U.S. NAVAL RESEARCH LABORATORY

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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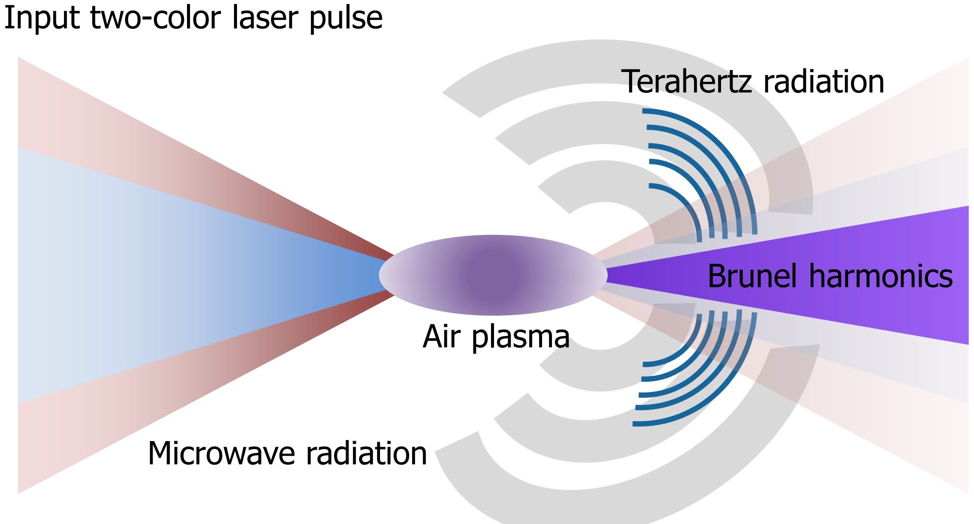
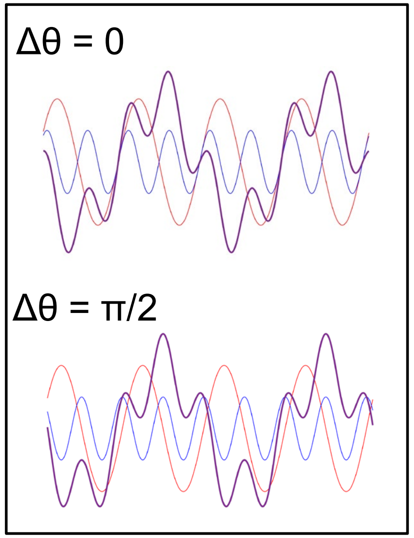
Controlling Tunneling Ionization of Air with Two-Color Pulses

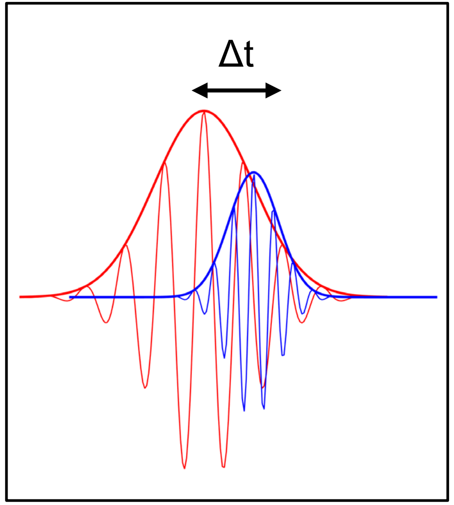
- Optical field ionization by intense laser pulses occurs in the tunneling or multiphoton regime – Keldysh theory
- For ionization of atmospheric air, traditional NIR sources ionize via multiphoton
- Long wavelengths are needed to drive tunneling ionization in air
- The liberation of electrons constitutes 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



Relative phase (intra-cycle)

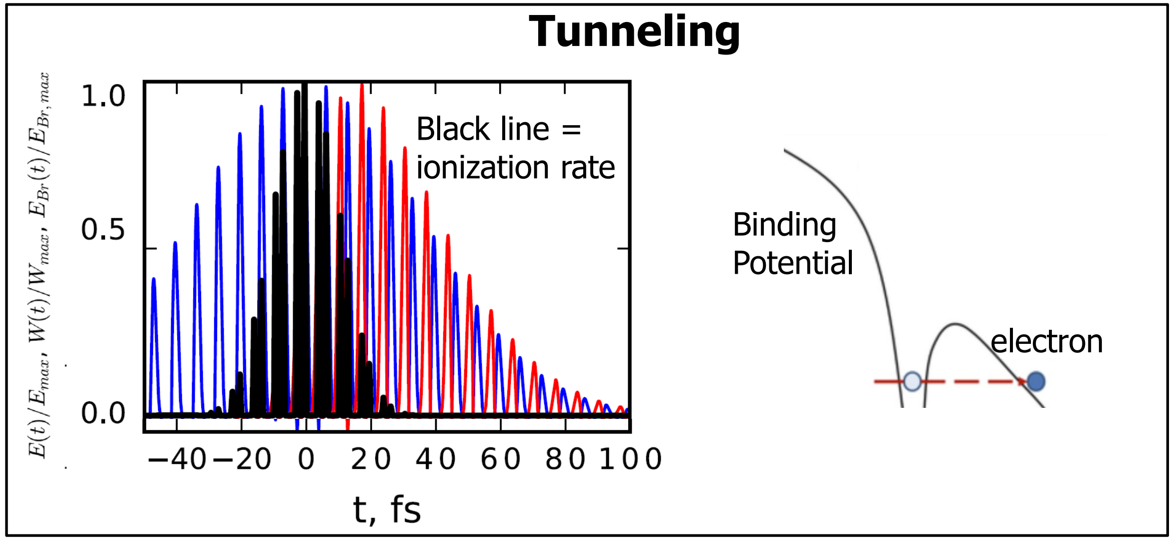


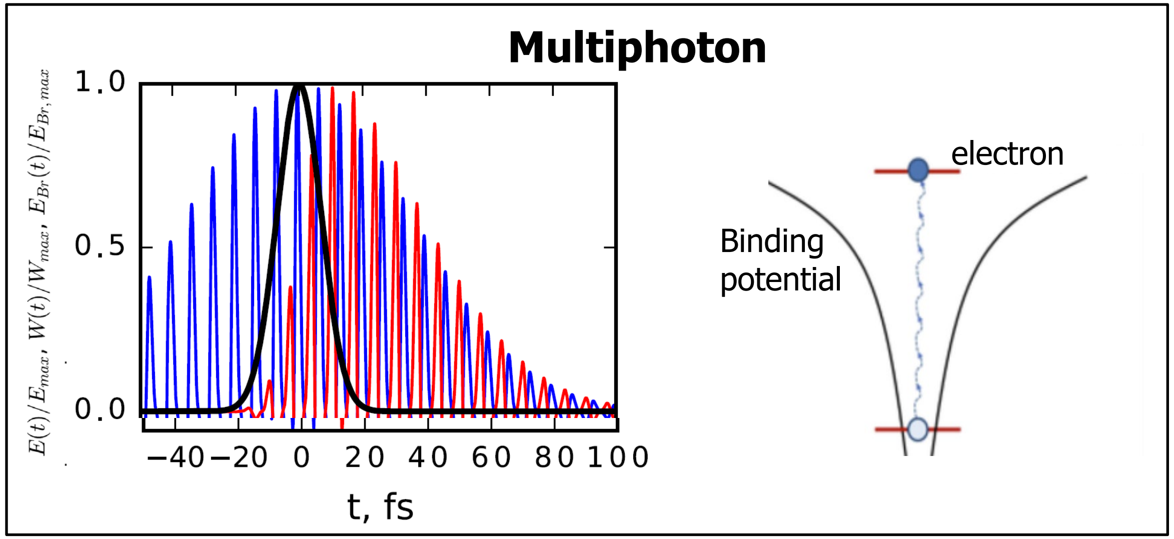
Envelope overlap (multiple cycles)



Ionization Currents Determine Secondary Radiation

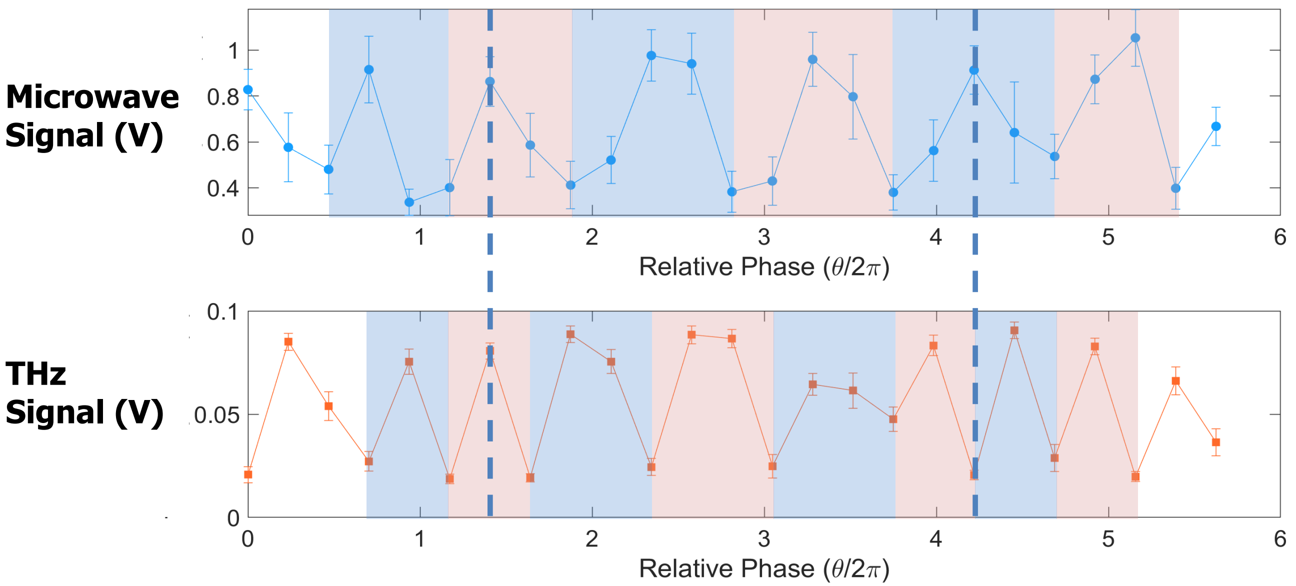
- Understand how ionization physics of air changes as a function of laser wavelength
- Use microwave, THz, and laser harmonics that radiate as a consequence of ionization currents (secondary radiation) to infer ionization physics
  - Amplitude and frequency spectrum
- Identify generation mechanism of microwave radiation with two-color pulses
- Link early and late plasma lifetime by combining secondary radiation measurements with plasma diagnostics
  - Interferometry and optical emission spectroscopy

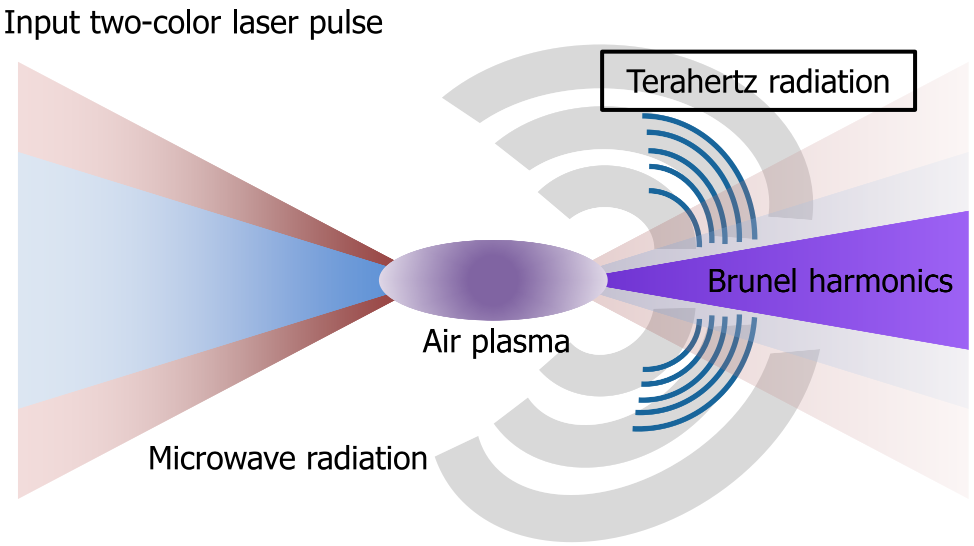

Tunneling


Multiphoton

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

Compare Relative Phase Dependence of Secondary Radiation





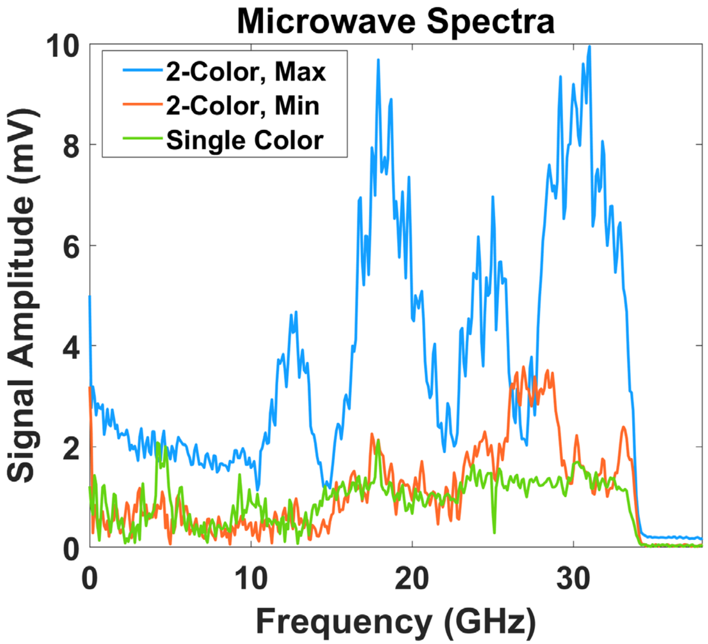
- Two-color scheme is common to generate THz in air, but doing it with LWIR fundamental would be unique (no publications yet as far as we know)

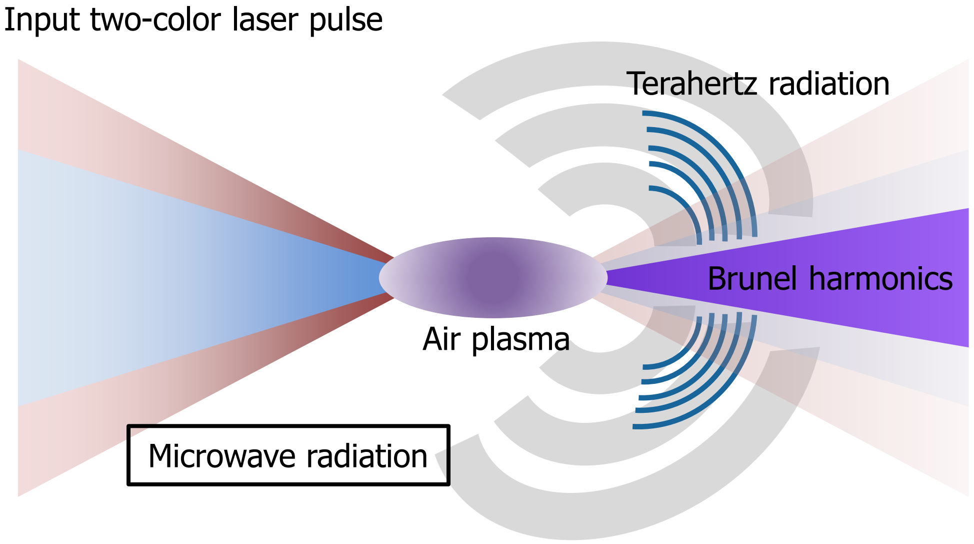
- Two-color NIR is in multiphoton regime, but Keldysh theory doesn't neatly apply.
  LWIR would be far into tunneling regime

- Two-color microwave generation is different from single-color, but mechanism is unclear.
  Going to tunneling regime may clarify physics.

Compare Relative Phase Dependence of Secondary Radiation

NRL PPD





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- Two-color NIR is in multiphoton regime, but Keldysh theory doesn't neatly apply here. LWIR would be far into tunneling regime
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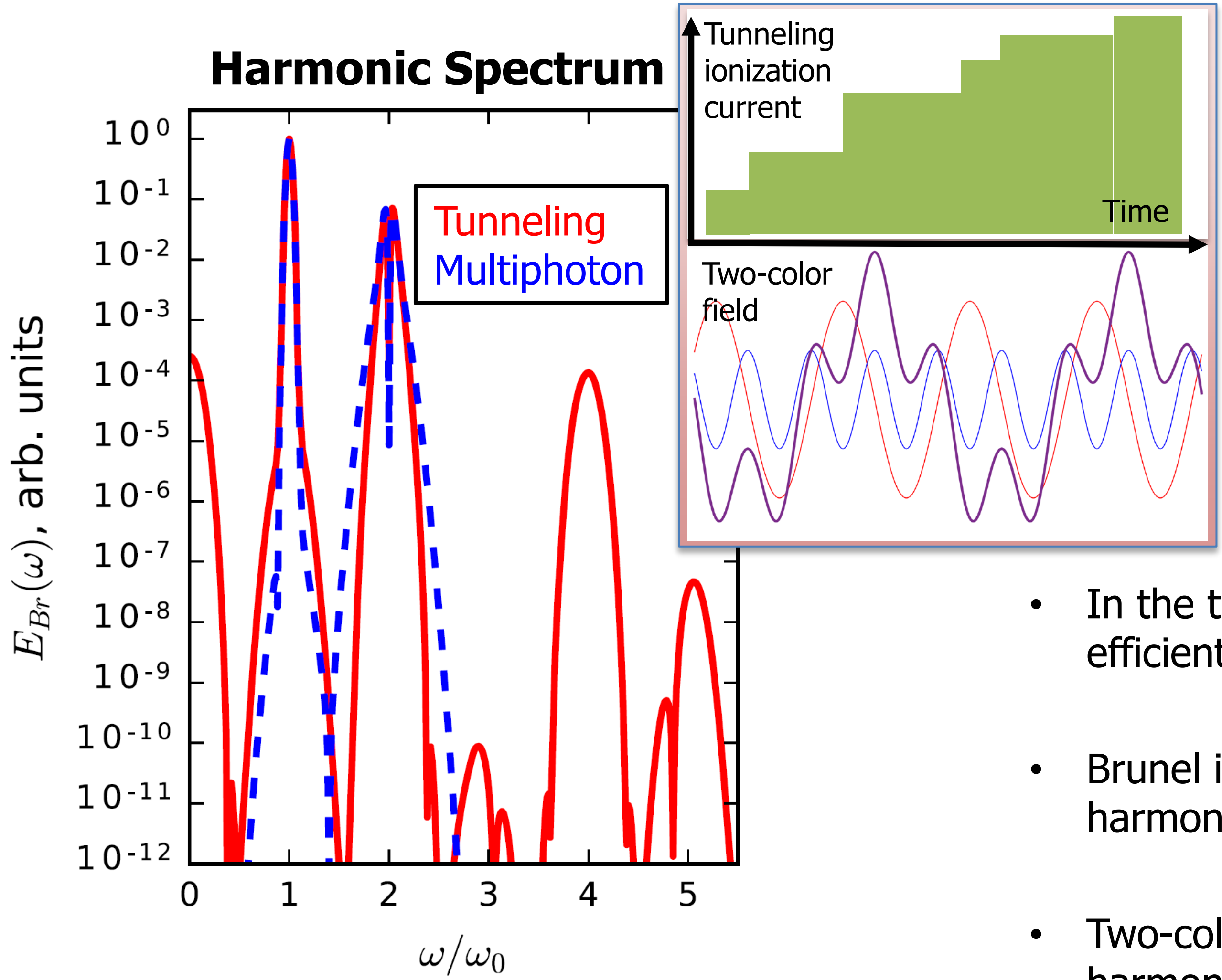
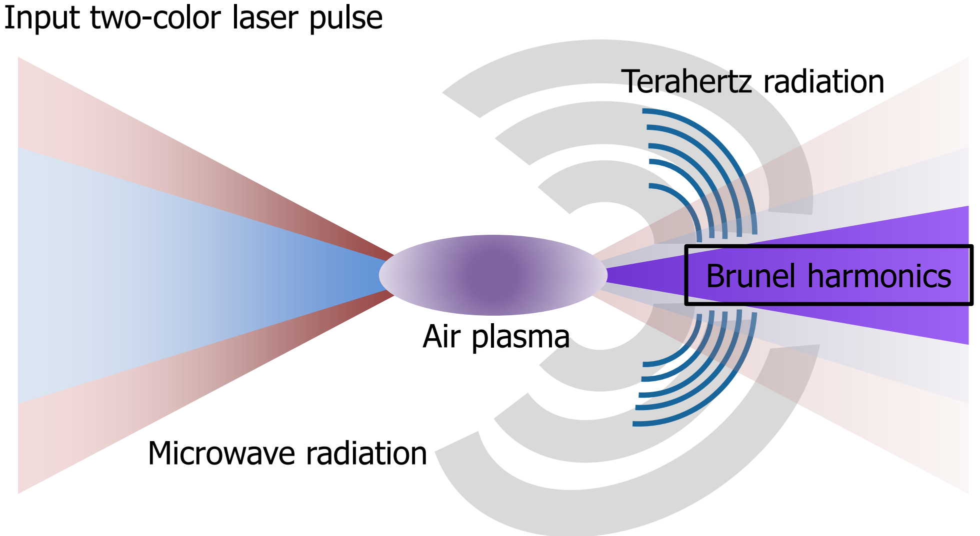
Long Wavelength Pulses Allow Additional Comparison with Brunel Harmonics



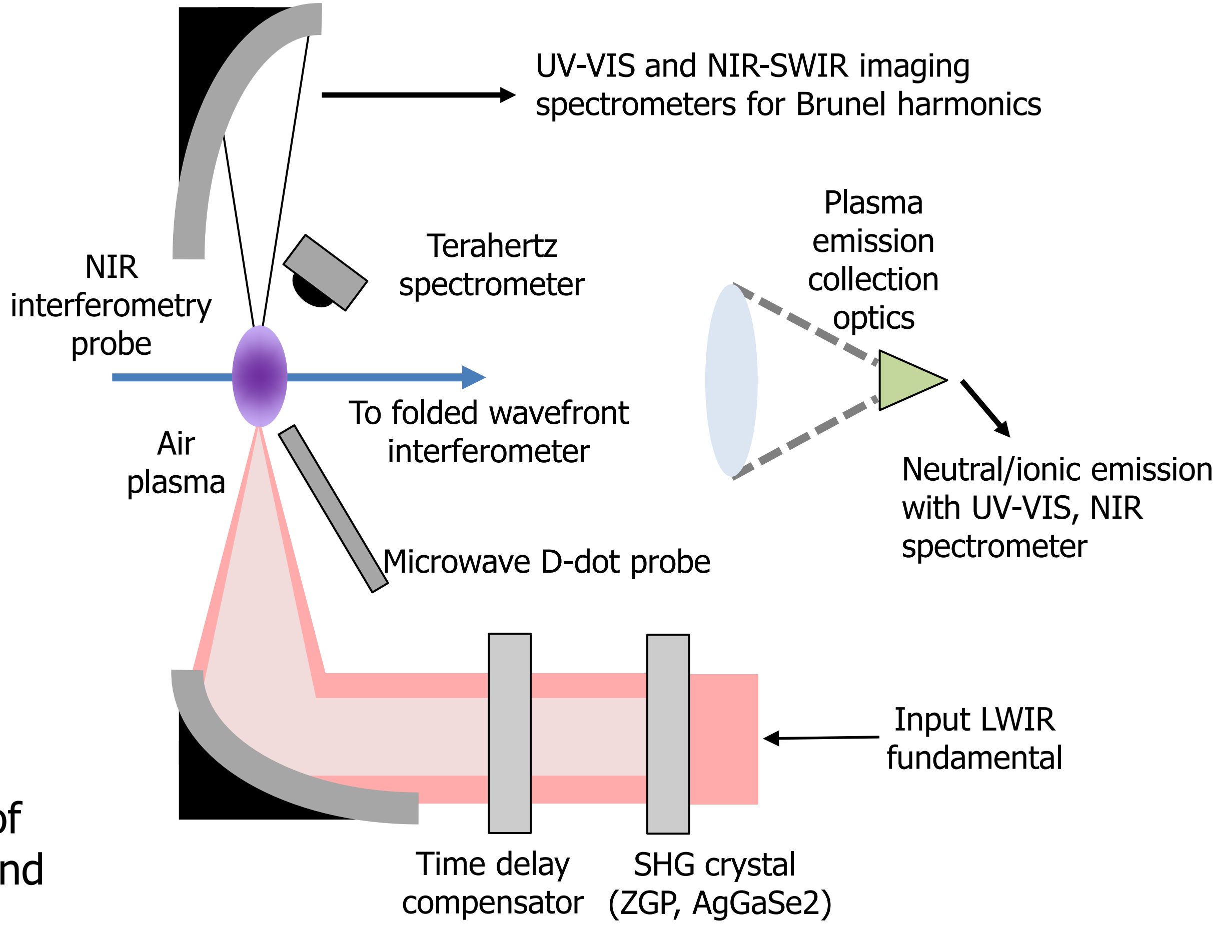
I. Babushkin, *J. Mod. Opt.* 64(10), 1078 (2017).



- In the tunneling regime, the ionization current efficiently generates harmonics via Brunel mechanism
- Brunel is not the same as recollision-driven high harmonic generation
- Two-color NIR pulses do not generate Brunel harmonics, would be strongly absorbed in air if they did

Experimental Goals and Setup

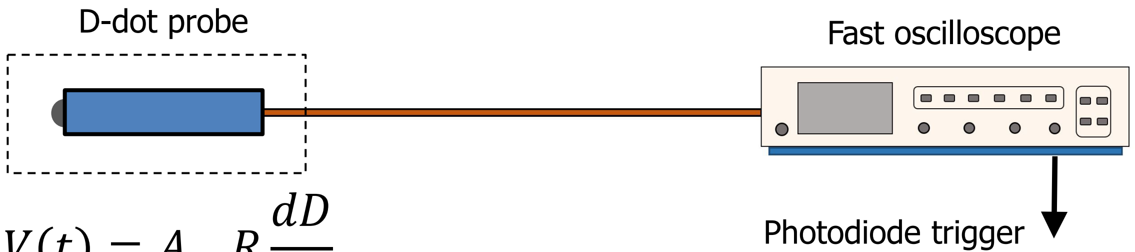
- Complete dataset = secondary radiation + plasma diagnostics as a function of two-color relative phase for single laser pulse format
- Deploy single-shot, triggerable measurements of:
    - Microwave radiation
    - THz radiation
    - Brunel harmonics
    - Plasma interferogram
    - Plasma optical emission spectrum
- Repeat experiment on NRL lasers: NIR (Ti:sapphire), SWIR + MWIR (OPA)
- Use relative phase dependence for quantitative points of comparison for simulations: SeaRay, CHMAIR/SPARC, and TurboWave



Microwave Pulse Detection

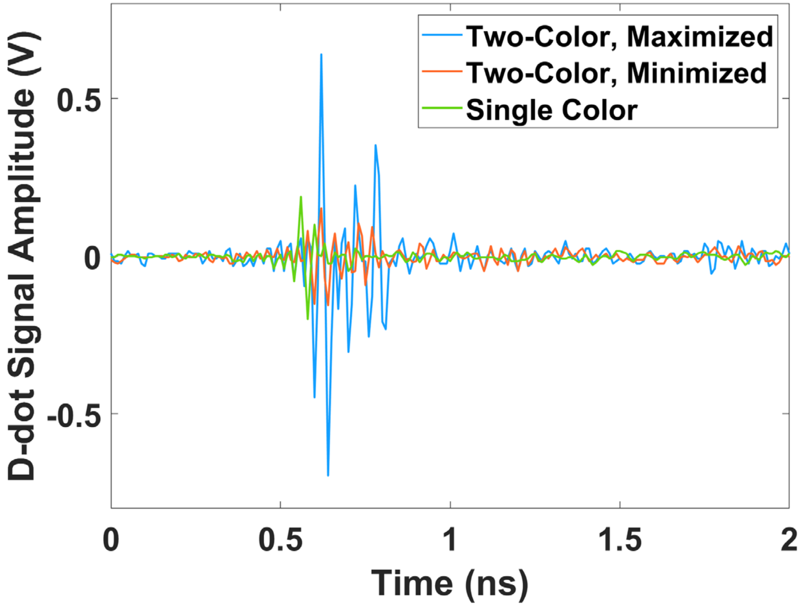
- Plasma currents radiate a broadband electromagnetic pulse that extends down to sub-GHz frequencies
- Detect microwaves with a high-frequency d-dot probe, sensitive to fields up to 50 GHz
- D-dot probe does not have a lower cutoff frequency unlike antenna
- Useful info contained in relative waveform amplitude, and the real part of the frequency spectrum

Microwave Receiver



$$V(t) = A_{eq} R \frac{dD}{dt}$$

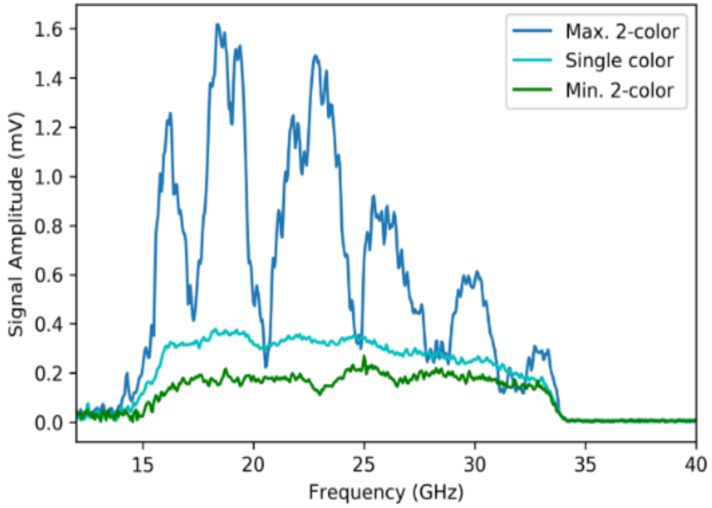
Microwave Waveforms



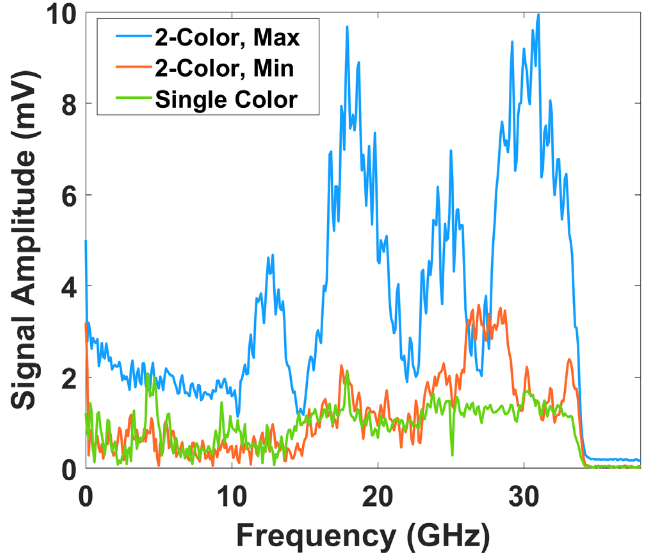
Modulations in Microwave Frequency Spectrum are Physical

- At the relative phase that maximizes microwave generation, we see very regular modulations of the microwave frequency spectrum
- Have also seen modulations with SWIR and NIR pulses
- Optimized amplitude is ~10X more peak power than single-color pulse
- Mechanism that explains single-color microwaves does not account for the spectral modulations
- Will modulations persist with tunneling ionization from LWIR driver?

SWIR, picosecond



NIR, femtosecond



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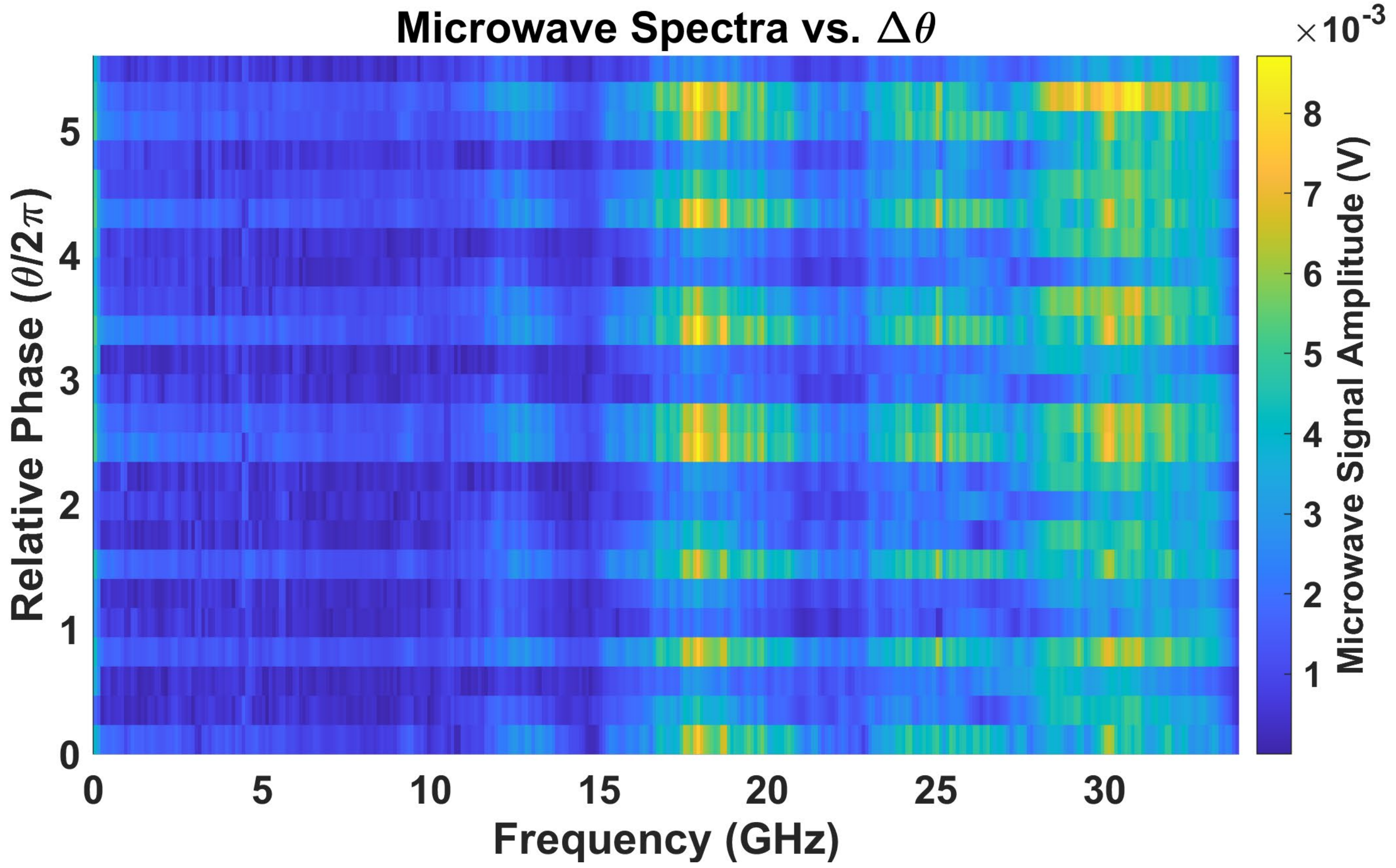
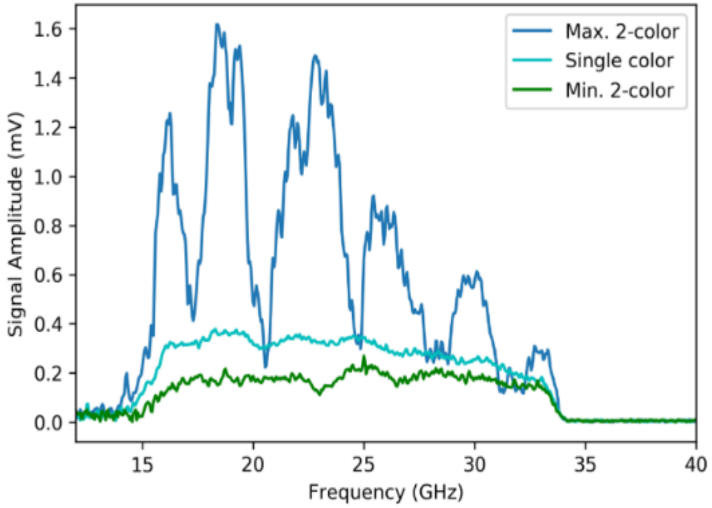
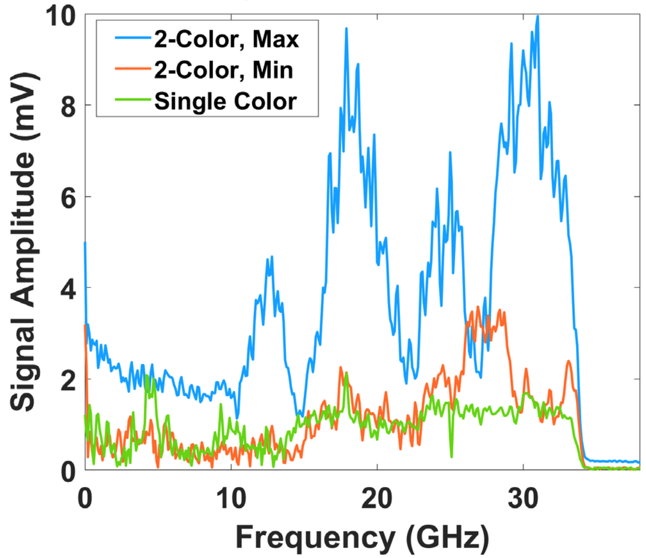


SWIR, picosecond



NIR, femtosecond



Implement Single-Shot THz Electro-Optic Sampling

Echelons generate NIR beamlets

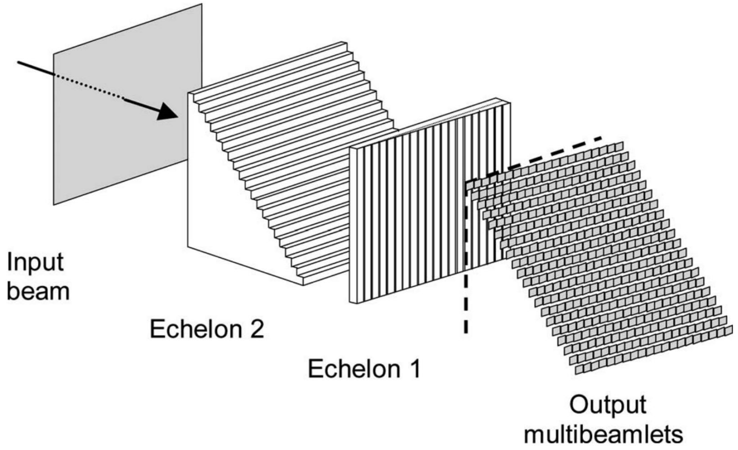
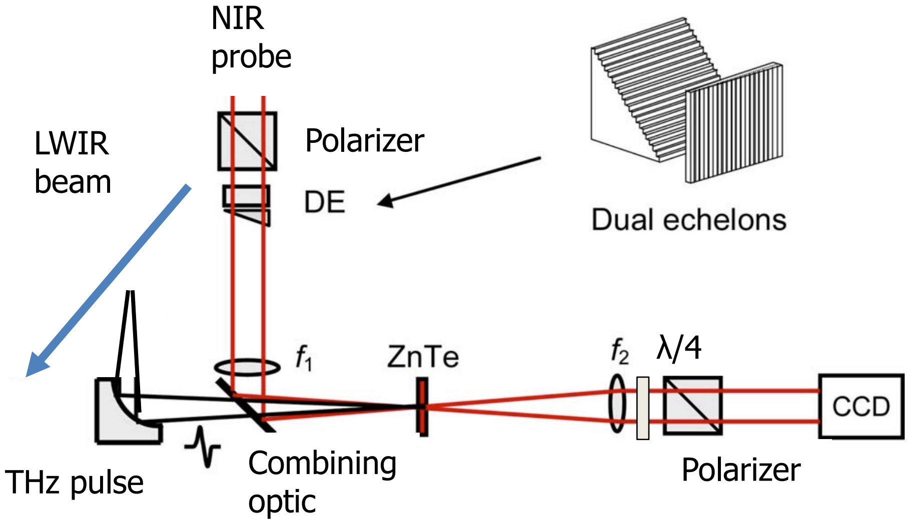


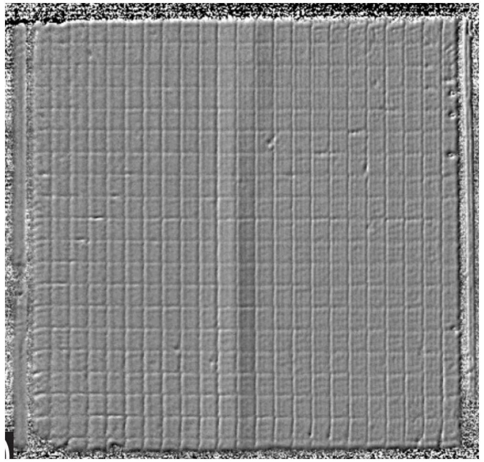
Fig. 1. Schematic of dual echelon optics capable of splitting an incoming single laser pulse into many incrementally delayed beamlets.

K.-Y. Kim, *Opt. Lett.* 32(14), 1968 (2007).

Image echelon through EO crystal on to camera



Read out THz waveform from intensity in boxes



Subtract CCD images with and without THz field

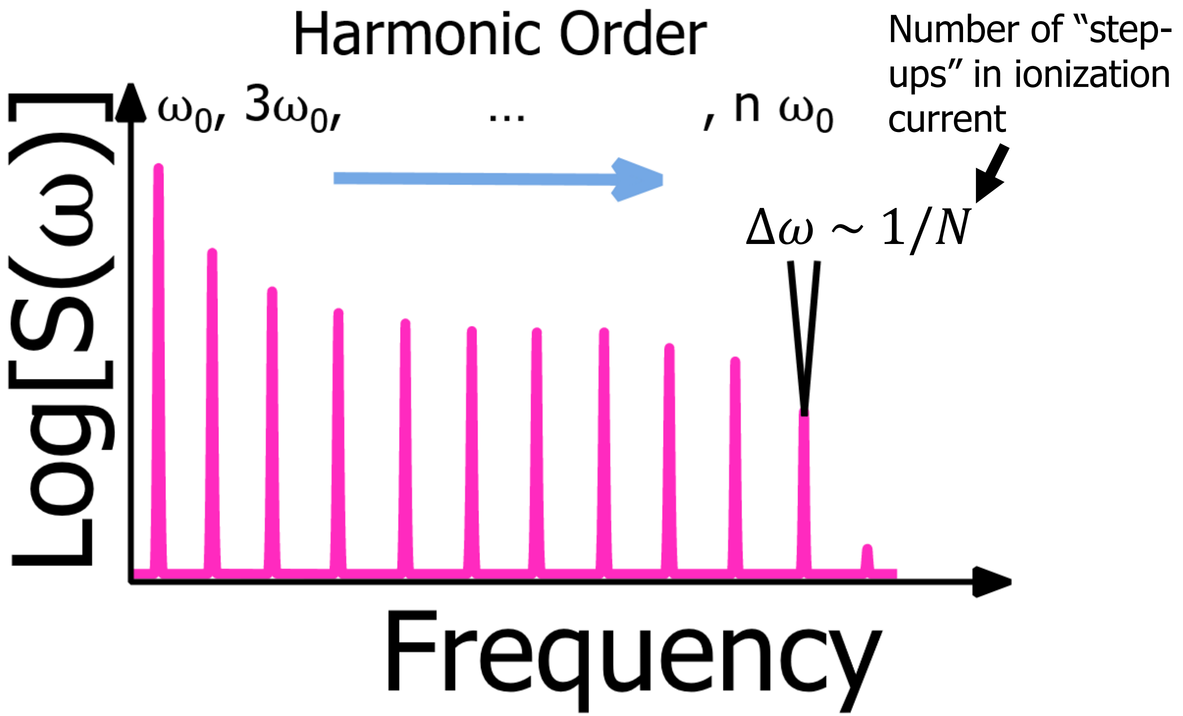
S. Teo, *Rev. Sci. Inst.* 86, 051301 (2015).

- Want to look for changes in THz spectrum due to relative phase
- Echelon-based electro-optic sampling obviates delay line scanning over many laser shots
- Sample ~2 ps THz field at 10’s fs resolution over ~10 ps time window
- Requires synchronized NIR laser pulses
- Build our own at NRL, and thoroughly test before ATF experiments

Spectrum of Brunel Harmonics Indicates Early Electron Dynamics

NRL PPD

- Need tunneling ionization to produce Brunel harmonics – cannot be done in air with NIR laser
- The relative amplitudes and linewidths of the Brunel harmonics contain valuable information about electron behavior within laser field
- Linewidths related to number of jumps in ionization current. Look for changes in linewidth with relative phase, and also look for trends with fundamental laser wavelength
- Primarily use NIR-SWIR spectrometer in ATF experiments, but will also look at higher order harmonics in visible range



Number of “step-ups” in ionization current

Harmonic Order	2	3	4	5	6	7	8	9	10	11	12	13	14
Wavelength (μm)	4.6	3.1	2.3	1.8	1.5	1.3	1.1	1.0	0.92	0.84	0.77	0.71	0.66

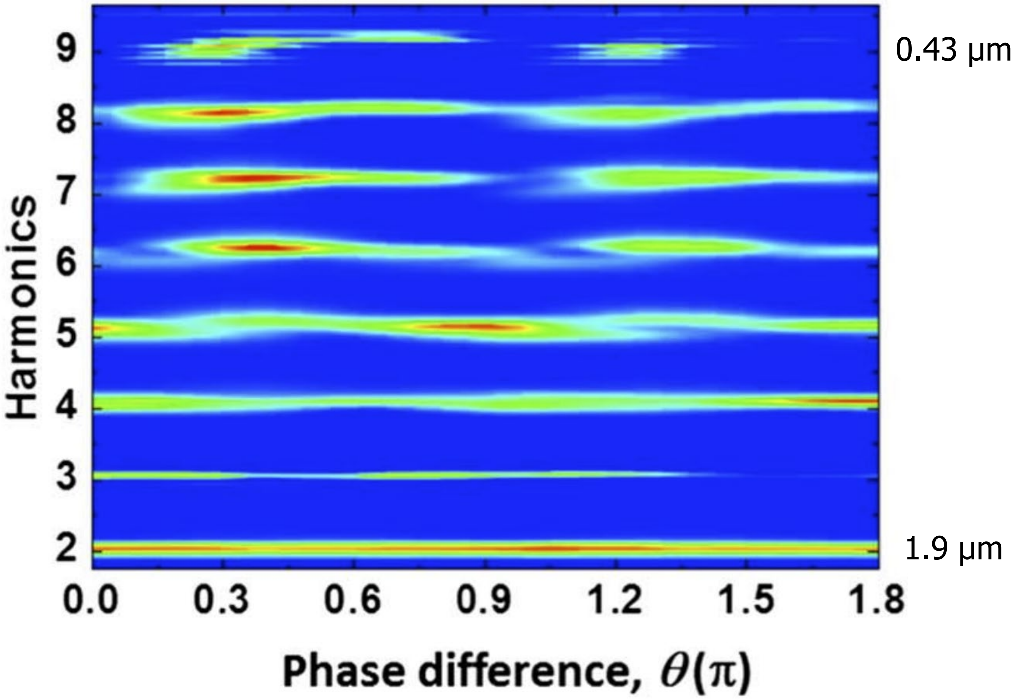
Spectrum of Brunel Harmonics Indicates Early Electron Dynamics

- Relative phase relationship between THz and Brunel harmonics can be simulated using carrier-resolving solver – SeaRay

- Difficult to discriminate low-order bound electron harmonics and Brunel harmonics

- Focus of year 2 experiments will be comparing Brunel and recollision-driven harmonic generation in air

1.9 + 3.9 μm pulse Brunel harmonics vs. relative phase, Experiment

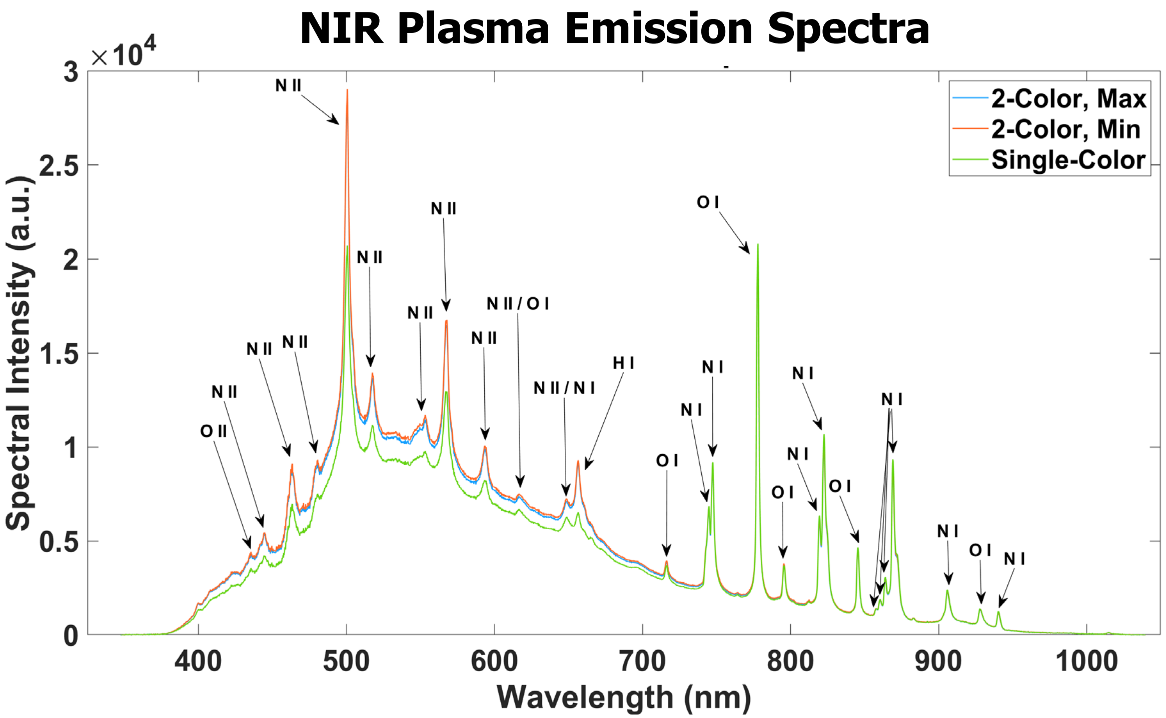


D. Jang, *Optica* 6(10), 1338 (2019).

Plasma Diagnostics: Interferometry and Optical Emission Spectroscopy

- Optical emission spectroscopy over large spectral range (e.g. 350-1100 nm) can give rough idea of electron kinetics
  - Compare emission lines to two-color pulses at other fundamental wavelengths
  - Population and decay of many excited states takes time period much longer than laser pulse, ionization currents
  - Benchmark air chemistry using CHMAIR/SPARC

- Interferometry to measure nominal plasma density and size:
  - Input an accurate electron density into simulations
  - If two-color field perturbs ionization rate in space and time, this may appear in interferogram fringes
  - Transverse probing



Summary

- Use the secondary radiation (microwaves, THz, and Brunel harmonics) to understand the role of ionization currents, and other potential collisional and collective charged particle dynamics excited by the two-color laser field.

- Link early and late plasma characteristics with concurrent measurements of the plasma size, density, and optical emission spectrum.

- Field single-shot diagnostics. Cameras for spectroscopy, interferometry, and oscilloscope for microwave receiver are triggerable. Need to build up echelon-based THz receiver.

- Integrate LWIR results in comparison study with NIR, SWIR, MWIR wavelengths

CO_2 Laser Requirements

NRL PPD

Configuration	Parameter	Units	Typical Values	Comments	Requested Values
CO_2 Regenerative Amplifier Beam	Wavelength	μm	9.2	*Wavelength determined by mixed isotope gain media*	
	Peak Power	GW	~3		
	Pulse Mode	---	Single		
	Pulse Length	ps	2		
	Pulse Energy	mJ	6		
	M^2	---	~1.5		
	Repetition Rate	Hz	1.5	*3 Hz also available if needed*	
	Polarization	---	Linear	*Circular polarization available at slightly reduced power*	
CO_2 CPA Beam	Wavelength	μm	9.2	*Wavelength determined by mixed isotope gain media*	*9.2 microns*
Note that delivery of full power pulses to the Experimental Hall is presently limited to Beamline #1 only.	Peak Power	TW	5	*~5 TW operation will become available shortly into this year's experimental run period. A 3-year development effort to achieve >10 TW and deliver to users is in progress.*	*< 100 GW*
	Pulse Mode	---	Single		*Single*
	Pulse Length	ps	2		*2 ps*
	Pulse Energy	J	~5	*Maximum pulse energies of >10 J will become available within the next year*	*~ 100-150 mJ*
	M^2	---	~2		
	Repetition Rate	Hz	0.05		*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	>1067		*<< 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		

Electron Beam Requirements – Not Applicable

NRL PPD

Parameter	Units	Typical Values	Comments	Requested Values
Beam Energy	MeV	50-65	*Full range is ~15-75 MeV with highest beam quality at nominal values*	
Bunch Charge	nC	0.1-2.0	*Bunch length & emittance vary with charge*	
Compression	fs	Down to 100 fs (up to 1 kA peak current)	*A magnetic bunch compressor available to compress bunch down to ~100 fs. Beam quality is variable depending on charge and amount of compression required.* *NOTE: Further compression options are being developed to provide bunch lengths down to the ~10 fs level*	
Transverse size at IP (σ)	μm	30 – 100 (dependent on IP position)	*It is possible to achieve transverse sizes below 10 um with special permanent magnet optics.*	
Normalized Emittance	μm	1 (at 0.3 nC)	*Variable with bunch charge*	
Rep. Rate (Hz)	Hz	1.5	*3 Hz also available if needed*	
Trains mode	---	Single bunch	*Multi-bunch mode available. Trains of 24 or 48 ns spaced bunches.*	

Special Equipment Requirements and Hazards

- Introduction of transmissive optics into LWIR beam:
  - Second harmonic crystal
  - Time delay compensator
  - Half waveplate to co-polarize harmonics (needed if using Type 1 SHG)

Experimental Time Request

CY2023 Time Request

Capability	**Setup Hours**	**Running Hours**
Electron Beam Only	N/A	
Laser* Only (in Laser Areas)	40	80
Laser* + Electron Beam		

Total Time Request for the 3-year Experiment (including CY2023-25)

Capability	**Setup Hours**	**Running Hours**
Electron Beam Only		
Laser* Only (in Laser Areas)	80	160
Laser* + Electron Beam		

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