Picosecond Optical Sum-Frequency Generation Enabling Fundamental Studies in Plasmas and at Interfaces with Materials

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University of Minnesota

Driven to Discover®

Insights from the Louvre Museum/ Musée du Louvre

- It’s all about **mixing colors** on the appropriate “support”

Leonardo da Vinci, Portrait of Lisa Gherardini, Wife of Francesco del Giocondo, known as the Mona Lisa

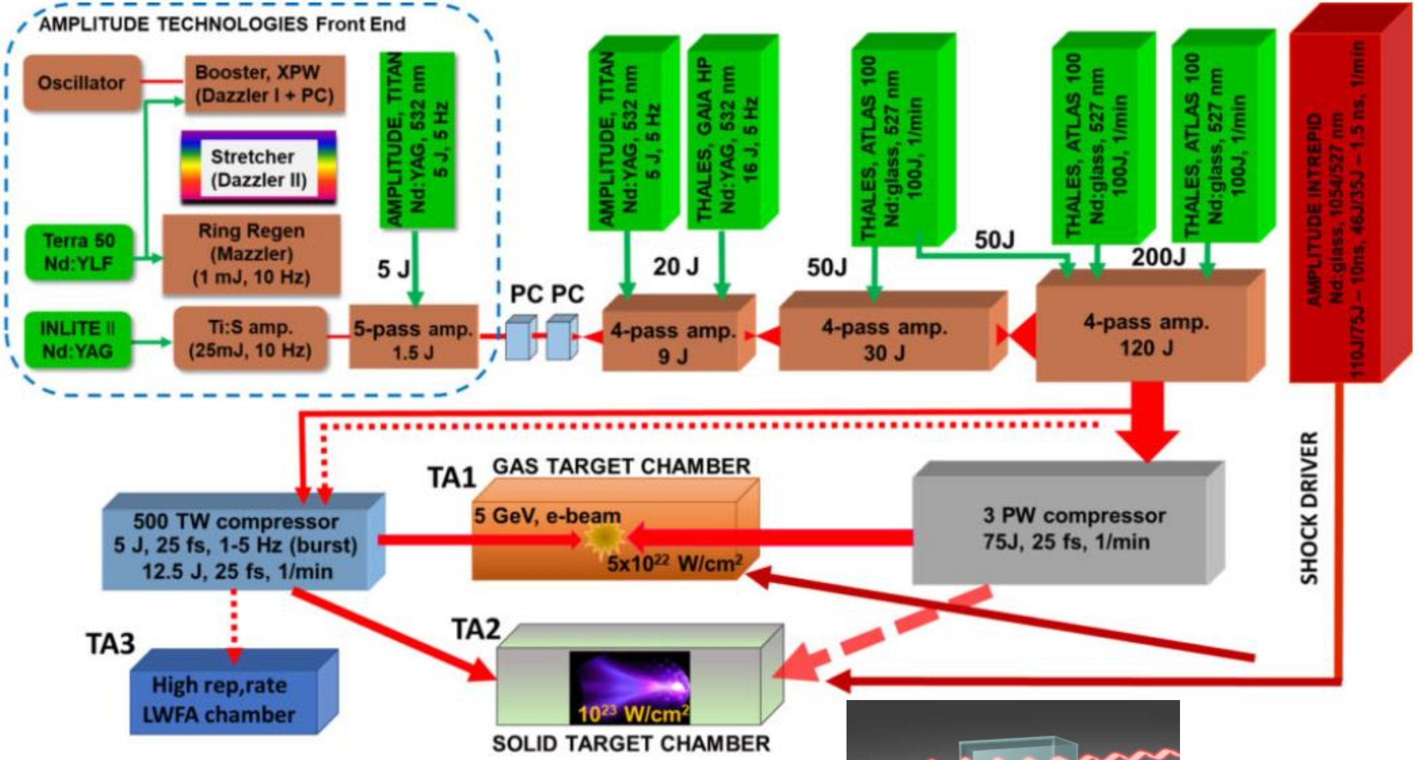
La Joconde

We should not underestimate the support quality requirements for achieving painting/sculpting masterpieces!

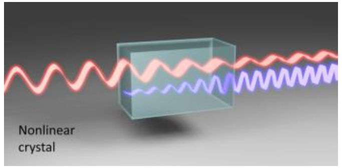
Aphrodite, known as the 'Venus de Milo'

La Vénus de Milo

The Support Matters: Example of ZEUS



Crystals without inversion symmetry
(BBO, LBO, KTP)

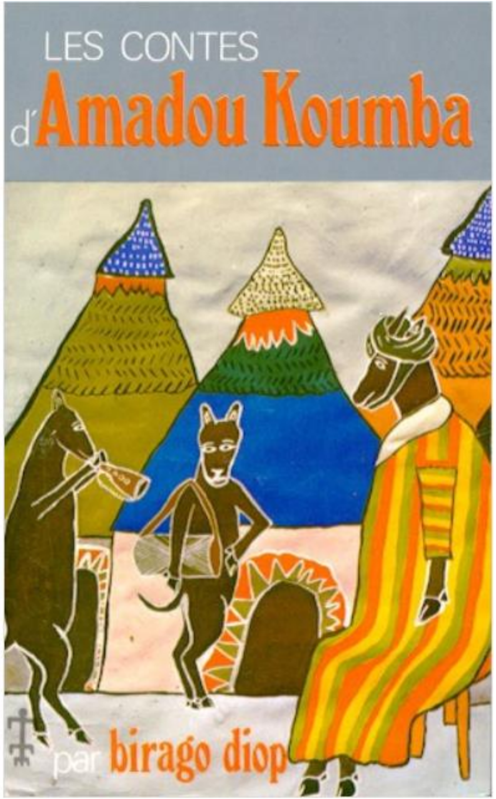


- Can we instead investigate the “reverse” process and look into characterizing the optics elements based on the properties of the light collected?

https://zeus.engin.umich.edu/about/laser-system/

French quote: “Good tools make good workers”

Establishing Communication



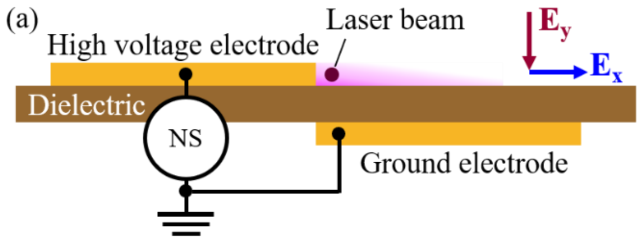
Can we engineer ways of having discussions with particles in a plasma? What would the language be?

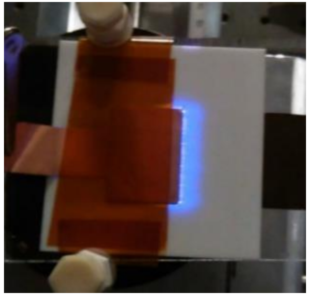
How about Mixing Colors (Painting) within Plasmas as Supports? **Nonlinear Spectroscopy**

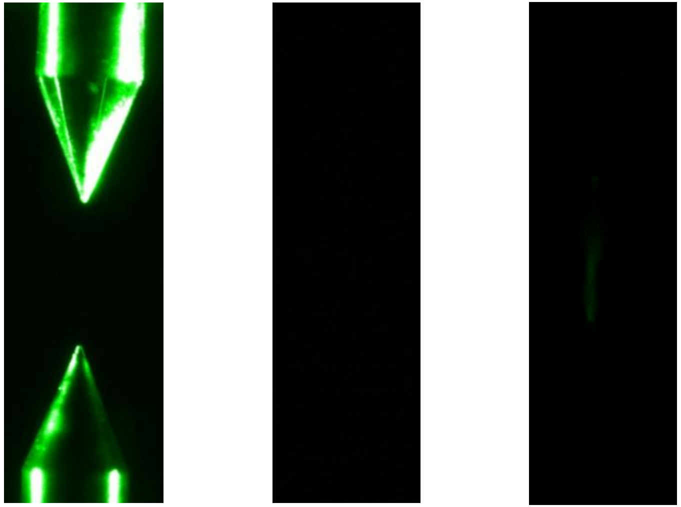
“Physics would be dull and life unfulfilling if all physical phenomena around us were linear. Fortunately, we are living in a nonlinear world. While linearization beautifies physics, nonlinearity provides excitement in physics.”

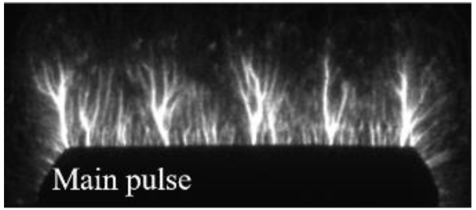
Y. R. Shen in “The Principles of Nonlinear Optics”

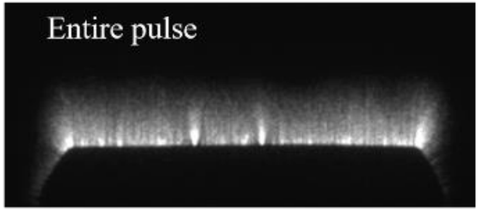
Overarching Scientific Questions (1)

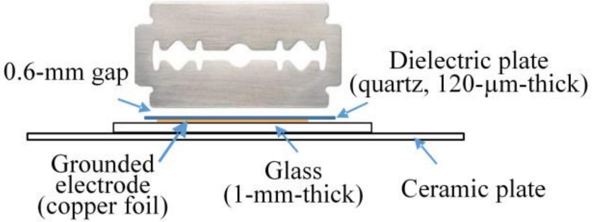


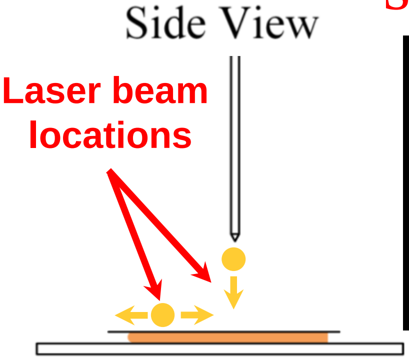




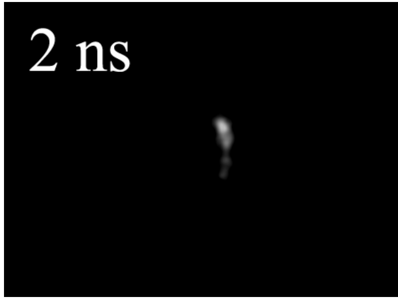








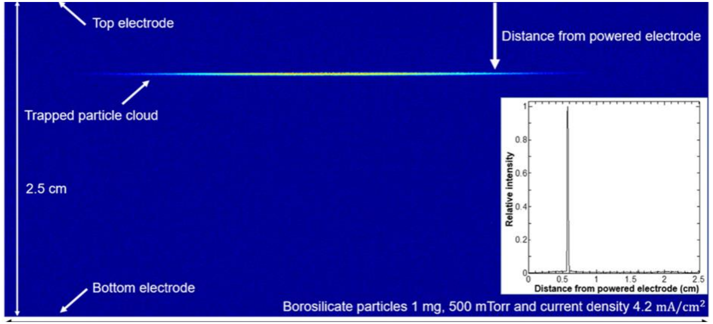
Side view, 2 ns gate



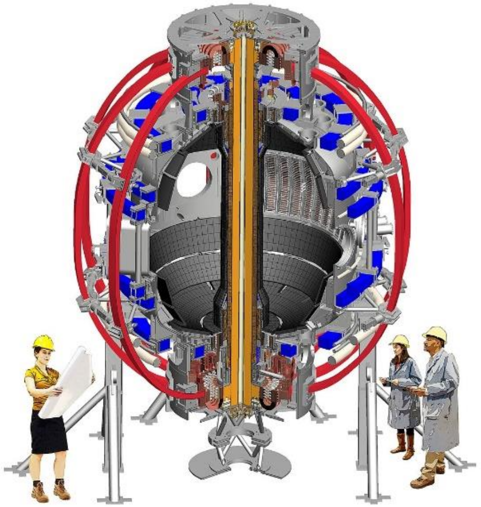
M. Simeni Simeni et al, PSST (2018) 104001

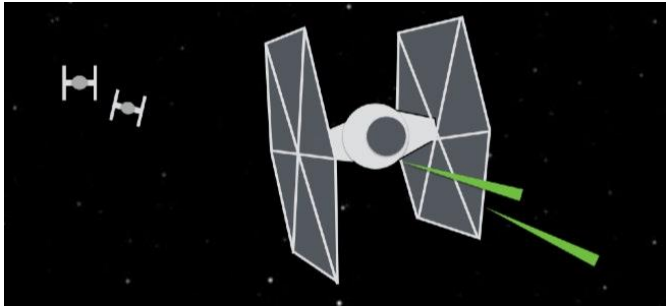
M. Simeni Simeni et al, 2017 J. Phys. D: Appl. Phys. 50 184002

Overarching Scientific Questions (2)

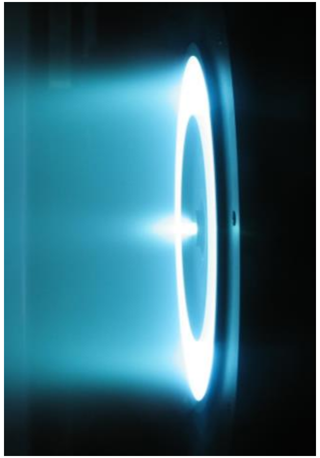


Spherical Tokamak at PPPL





Star Wars twin ion engine fighters



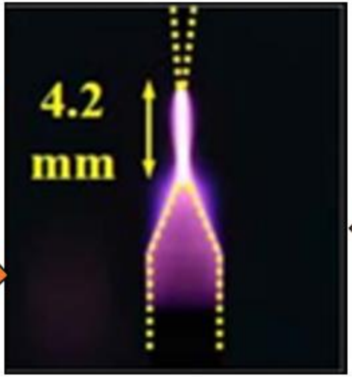
- Transitions in confinement regimes (L-H) at the walls of tokamak plasmas
- Charge particles dynamics in Hall-effect thrusters and dusty plasmas

Ono et al, Plasma Sources Sci. Technol. 30 (2021) 095014

Broader Scientific and Societal Context

Flow Control
Mixing Enhancement

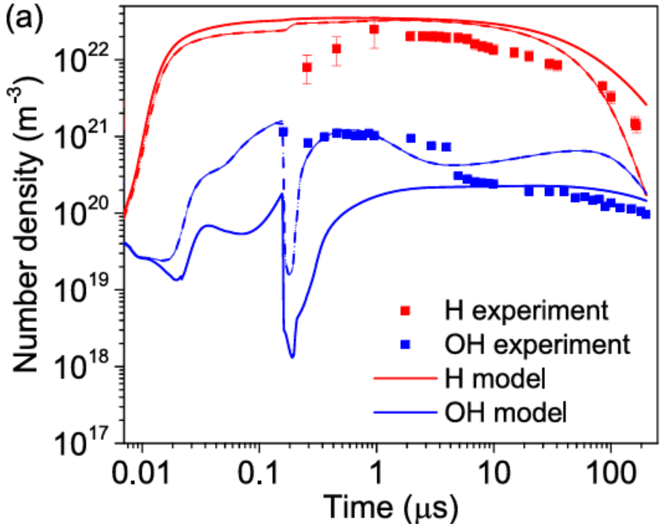
Gas cleaning
Removal of VOCs and NOx

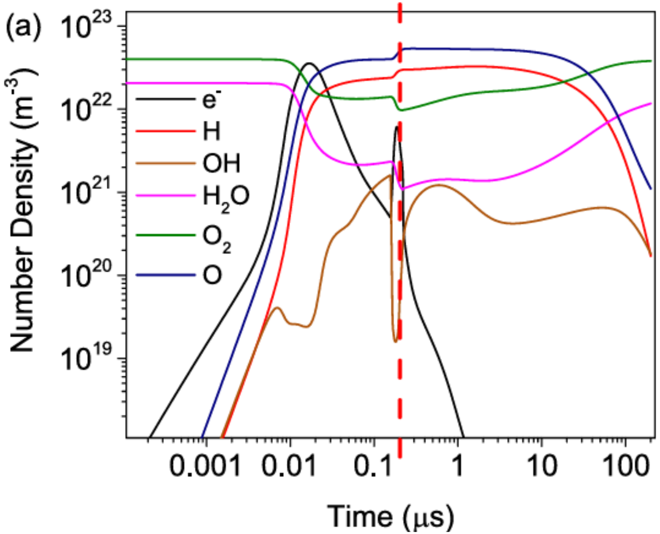


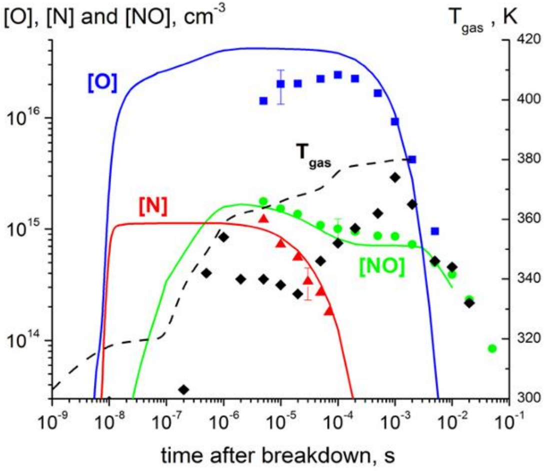
Combustion
Ignition & Lean-flame operation

Drug delivery
Electroporation

Spatio-Temporal Measurements of Electric Fields and Species Number Densities







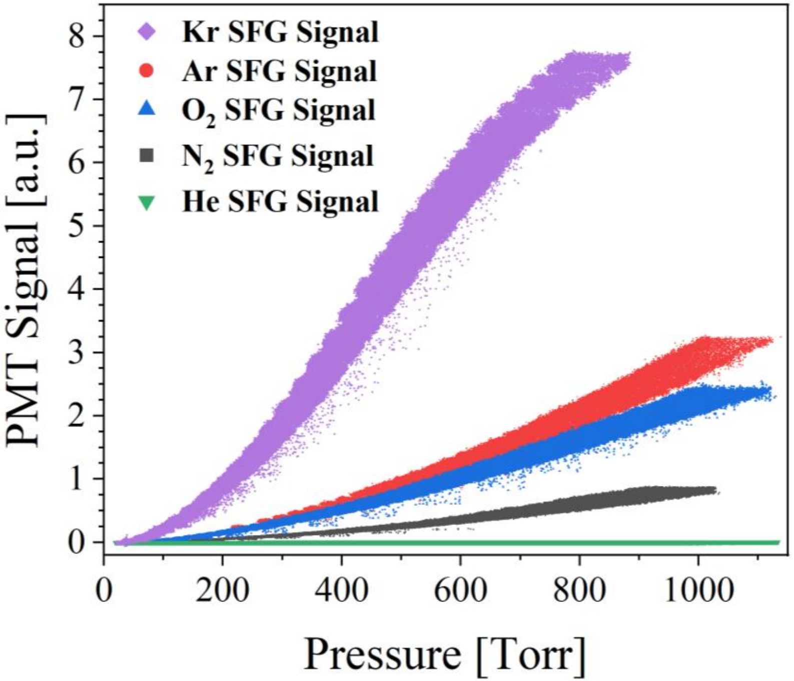
- Complex chemistry generated upon electron impact on heavy particles
- Validation of models involving 1000 reactions and hundreds of species

Can we use a single laser-based technique number densities measurements for all the species?

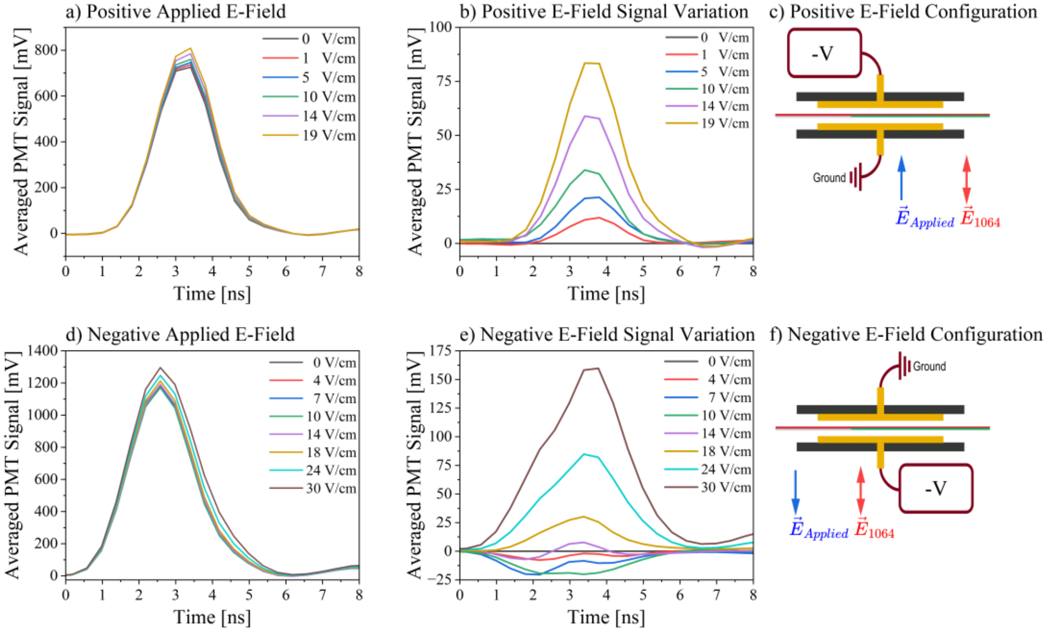
Y. Luo et al, J. Phys. D: Appl. Phys. 52 (2019) 044003
I. Shkurenkov et al, PSST 23 (2014) 065003

Outline

Three-Wave Mixing for Number Density Measurements



Sub-1V/cm E-Field Measurements



 G. LaCombe, J. Wang, J. Rouxel, et al, Opt. Lett 49 (23), 6717-6720

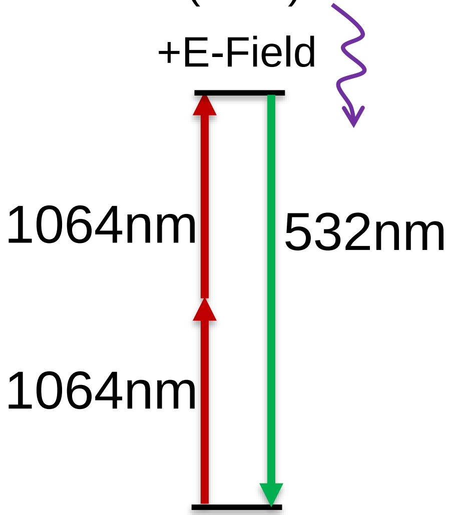
G. LaCombe, J. Wang, K. Frederickson, et al, under review at PSST

Three-Wave Mixing for Number Density Measurements at the Picosecond Timescale

4-Wave Mixing

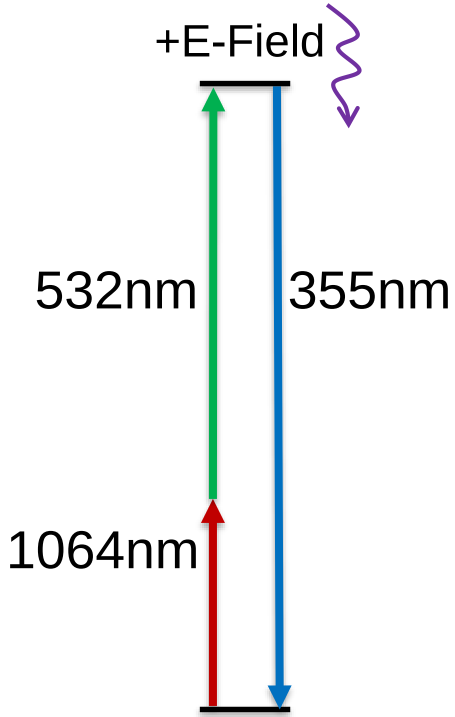
Second Harmonic Generation

(**SFG**)



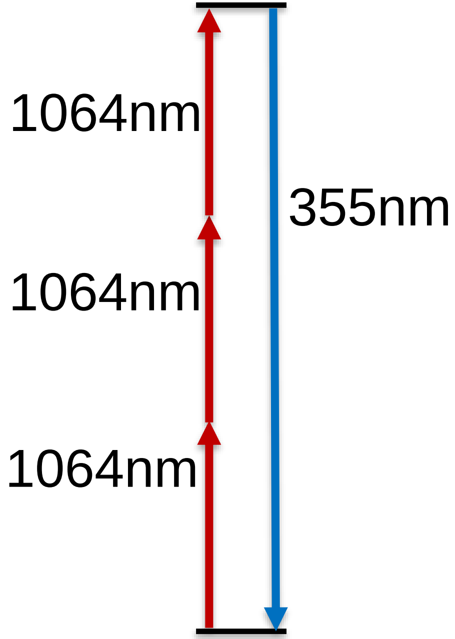
Sum Frequency Generation

(**SFG**)



Third Harmonic Generation

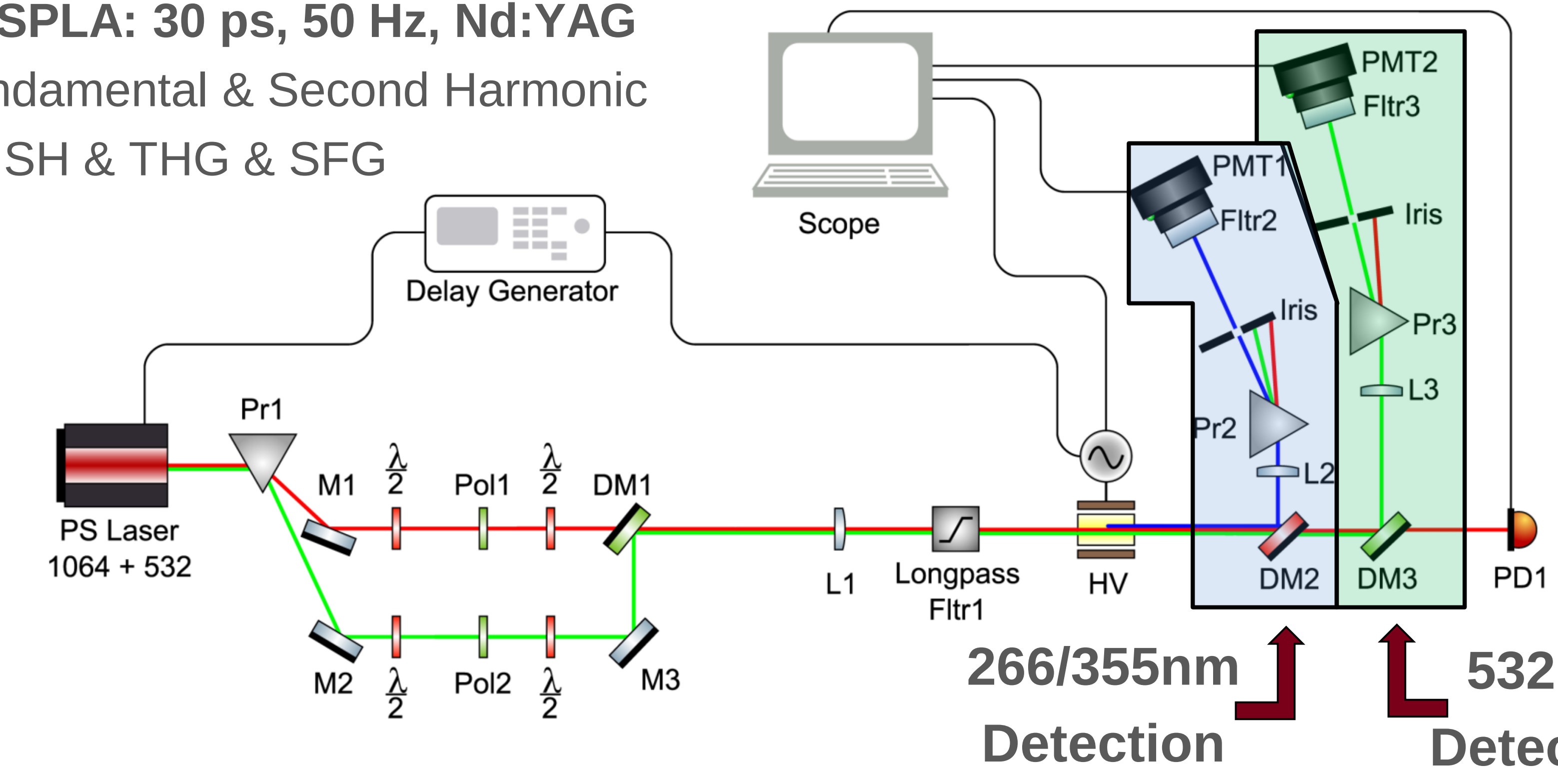
(**THG**)



$I_{532} \propto \left|\chi^{(3)}\right|^2 N^2 E_{ext}{}^2 I_{1064}{}^2$	$I_{355} \propto \left|\chi^{(3)}\right|^2 N^2 E_{ext}{}^2 I_{1064} I_{532}$	$I_{355} \propto \left|\chi^{(3)}\right|^2 N^2 I_{1064}{}^3$

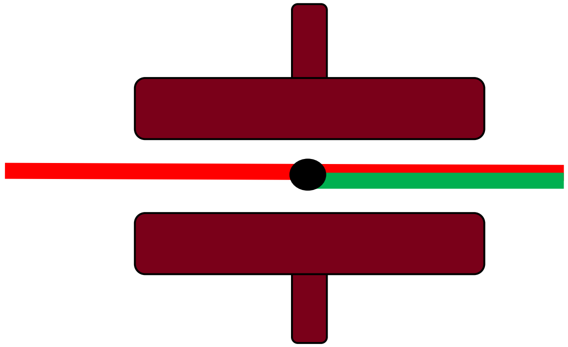
Experimental Setup

- **EKSPLA: 30 ps, 50 Hz, Nd:YAG**
- Fundamental & Second Harmonic
- EFISH & THG & SFG



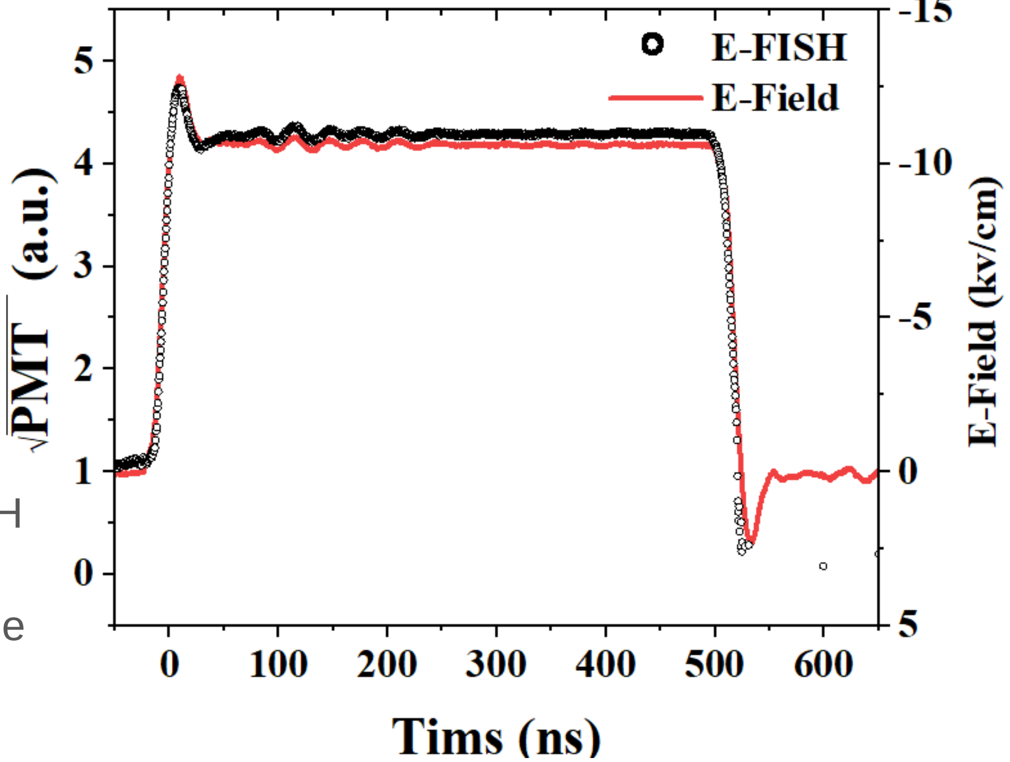
Initial Testing: EFISH

High voltage ns pulse



Ground

Measurements of a sub-breakdown ns High Voltage Pulse



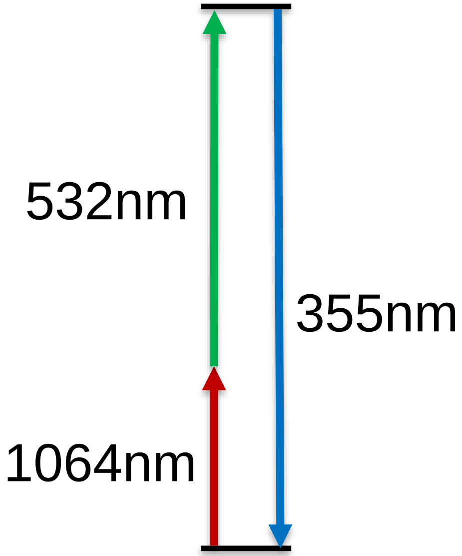
- Sub-breakdown pulse to characterize EFISH response
- Agreement between EFISH and high voltage probe

3-Wave Mixing Process

Process for non-linear generation to occur without an electric field or 4th wave

- Require non-centrosymmetric medium
  - Non-linear Crystals
  - Interfaces between mediums
- In the gas phase 3 wave mixing is forbidden due to the electric dipole approximation

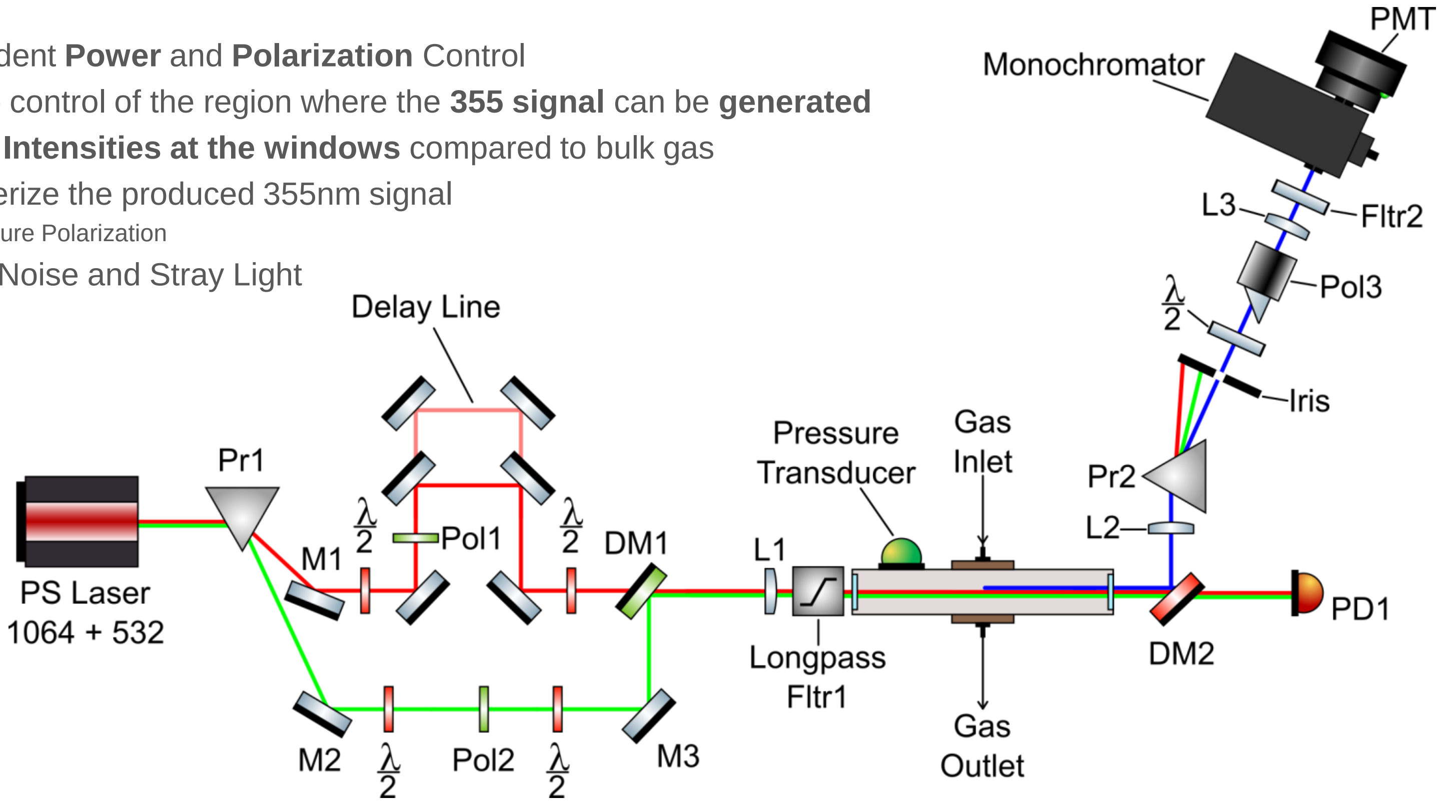
Sum Frequency Generation (**SFG**)



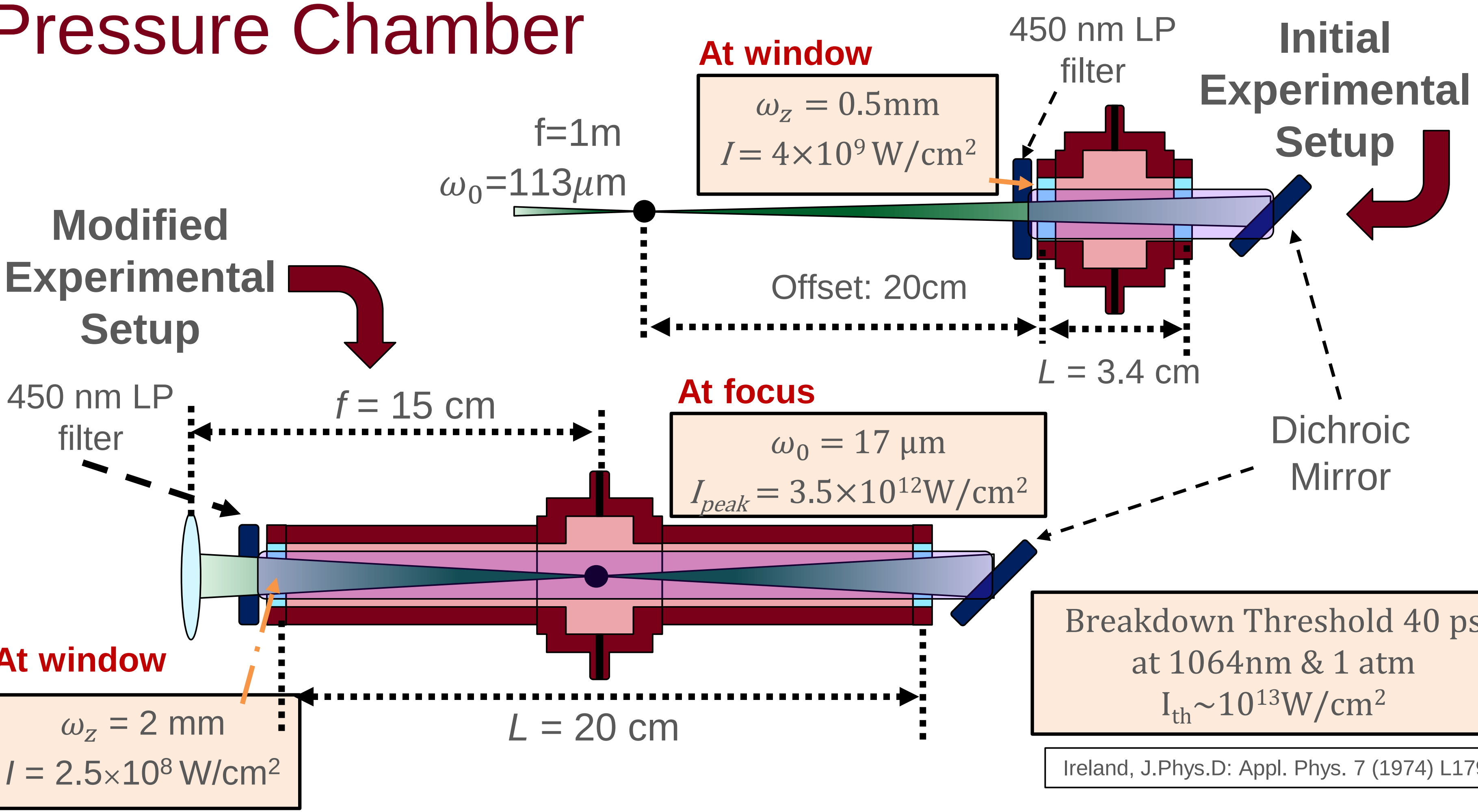
$$\boxed{I_{355} \propto \left|\chi^{(2)}\right|^2 N^2 I_{1064} I_{532}}$$

Sum Freq. Experimental Setup

- Independent **Power** and **Polarization** Control
- Increase control of the region where the **355 signal** can be **generated**
- **Reduce Intensities at the windows** compared to bulk gas
- Characterize the produced 355nm signal
  - Measure Polarization
- Reduce Noise and Stray Light



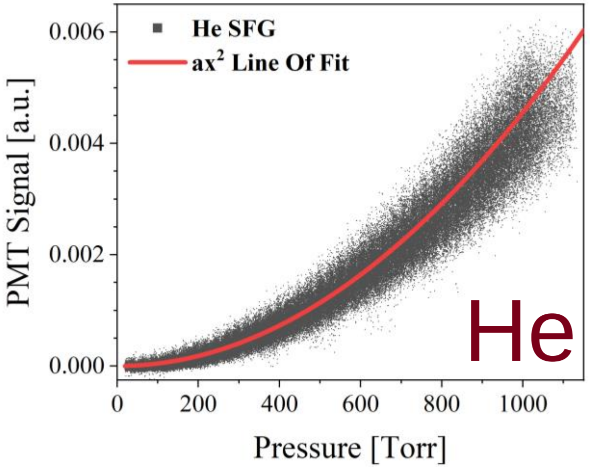
Pressure Chamber



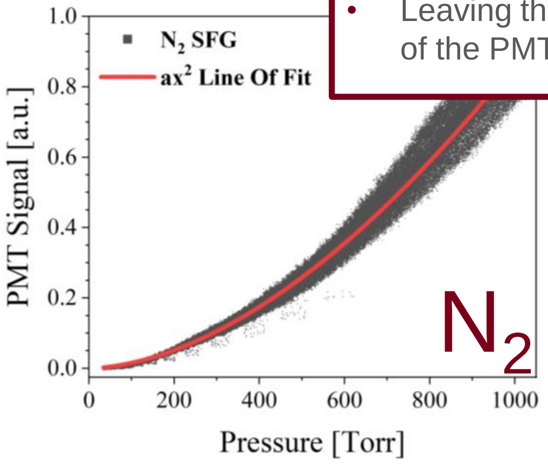
Breakdown Threshold 40 ps at 1064nm & 1 atm
$\text{I}_{\text{th}} \sim 10^{13}\text{W/cm}^2$

Ireland, J.Phys.D: Appl. Phys. 7 (1974) L179

Initial Pressure Results in He, N_2, & Kr

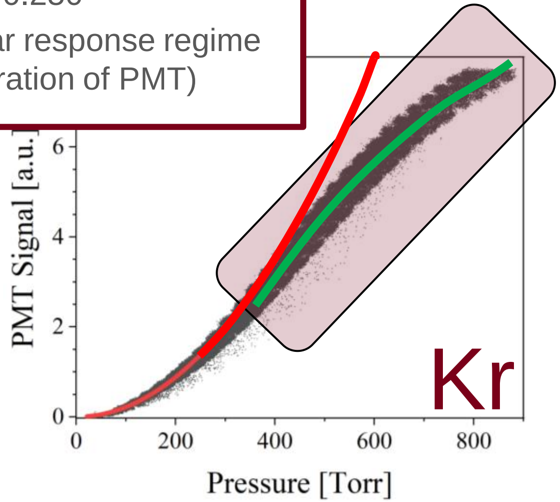


- PMT Gain: **0.571**



- PMT Gain: **0.325**

- Minimum Gain of 0.250
- Leaving the Linear response regime of the PMT (Saturation of PMT)



- PMT Gain: **0.251**

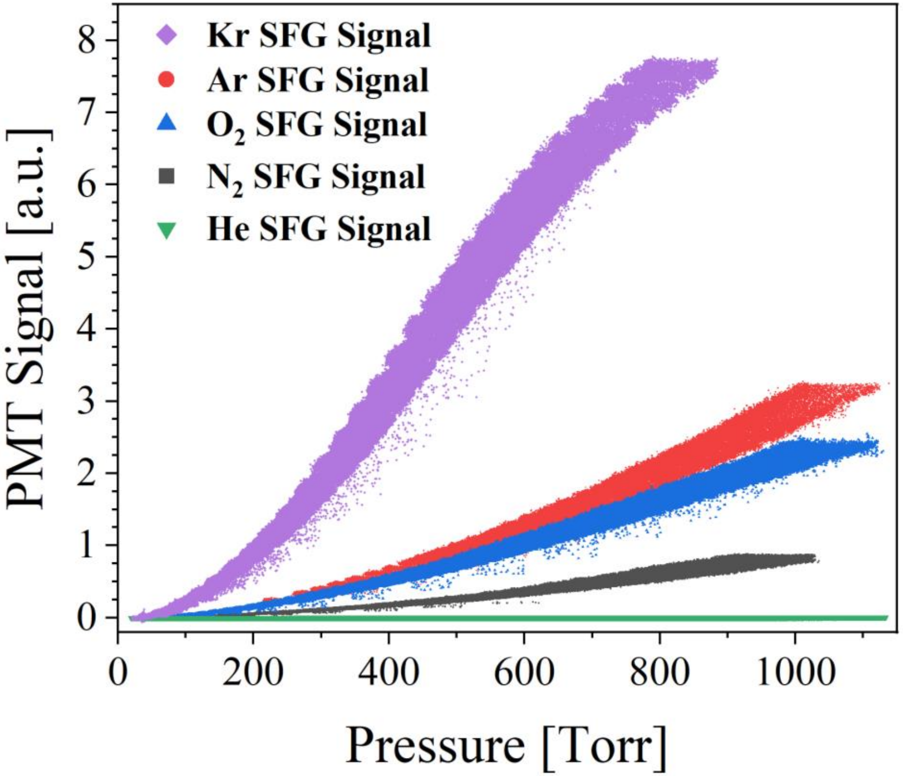
- Measuring a nonlinear effect $\propto N^2$
- Capability for pressure at 20 torr

All cases:

- 1064nm Pulse Energy: **0.88mJ**
- 532nm Pulse Energy: **0.06mJ**

SFG Pressure Measurements in Gas Variations

SFG vs P in gases



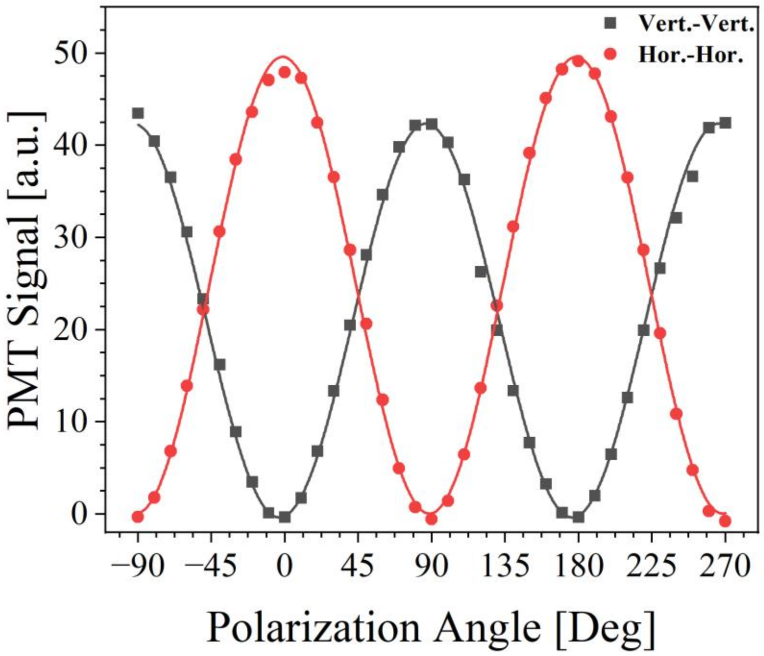
$$\left|\chi^{(2)}\right| \propto \gamma$$

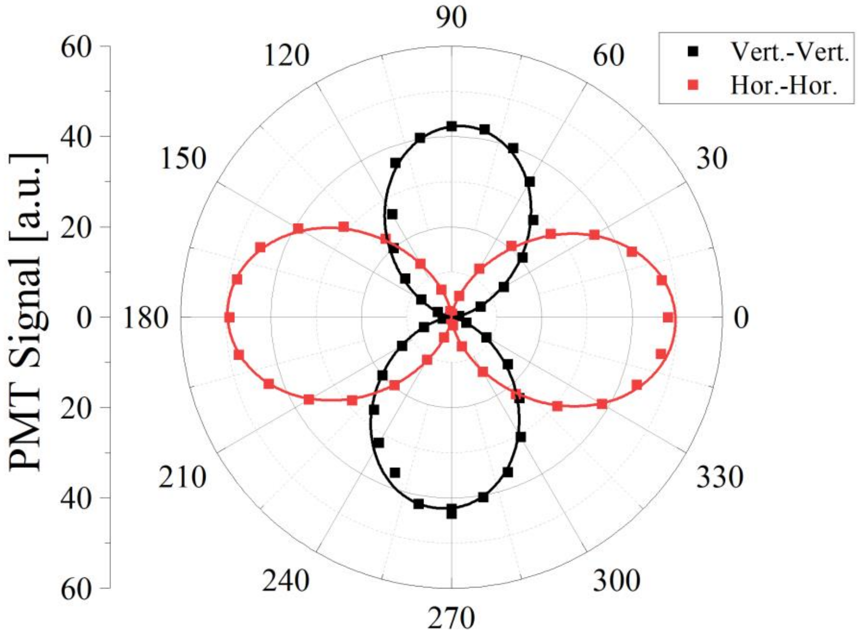
Species	Nonlinear hyperpolarizability γ (a.u.)
Kr	77
Ar	32
O_2	31
N_2	21
He	1

Finn and Ward (1971), Phys. Rev. Lett. 26, 285

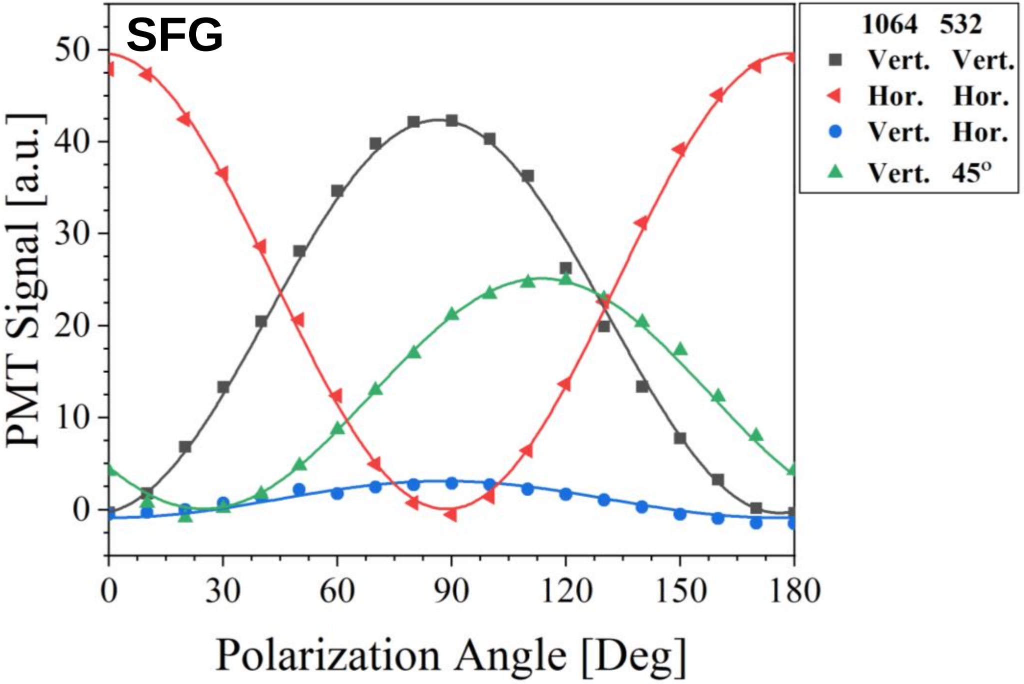
- **Measured signal intensities agree with values from the literature**
- **Monotonically increasing w/ pressure without an applied electric field**

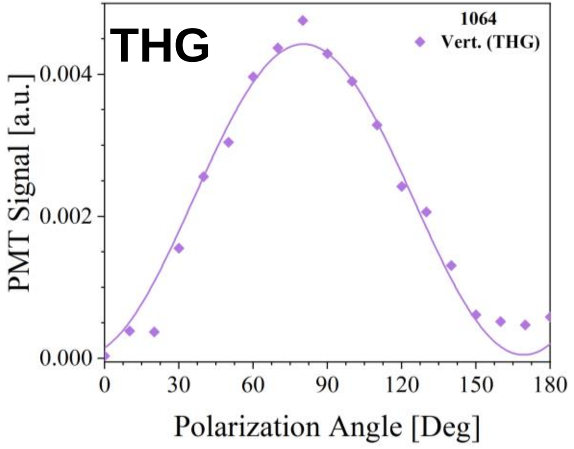
SFG Polarization V-V vs H-H





SFG Polarization





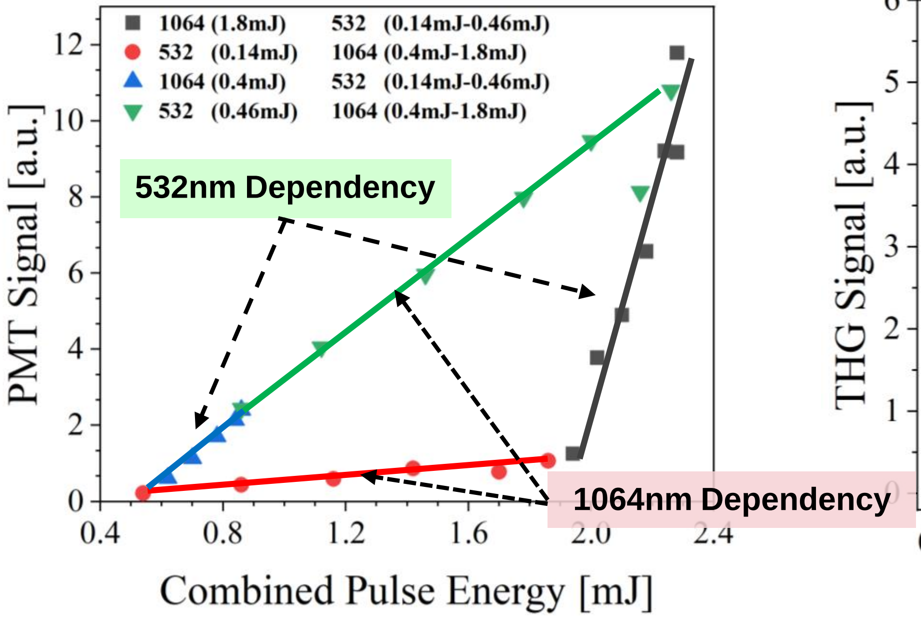
Beam Polarization

	1064 nm	532 nm	355 nm
SFG	Vertical	Vertical	Vertical
SFG	Vertical	Horizontal	Vertical
THG	Vertical	N/A	Vertical
SFG	Vertical	45 Degrees	~20 Off Vertical
SFG	Horizontal	Horizontal	Horizontal

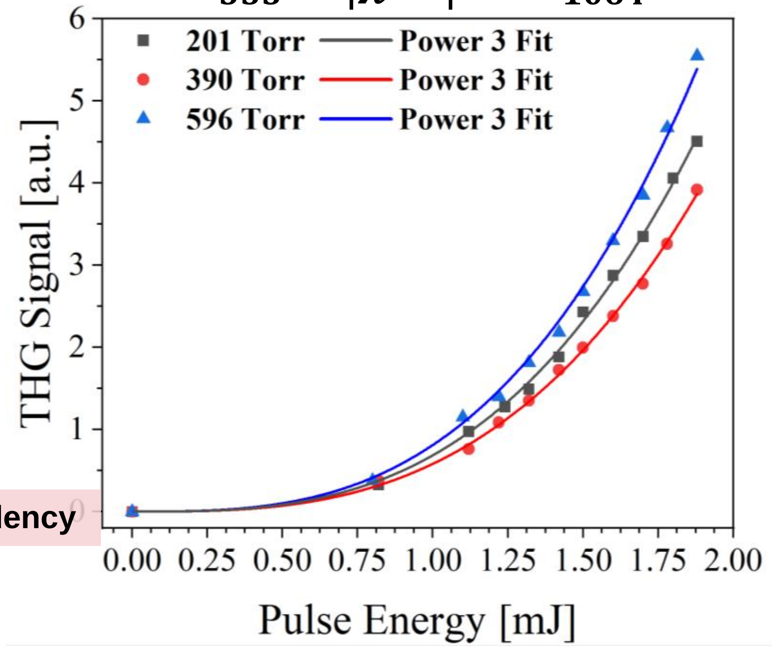
- Increased SFG signal with the addition of 532nm light
- SFG polarization related to both incident lasers polarizations

SFG Power

$$I_{355} \propto \left|\chi^{(2)}\right|^2 N^2 I_{1064} I_{532}$$



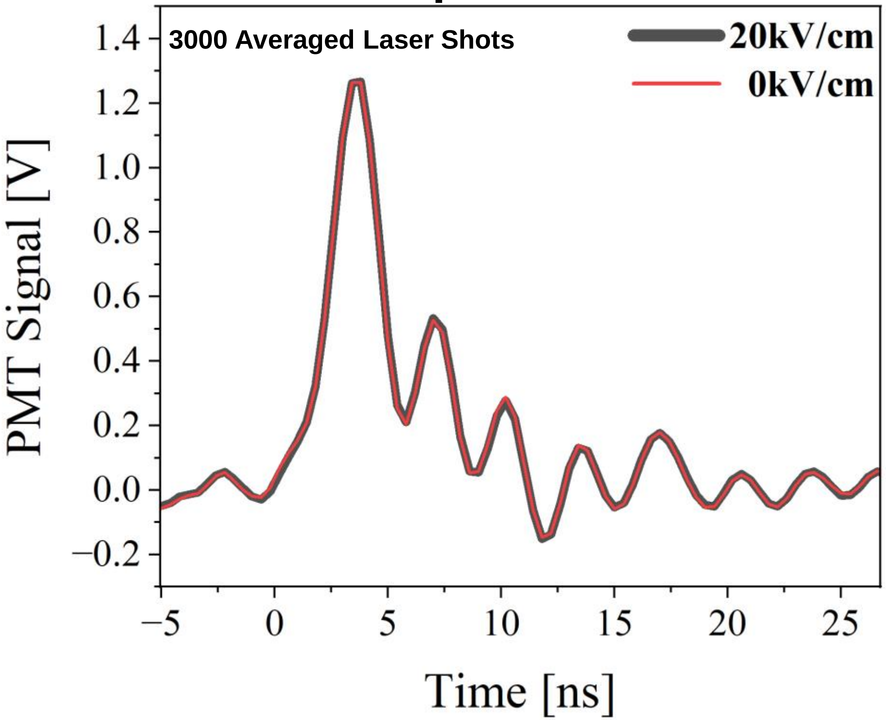
$$I_{355} \propto \left|\chi^{(3)}\right|^2 N^2 {I_{1064}}^3$$



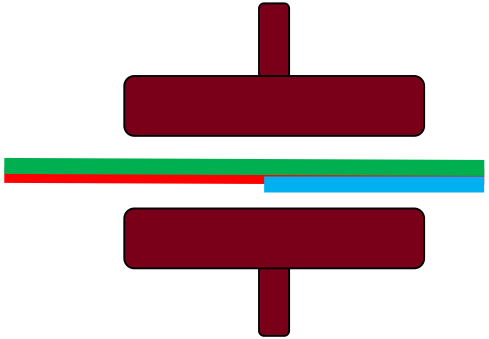
- SFG shows a linear relation to E_L
- THG ~ E_L^3

SFG Electric Field Response

E-Field Response PMT Trace



Applied external E-Field (DC)



$$\boldsymbol{I_{355} \propto \left|\chi^{(2)}\right|^2 N^2 I_{1064} I_{532}}$$

$$\cancel{I_{355} \propto \left|\chi^{(3)}\right|^2 N^2 E_{ext}{}^2 I_{1064} I_{532}}$$

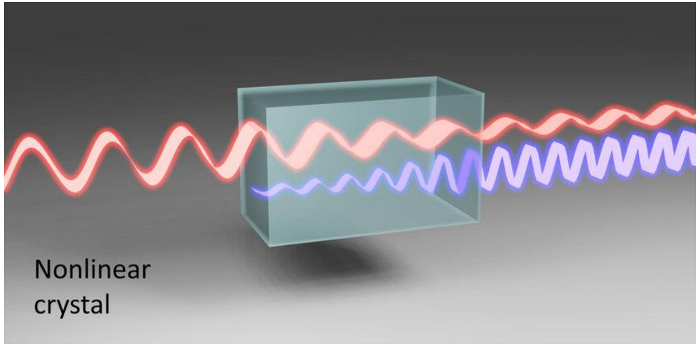
- **SFG signal independent from E-Field**
- **SFG is 3 wave mixing, $\chi^{(2)}$, process**

Topical Summary

- Non-resonant 3-Wave mixing in the gas phase is possible although not predicted by current formalisms/models
- It is observed in virtually all gases
- The measured signal at 355 nm is proportional to the intensities of the incident 1064 and 532 nm beam
- The polarization of the signal at 355 nm depends on the polarizations of both incident beams
- The signal is totally non-dependent on any externally-applied E-Field
- The mechanism leading to the occurrence of this phenomenon is still a mystery. In theory, a symmetry-breaking mechanism is needed

Sub-1 V/cm Homodyne Picosecond E-FISH-based Electric Field Measurements with Sensitivity to the Field Polarity

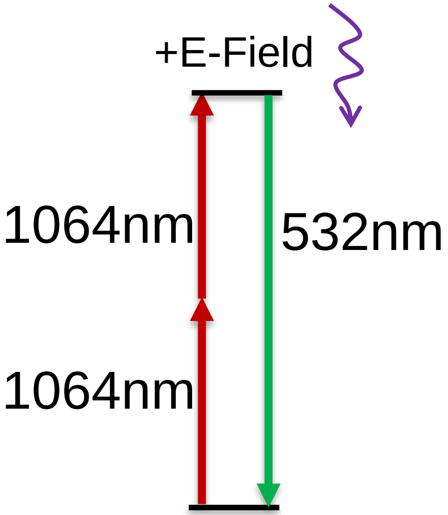
Second Harmonic Generation Sources



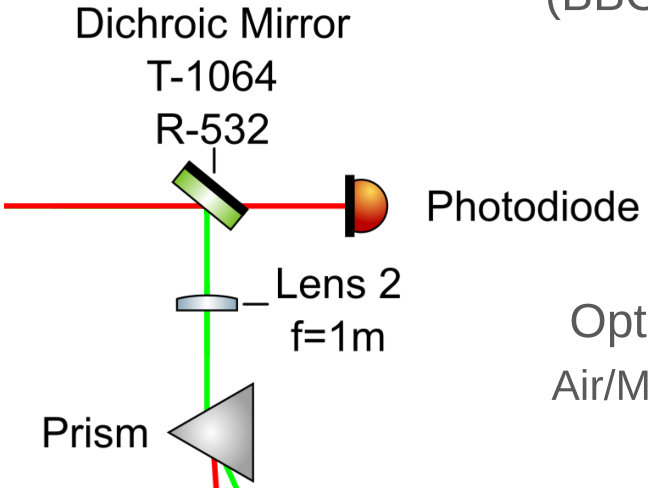
Crystals without inversion symmetry

(BBO, LBO, KTP)

E-FISH



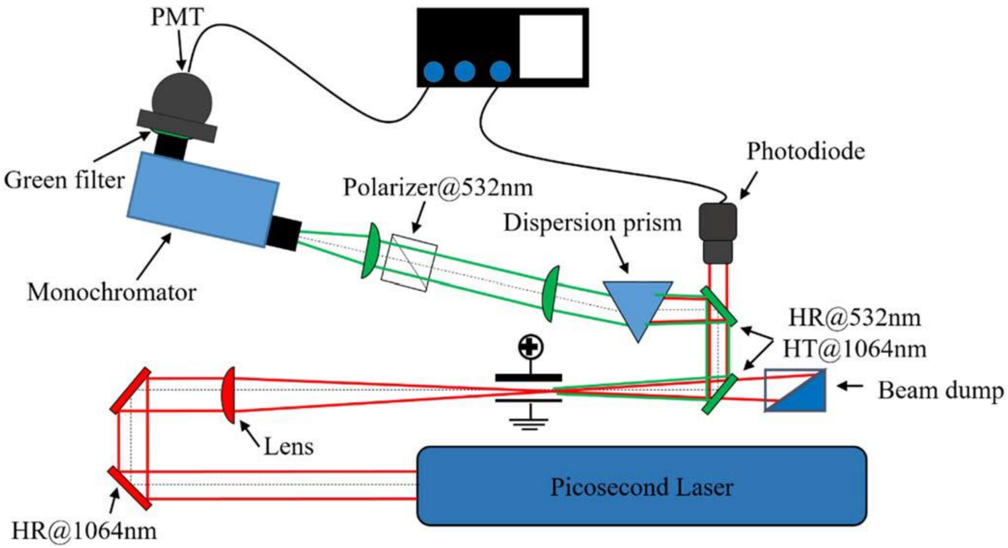
$$I_{532} \propto \left|\chi^{(3)}\right|^2 N^2 E_{ext}{}^2 I_{1064}{}^2$$

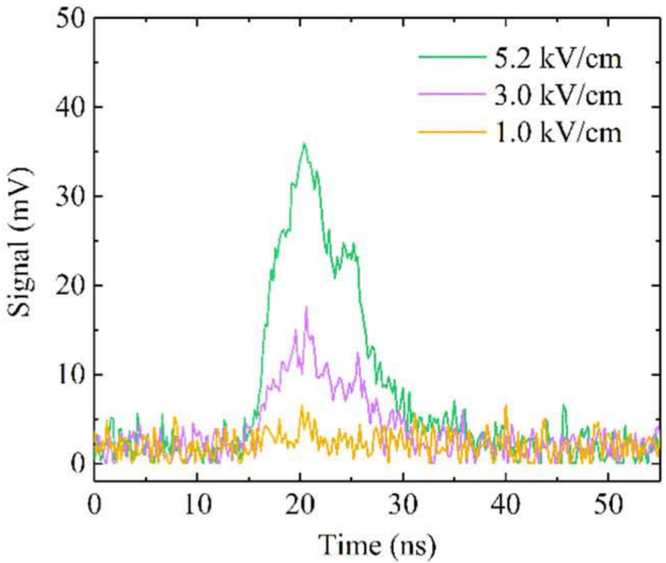


Optical Interfaces

Air/Material Boundary

Previous E-FiSH Results

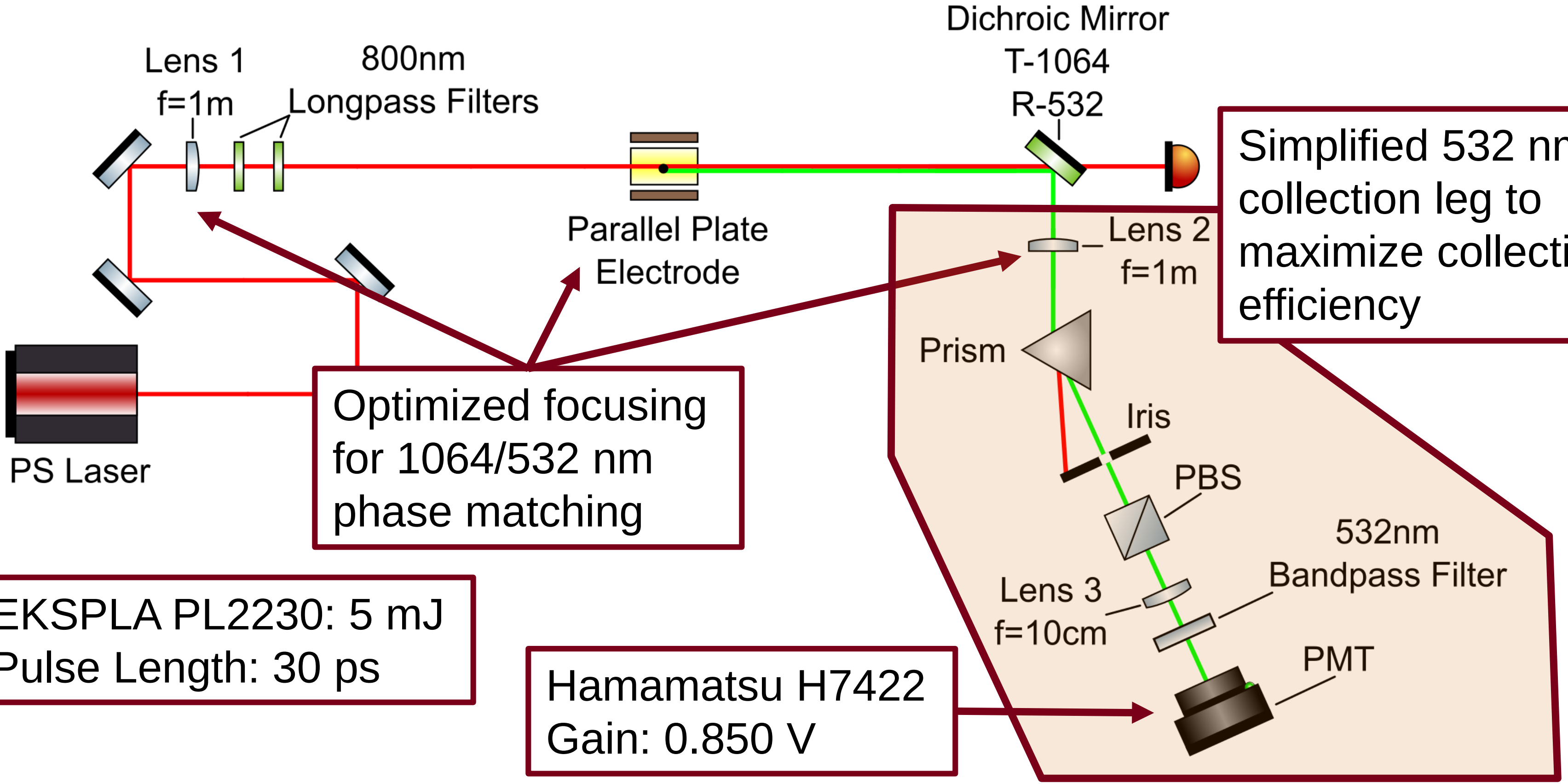




- High Complexity to ensure measurement of only second harmonic
- Signals at 1 kV/cm comparable to the noise

M Simeni Simeni *et al* 2018 *Plasma Sources Sci. Technol.* **27** 104001

E-FISH Experimental Setup



Simplified 532 nm collection leg to maximize collection efficiency

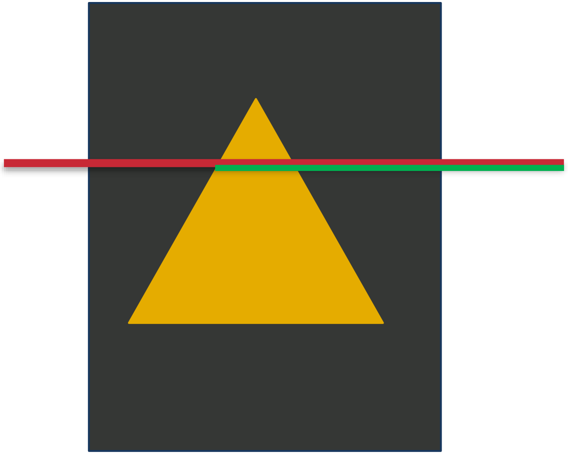
Optimized focusing for 1064/532 nm phase matching

EKSPLA PL2230: 5 mJ
Pulse Length: 30 ps

Hamamatsu H7422
Gain: 0.850 V

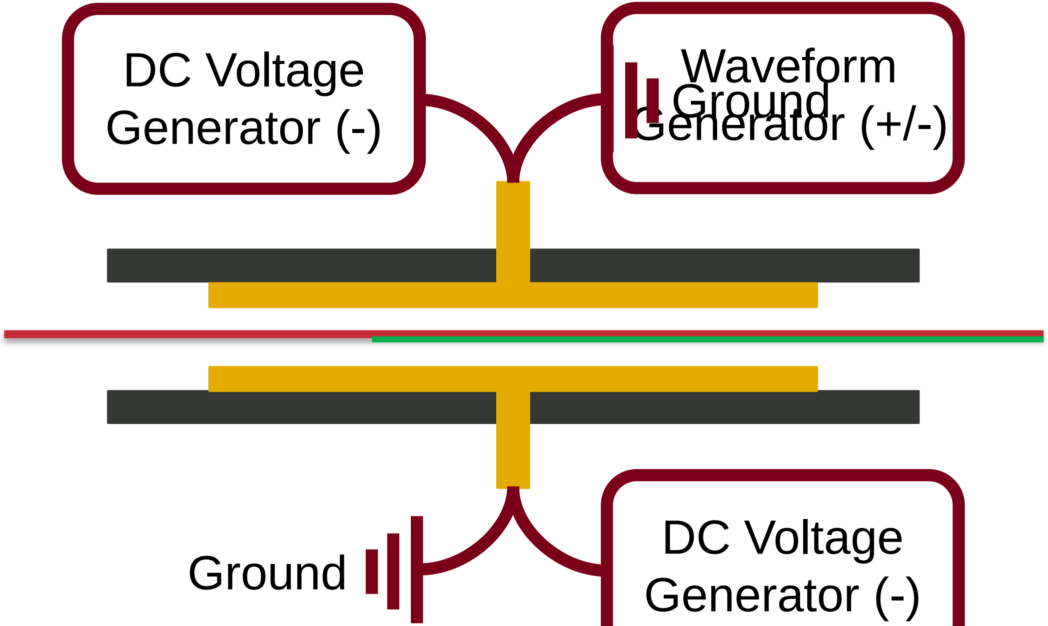
Electrode Setup

Top-down
Cross-section



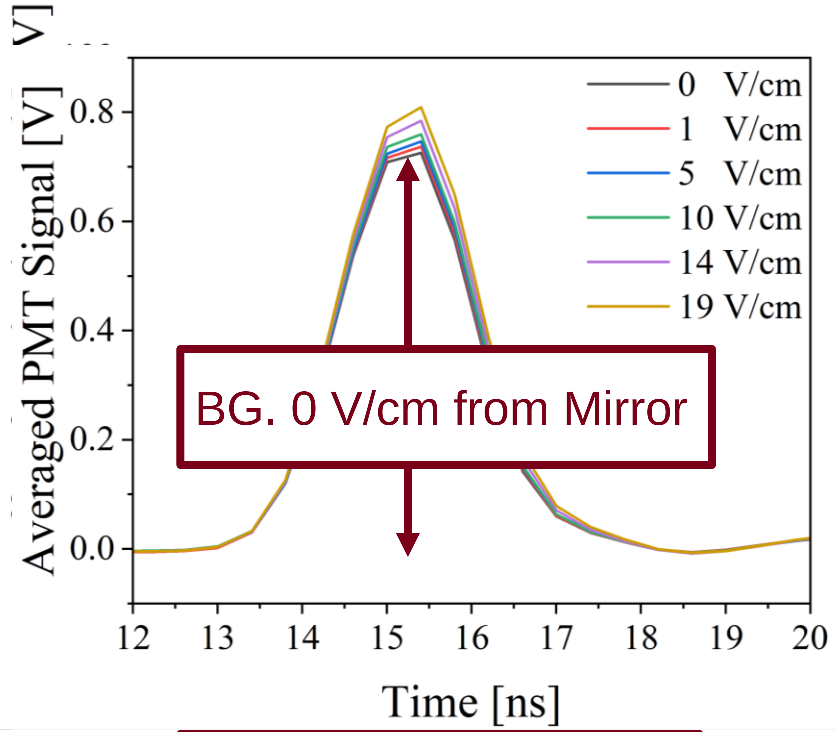
Effective Electrode
Length: **3.53 cm**

Side-on
Cross-section

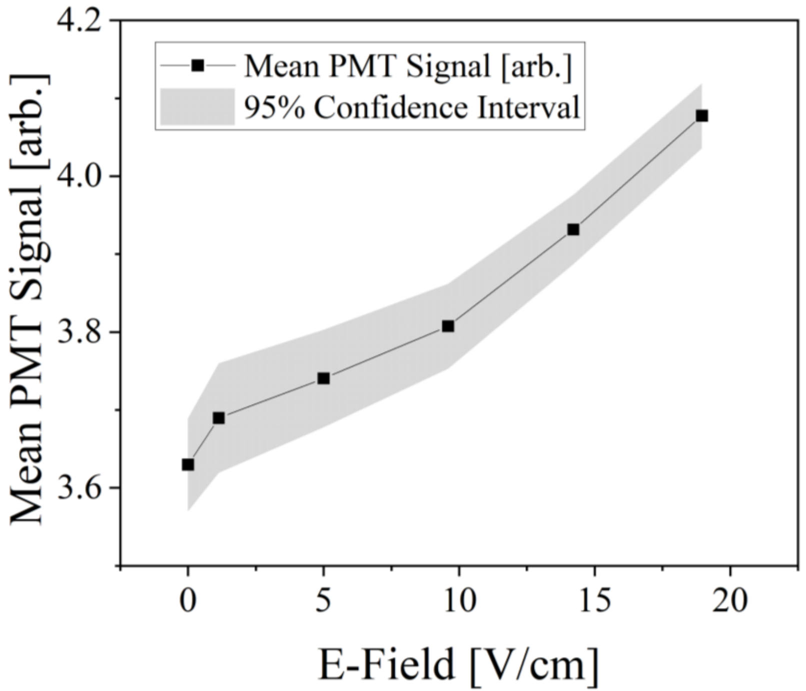


Initial DC Test Results (Pos. E-Field)

Average of over 19,000 Laser shots for each voltage

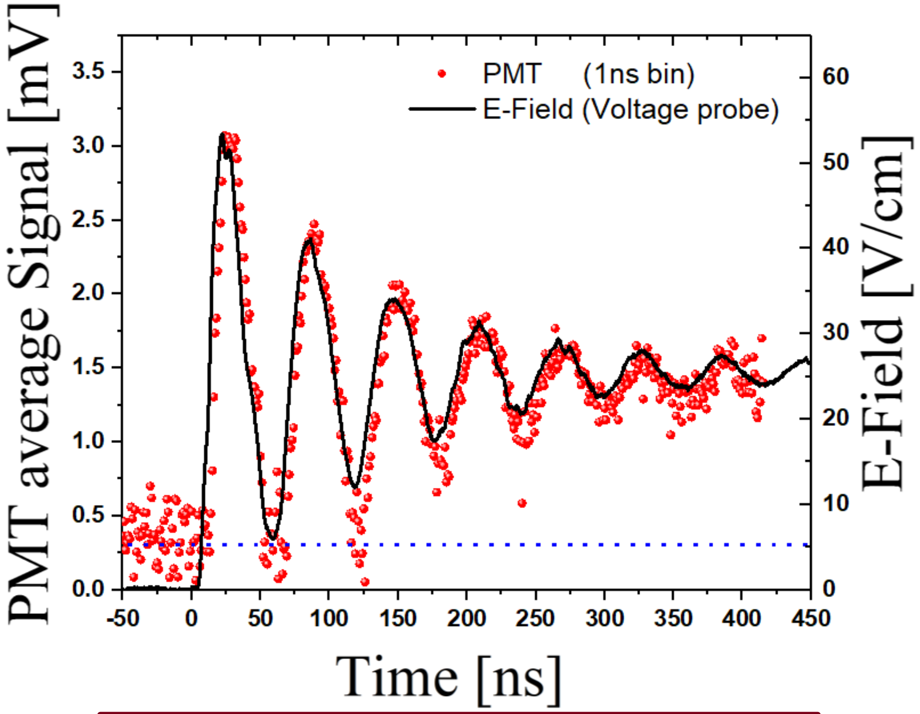


Averaged PMT Traces

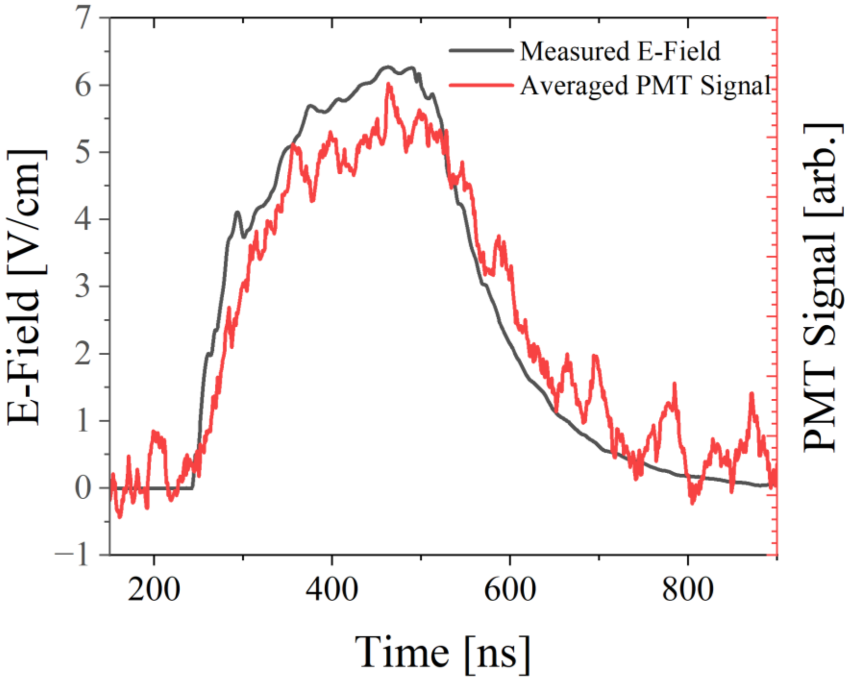


Integrated Signal

Time Resolved Data, High Voltage Pulse

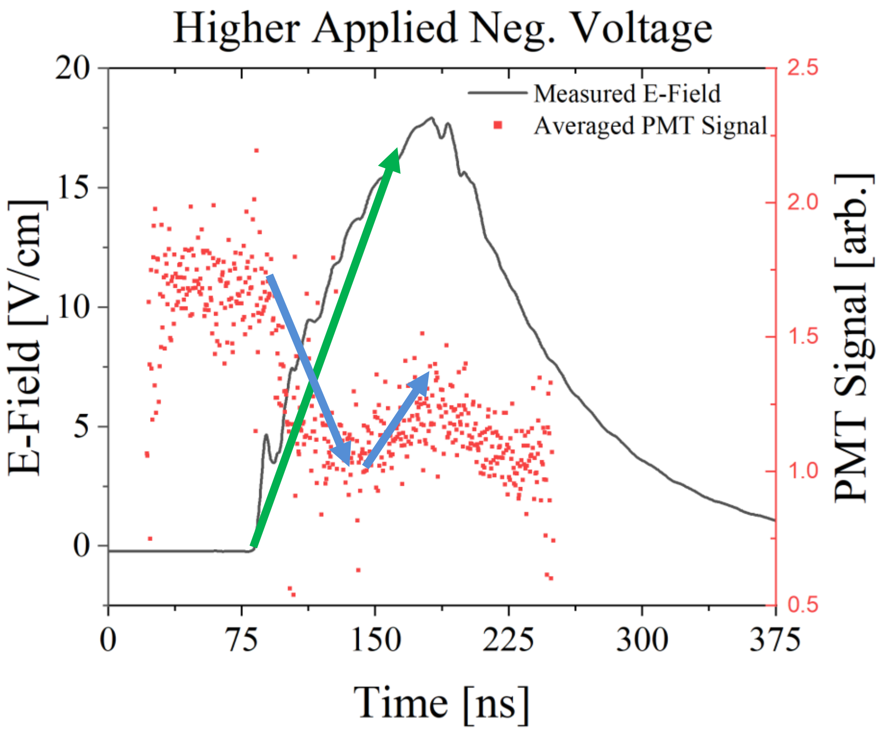


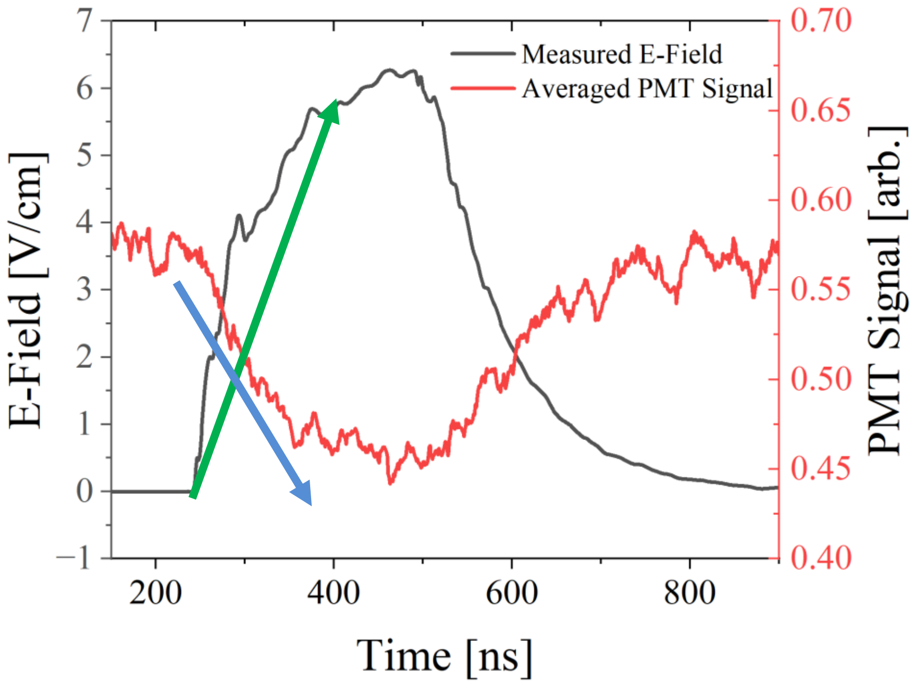
Pulse from high voltage source



Pulse from Delay Generator

True Waveform Shape (Neg. E-Field)

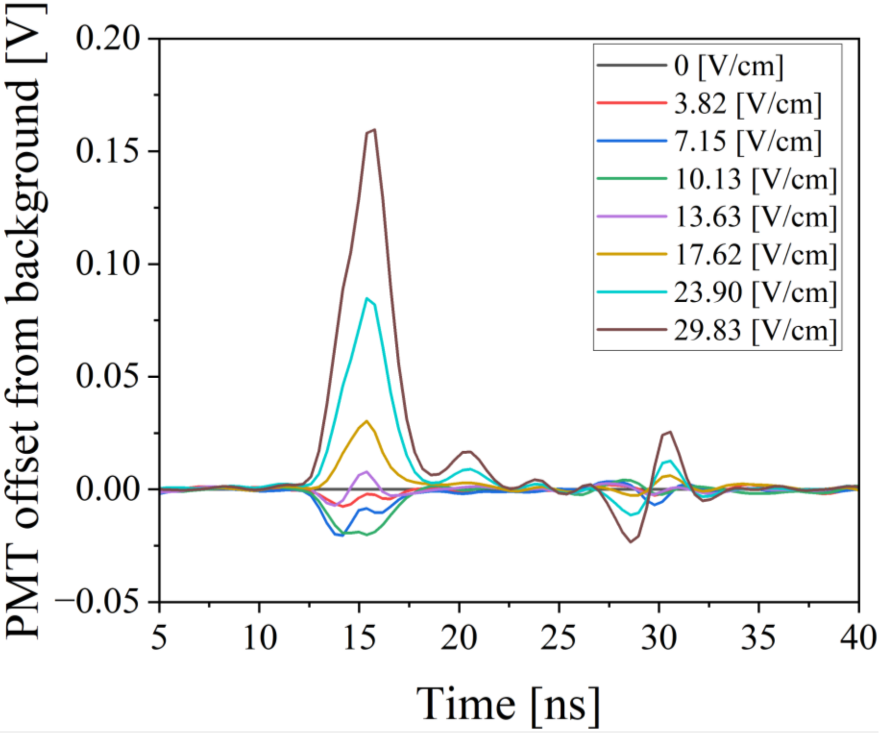


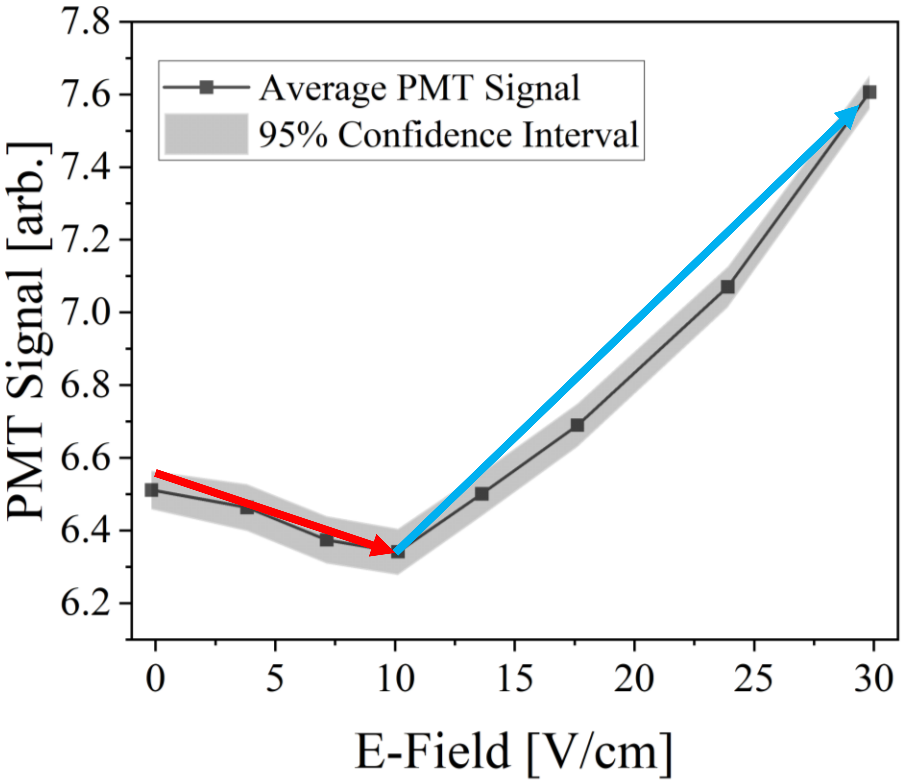


Inverted E-Field DC Results

(Neg. E-Field)

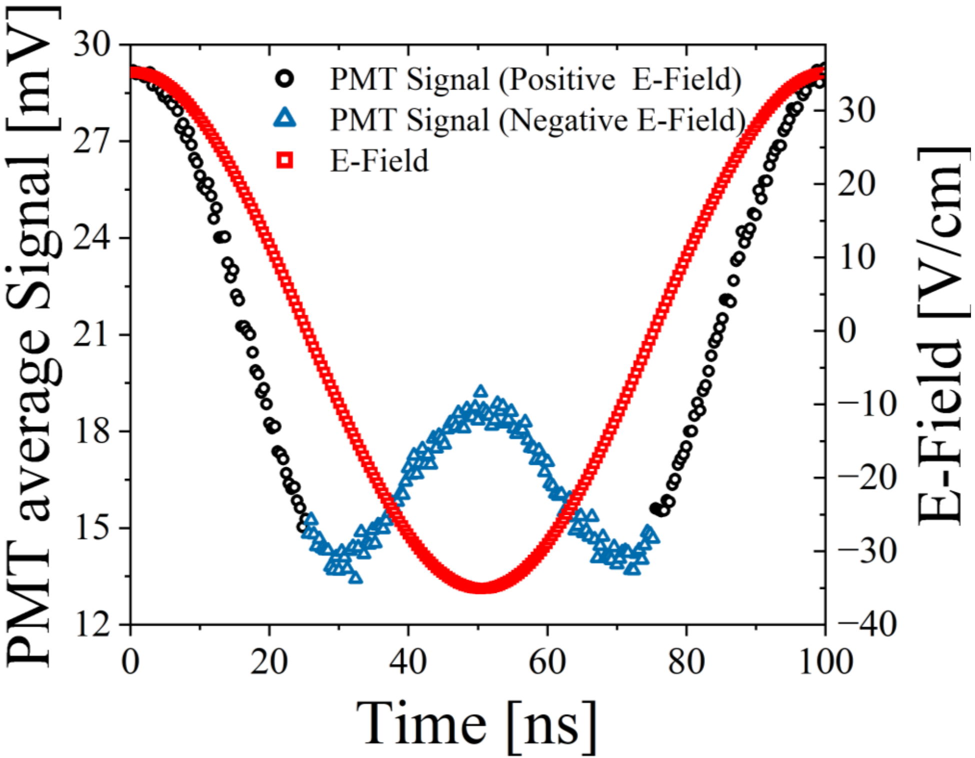
Average of at least 29,000 Laser shots for each voltage



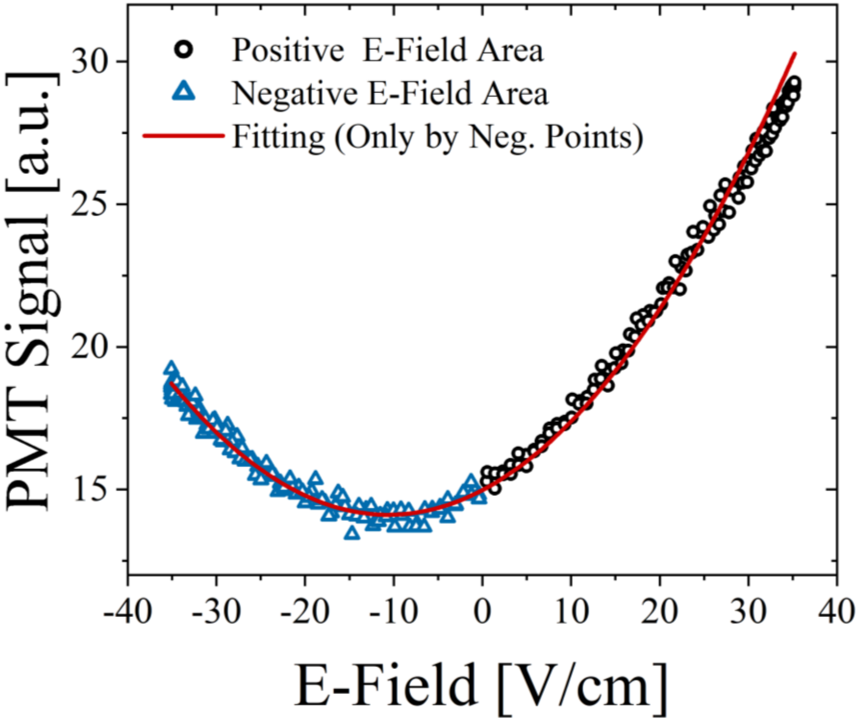


Sine Wave Test Results

Average of 3000 Laser shots per 0.4 ns bin

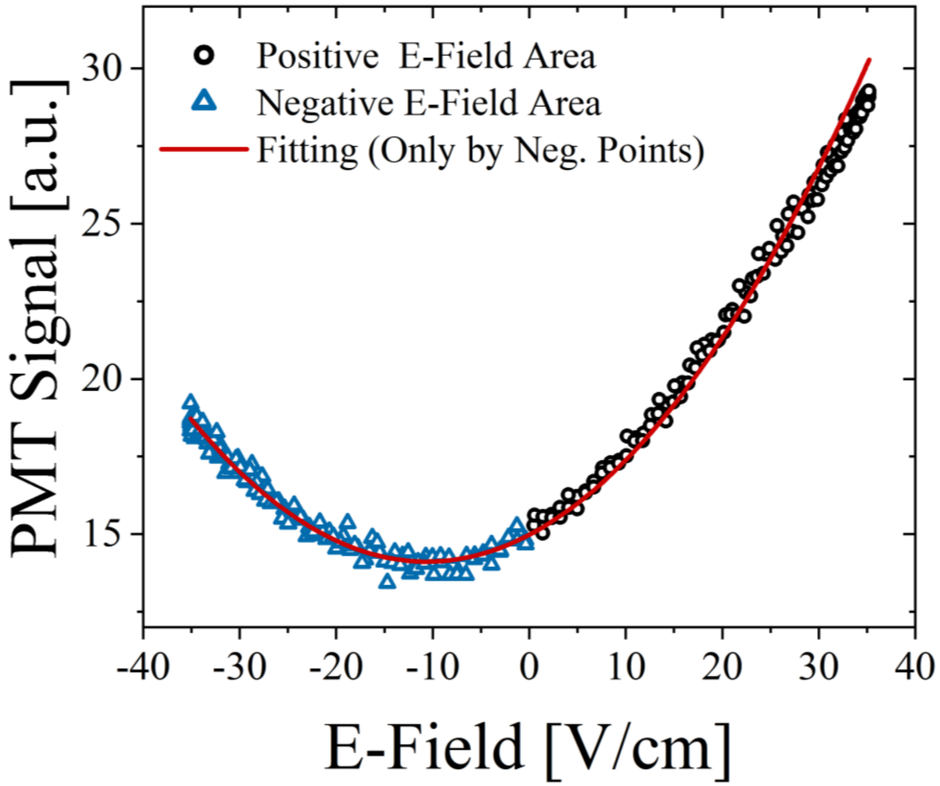


Time-Resolved Sine Wave



E-Field Dependence of PMT Signal

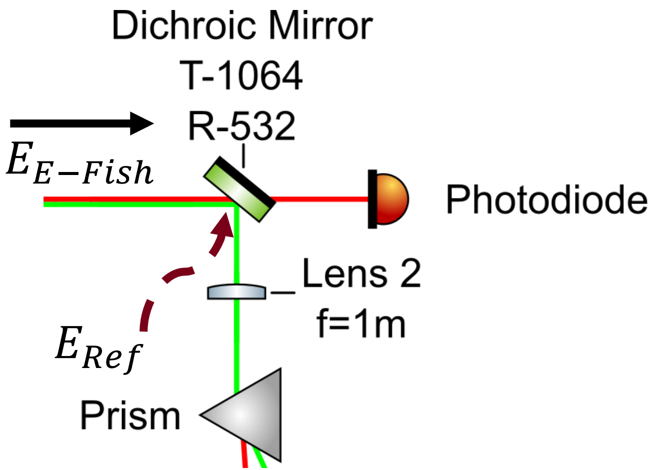
Homodyne Detection



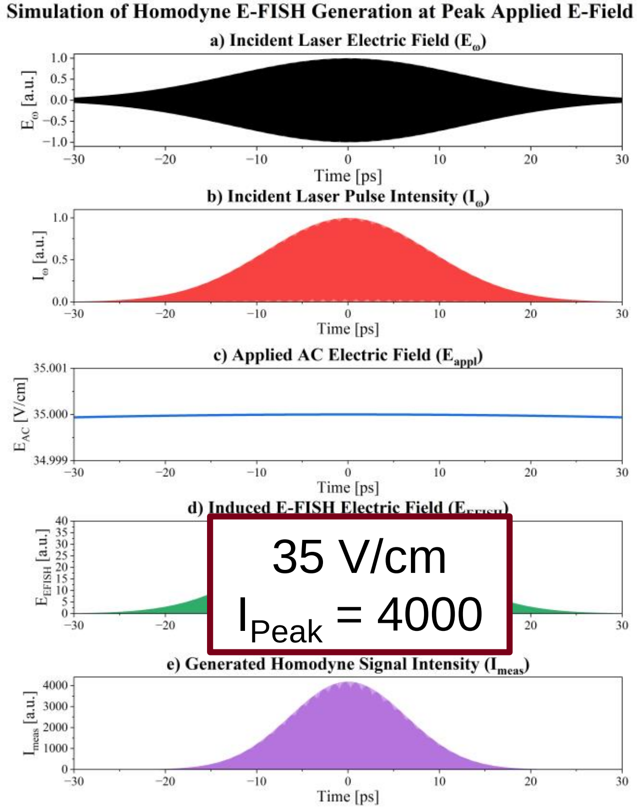
- Homodyne detection measures the polarity of the E-Field.

$$I_{pmt} \propto \left(E_{Ref} + E_{E-FISH}\right)^2$$

$$E_{Ref}^2 + \boxed{E_{Ref}E_{E-FISH}} + E_{E-FISH}^2$$



Homodyne Simulations



35 V/cm

I_{Peak} = 4000

Laser Electric Field Profile

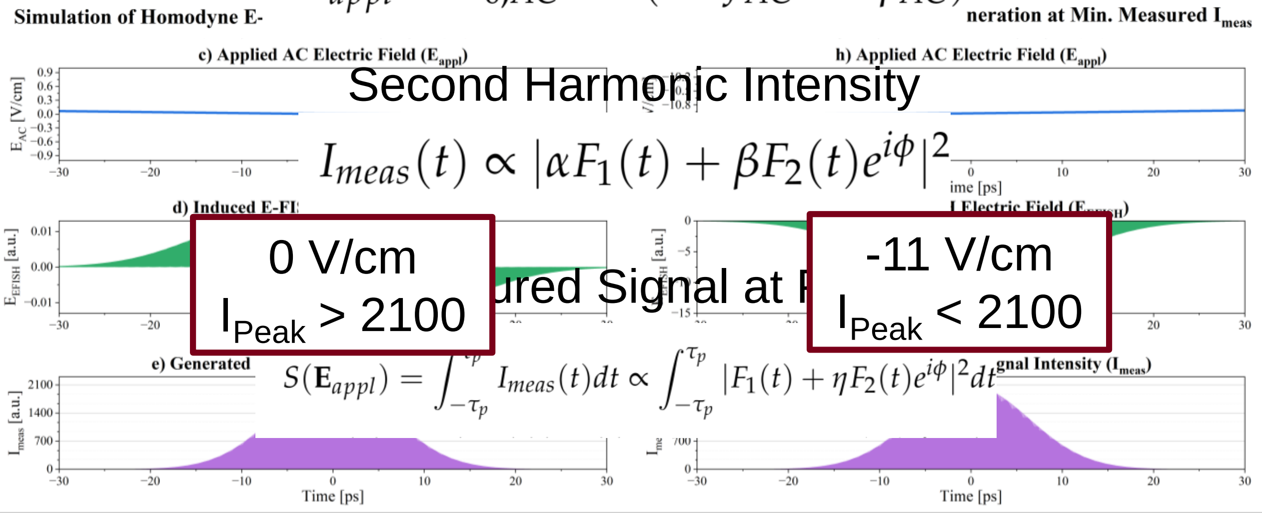
$$\mathbf{E}_\omega = E_{0,\omega}\, g(t)\ \cos(\omega_0 t)$$

Gaussian Temporal Profile

$$g(t) = e^{-4\ln 2\left(\frac{t}{\tau_p}\right)^2}$$

External Electric Field

$$\mathbf{E}_{appl} = E_{0,AC}\ \cos(2\pi f_{AC}\, t + \phi_{AC})$$



Second Harmonic Intensity

$$I_{meas}(t) \propto |\alpha F_1(t) + \beta F_2(t) e^{i\phi}|^2$$

0 V/cm

I_{Peak} > 2100

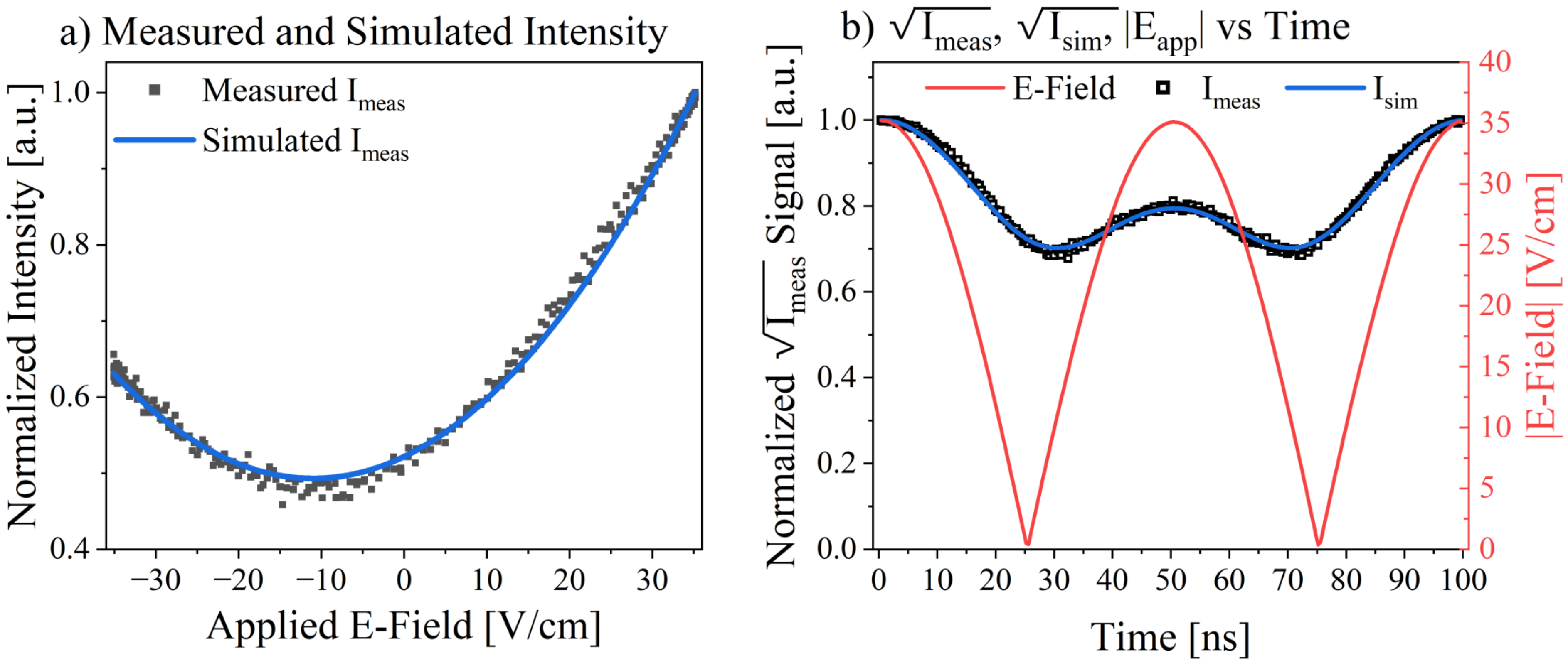
ured Signal at F

-11 V/cm

I_{Peak} < 2100

$$S(\mathbf{E}_{appl}) = \int_{-\tau_p}^{\tau_p} I_{meas}(t)dt \propto \int_{-\tau_p}^{\tau_p} |F_1(t) + \eta F_2(t)e^{i\phi}|^2 dt$$

Homodyne Simulations

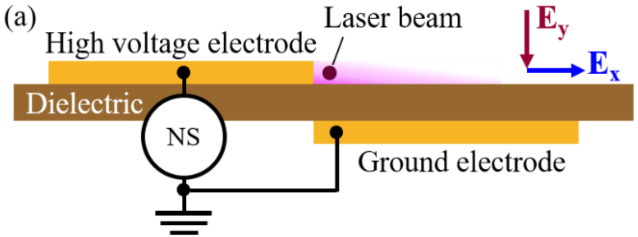


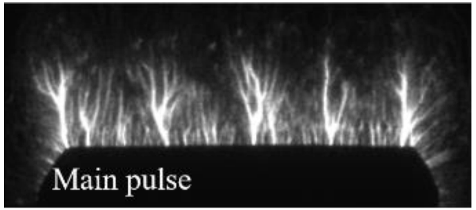
E-FISH Improvements

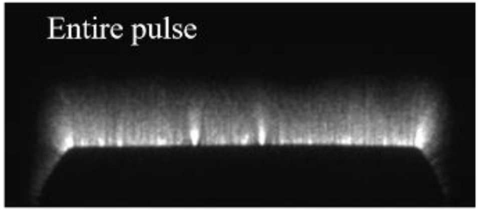
- Optimizations:
  - Homodyne enhancement
  - Optics for reference wavelength
  - Focusing to maximize signal generation
  - Signal collection efficiency
- Measurement Capability:
  - E-Fields down to **1 V/cm at 1 atm**
  - **Measure Sign of E-Fields**

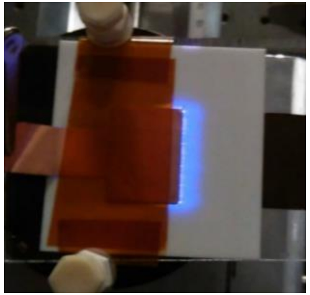
In-situ Characterization of Semiconductor Materials using Surface-enhanced SHG

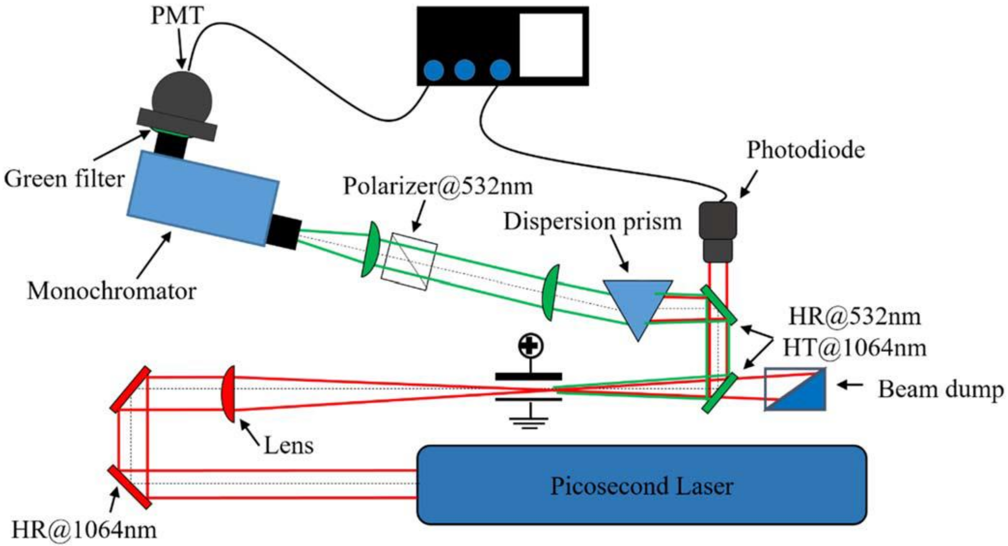
Transmission Setup for Surface Measurements











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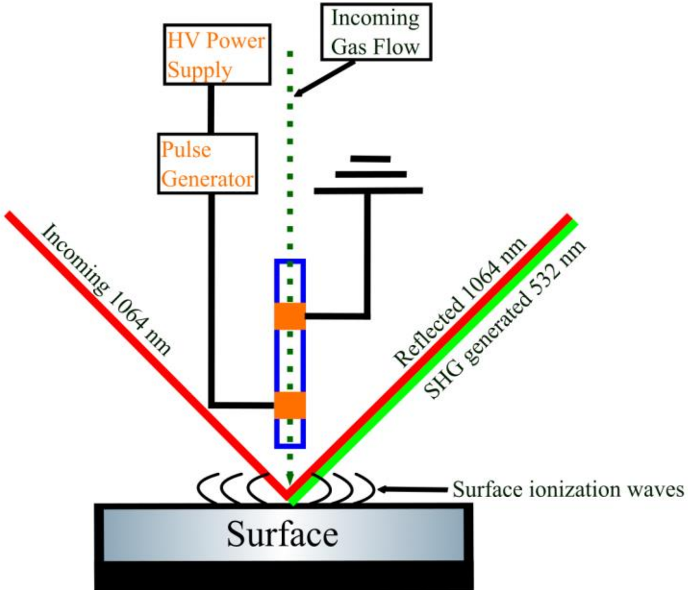
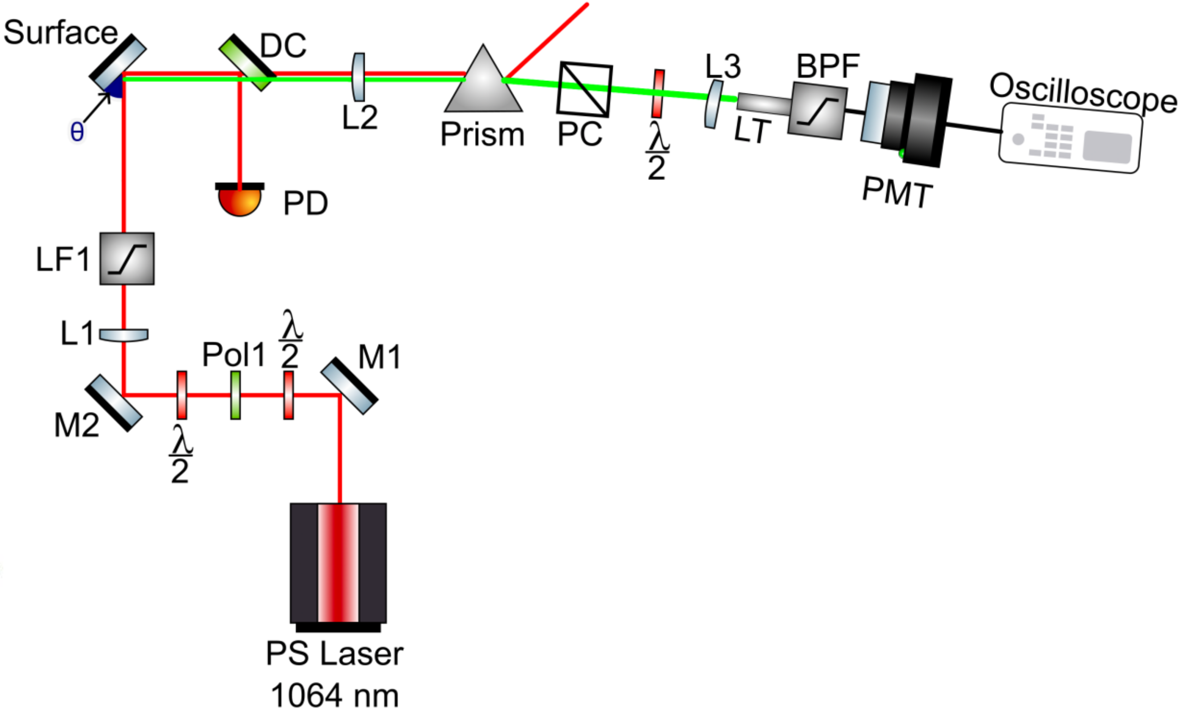
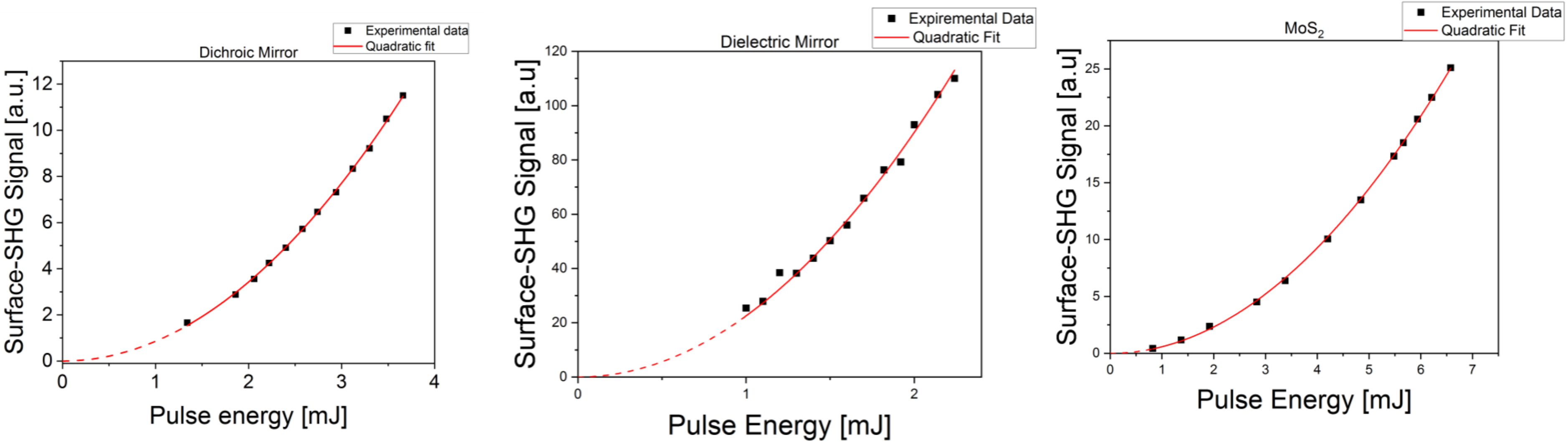
Reflection Setup for Surface Measurements





Preliminary Results for surfaces: Intensity



- Expected quadratic dependence of the SHG signal with the incident laser pulse energy
- Different surfaces lead to different intensities

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Questions

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