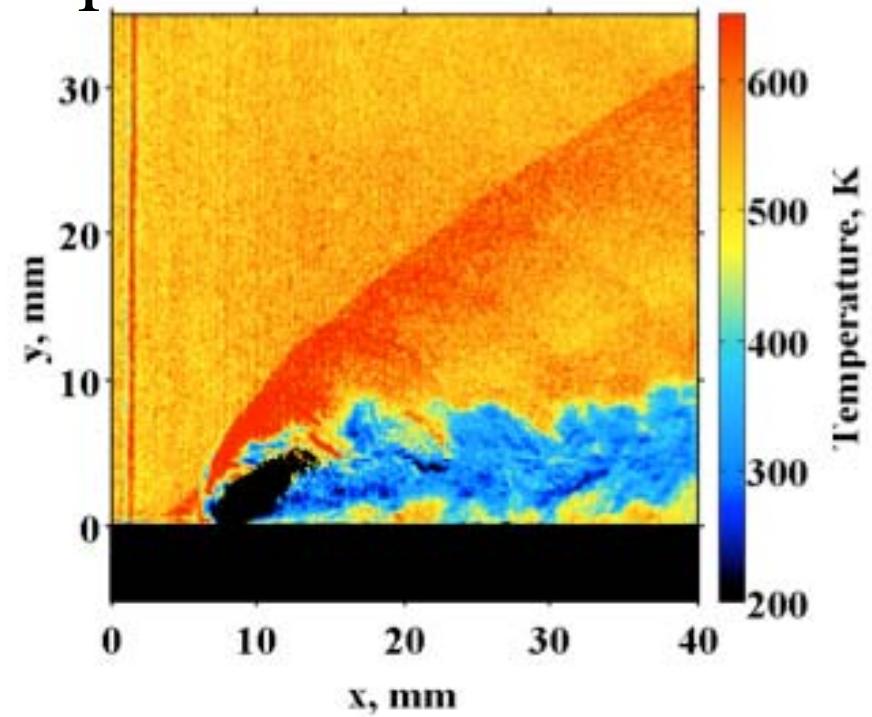
Jet in Crossflow: Temperature Imaging

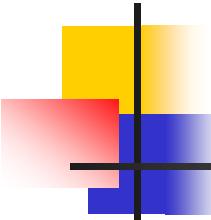
$M = 2.3$   
 $P = 1 \text{ bar}$

Raw Signal



Temperature





## 9. The Future for PLIF Imaging in Complex Flows

- Applications
  - Non-ideal effects in shock tubes (shock/boundary layer)
  - IC-Engines (EGR, T, radicals, pollutants)
  - SCRAMJETS (mixing, flame structure)
  - Fluid dynamics (mixing, turbulent combustion)
  - Plasma-enhanced combustion (flame structure, species)
- Species
  - Tracers (toluene, ketones, NO)
  - Naturally present (CO<sub>2</sub>, UHC, OH, NO)
- Strategies
  - High-speed PLIF
  - Multi-parameter imaging (T, species)
  - IR PLIF

# Quantitative Laser Diagnostics for Combustion Chemistry and Propulsion

## Closing Comments: The Future

- Use of laser diagnostics will become increasingly routine
- Availability of packaged systems will continue to grow
- Availability and cost of mid-IR TDLAS will improve
- Sensitivity improvements will be found (e.g. CEAS)
- Use of hirep PLIF systems will become more common
- New PLIF tracers and techniques employing traces will be found
- Laser diagnostics will play a key and expanding role in future research on combustion
- **The future for laser diagnostics is bright!**
- **Thanks for attending this short course!**
- **Best wishes for your future!**