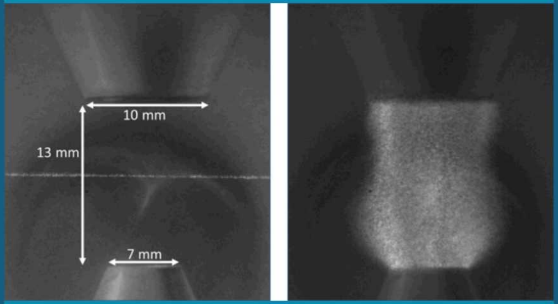
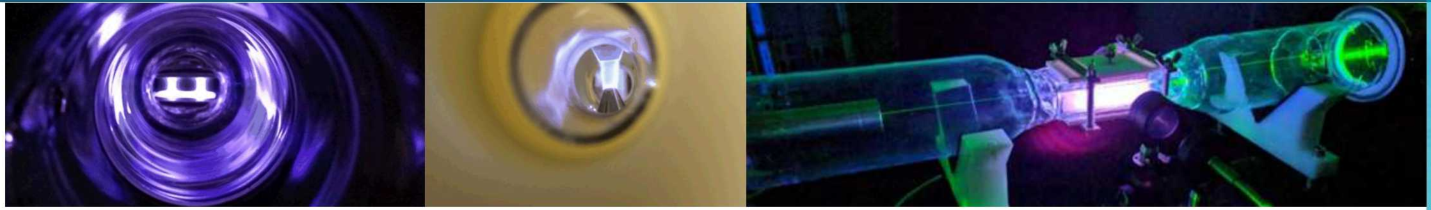
SAND2019-8086C

Electric Field Induced Second Harmonic Generation Using Ultrafast Lasers for Measurements in Pulsed Nanosecond Plasmas





PRESENTED BY

Benjamin M. Goldberg

PRINCETON UNIVERSITY

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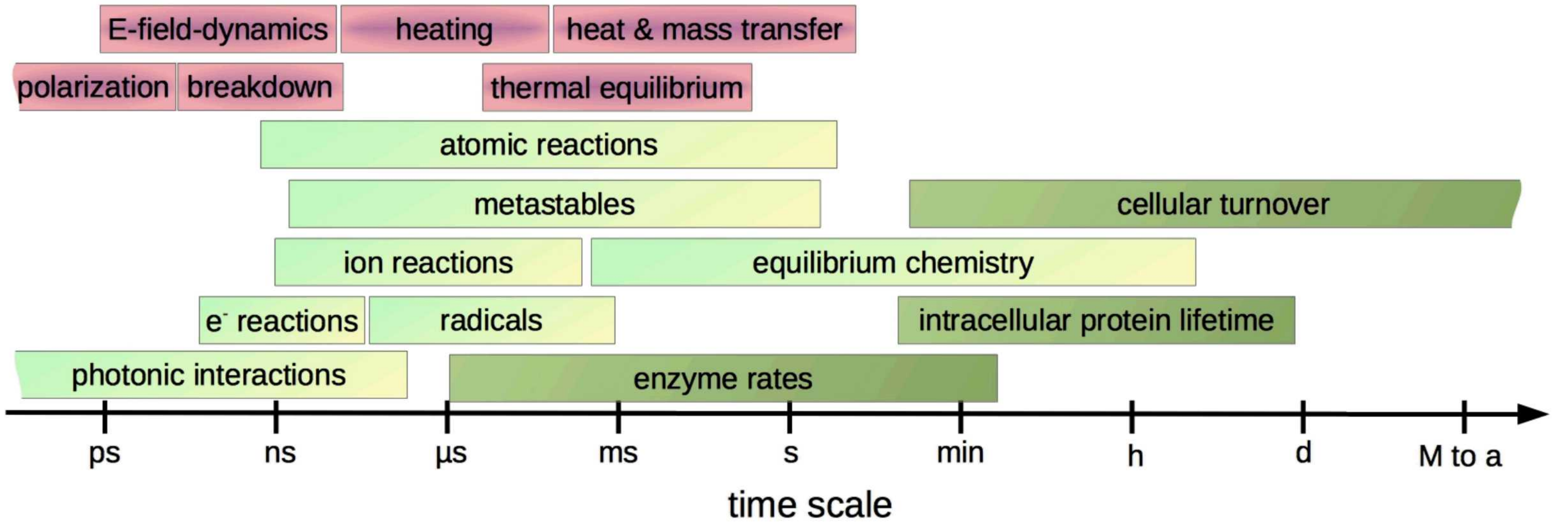
Importance of Reduced Electric Field (E/N)



Reuter, S et. al., *J. Phys D* **2018**

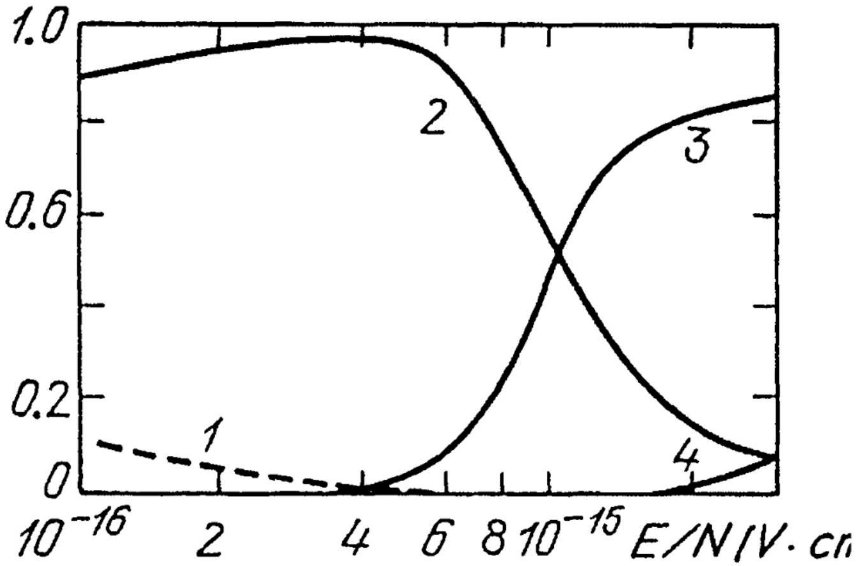
- **In order to understand how we can use the plasma we must have an in depth understanding of fundamental plasma processes**

Electron collisions with neutrals mainly excite internal degrees of freedom

- Leads to non-equilibrium energy distributions

By controlling E/N, we can choose where that energy goes

- Rotational (< 1Td), Vibrational (1-50 Td), Electronic (50 – 100 Td), Ionization (100 Td)



Yuri P Raizer. Gas Discharge Physics

Noninvasive Electric Field Measurement Methods

OES

- Field inferred by monitoring plasma emission
- Requires specific model depending upon gas composition

LIF Dip

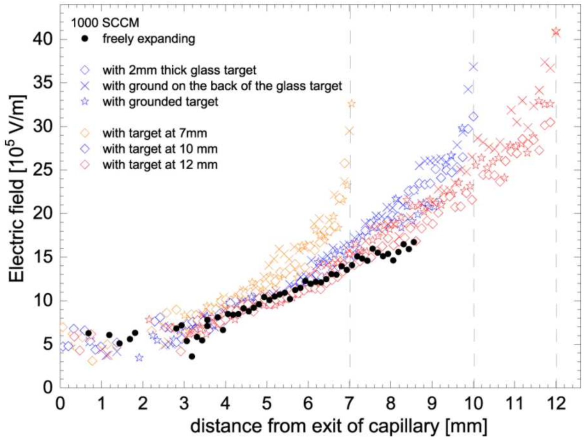
- Able to generate 2D field maps with sensitivity of ~10V/cm
- Pressure limited to ~1 Torr due to pressure broadening

E-Field CARS

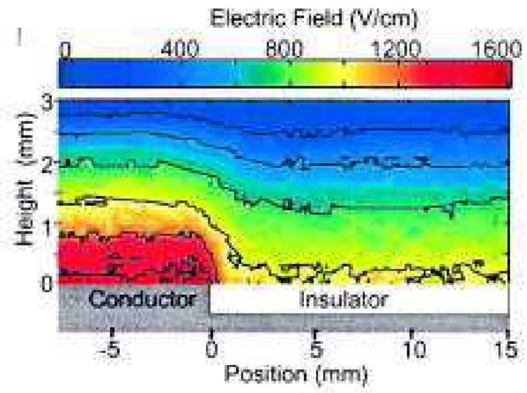
- 4-wave mixing technique for sub-nanosecond measurements
- Resonant technique which has only been demonstrated in H_2 and N_2

Second Harmonic Generation

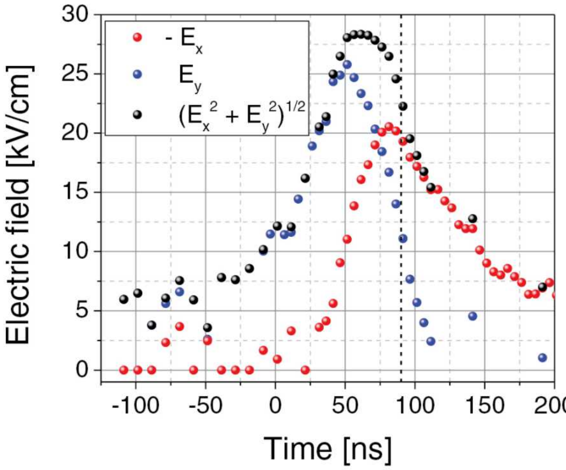
- Non-resonant method with sub-nanosecond resolution
- Signal scales as E^2
- Sensitivity to electric field vectors



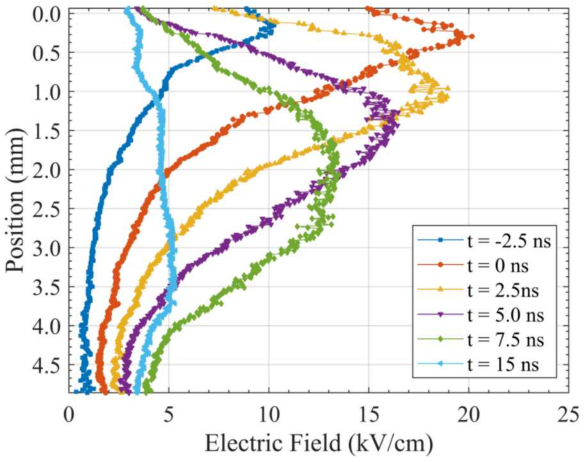
A. Sobota et al, PSST 2019



E. V. Barnat et al Appl. Phys. Lett. 2004



M. Simeni Simeni et al J.Phys. D 2017



B. M. Goldberg et al, Optics Lett 2019

Electric Field Induced Second Harmonic Generation (E-FISH)

In a centrosymmetric medium, second harmonic generation is not possible

- **By applying an electric field, that symmetry is destroyed allowing for SHG**

For SHG, define second order polarization to be: $\boldsymbol{P}^{(2)}(t) = \chi^{(2)}\varepsilon_0\boldsymbol{E}^2(t)$

- $\boldsymbol{P}$: Induced polarization
- χ: Electric Susceptibility
- ε_0: Permittivity of vacuum
- $\boldsymbol{E}$: Electric Field

Assuming an oscillating electromagnetic field of frequency ω:

- $E(t) = E_0 e^{-i\omega t} + E_0^* e^{+i\omega t}$

$\Rightarrow P^{(2)}(t) = \left(2\varepsilon_0\chi^{(2)}E^*E\right) + 2\varepsilon_0\chi^{(2)}\left(E_0^2 e^{-2i\omega t} + c.c.\right)$

E-FISH is described as a third order nonlinear process:

- $P^{(2\omega)} = \frac{3}{2}N\chi^{(3)}(-2\omega, 0, \omega, \omega)E_{Ext}E_{Pump}E_{Pump}$
- $\Rightarrow \boldsymbol{I}^{(2\omega)} = \boldsymbol{A} \cdot \boldsymbol{N}^2(\boldsymbol{E}_{Ext})^2\left(\boldsymbol{I}_{Pump}\right)^2$

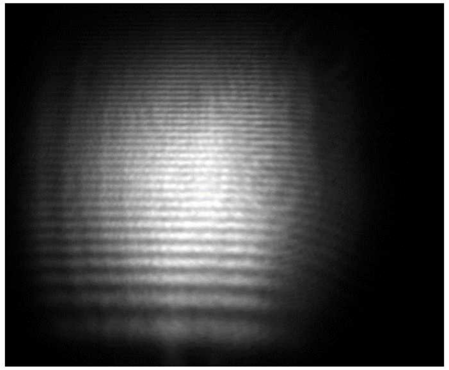
- $P^{(2\omega)}$: Induced polarization at 2ω
- N: Number Density
- $\chi^{(3)}$: Nonlinear Susceptibility (species dependent)
- E_{Ext}: Electric Field to be measured
- E_{Pump}: Electric field of incident laser

Benefits of E-FISH Method:

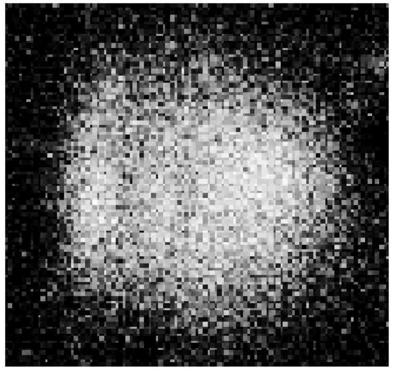
- Signal scales as N^2
- Signal scales as E^2
- Time resolution determined by pump beam duration
- Spatial resolution determined by beam focusing parameters
- Non-resonant method works in any species and gas mixtures

Dogariu, A., *Physical Review Applied*, 7(2), 024024.

800 nm



400 nm



Early E-FISH Work

Initial similar work completed in 1960's and 70's

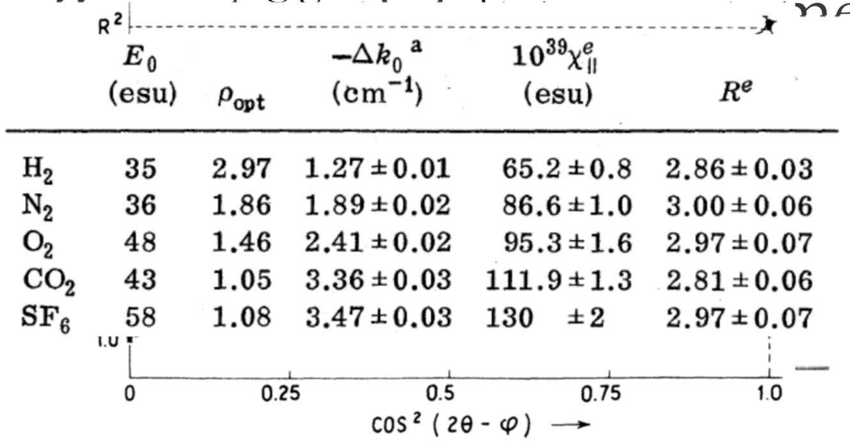
- Used nanosecond pulsed lasers, limiting signal generation
- Measured nonlinear susceptibility polarization components
- Measured nonlinear susceptibilities of gasses against known electric fields
- More recently used for detection of Terahertz waves
- **We use a more general approach to measure an externally applied field**

...nents in plasmas:

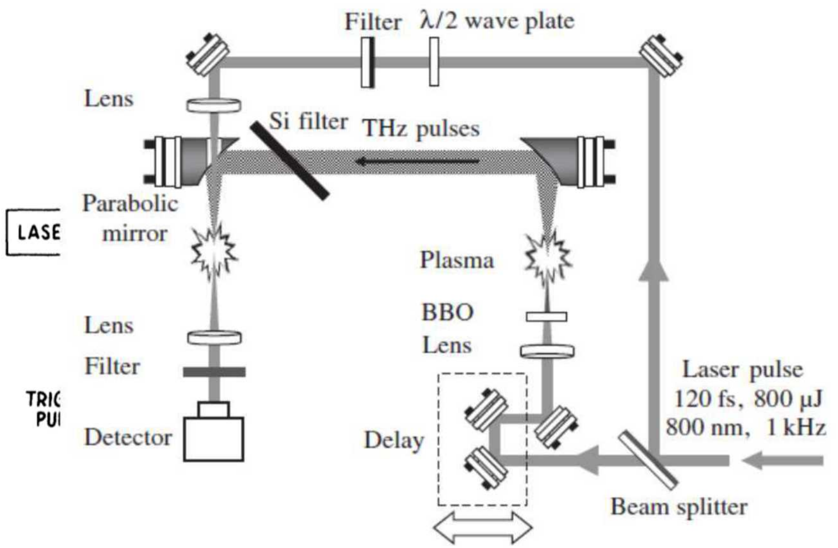
2019

d Flame, 2019

Second Harmonic Signal vs λ/2 Orientation



	E_0 (esu)	ρ_{opt}	$-\Delta k_0$ [a] (cm^{-1})	$10^{39}\chi^e_{\parallel}$ (esu)	R^e
H_2	35	2.97	1.27 ± 0.01	65.2 ± 0.8	2.86 ± 0.03
N_2	36	1.86	1.89 ± 0.02	86.6 ± 1.0	3.00 ± 0.06
O_2	48	1.46	2.41 ± 0.02	95.3 ± 1.6	2.97 ± 0.07
CO_2	43	1.05	3.36 ± 0.03	111.9 ± 1.3	2.81 ± 0.06
SF_6	58	1.08	3.47 ± 0.03	130 ± 2	2.97 ± 0.07



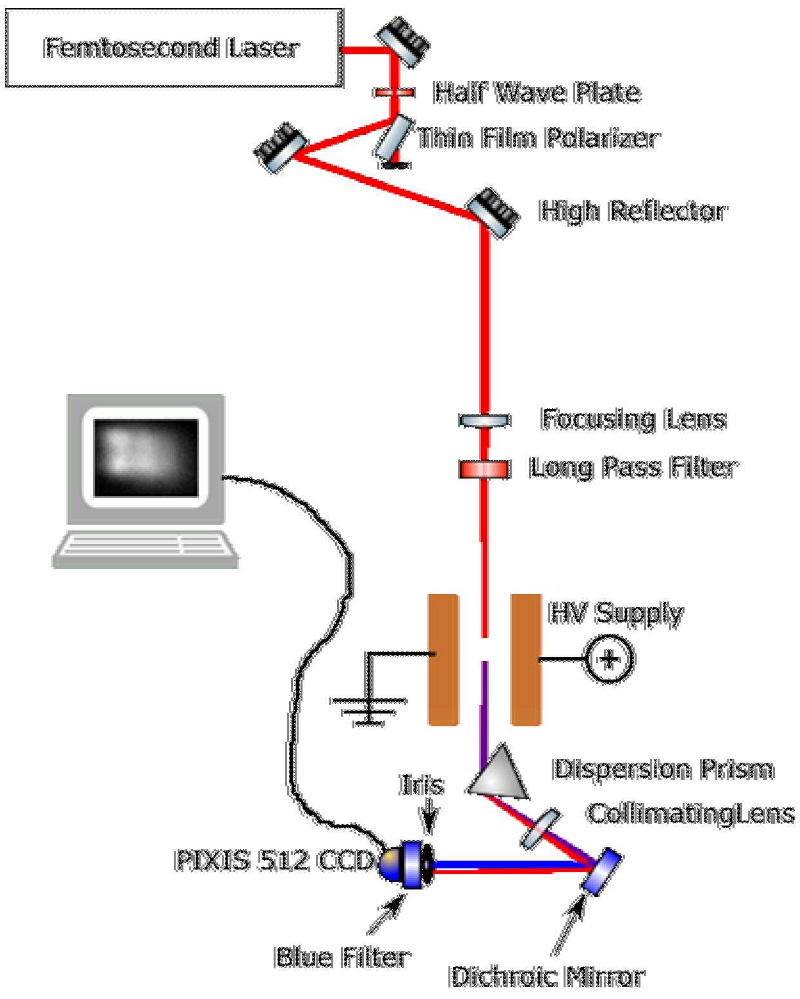
Typical Experimental Schematic

Most experiments utilize either femto- or picosecond lasers
- Fs Laser: 7 mJ per pulse at 1 kHz with 50 fs pulses and ~15 nm bandwidth
- Ps Laser: 30 mJ per pulse at 10 Hz with 100 ps pulses and frequency tunable

Detection accomplished with either PMT or CCD depending upon desired parameters
- 1-D sensitivity has been achieved using cylindrical focusing and ICCD

Key experimental difficulties:
- High intensity lasers generate SHG on upstream optics which needs to be filtered
- White light from multiphoton ionization can damage detection equipment
- Cannot rely upon simple spectrometer to separate pump from SHG signal

Discharges studied:
- Bare metal electrodes in 100 Torr room air
- Atmospheric pressure plasma jet with Argon core flow and N2 co flow
- Dielectric barrier discharges in Fuel and Oxygen containing mixtures



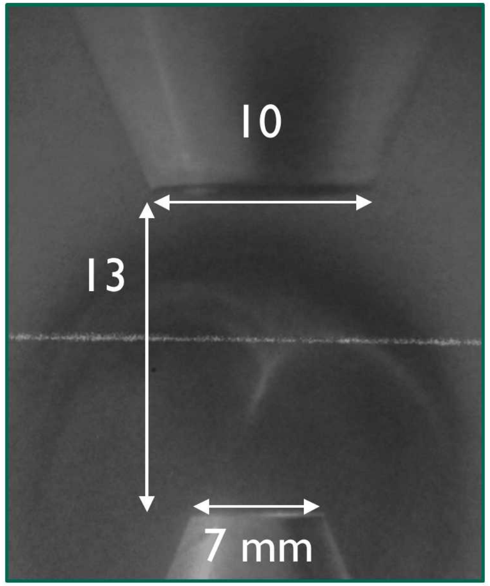
Measurements with Picosecond Laser in 100 Torr Room Air

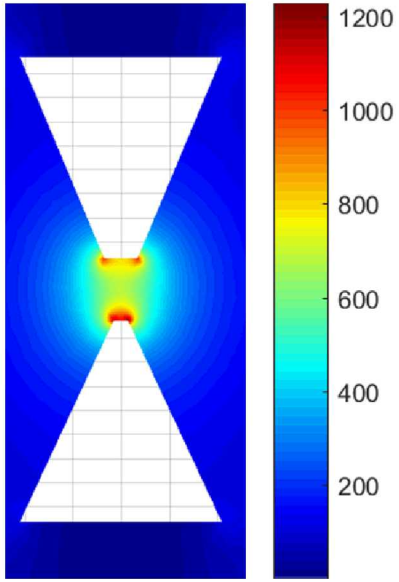
Used different electrode cross sections to increase local field at anode

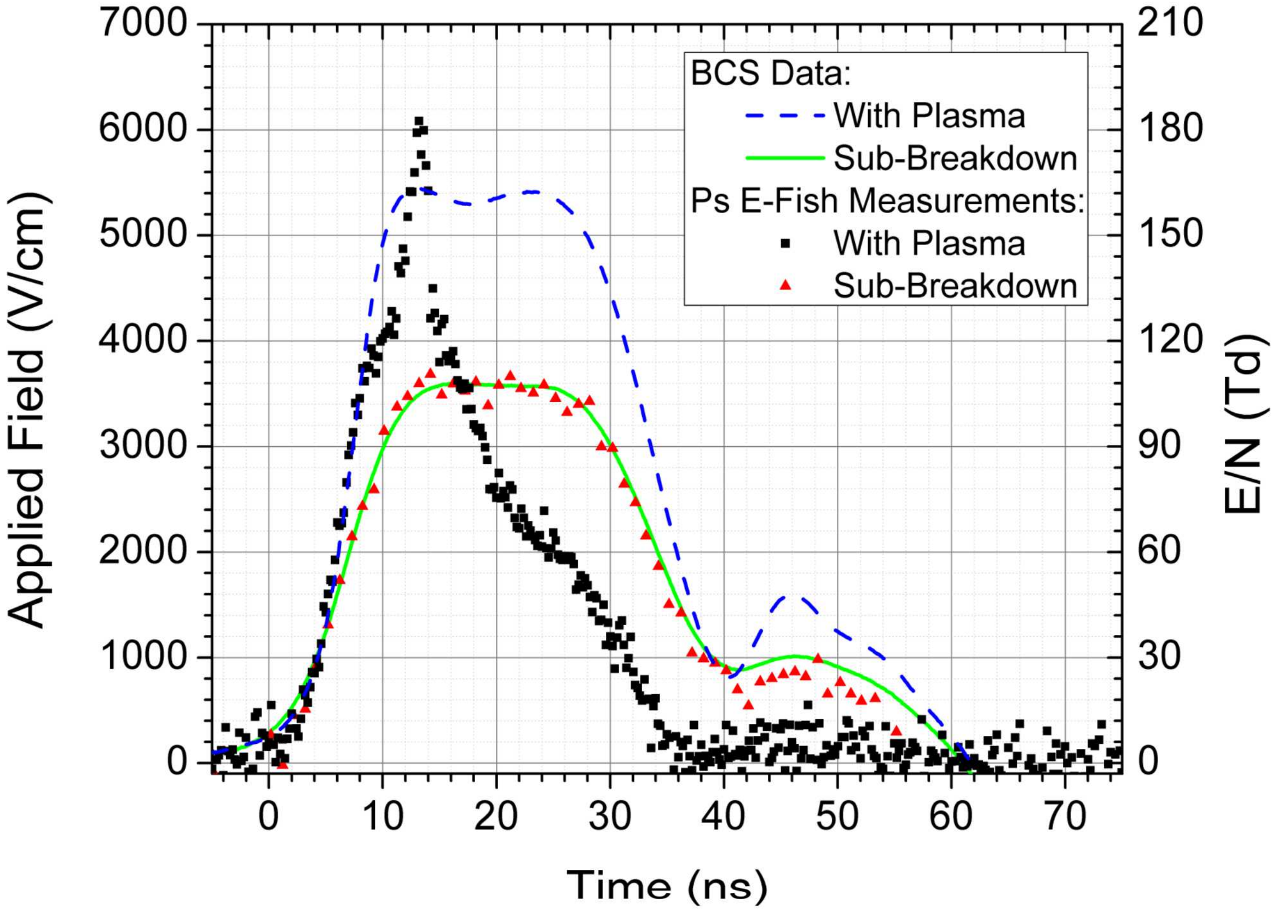
- Probe region is center of discharge gap

Electric field follows the applied voltage until breakdown occurs

- Kink in field corresponds to initiation of breakdown
- Field continues to rise as plasma fills the gap
- Field begins to fall once gap is filled with highly conductive plasma





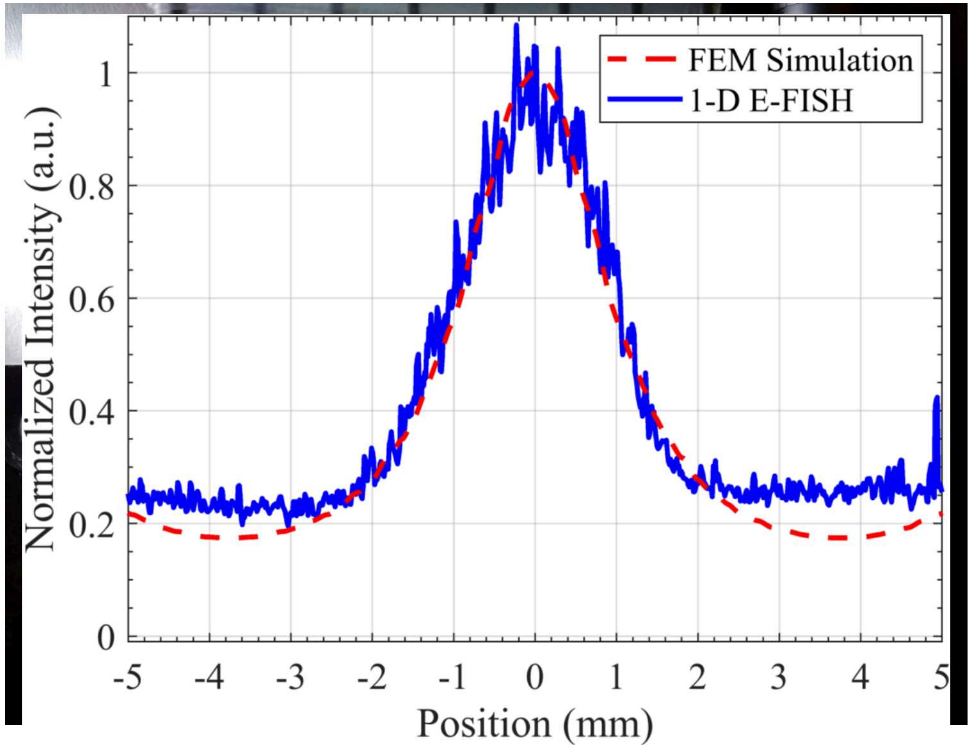


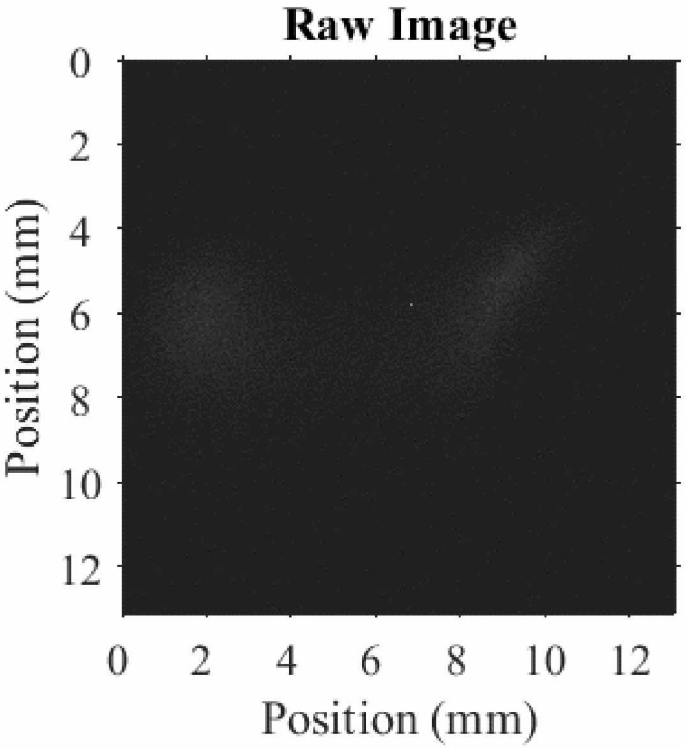
B. M. Goldberg et al, *Applied Physics Letters,* 2018

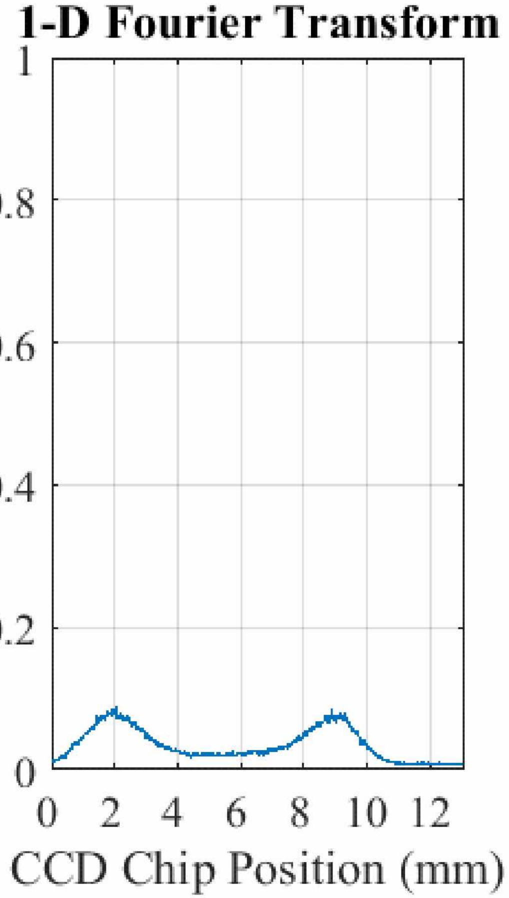
Cylindrical Focusing for 1-D Sensitivity

Alternatively, we can focus into a laser sheet to reduce the net intensity

- Using a camera for detection allows for 1-D measurement sensitivity
- Detect SHG signal using PI-MAX4 ICCD with pixels size of 12.8 x 12.8 μm
- Scanned a localized field across laser sheet to determine spatial sensitivity





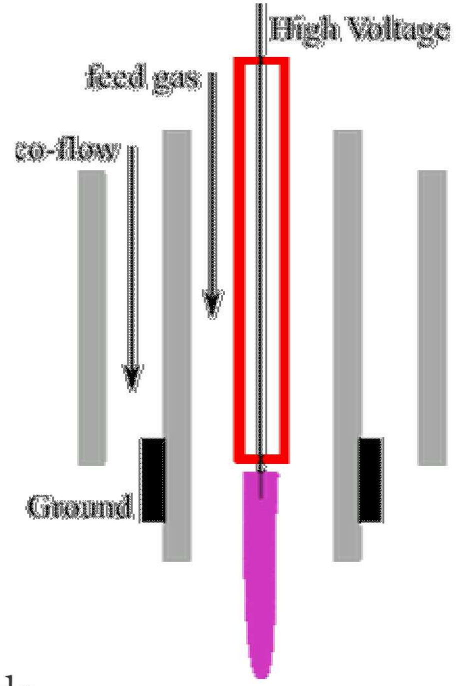


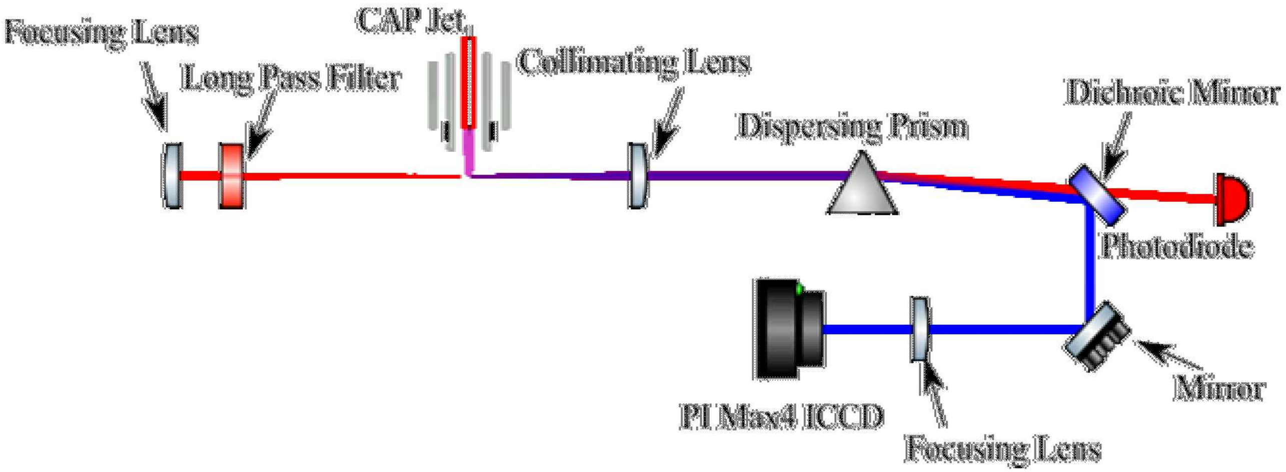
Resolution is ~50 μm

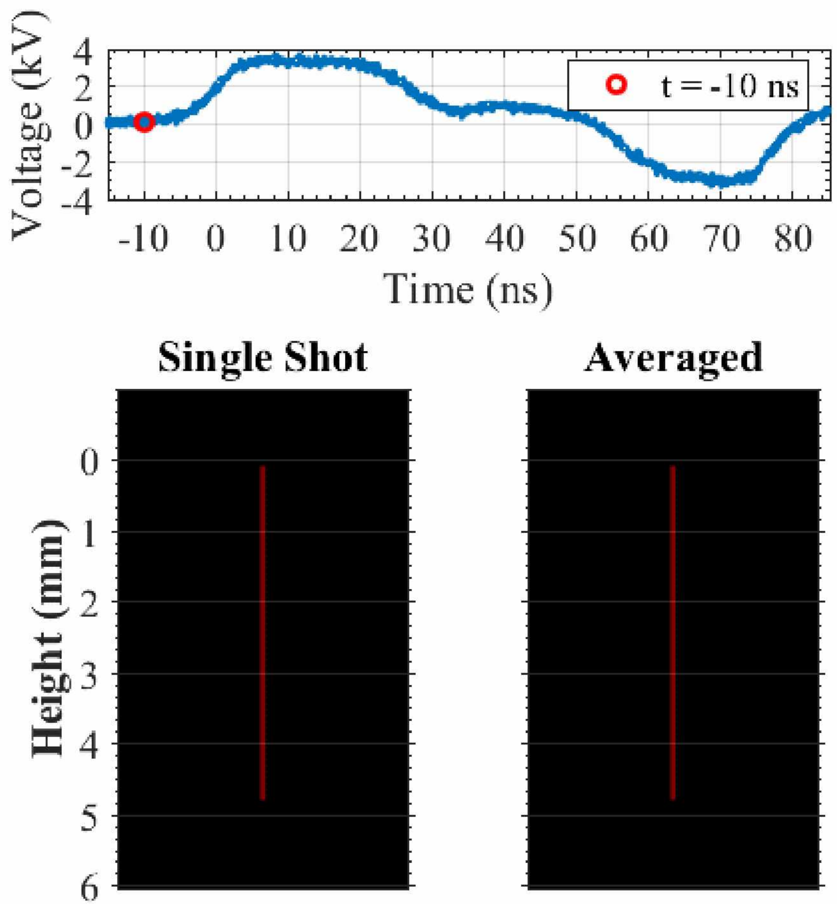
Cold Atmospheric Pressure Plasma Jet

Laminar Argon core flow with N_2 co-flow
- Streamer front dimensions are ~30-100 μm
- Plasma travels as an ionization wave, creating spatially and temporally evolving fields
- Measurement volume: 10 μm x 220 μm x 2 cm

FID GmbH FPG 20-10 NM
- 3.7 kV applied with FWHM of ~30 ns
- Run at 1 kHz synchronized with fs laser
- Time t=0 defined to be ½ peak voltage amplitude







3 ns ICCD gate with 1 ns time steps

Red line represents laser probe region

CAP Calibration Routine

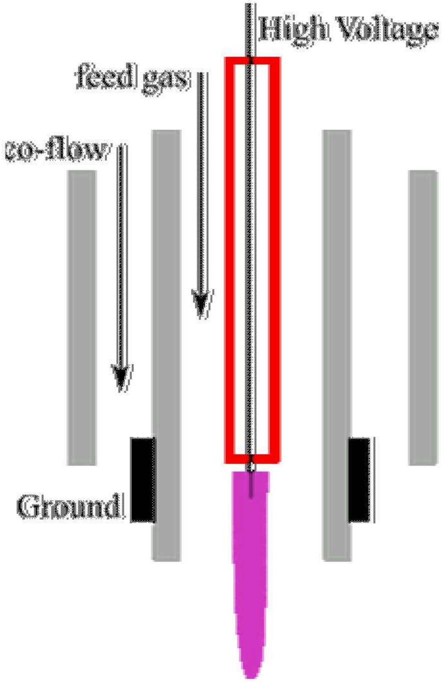
CAP does not have a well defined ground plane making calibration difficult

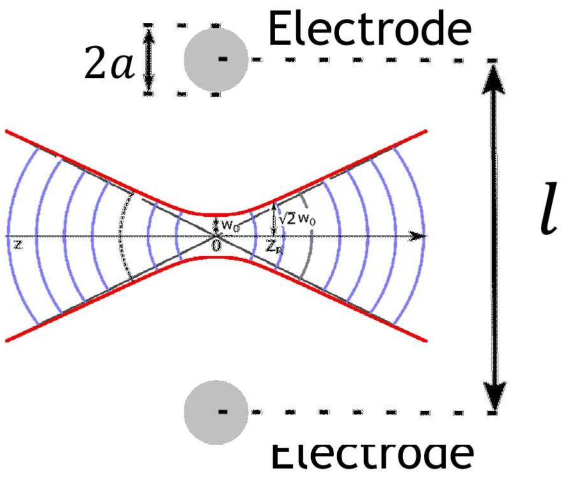
Key concerns:

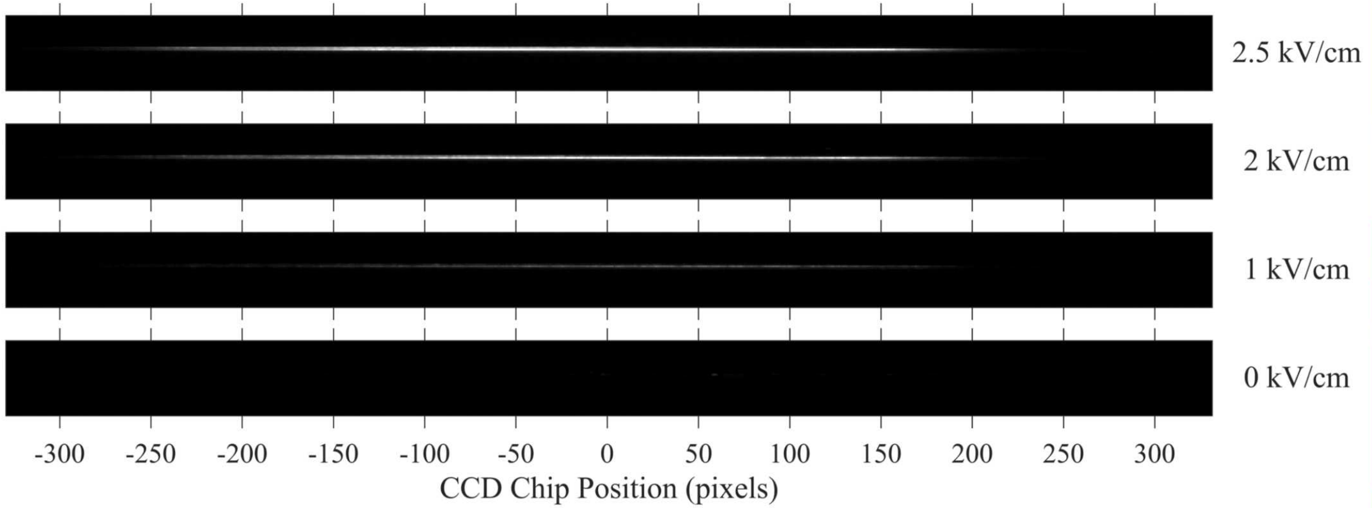
- Different gas mixture fractions
- Localized field enhancements due to dielectrics
- "Extent" of calibration field compare with field due to plasma jet
- Systematic errors such as Gaussian pump beam intensity

Use cylindrical electrodes using short focal length lens

- Measured and accounted for gas species using Rayleigh scattering
- Removed jet output to remove dielectrics
- Measured confocal parameter to be 220 μm and coherence length to be ~1 cm
- Pixel-by-pixel calibration map





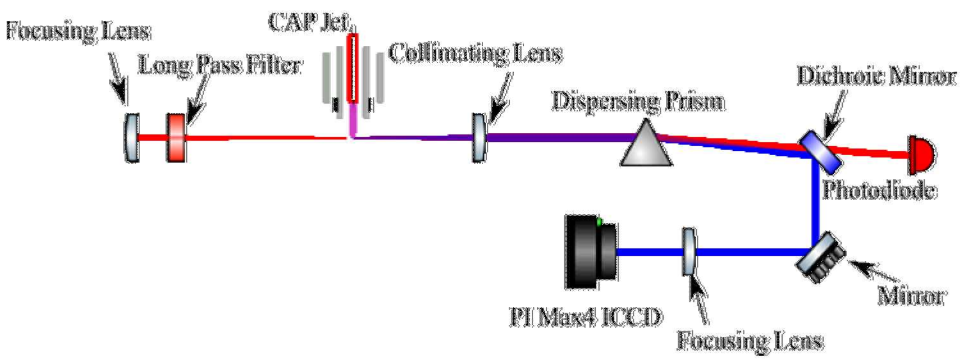


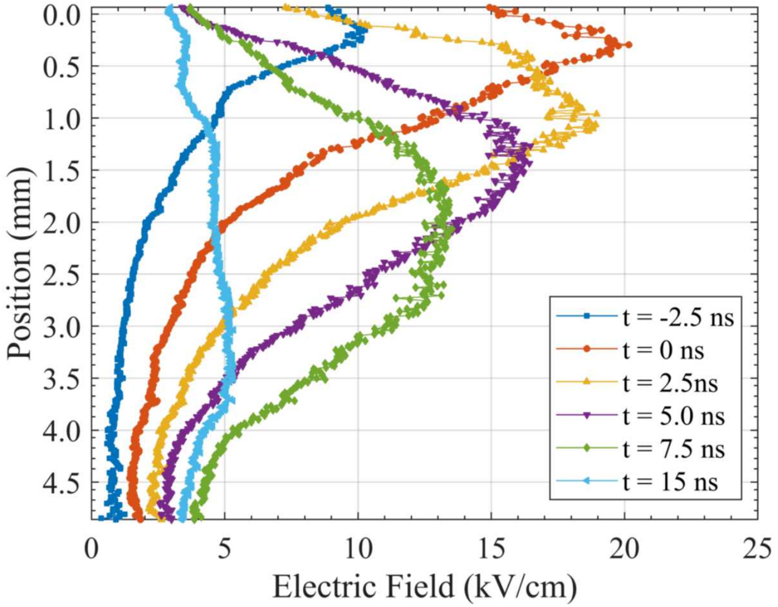
Time Resolved Electric Field Measurements

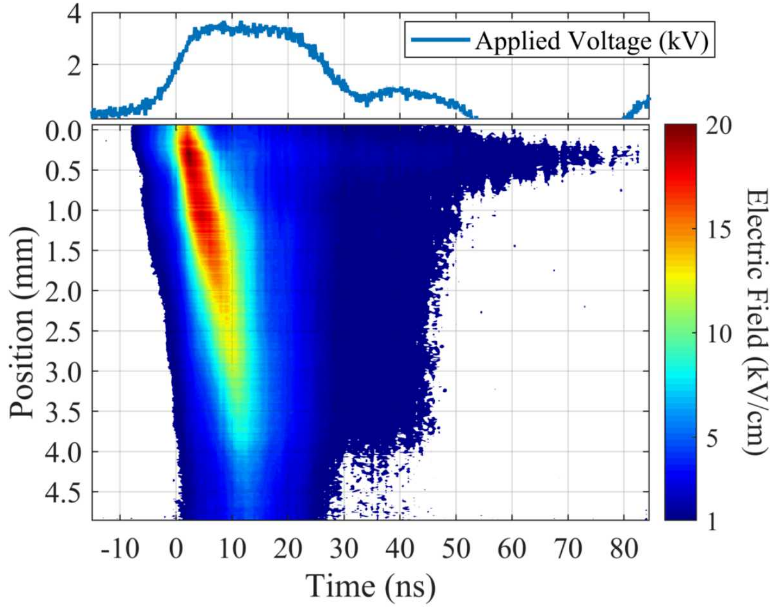
Individual laser shots collected and analyzed in a post processing routine
- Time bins of 500 ps used
- ~100 shots per bin

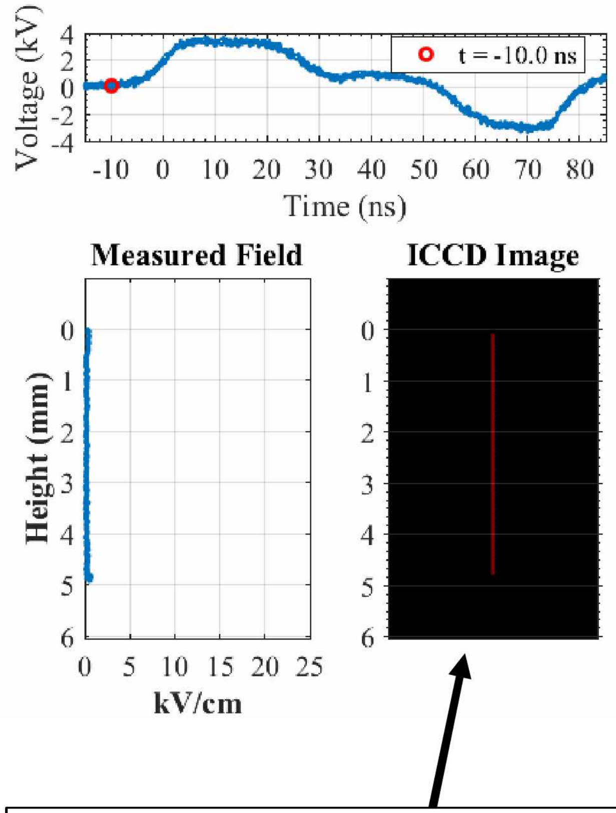
Ionization wave propagation clearly visible
- Wave speed ~0.3 mm/ns, consistent with ICCD images

Peak field ~20 kV/cm, much lower than expected
- Likely due to spatial sensitivity as plasma filaments move



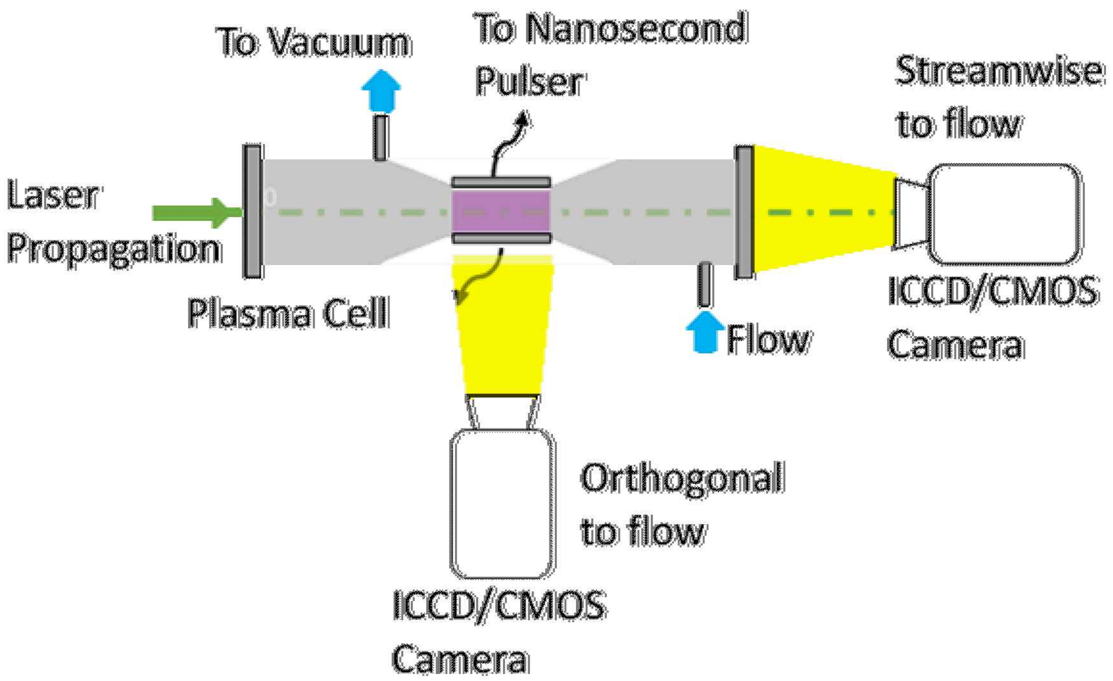






3 ns gate with 1 ns time steps
- Averages represent 16 images
- Red line represents laser probe

DBD with Fuel and Oxidizer

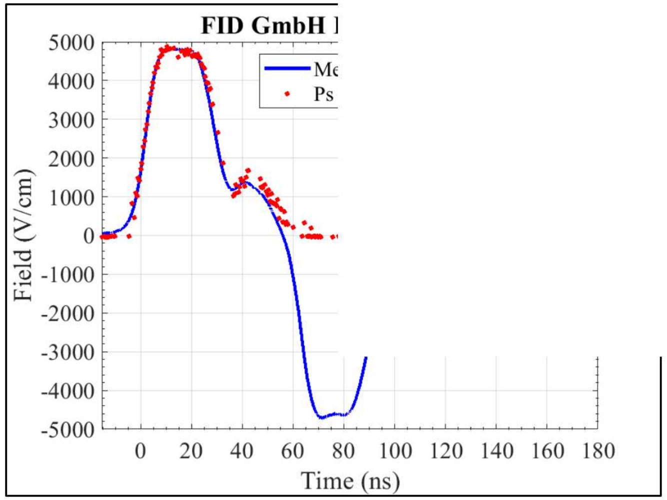


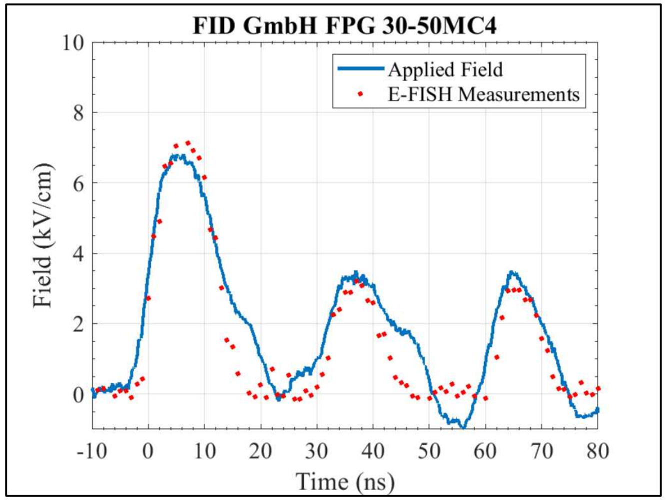
Reactor

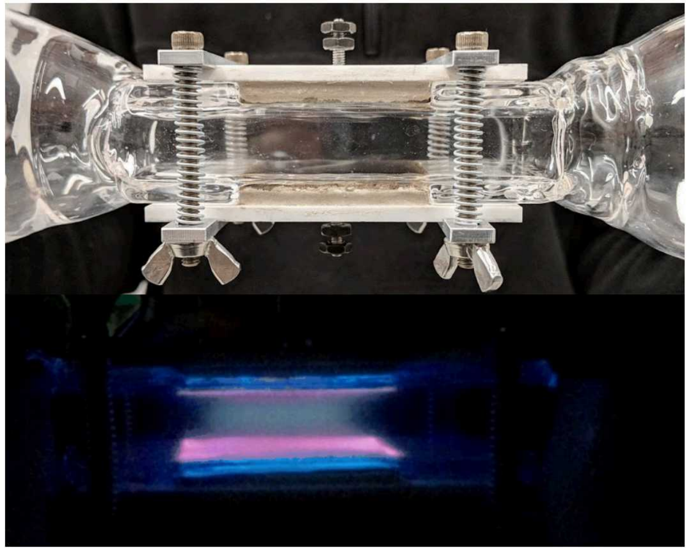
- Pressure: 60 Torr
- Initial Temperature: 293 K
- Gap distance: 14 mm
- Quartz double dielectric barrier: 1.6 mm
- Argon bath gas with CH_4 and/or O_2

Two pulsers used:

- FID GmbH FPG 20-10 NM
  - Applied Voltage: ~7 kV
  - Pulse Duration: ~30 ns FWHM
  - Continuous 1 kHz synchronized with fs laser
- FID GmbH FPG 30-50MC4
  - Peak Voltage: ~10 kV
  - Pulse Duration: ~12 ns FWHM
  - Pulse burst mode: 150 pulses, 30 kHz

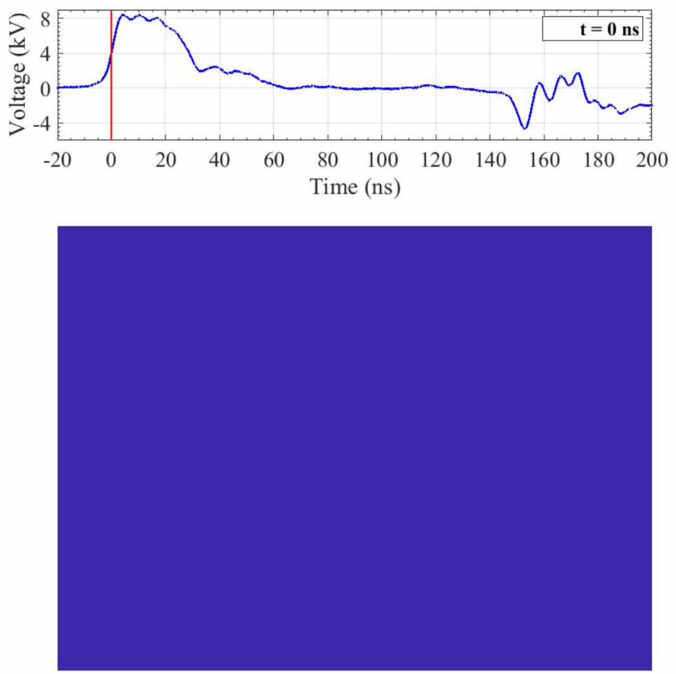




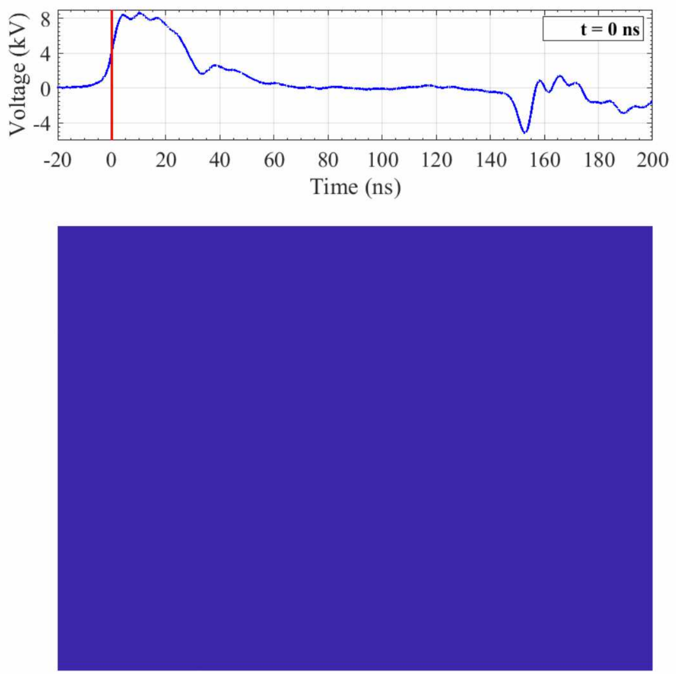


1 kHz – Time Resolved Imaging

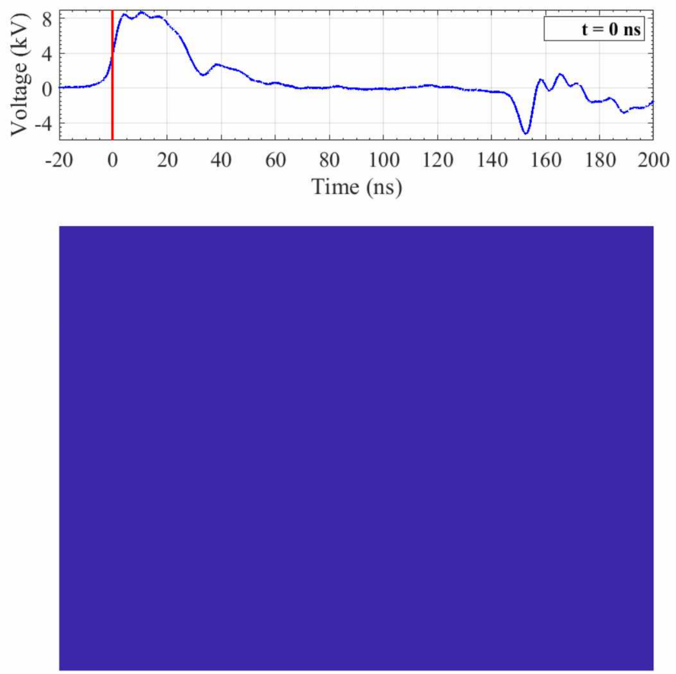
0.9 Ar + 0.1 CH_4



0.8 Ar + 0.2 O_2



0.7 Ar + 0.1 CH_4 + 0.2 O_2



Averages of 5 shots per frame

Plasmas appear to be uniform, although radial field appears as well

Breakdown occurs on reflected pulse as well

Camera gate: 1 ns

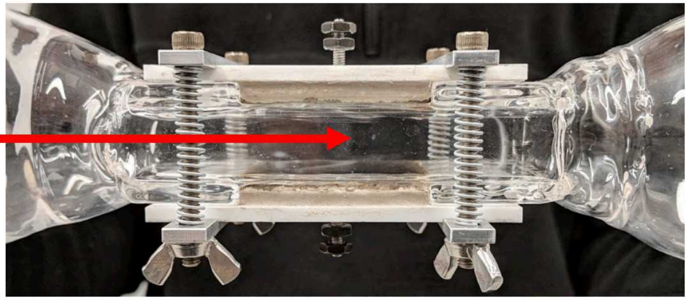
All videos on same intensity scale

1 kHz – Calibration Results

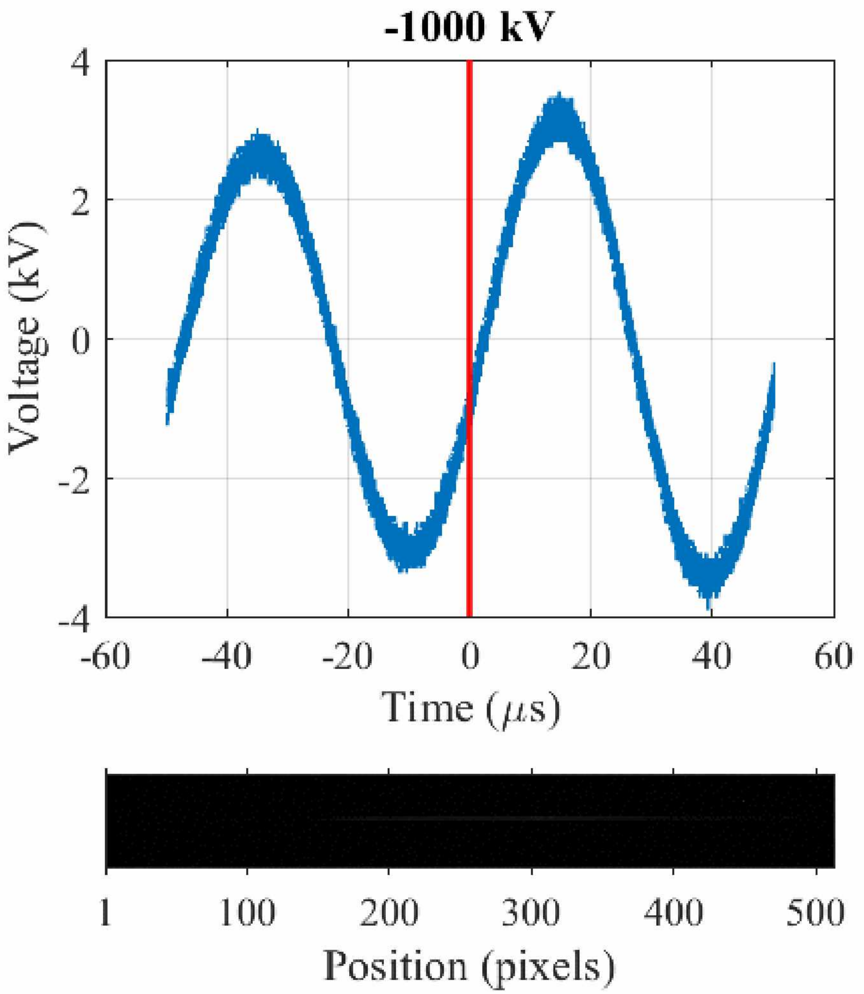
NOTE - PIXIS 512 CCD Used with 500 ms gate

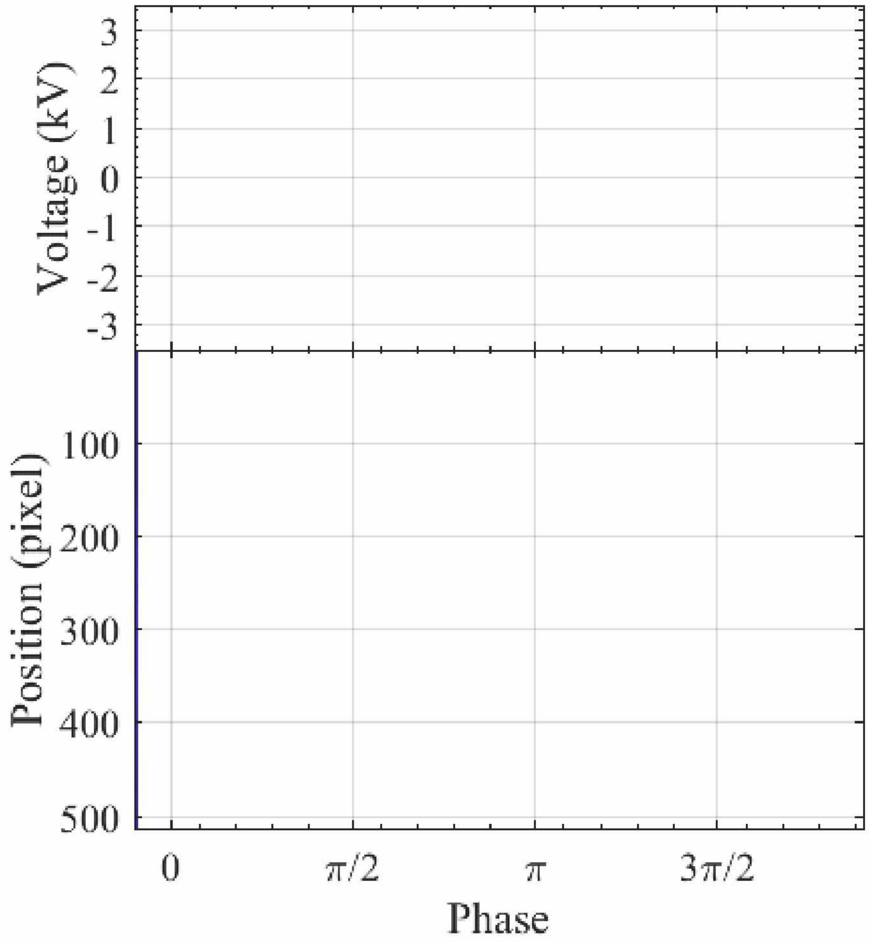
$I^{(2\omega)} \propto A \cdot N^2 (E_{Ext})^2 \left(I_{Pump}\right)^2$

Laser Propagation



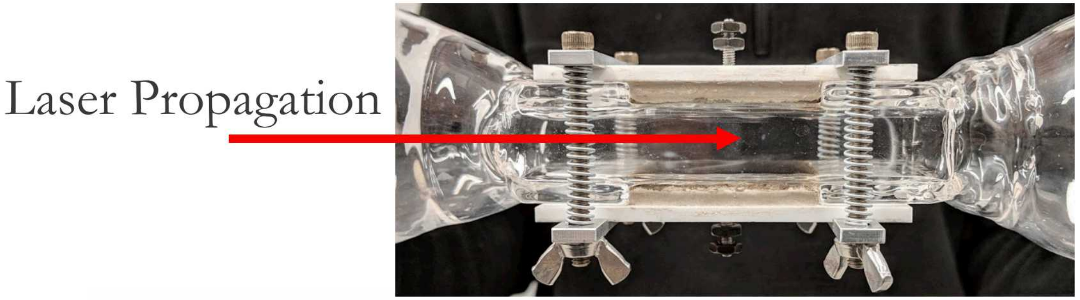
Signal sensitive to E_{Ext}^2 → can not determine +/- field



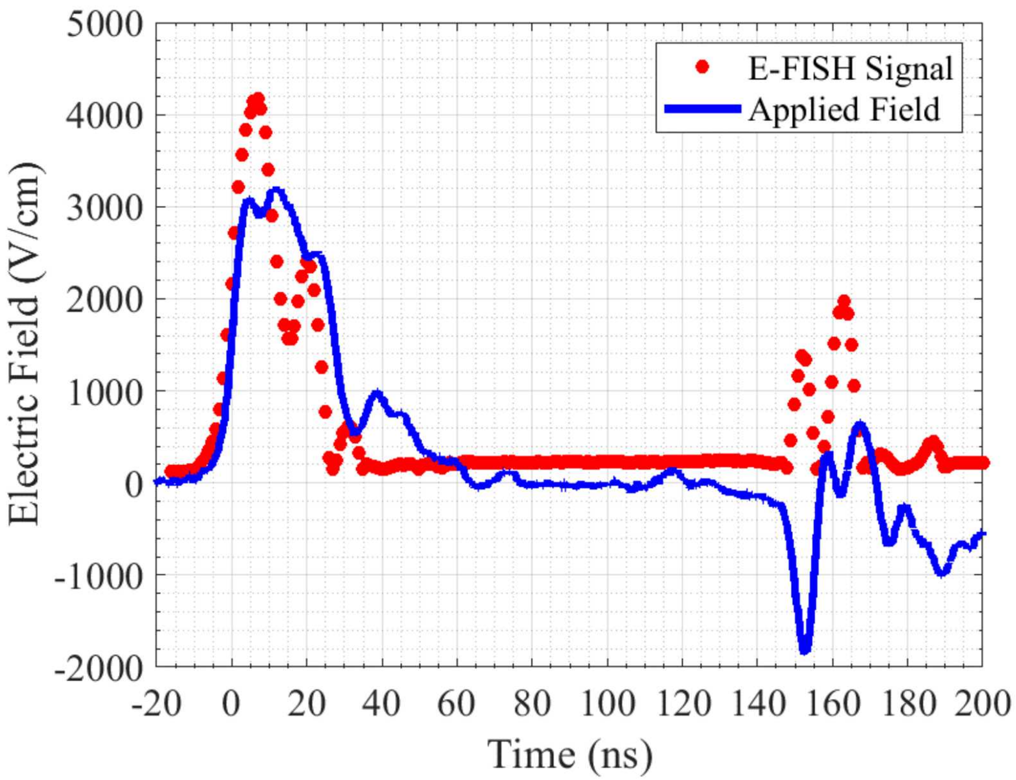


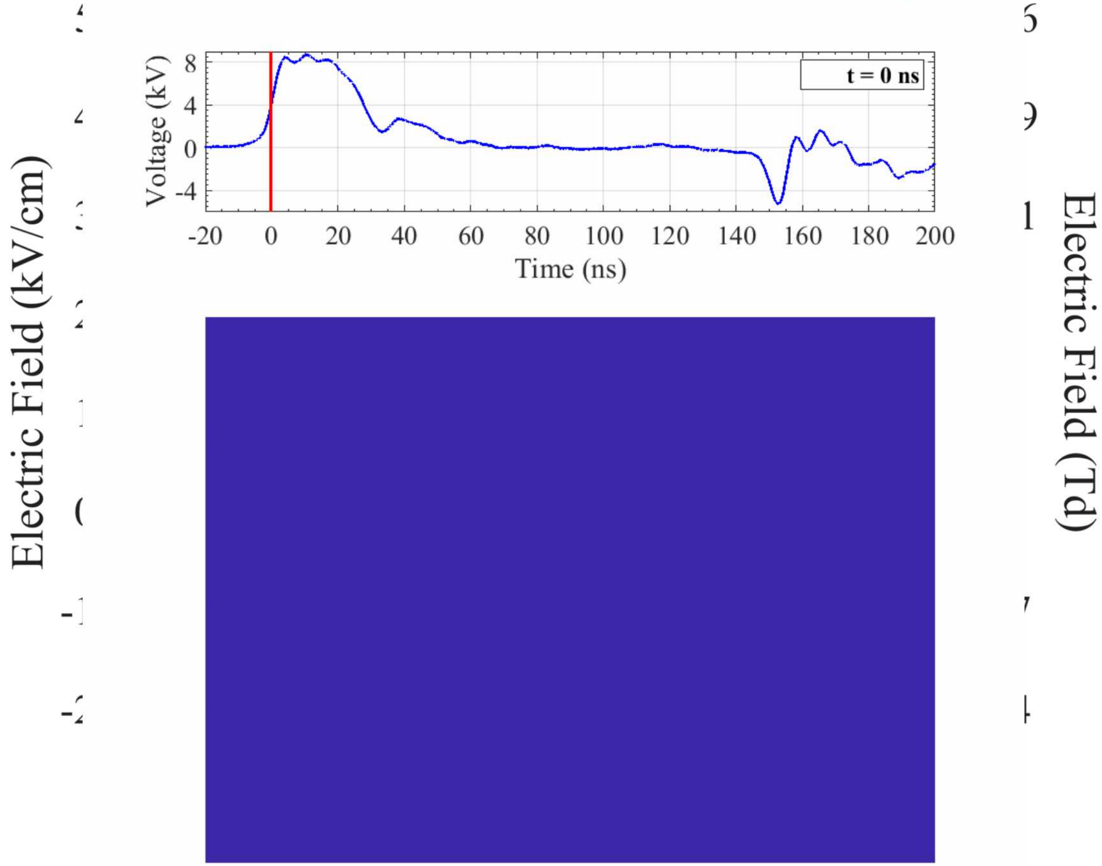
1 kHz – Electric Field Results

NOTE - PIXIS 512 CCD Used with 500 ms gate



0.7 Ar + 0.1 CH_4 + 0.2 O_2



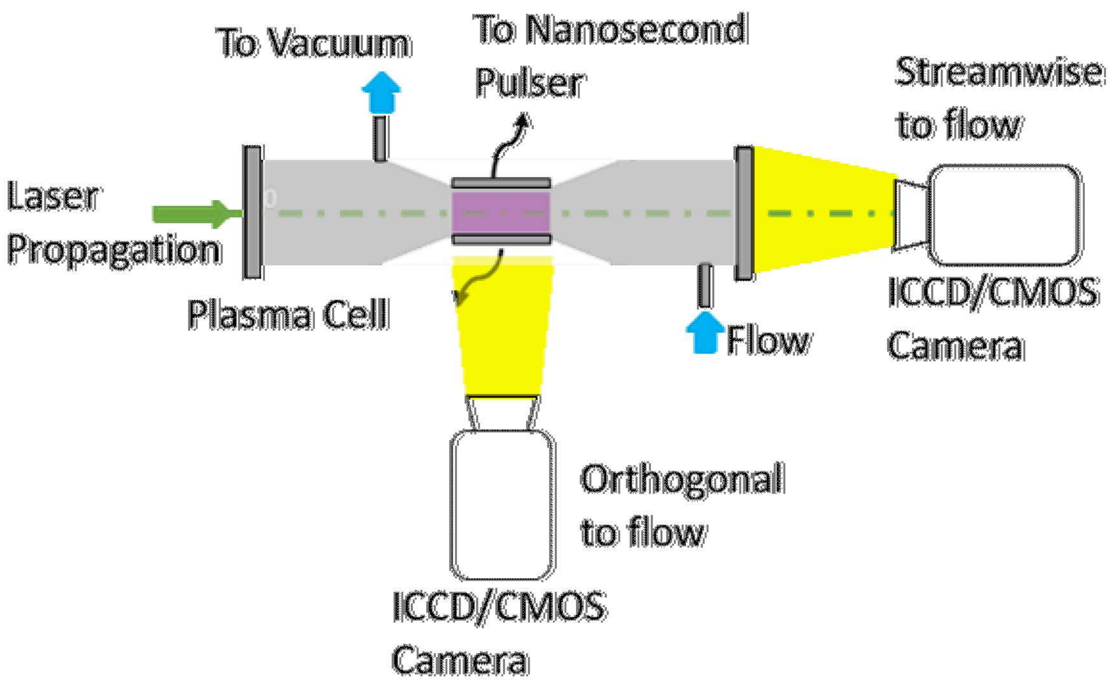


Burst Mode - Time Integrated Imaging

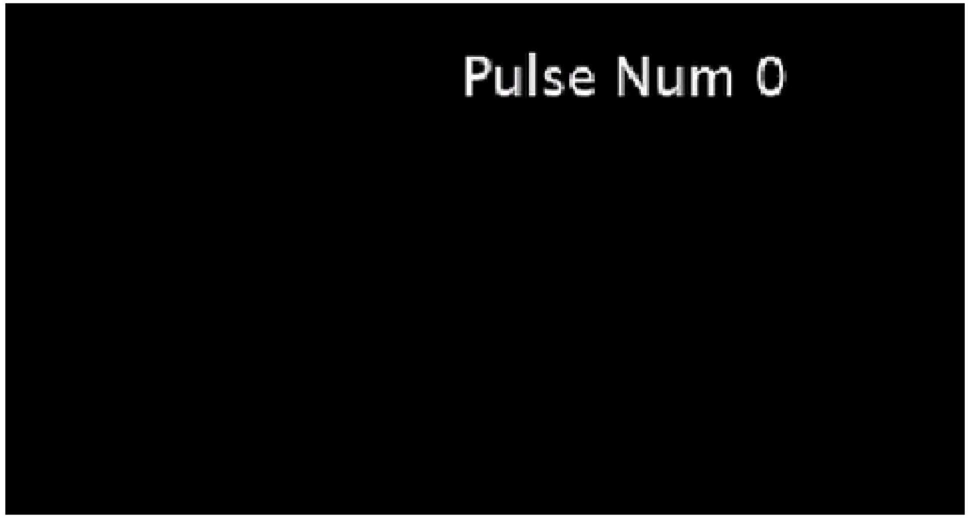
Using high speed sCMOS cameras, we can image each pulse in a burst

Filaments begin forming around pulse 100

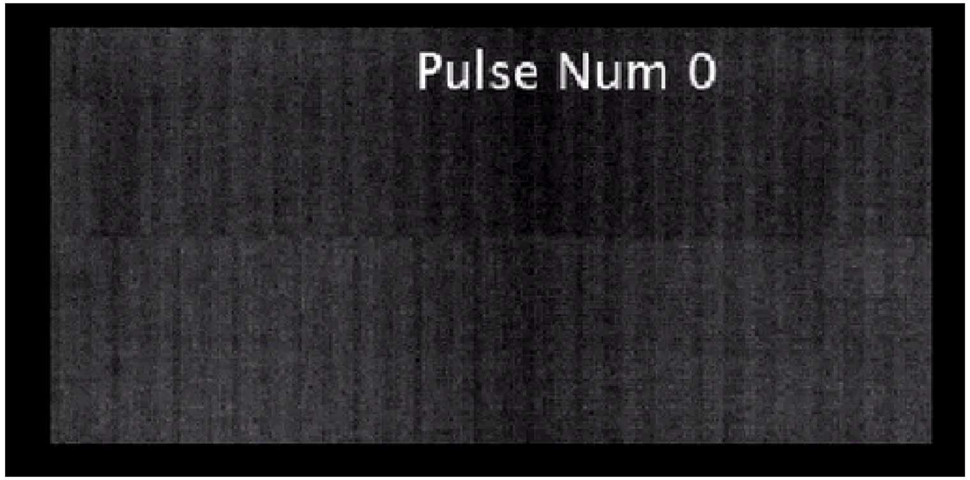
- Within a given burst, filaments are stationary
- ****Note** movement in flow direction due to convection**



6.6% CH_4 13.4% O_2 80% Ar
End View: Streamwise direction

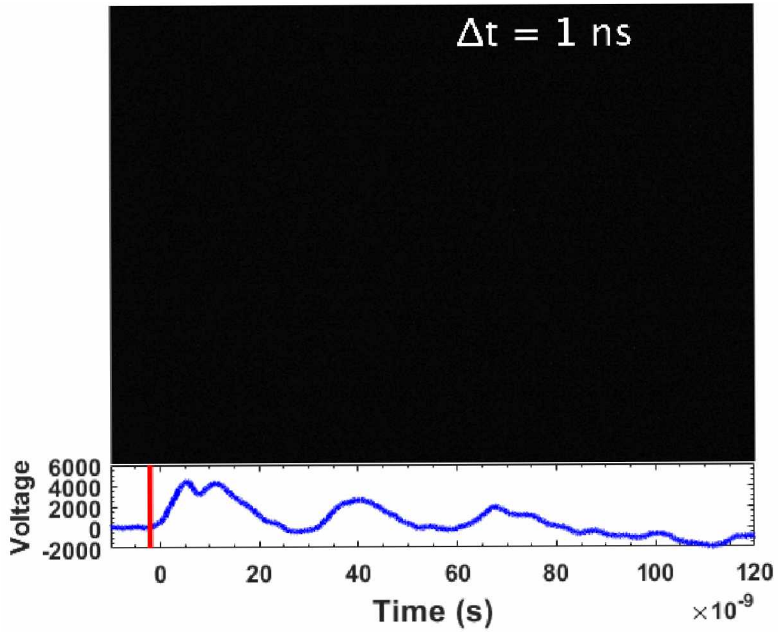


Side View: Orthogonal direction

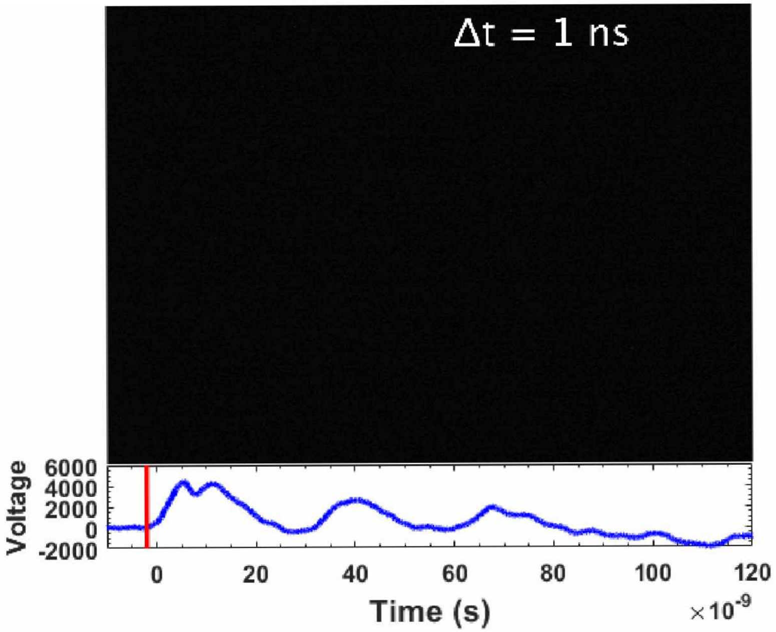


Burst Mode - Time Resolved Imaging

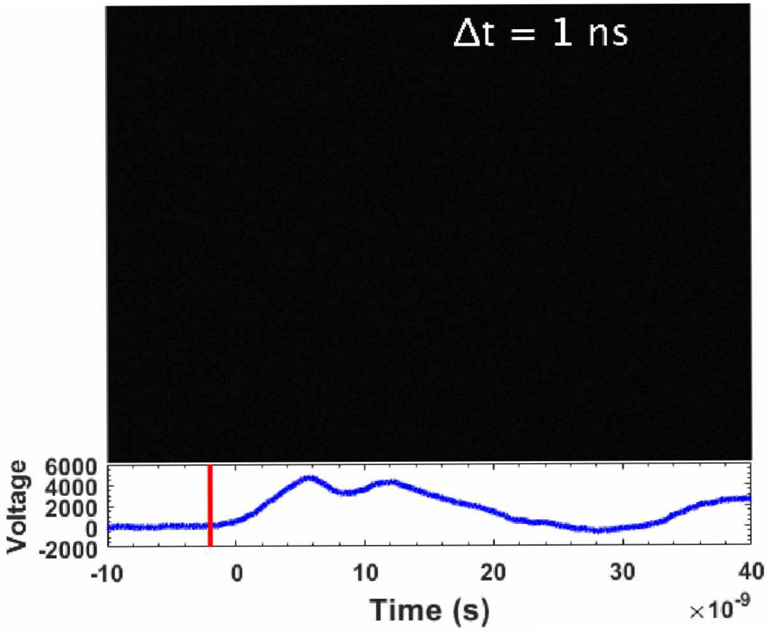
93.3% Ar + 6.6% CH_4
Pulse 150



86% Ar 13% O_2
Pulse 150



6.6% CH_4 13.4% O_2 80% Ar
Pulse 150

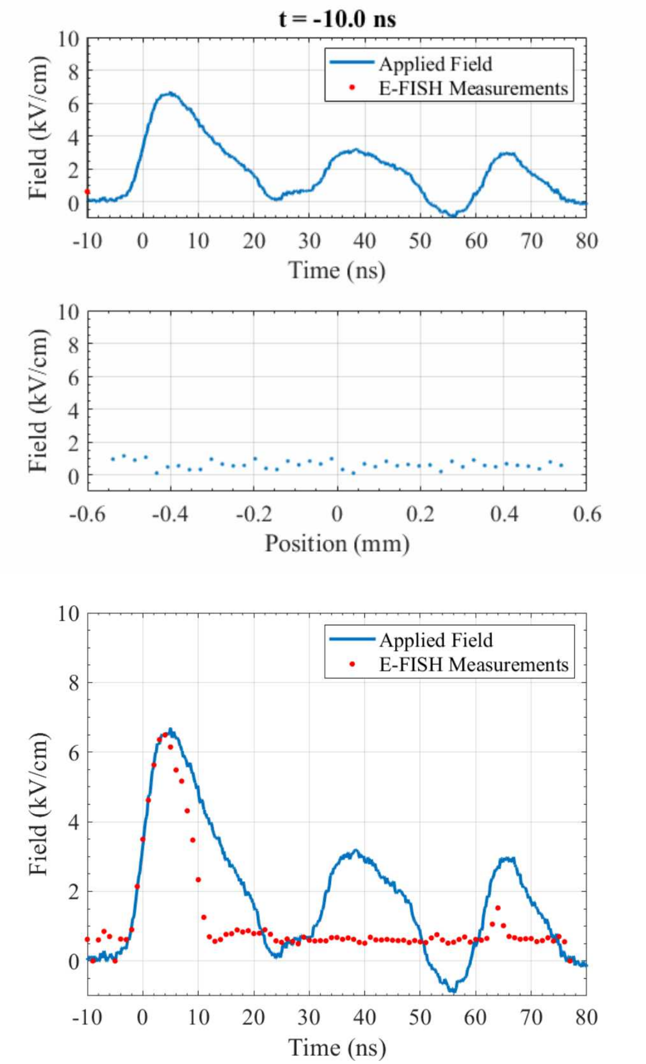


Nanosecond resolution of single pulses
- 1 ns gate, but only 10 Hz camera repetition rate
- Image not from the same burst: uncorrelated shot to shot

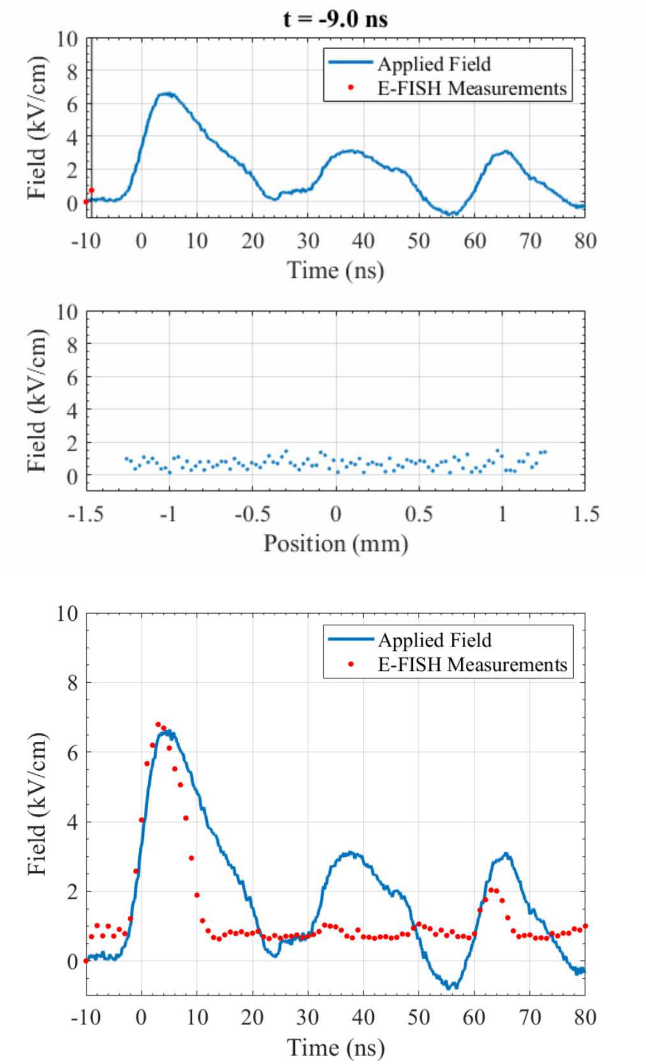
Diffuse discharge with just O_2 or CH_4 addition, but significant filamentation with both present
- Filaments stationary within a given burst, but move from burst to burst

Burst Mode – Averaged Electric Field Results

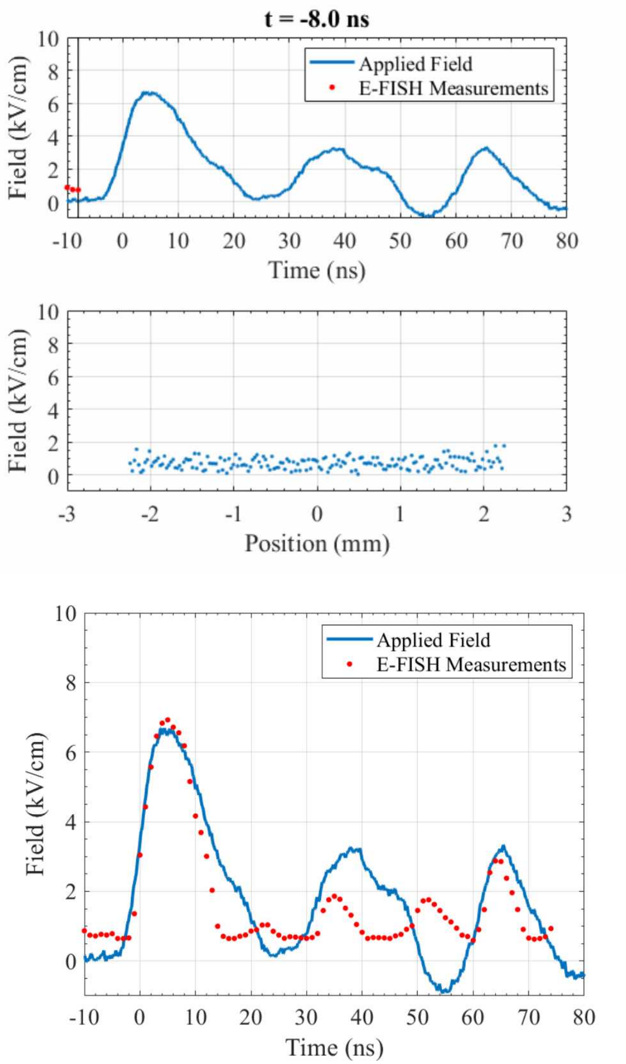
93.3% Ar + 6.6% CH_4
Pulse 150



86% Ar 13% O_2
Pulse 150



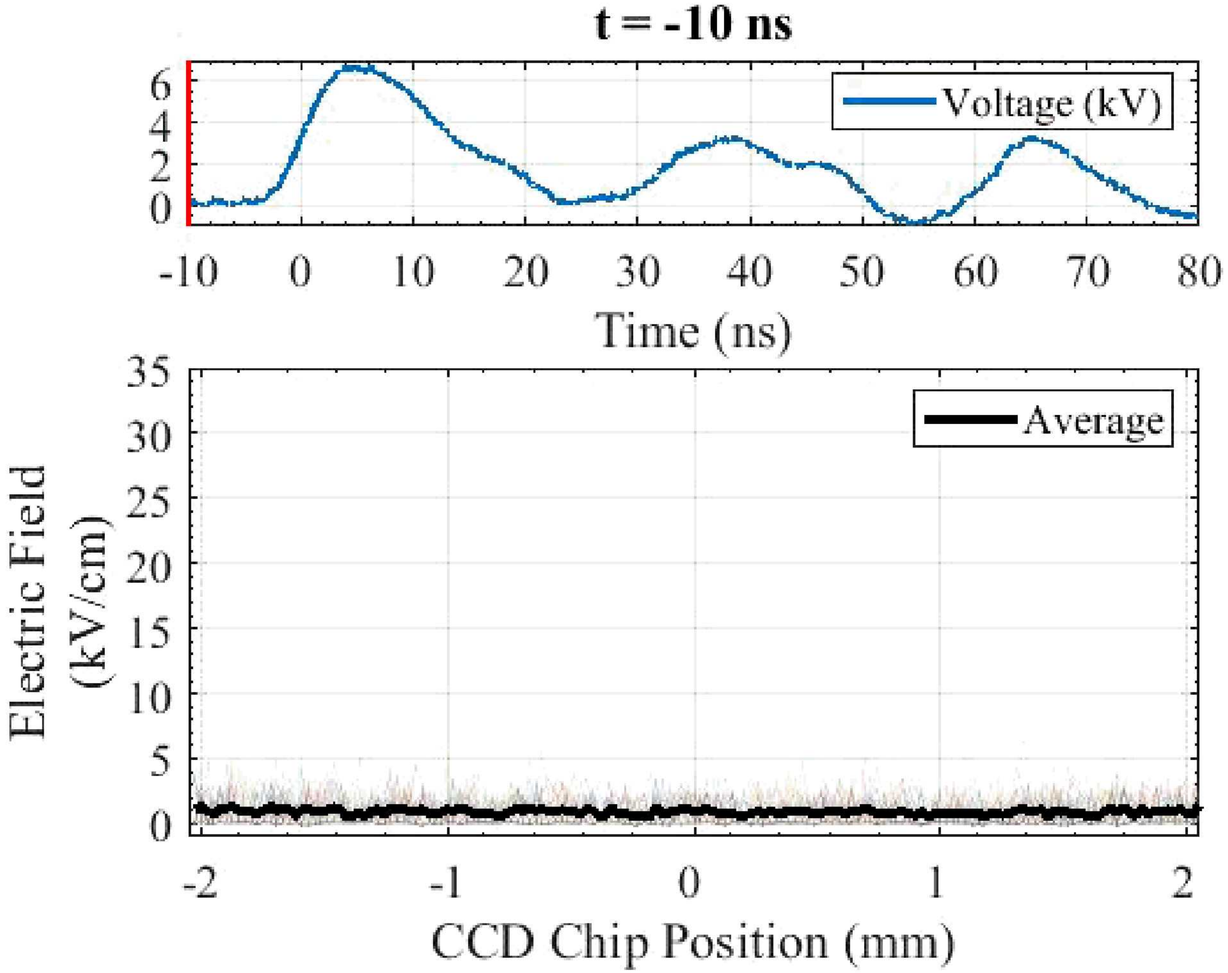
6.6% CH_4 13.4% O_2 80% Ar
Pulse 150



Single Shot Measurement Capability

Data presented was an average of ~100 individual laser shots

- We can look at single shot data for streamer formation!



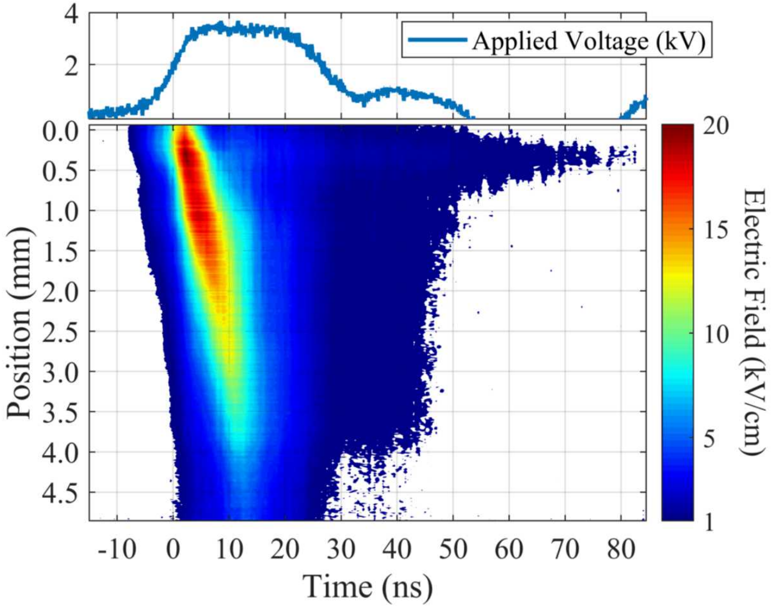
Conclusions

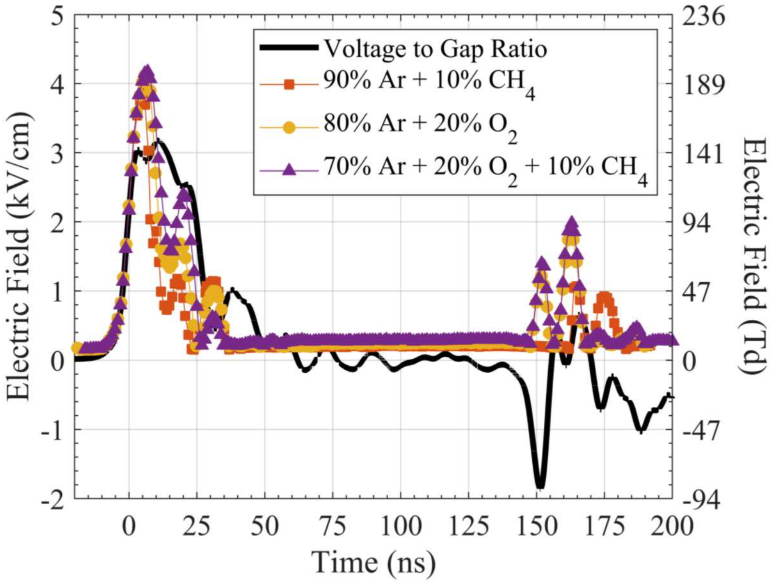
Electric field induced second harmonic generation is a unique method for non-invasive electric field measurements

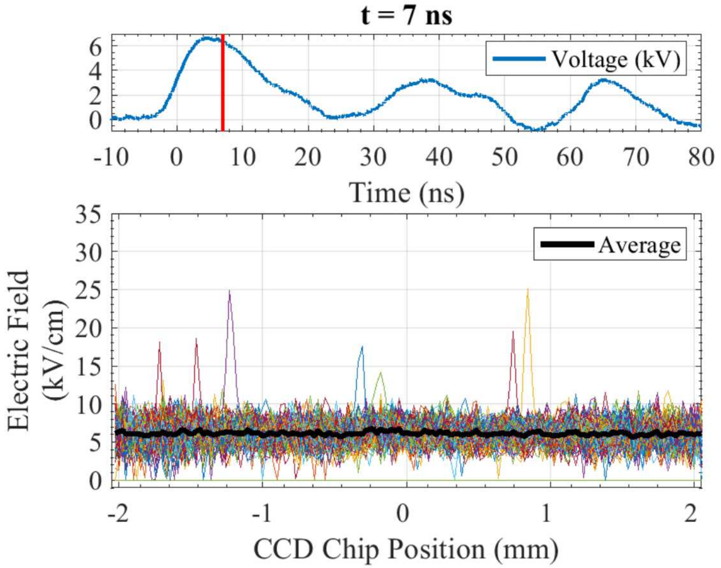
- Key benefits include: time response, field vector sensitivity, and species independent measurements
- Measurements have been demonstrated using femtosecond and picosecond lasers

Cylindrical focusing can be used for 1-D spatial sensitivity

- Measurements in a cold atmospheric plasma jet demonstrated an ionization wave travelling with ~0.3 mm/ns
- Measurements in DBD demonstrated a uniform diffuse discharge
- Single shot measurements can be used to observe localized streamer formation for plasma instabilities







Current Work – Looking at gas surface interactions using SFG

SHG is a specialized form of **S**um **F**requency **G**eneration

- At the surface interface, inversion symmetry is broken, allowing for SFG
- Used to measure vibrational spectra of molecules at interfaces

$$I_{SFG} \propto \left| \underbrace{\left|\chi_R^{(2)}\right| e^{i\delta}}_{\text{Resonant Contribution}} + \underbrace{\left|\chi_{NR}^{(2)}\right| e^{i\varepsilon}}_{\text{Non-resonant Contribution}} \right|^2$$

Key benefits to SFG:

- Signal generation localized to interfacial molecules between gaseous phase and bulk media
- Can measure the degree of order of interfacial molecules
- Can measure the orientation of the molecul
- Can determine the average tilt angle of the interfacial molecules

$$\chi_R^{(2)} = \frac{B}{\omega_v - \omega_{IR} - i\Gamma}$$

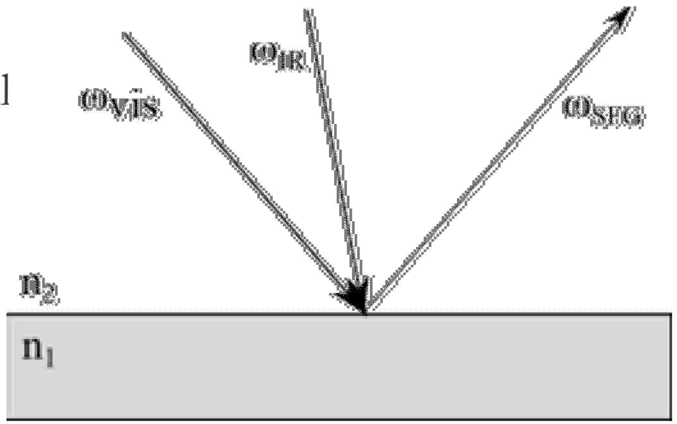
$\chi_R^{(2)}$: Resonant susceptibility

B: Strength of vibrational mode

ω_v:Frequency of mode

ω_{IR}: Probe Frequency

Γ: Resonant linewidth



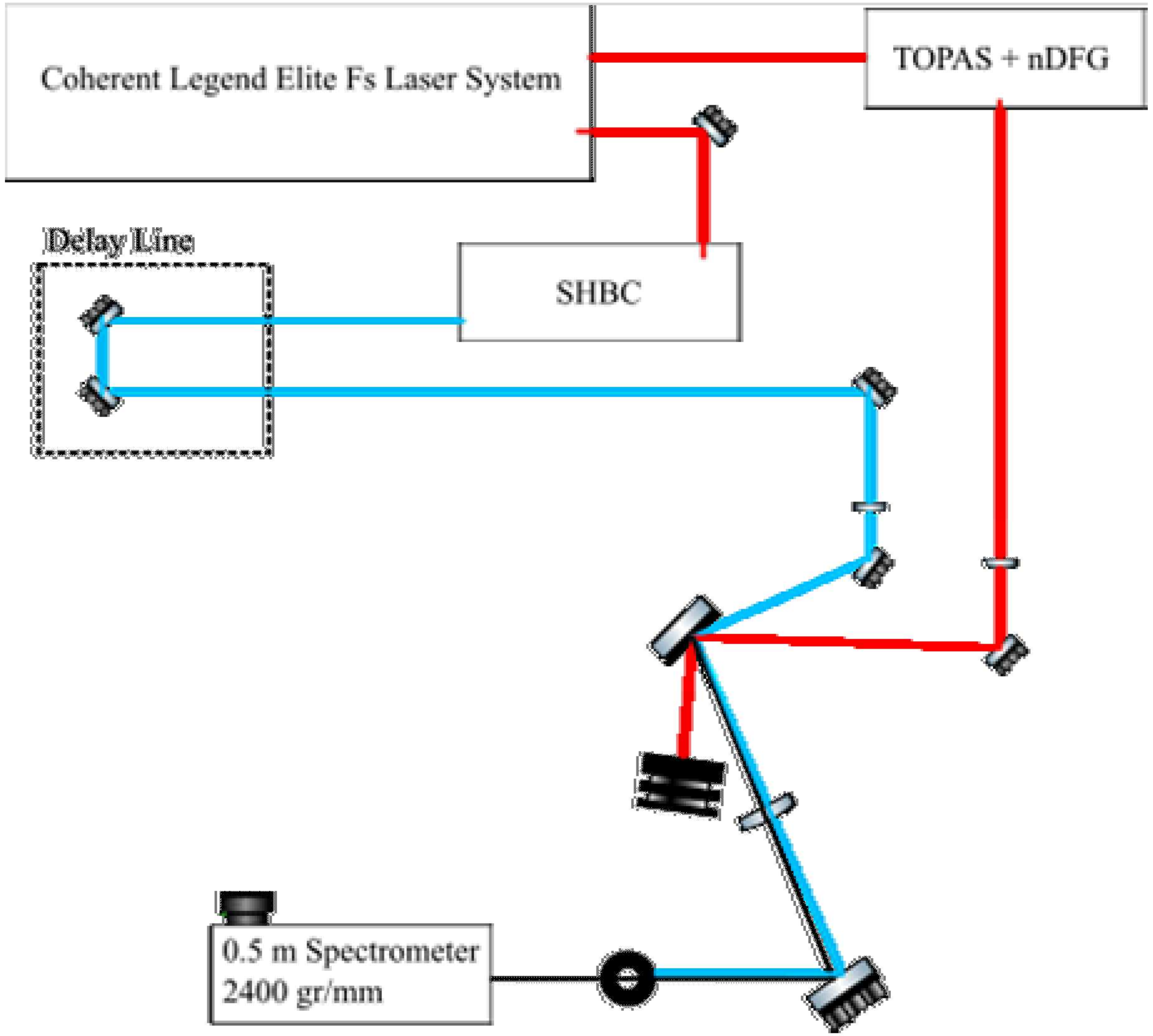
Resonant enhancement occurs when $\omega_{IR} = \omega_v$

Experimental Schematic for SFG

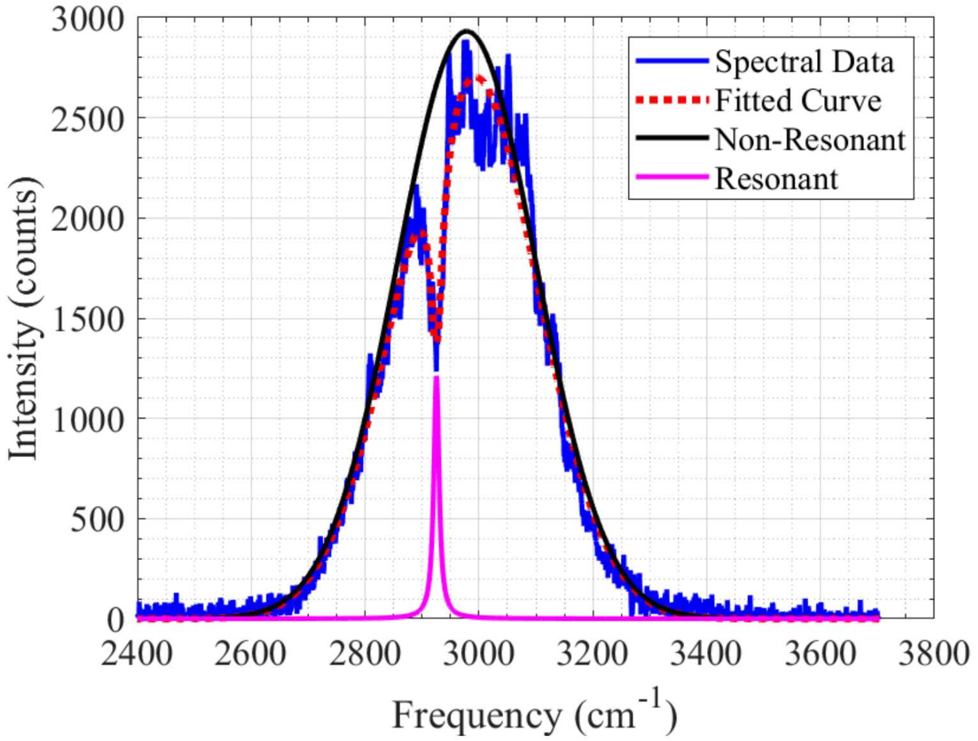
Coherent Legend Elite FS laser system

- Use 1 leg to pump a Light Conversion High Energy TOPAS + nDFG
  - frequency tunable pulse from ~2-10 μm with ≥10 μJ per pulse
  - 50 fs pulse with ~200 cm^{-1} bandwidth
- Generate visible light using second harmonic bandwidth compression
  - Split single pulse into two pulses with equal but opposite orientation of chirp
  - Beams recombined on SHG crystal, generating a frequency narrow pulse with ~6 ps duration

Signal detection done with Newton iStarr ICCD



Initial Results – CH Stretch on Gold Surface



$$I_{SFG} \propto \left| \left|\chi_R^{(2)}\right| e^{i\delta} + \left|\chi_{NR}^{(2)}\right| e^{i\varepsilon} \right|^2$$

Assume:

- $\left|\chi_R^{(2)}\right|$ is given by a Lorentzian
- $\left|\chi_{NR}^{(2)}\right|$ is given by a Gaussian

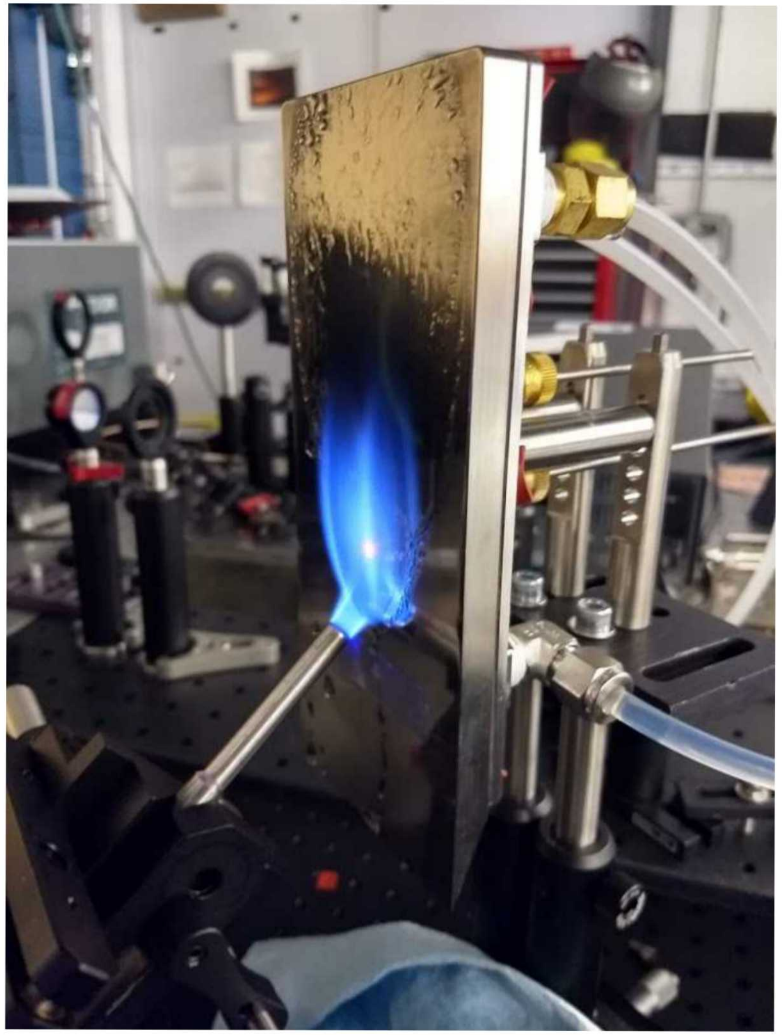
Used 10 second camera gate with 50 averaged images
→ 500,000 laser shots averaged!!

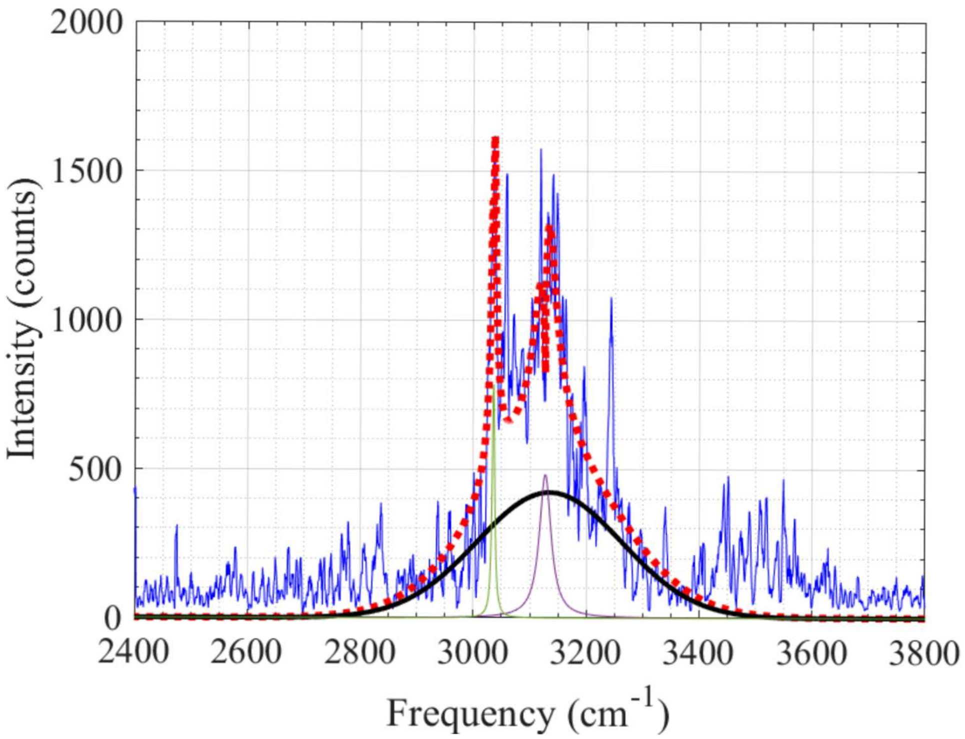
Nonlinear LSQ fit for following parameters:

- Resonant frequency (constrained by known CH stretches)
- Resonant linewidth (constrained by realistic values)
- Resonant amplitude
- Phase between resonant and non-resonant
- Non-resonant amplitude

Measured and calibrated non-resonant frequency and linewidth using polystyrene calibration film

Initial Results – Measurements on a Steel Wall with a Flame





Acknowledgments

Collaborators Include:

- Professor Richard Miles (Princeton and Texas A&M), Professor Yiguang Ju (Princeton), & Professor Egemen Kolemen (Princeton and Princeton Plasma Physics Laboratory)
- Dr. Stephan Reuter (Polytechnique Montréal) and Dr. Arthur Dogariu (Princeton)
- Aric Rousso (Princeton) and Timothy Chen (Princeton)
- Luka Hansen (Kiel University) and David Feng (Princeton)
- Dr. Christopher Kliewer (Sandia National Labs)

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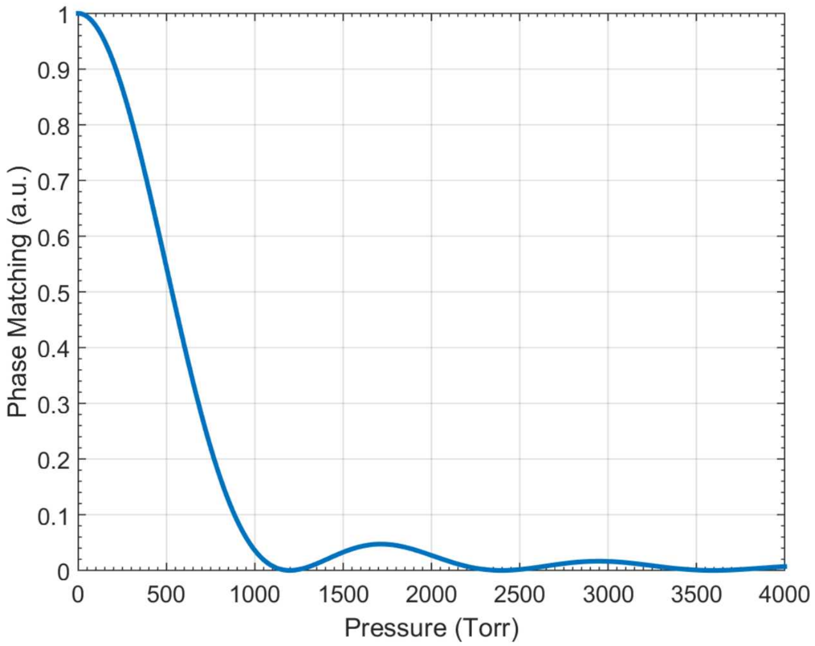
Thank you for your attention

Questions?

Phase Matching



In transparent media, the index of refraction increases as a function of frequency

For second harmonic generation:

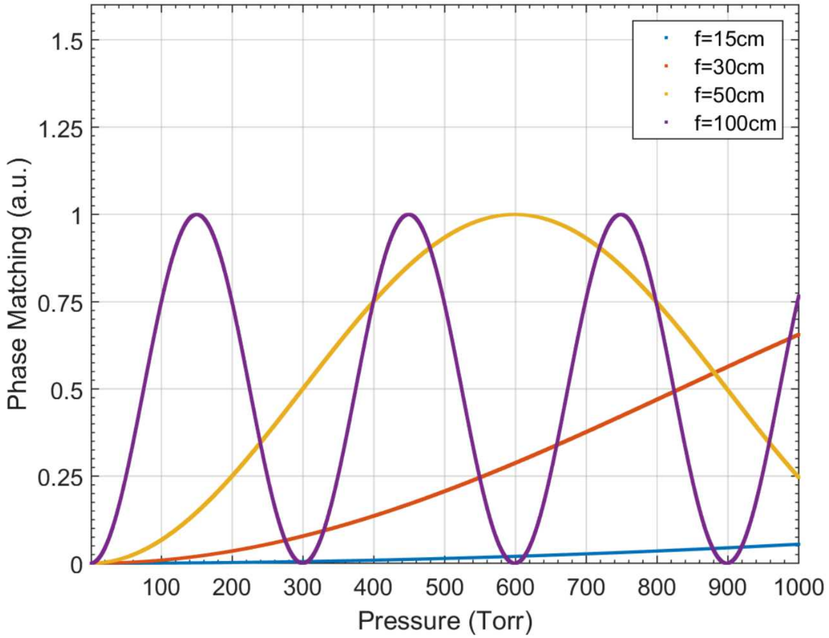
- $\Delta k = 2 \cdot \frac{n_{Pump}\omega_{Pump}}{c} - \frac{n_{SHG}\omega_{SHG}}{c} \neq 0$
- $I_{SHG} \propto sinc^2\left(\frac{\pi \Delta k L}{2}\right)$

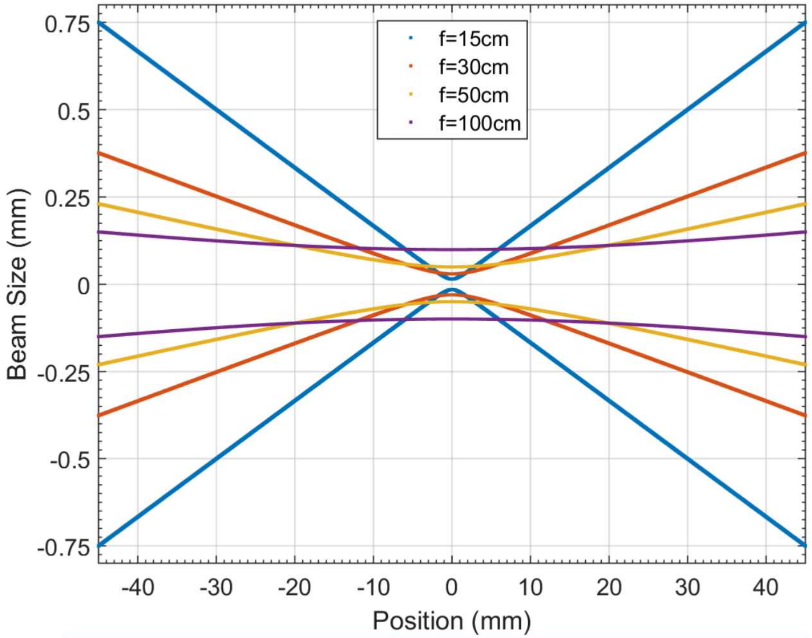
If measurement length is greater than $^2/_{\Delta k}$, efficiency goes down as second harmonic goes out of phase with driving polarization

$$z_R = \frac{\pi w_0^2}{\lambda} = \frac{4\lambda}{\pi}\left(\frac{f}{D}\right)^2$$

Robert W Boyd. Nonlinear optics

Phase Matching





$$z_R = \frac{\pi w_0^2}{\lambda} = \frac{4\lambda}{\pi}\left(\frac{f}{D}\right)^2$$

$$I^{2\omega} \propto N^2 \mathrm{sinc}^2\left(\frac{L}{L_{coh}}\right)$$

Focal Length	Rayleigh Range
150 mm	0.894 mm
300 mm	3.575 mm
500 mm	9.93 mm
1000 mm	39.73 mm

Benefits of E-FISH Method

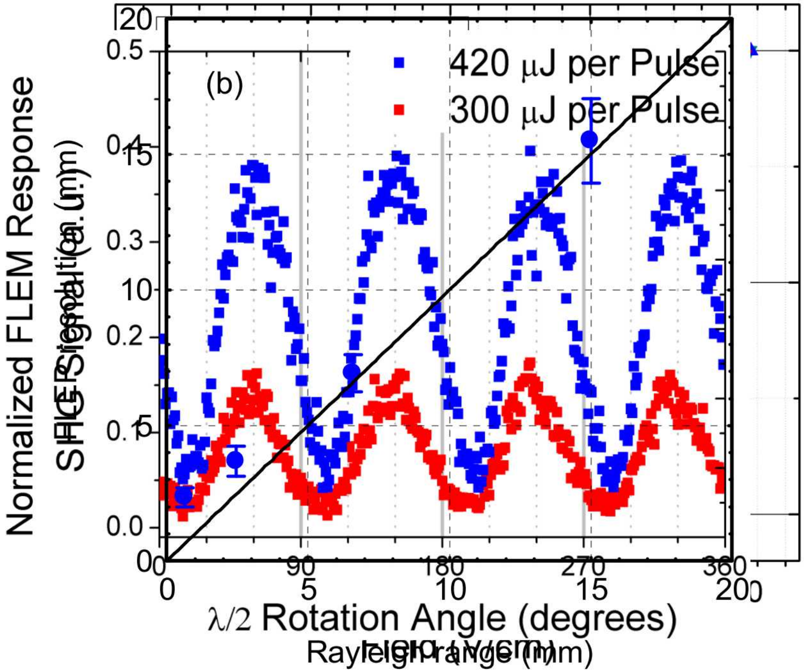
Quadratic Dependence upon Electric Field
- Sensitive down to 100's of V/cm

Time Resolution
- Fs laser acts as a δ function compared with ns changes in fields

Measurement length
- Determined by beam focusing parameters allowing for sub-mm spatial resolution

Species Independence
- Non-Resonant technique that can be used in any gaseous mixture

Field Vector Sensitivity
- Nonlinear susceptibility polarization components indicate the ability to measure field vector components



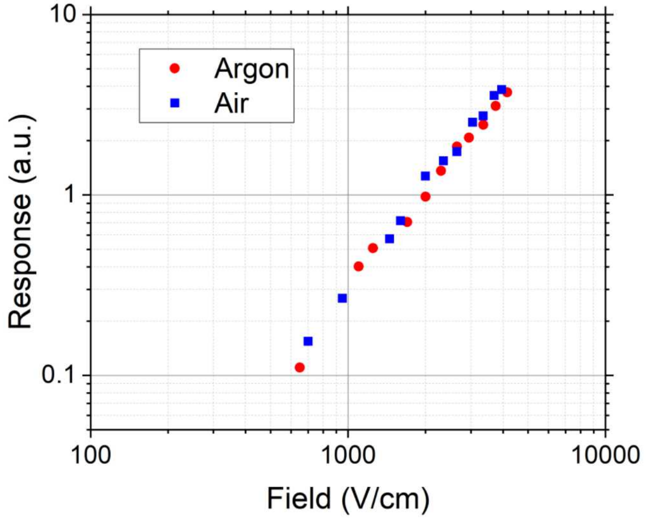
Dogariu, A., *Physical Review Applied*, *7*(2), 024024.

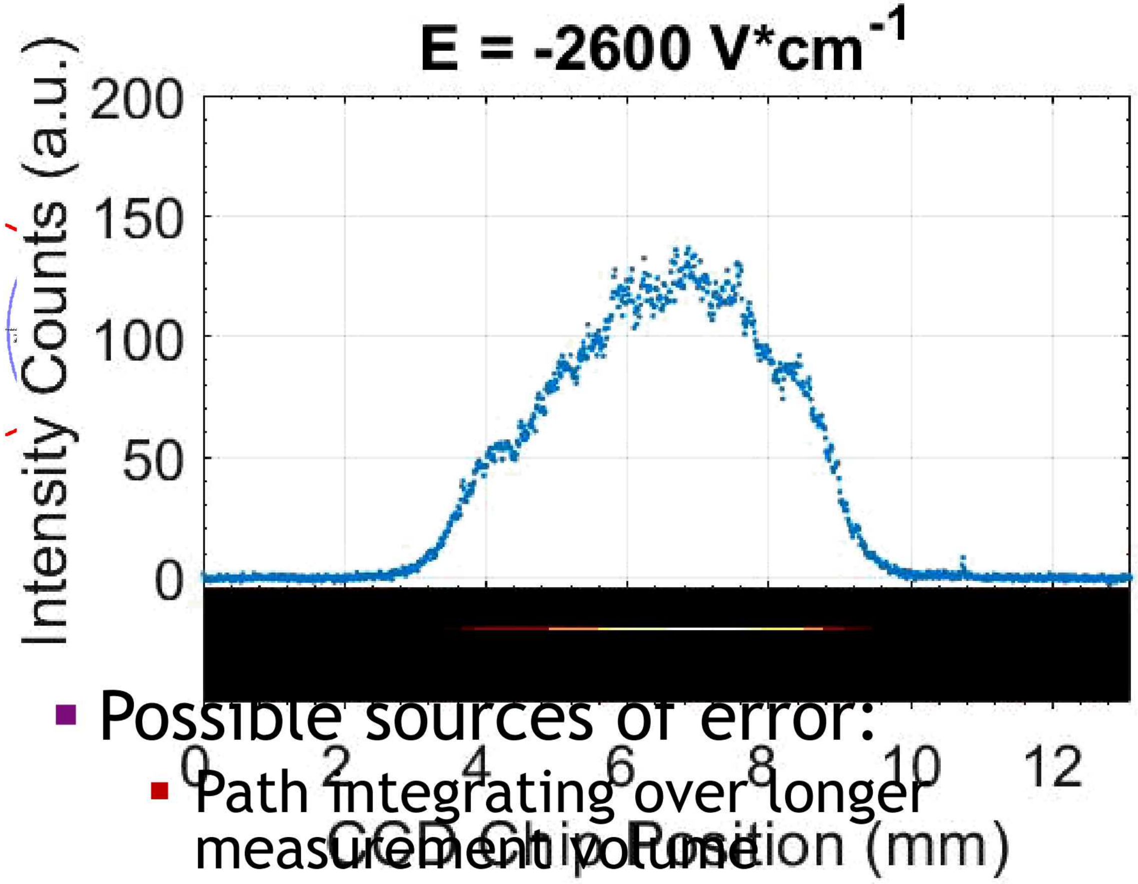
CAP - Calibration

APPJ does not have well defined ground

Calibration completed between cylindrical electrodes

- $E(z) = \frac{2 \cdot V_{Applied}}{d \cdot \cosh^{-1} l/_{2a}} \cdot \frac{1}{\left[1+4^{(z-z_0)^2}/_{d^2}\right]}$



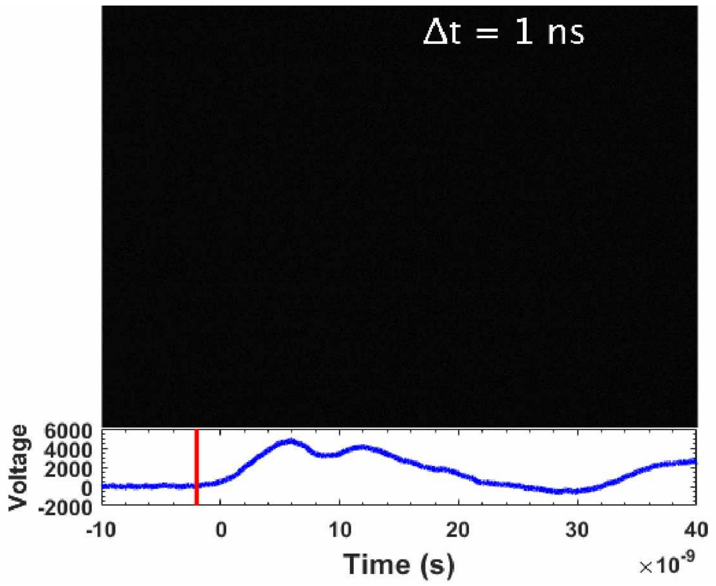


- Possible sources of error:
  - Path integrating over longer measurement volume
  - Different susceptibilities of different gases

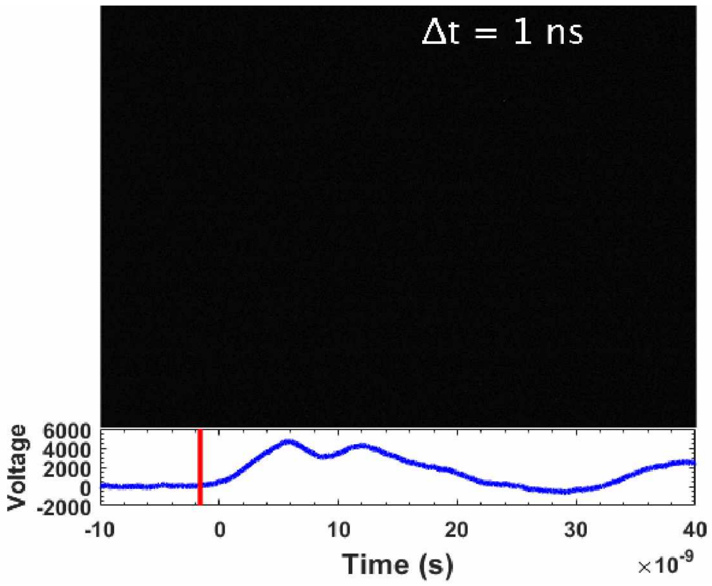
$$I^{(2\omega)} \propto A \cdot N^2 (E_{Ext})^2 \left(I_{Pump}\right)^2$$

Burst Mode - Time Resolved Imaging

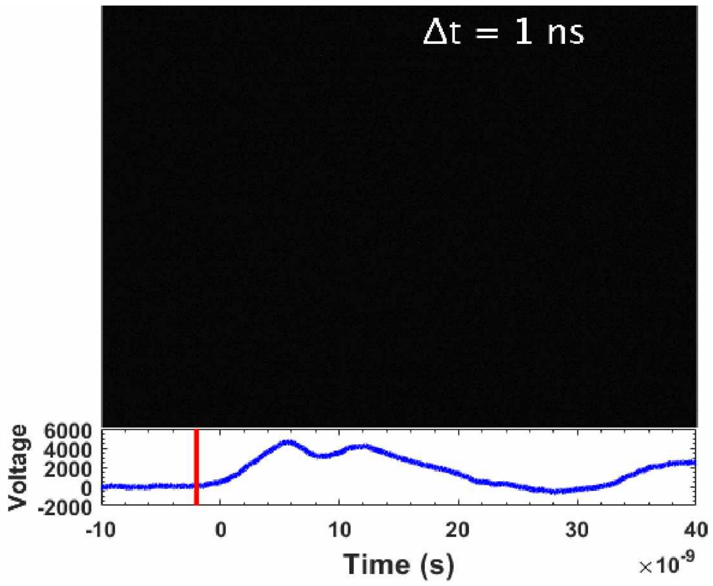
6.6% CH_4 13.4% O_2 80% Ar
Pulse 25



6.6% CH_4 13.4% O_2 80% Ar
Pulse 100



6.6% CH_4 13.4% O_2 80% Ar
Pulse 150

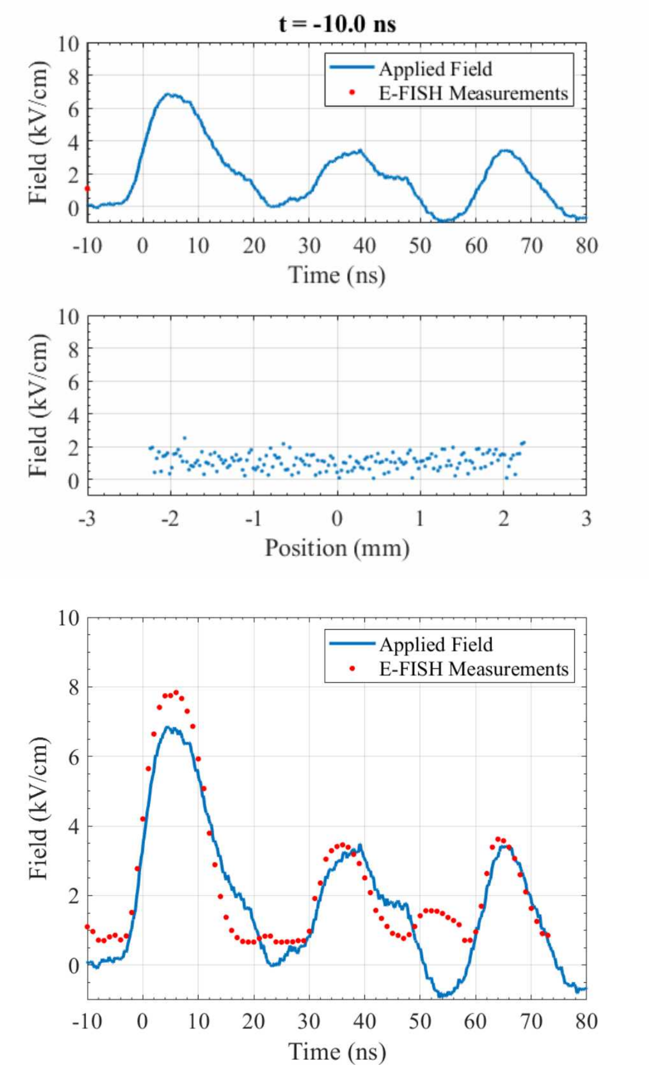


Nanosecond resolution of single pulses
- 1 ns gate, but only 10 Hz camera repetition rate
- Image not from the same burst: uncorrelated shot to shot

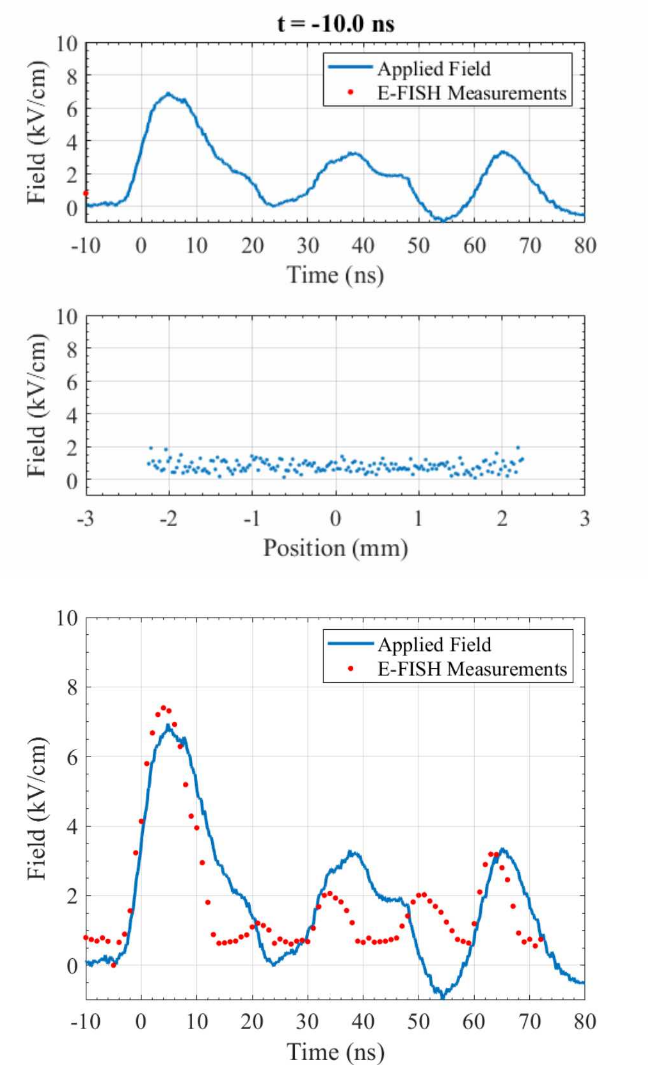
Shows evidence of significant filaments by pulse 100
- Non-uniformity moves from burst to burst

Burst Mode – Averaged Electric Field Results

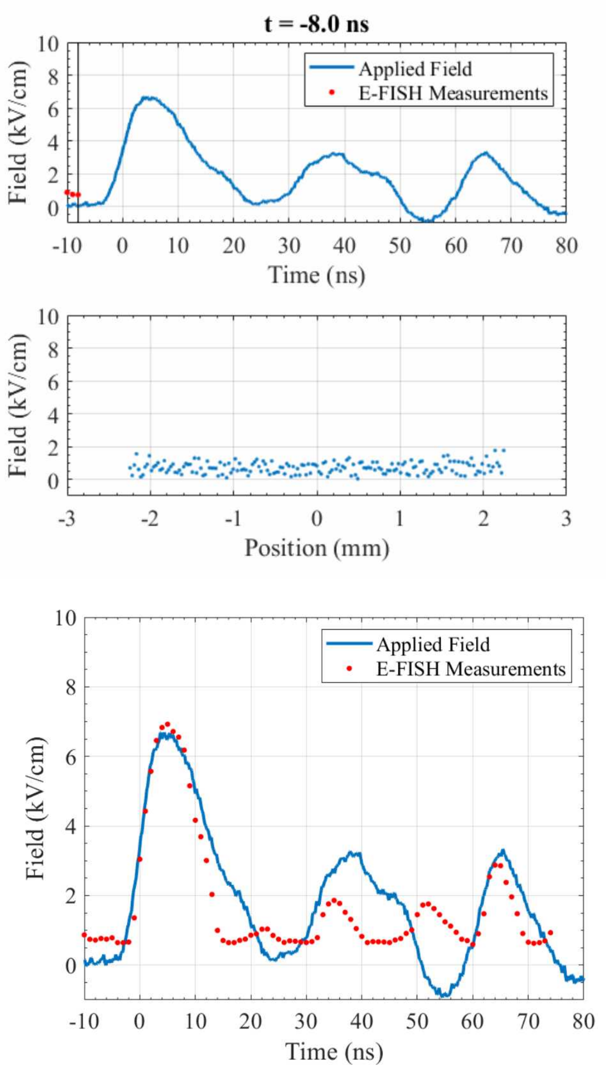
6.6% CH_4 13.4% O_2 80% Ar
Pulse 25



6.6% CH_4 13.4% O_2 80% Ar
Pulse 100



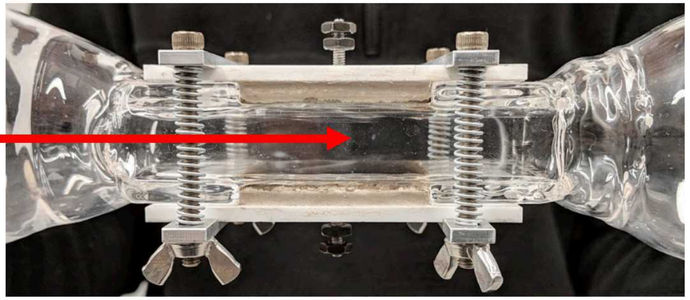
6.6% CH_4 13.4% O_2 80% Ar
Pulse 150



1 kHz – Electric Field Results

NOTE - PIXIS 512 CCD Used with 500 ms gate

Laser Propagation



0.7 Ar + 0.1 CH_4 + 0.2 O_2