U.S. NAVAL RESEARCH LABORATORY

AE128: Ionization Currents and Secondary Radiation from Two-Color Pulses at Long Laser Wavelengths

Proposal # 312798

Funding Status: NRL Base Program, Received

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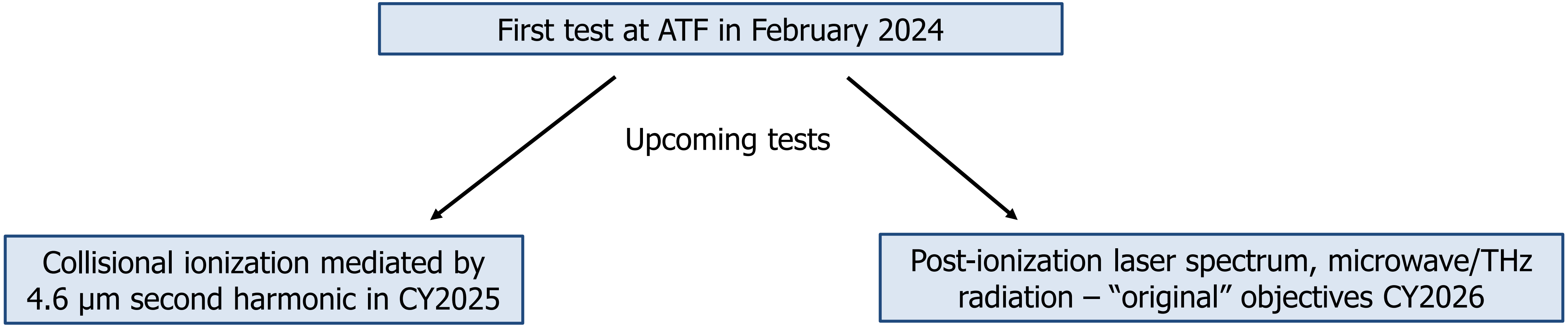
Summary of Progress

- Study ionization physics of air for short-duration LWIR laser pulses
- Use two-color LWIR pulses to create controllable laser electric field waveform, and measure how the initial laser-air interaction changes as a function of the two-color harmonic relative phase



Originally Proposed Objectives for AE128

NRL PPD

- **Map wavelength dependence of air ionization**
- Understand ionization mechanisms and optical nonlinearities:
  - Plasma secondary radiation, electron density, plasma optical emission spectrum
  - Infer changes to laser electric field from forward scattered spectrum– rotational/vibrational Raman scattering, self and cross (ω/2ω) phase modulation, low-order harmonic generation
- Plasma secondary radiation = microwaves, THz, Brunel harmonics
- Integrate LWIR results in comparison study with NIR, SWIR, MWIR wavelengths with different lasers at NRL (Ti:S and OPA)

Originally Proposed Objectives for AE128

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After our first test at ATF, we have expanded the project objectives and will return to the originally proposed experiments in 2026

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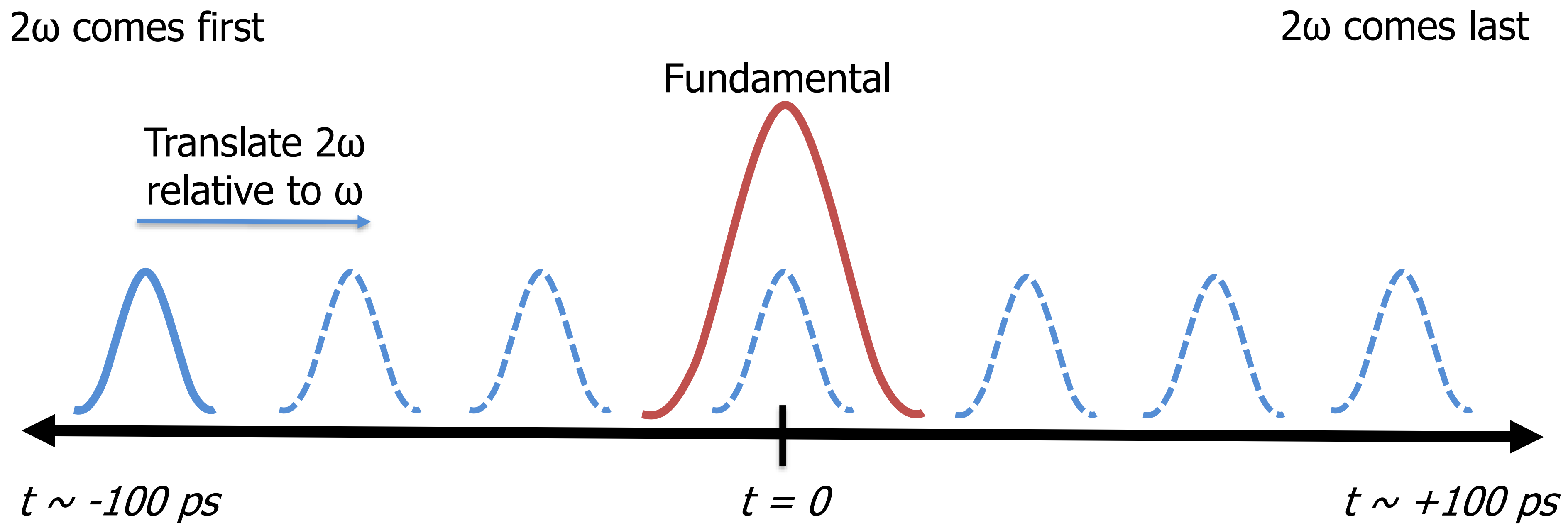
- Integrate LWIR results in comparison study with NIR, SWIR, MWIR wavelengths with different lasers at NRL (Ti:S and OPA)

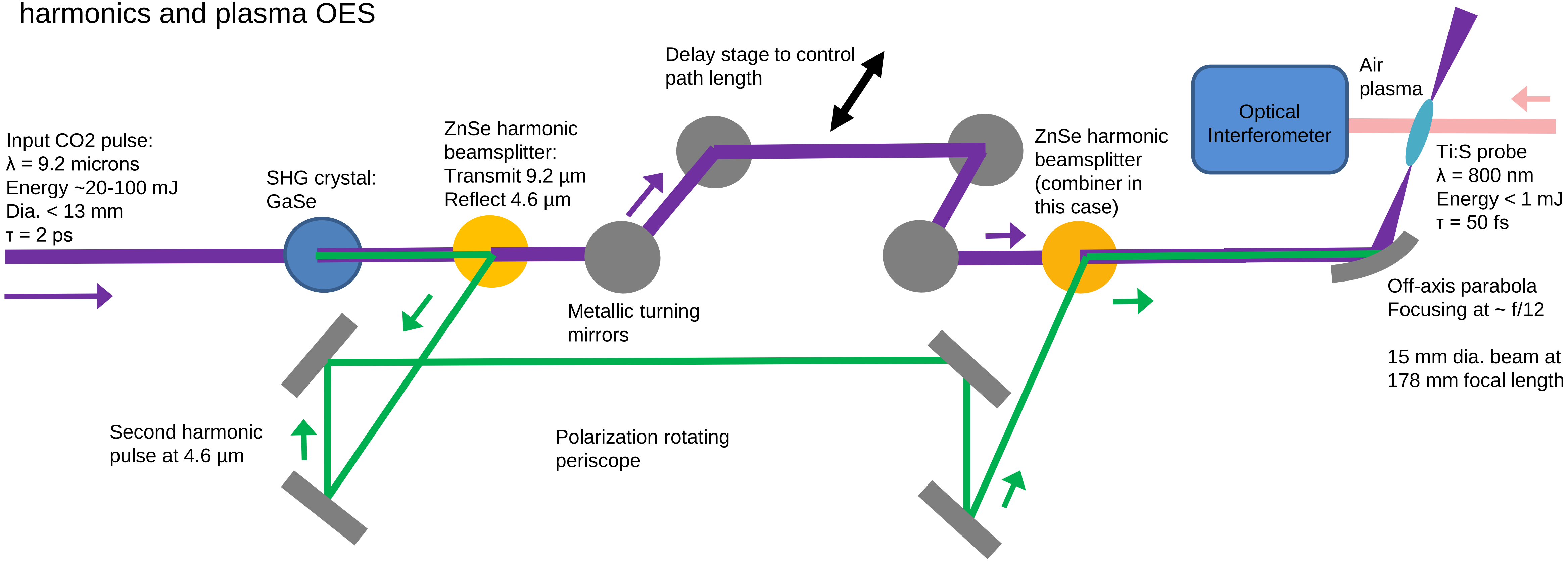
First BNL Experiments February 2024



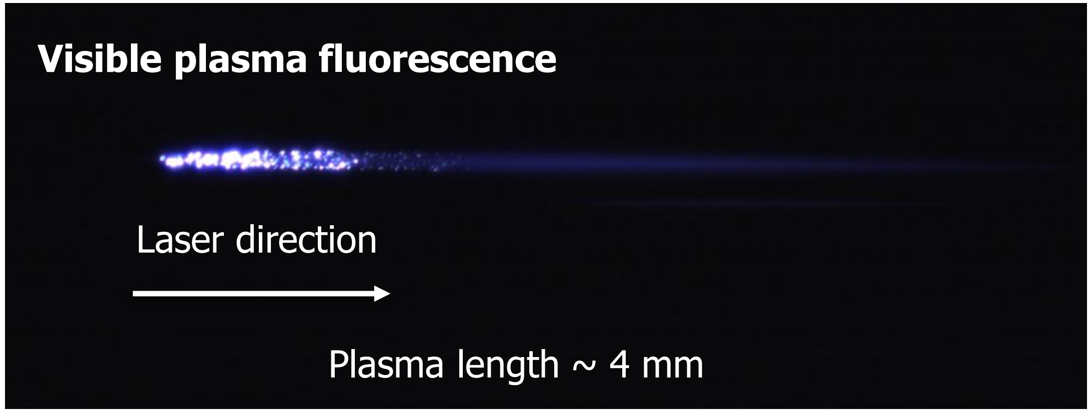
- Impose time delay between ω and 2ω
- Focus at *f/12* to make an air spark
- Maximum pulse energy into optical setup ~150 mJ, well below full compressed energy

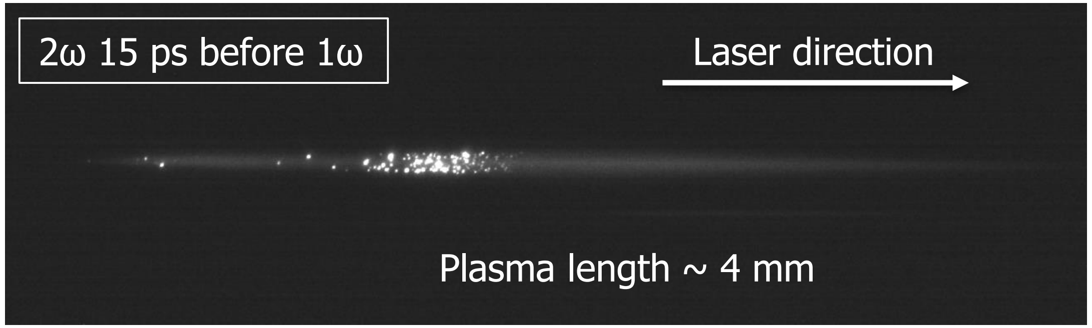
First BNL Experiments February 2024

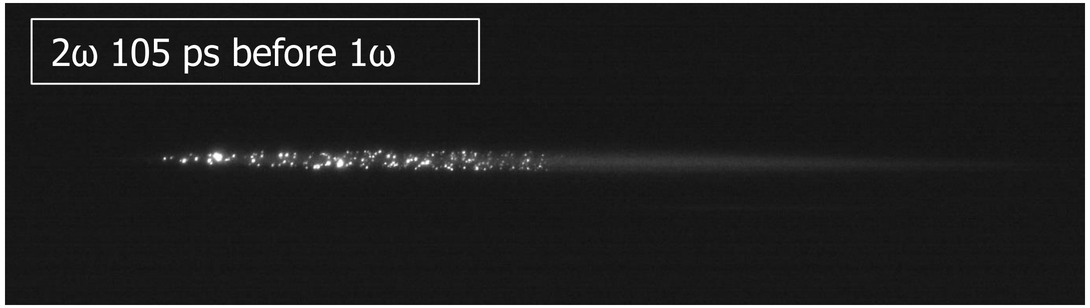
- Second harmonic generation by type-1 phase matching in GaSe
- Optical setup co-polarizes 1ω and 2ω fields by rotating 2ω polarization by 90 degrees
- Other diagnostics viewing the air plasma: d-dot probe, visible camera, spectrometers for laser harmonics and plasma OES



Second Harmonic Field Mediates Collisional Ionization



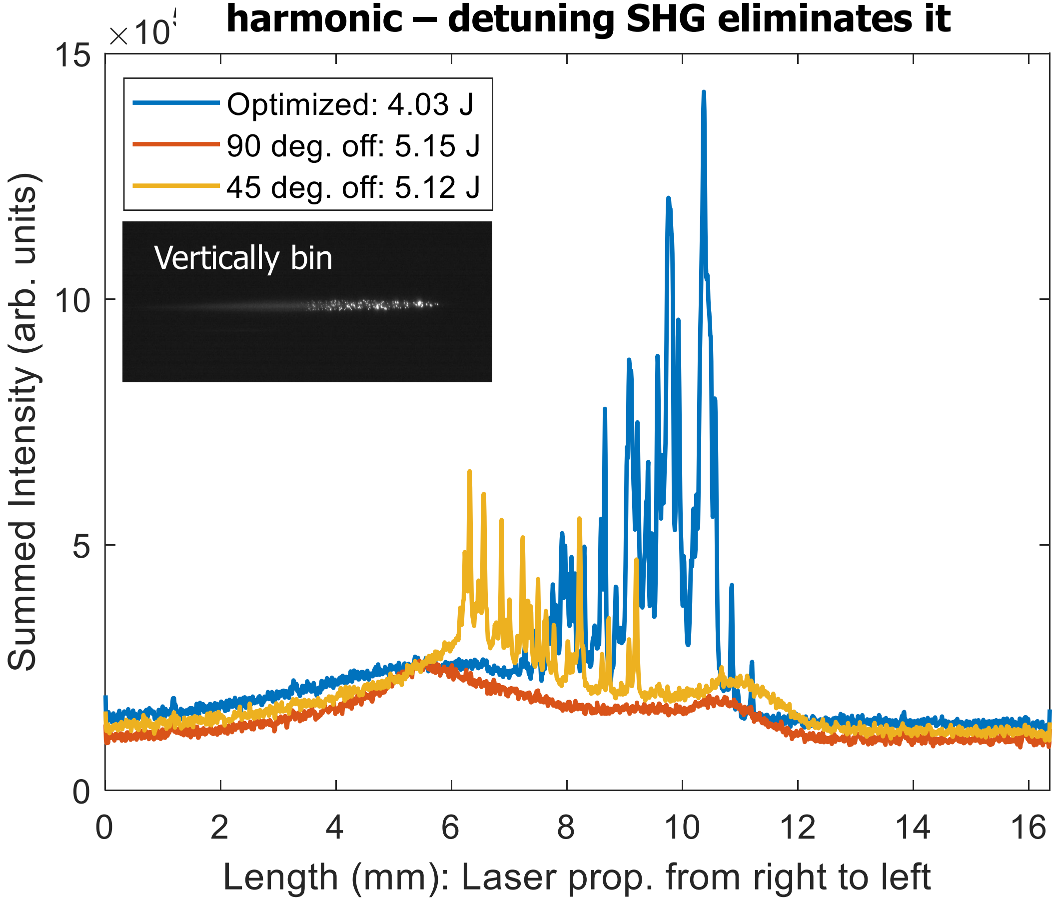




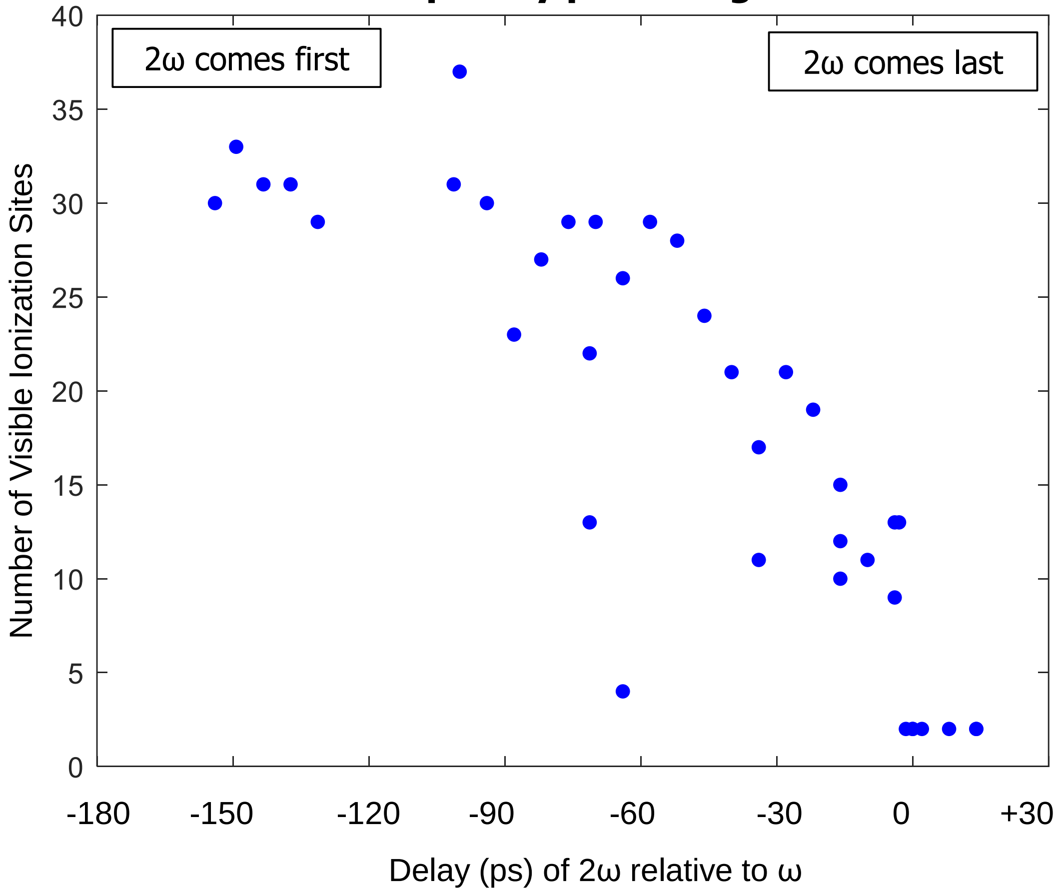
- Two-color LWIR pulse causes combined tunneling and collisional ionization – suggested by visible plasma fluorescence
- Collisionally ionized plasmas are hotter and denser than optical field ionized ones
- Distribution of collisional ionization seeds changes on 10's ps scale with delay between 1ω and 2ω

Second Harmonic Field Mediates Collisional Ionization

Collisional ionization requires second harmonic – detuning SHG eliminates it



Collisional ionization requires 2ω temporally preceding ω



Plans for Collisional Ionization Study

NRL PPD

- Mechanism by which 4.6 μm second harmonic wave mediates collisional air ionization demands further study

- Dedicate next test at ATF to this phenomenon

- Replicate experiment on NRL CO_2 laser
    - Use ZGP for second harmonic generation instead of GaSe
    - Debug diagnostics: optical emission spectroscopy, optical interferometry, coherent microwave scattering

- Theory and simulation to explain collisional ionization
    - CHMAIR+SPARC – simulate air chemistry in time (and space)
    - TurboWave – simulate laser interaction

- Vary pressure and gas species to enhance/diminish collisional ionization

Summary of Progress

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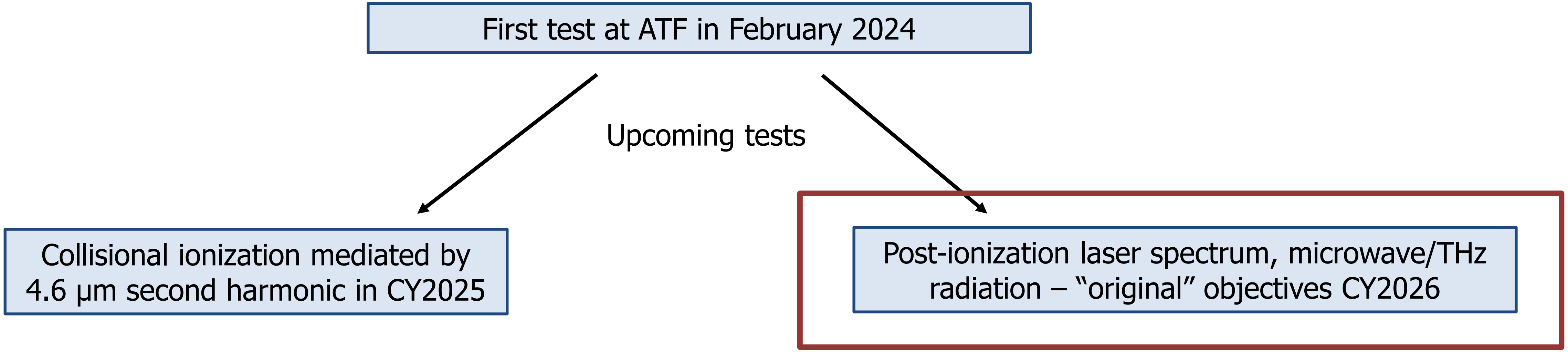
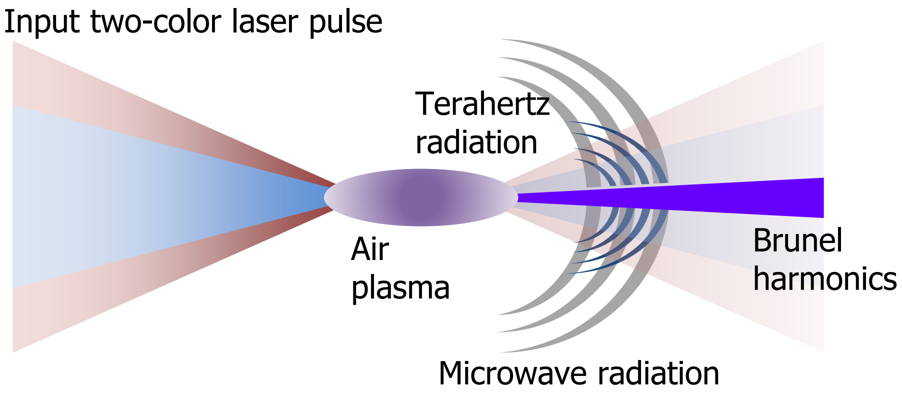
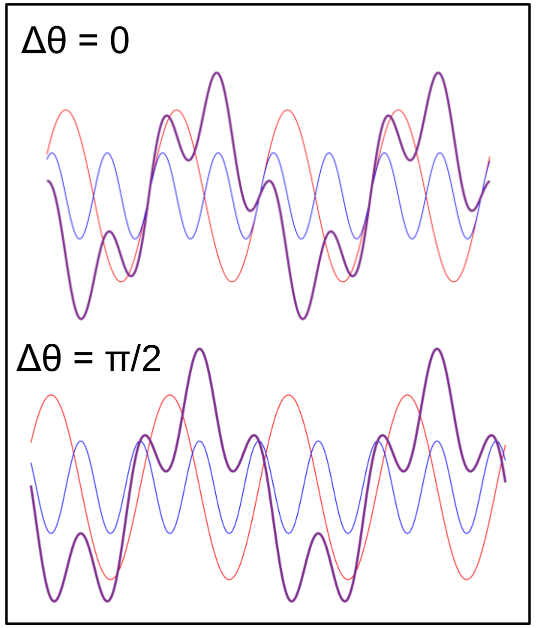


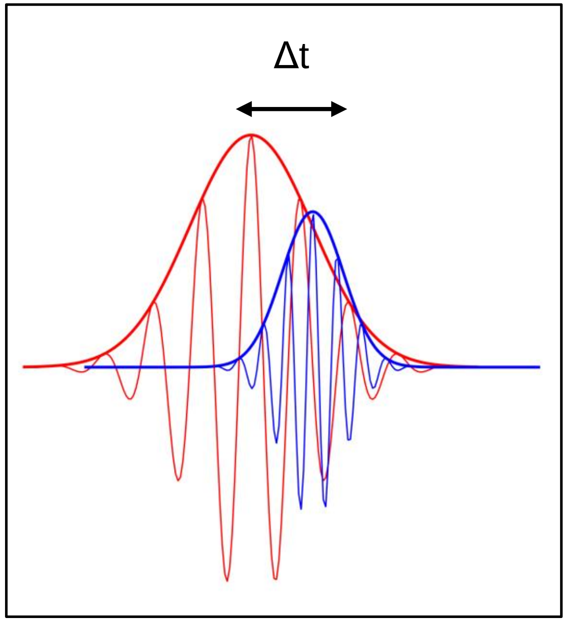
Coherent superposition of ω + 2ω



Relative phase (intra-cycle)



Envelope overlap (multiple cycles)



- Use a two-color pulse to coherently control ionization currents – make it easier to interpret diagnostics based on secondary radiation
- Secondary radiation = microwaves, THz, and Brunel harmonics
- Link early and late plasma characteristics with concurrent measurements of the plasma size, density, and optical emission spectrum

Plans for Ionization Currents/Secondary Radiation Study

NRL PPD

- Troubleshoot coherent ω + 2ω superposition using NRL CO_2 laser
- Simplify optical setup with custom crystal optics
  - Find dual LWIR wavelength waveplate to co-polarize harmonics
  - Find suitable transparent material for relative phase control either using birefringence or GVD
- Single-shot THz diagnostic under development in collaboration with AFRL/UNM
- Develop spectrometer capabilities/alignment procedure to cover VIS-LWIR
  - In 2024 test, did not observe Brunel harmonics or nonlinear harmonic generation
  - 2.2 μm was the longest wavelength we could measure at the time
  - Harmonic generation may require softer focus to allow phase-matched build-up
- Target CY2026 for these experiments

Products Delivered

Conference Presentations:

- "Ionization of Air by Intense Long Wave Infrared Two-Color Laser Pulses", Directed Energy Professional Society Science and Technology Symposium, May 2024

Journal Articles:

- LWIR Collisional Ionization Mediated by MWIR Second Harmonic Field, *In prep.*

CO_2 Laser Requirements

NRL PPD

Configuration	Parameter	Units	Typical Values	Comments	Requested Values
CO_2 Regen. Amplifier	Wavelength	μm	9.2	*Wavelength determined by mixed isotope gain media*	*9.2 μm*
	Peak Power	GW	~3		*~3*
	Pulse Mode	---	Single		
	Pulse Length	ps	2		*2*
	Pulse Energy	mJ	6		*6 mJ*
	Repetition Rate	Hz	1.5	*3 Hz also available if needed*	*~1 Hz*
CO_2 CPA Beam	Wavelength	μm	9.2	*Wavelength determined by mixed isotope gain media*	*9.2 μm*
Note that delivery of full power pulses to the Experimental Hall is presently limited to Beamline #1 only.	Peak Power	TW	2-3	*Up to 5 TW operation will be available in a limited number of shots upon the user's request.*	*< 100 GW*
	Pulse Mode	---	Single		*Single*
	Pulse Length	ps	2	*3-year development effort to achieve <500 fs at >10 TW and deliver to users is in progress.*	*2 ps*
	Pulse Energy	J	~5	*10J will be available in a limited number of shots upon the user's request.*	*~100-150 mJ*
	Strehl Ratio	---	~0.5	*Recommended conservative estimate subject to verification.*	
	Repetition Rate	Hz	0.01	*Burst operation at up to 0.05 Hz for a limited period is possible upon user's request. This regime should be avoided to extend the lifetime of the HV spark gaps in the amplifier's PFN*	*0.05*
	Polarization		Linear	*Adjustable linear polarization along with circular polarization can be provided upon request*	*Linear*

Other Experimental Laser Requirements

Ti:Sapphire Laser System	Units	Stage I Values	Stage II Values	Comments	Requested Values
Central Wavelength	nm	800	800	*Stage I parameters are presently available and setup to deliver Stage II parameters should be complete during FY22*	*800 nm*
FWHM Bandwidth	nm	20	13		
Compressed FWHM Pulse Width	fs	<50	<75	*Transport of compressed pulses will initially include a very limited number of experimental interaction points. Please consult with the ATF Team if you need this capability.*	*~ 50 fs*
Chirped FWHM Pulse Width	ps	≥50	≥50		
Chirped Energy	mJ	10	200		
Compressed Energy	mJ	7	~20	*20 mJ is presently operational with work underway this year to achieve our 100 mJ goal.*	*< 7 mJ*
Energy to Experiments	mJ	>4.9	>80		*< 4 mJ*
Power to Experiments	GW	>98	>1000		*<< 100 GW*

Nd:YAG Laser System	Units	Typical Values	Comments	Requested Values
Wavelength	nm	1064	*Single pulse*	
Energy	mJ	5		
Pulse Width	ps	14		
Wavelength	nm	532	*Frequency doubled*	
Energy	mJ	0.5		
Pulse Width	ps	10		

Special Equipment Requirements and Hazards

NRL PPD

- CO_2 laser
  - Keep 13 mm Teflon limiting aperture in place
- Ti:S laser for interferometry probe
- **New: Small vacuum/pressure vessel, gas cylinders**
- Microwave scattering diagnostic: already on ESR, but we may need to increase the power transmitted in free space

Experimental Time Request

NRL PPD

CY2025 Time Request

Capability	Setup Hours	Running Hours
Electron Beam Only		
Laser* Only (in Laser Areas)	60	60
Laser* + Electron Beam		

Total Time Request for the Experiment remaining duration (incl. CY2025)

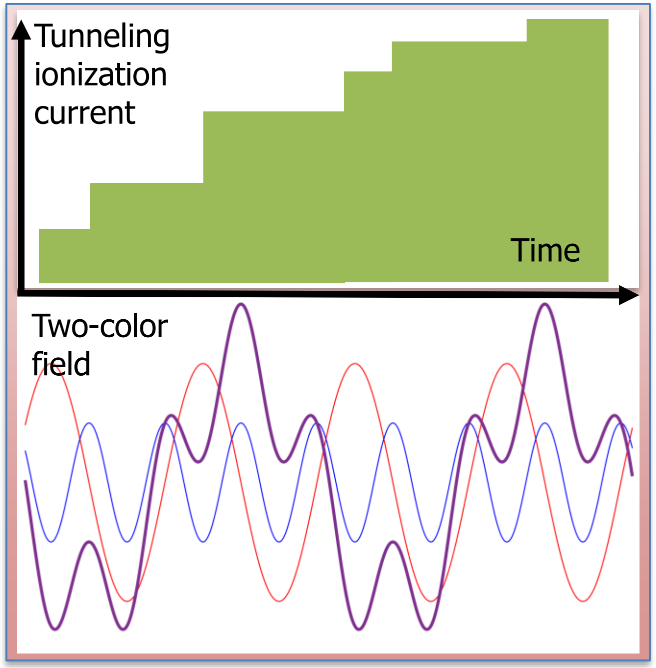
Capability	Setup Hours	Running Hours
Electron Beam Only		
Laser* Only (in Laser Areas)	120	120
Laser* + Electron Beam		

* Laser = Near-IR or LWIR (CO_2) Laser

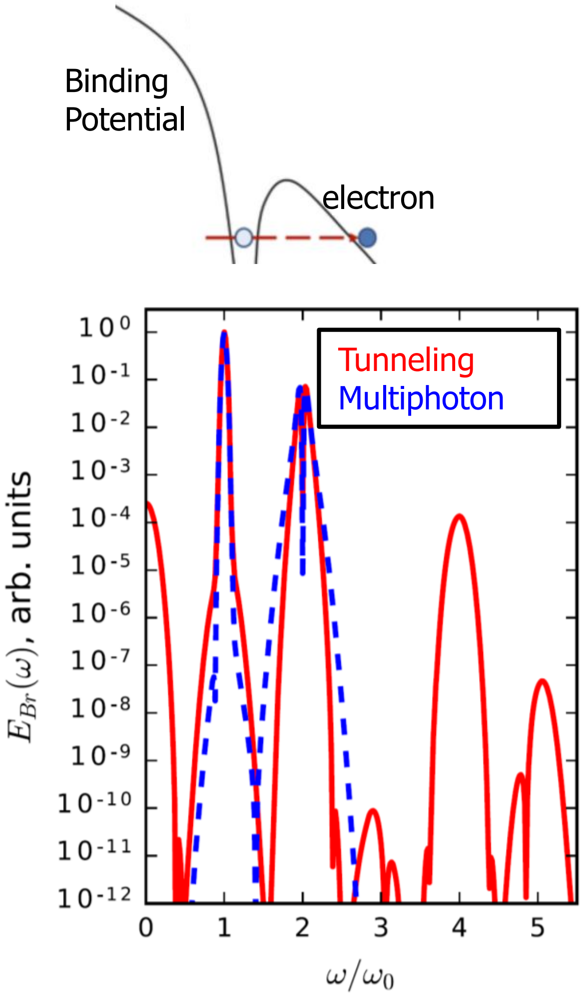
Back-up slides

Ionization Currents Determine Secondary Radiation

- Optical field ionization by intense laser pulses occurs in the tunneling or multiphoton regime – Keldysh theory
- For ionization of atmospheric air, traditional NIR sources ionize via multiphoton
- Long wavelengths are needed to drive tunneling ionization in air
- The liberation of electrons constitutes an ionization current, whose time dependence can be step-like (tunneling) with each laser field maximum, or vary slowly with the laser envelope (multiphoton)
- The relative phase of a two-color pulse changes the shape of the laser electric field waveform, and thus the ionization currents



Tunneling

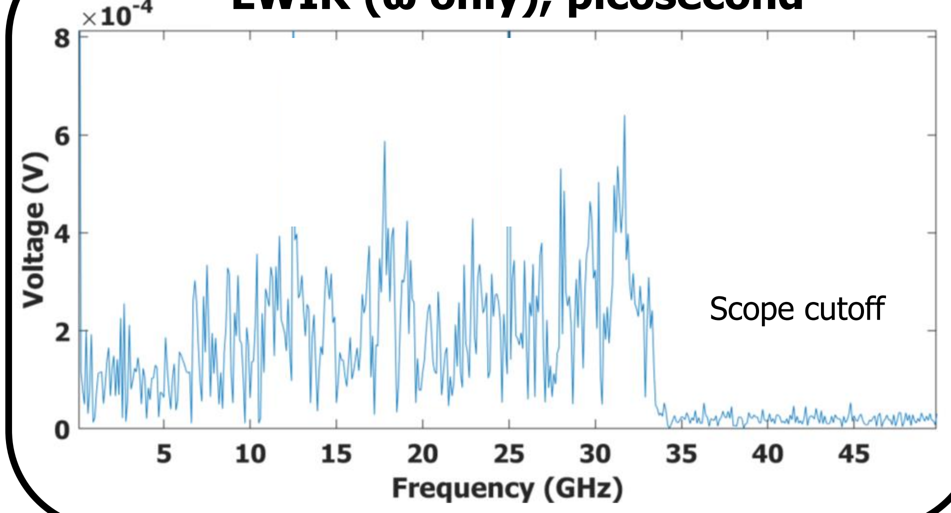


I. Babushkin, *J. Mod. Opt.* 64(10), 1078 (2017).

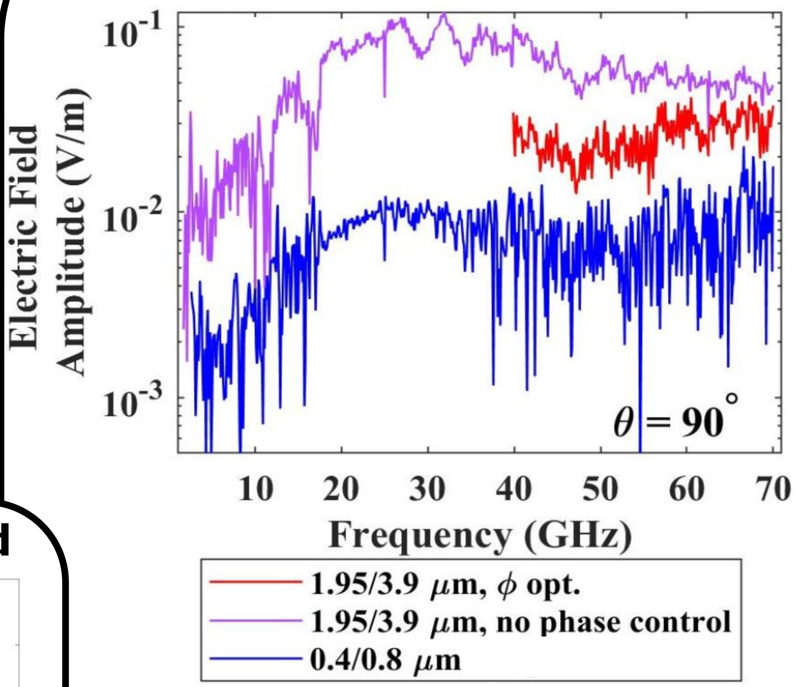
Wavelength Scaling of Two-Color Microwave Generation Underway

NRL PPD

LWIR (ω only), picosecond

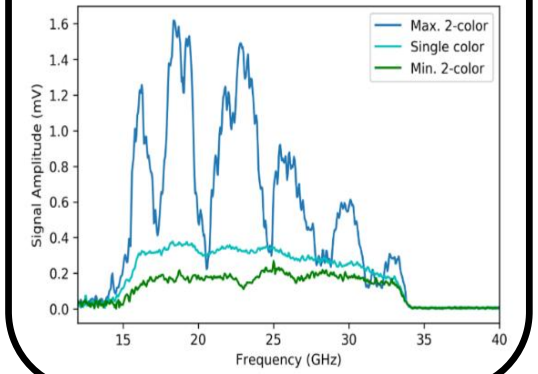


MWIR vs NIR, femtosecond

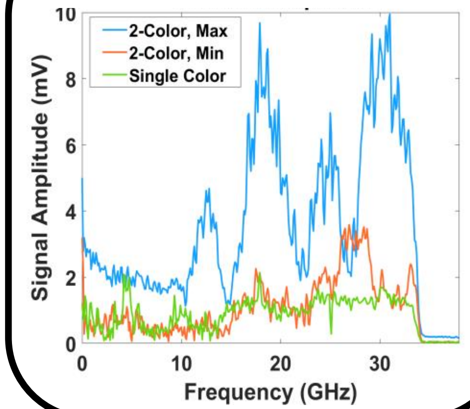


Englesbe, A. Dissertation, U. Michigan (2019).

SWIR, picosecond



NIR, femtosecond



- Progressing toward complete wavelength scaling from NIR to LWIR
- Have some two-color microwave generation data for 1ω = 0.8, 1.5, 3.9, and 9.2 μm
- Different peak powers, focal geometries, microwave receivers
- Generally speaking, ponderomotive scaling holds

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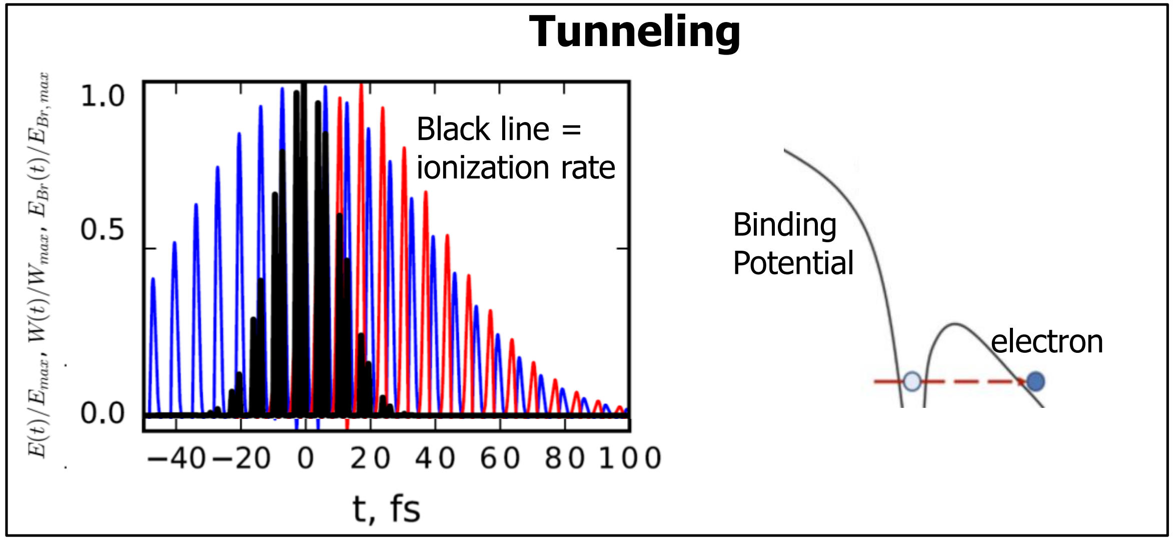

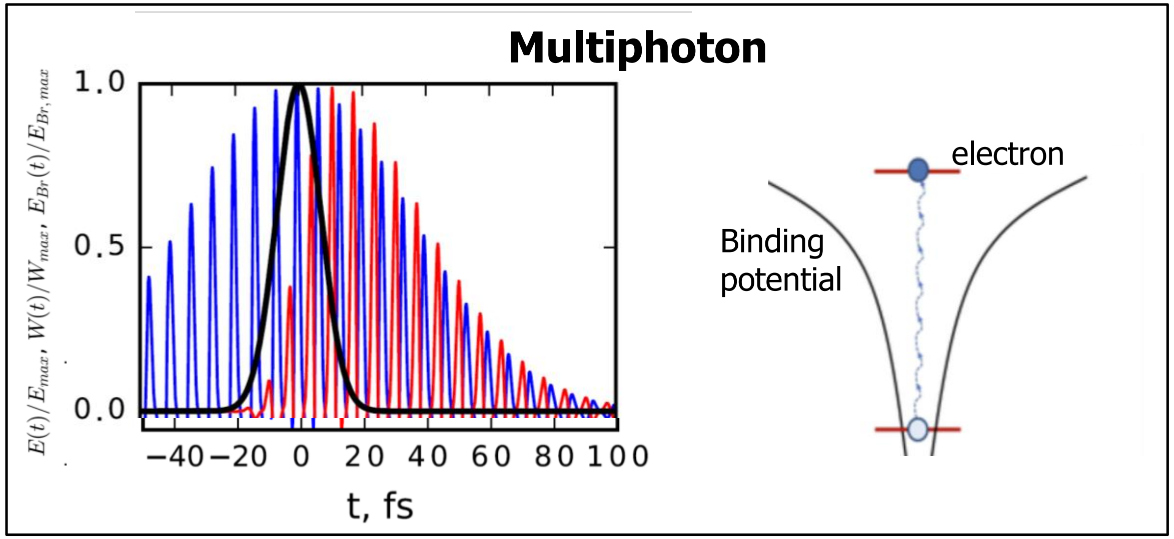

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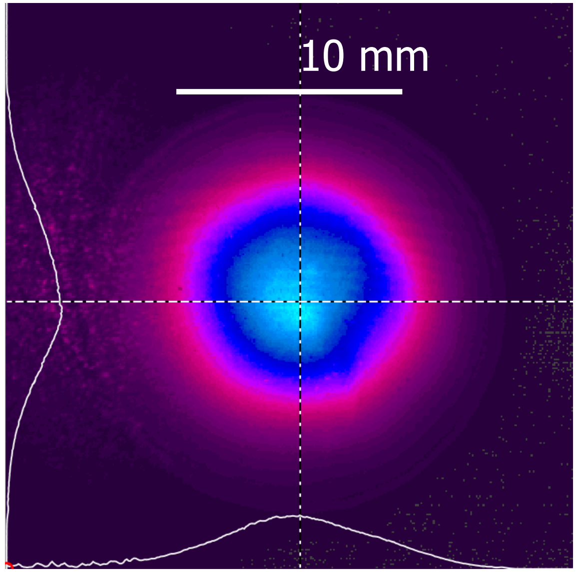
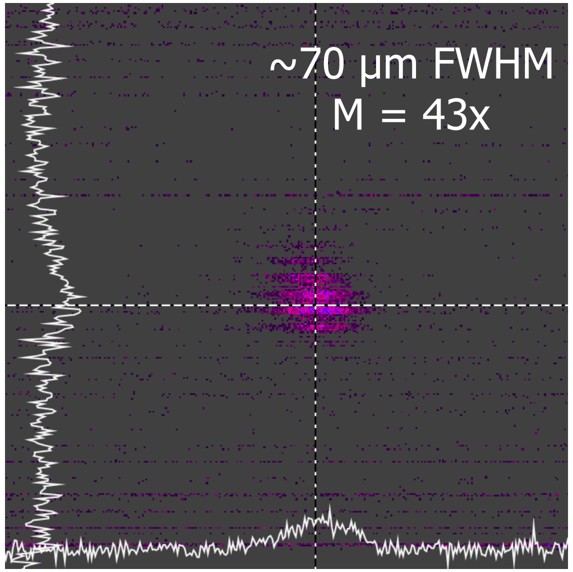
Generated Two-Color LWIR pulses GaSe for SHG

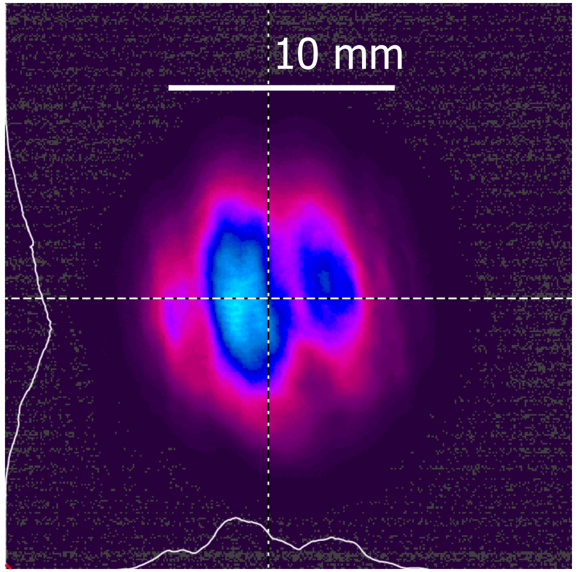
Input 9.2 μm Profile



OAP Focal Spot



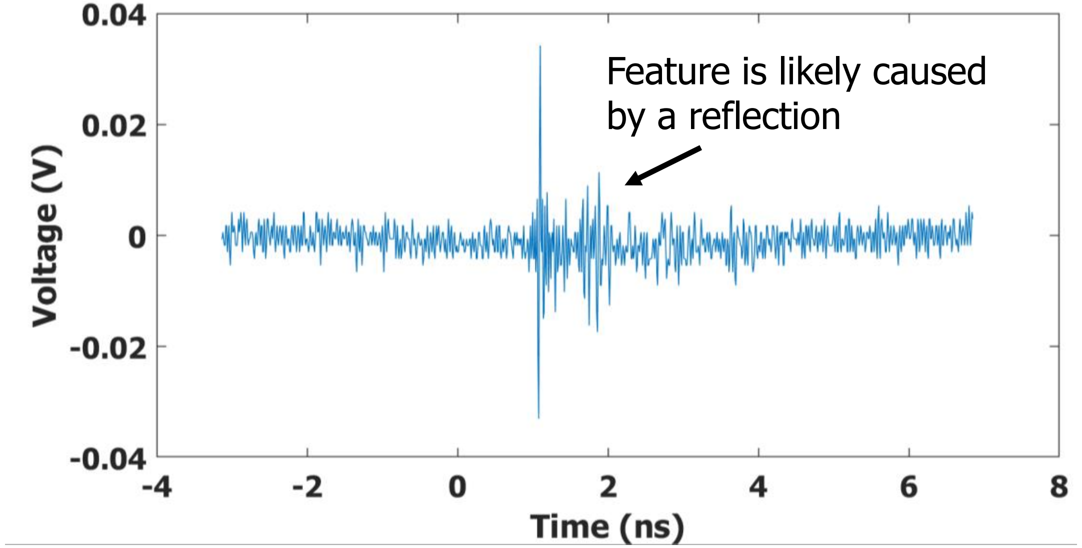
Input 4.6 μm Profile



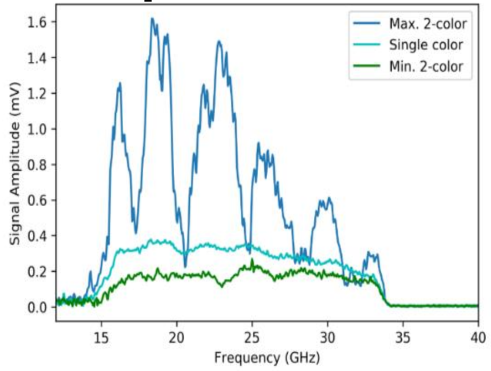
- Good input beam quality led to ~70 μm FHWM focal spot
- Keldysh parameter ~ 0.04, focal intensity ~ 10^{14} W/cm^2
- Achieved sufficient SHG conversion efficiency (~7%) for two-color pulse
- GaSe crystal quality is not great - note structure in 2ω profile
- ~50% energy throughput due to reflections – lose ~30% pulse energy on each crystal surface

Two-Color Enhancement Absent for Microwave Radiation

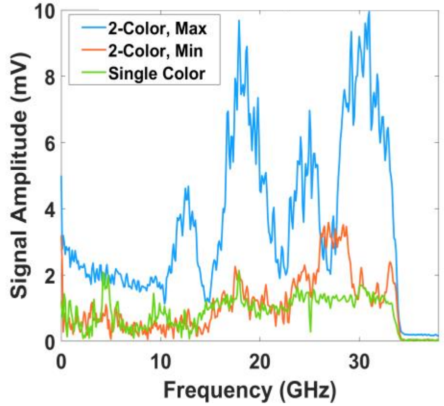
- Microwave amplitude is large considering 15 GW peak power (~30 mJ going into focusing optic in 2 ps pulse) – expected from ponderomotive scaling
- With a NIR laser pulse peak power of 15 GW in similar focusing geometry microwave peak power would probably not exceed noise floor without amplification
- When two-color pulse is properly phased, observe amplitude enhancement and spectral modulations (see NIR, SWIR examples)
- We did not see amplitude enhancement or change in spectrum during delay stage scans
- Careful analysis required for meaningful comparisons between laser wavelengths



SWIR, picosecond



NIR, femtosecond



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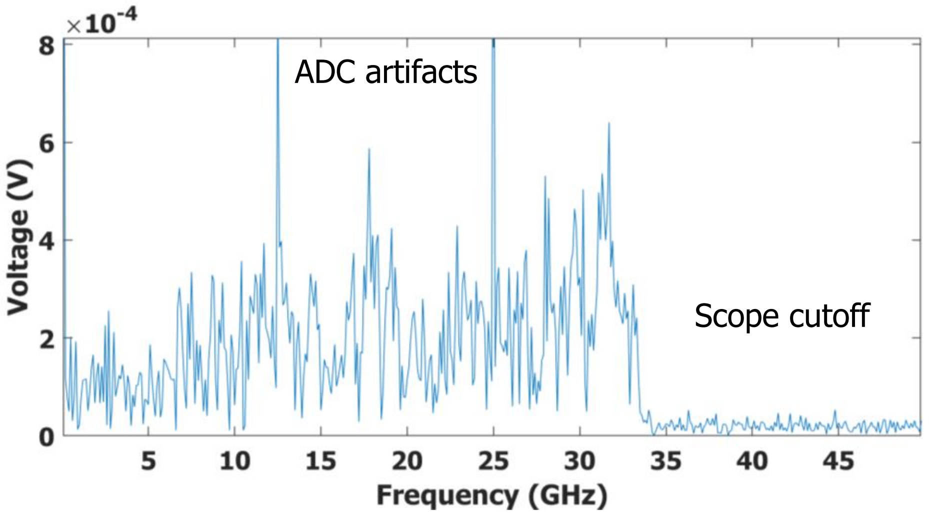
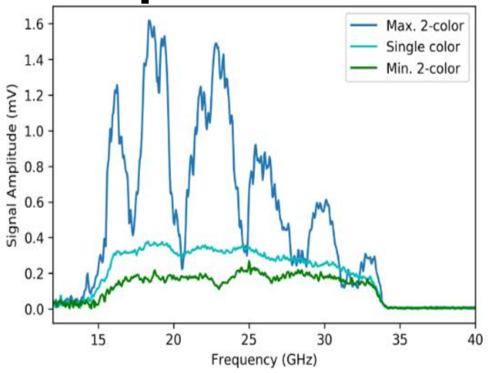


SWIR, picosecond



NIR, femtosecond