

Harmonic Generation in Reflection from Plasma Mirrors

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National Science Foundation
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Outline

- 1) Introduction to plasma optics and their applications
- 2) Relativistic harmonic generation from plasma mirrors: overview and scaling laws
- 3) Cascaded plasma mirrors for enhanced harmonic generation
- 4) Modeling harmonic generation from plasma mirrors with particle-in-cell simulation
- 5) Harmonic source with controlled polarization state
- 6) Summary of significant results



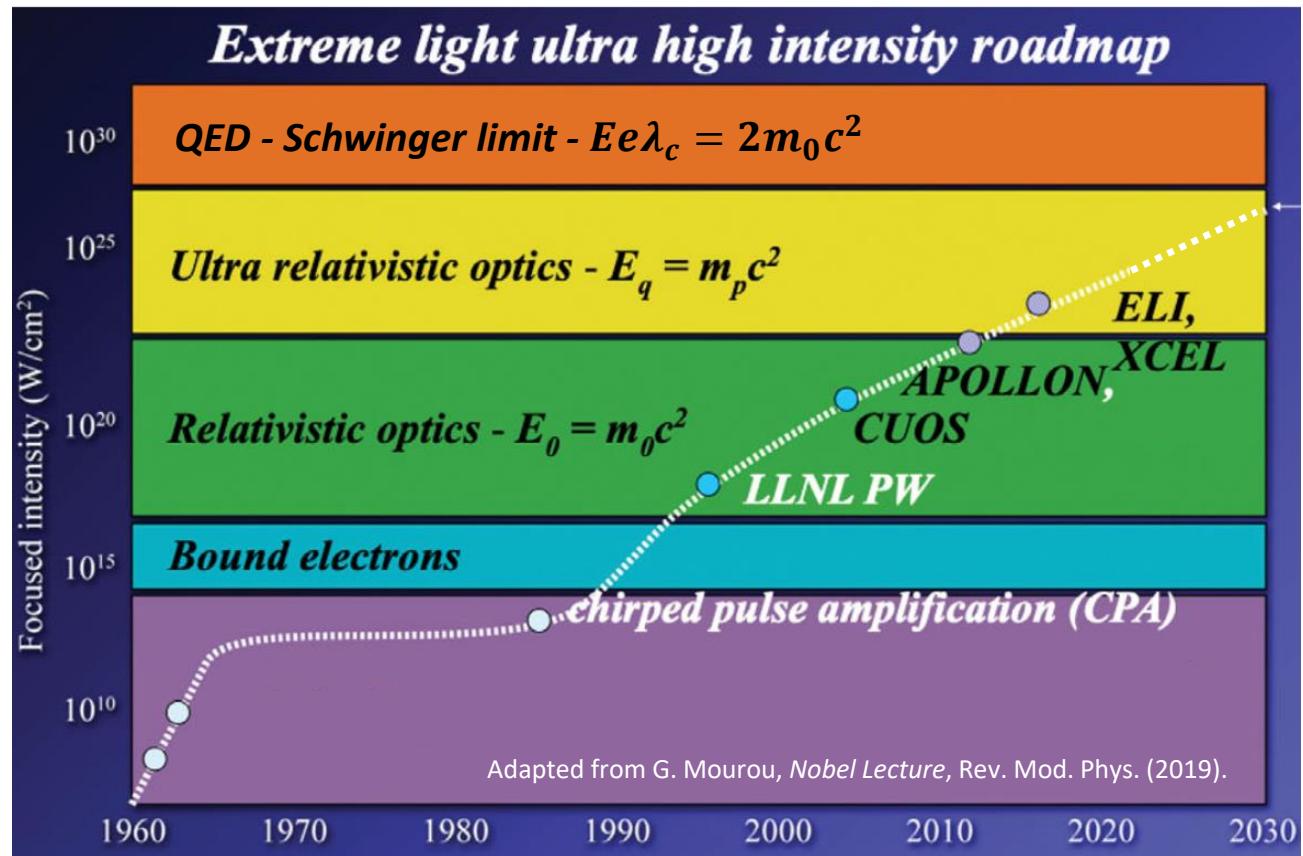
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A demand for plasma-based optics

- The discovery of chirped-pulse amplification in 1985 has led to the development lasers capable of reaching terawatt and petawatt peak powers, with focused intensities exceeding 10^{23} W/cm^2 [1, 2]
- Increasing the laser intensity unlocks new regimes of physics, allowing for novel light-matter investigations ranging from bound electrons at 10^{14} W/cm^2 to nonlinear QED effects at 10^{29} W/cm^2
- To manipulate light of ionizing intensities we need plasma-based optical components, such as plasma mirrors [3, 4], waveplates [5, 6], q-plates [7], beam combiners [8], amplifiers [9, 10], lenses [11], and gratings [12]
- Integrating these components into high-power beam lines is essential for the future of extreme light-matter interactions



[1] D. Strickland and G. Mourou, *Opt. Commun.* (1985).

[2] J. W. Yoon et al. *Opt. Express* (2019).

[3] M. M. Murnane, et al. *Phys. Rev. Lett.* (1989).

[4] M. R. Edwards and J. M. Mikhailova, *Sci. Rep.* (2020).

[5] P. Michel, et al. *Phys. Rev. Lett.* (2014).

[6] D. Turnbull et al. *Phys. Rev. Lett.* (2016).

[7] K. Qu, et al. *Phys. Rev. E* (2017).

[8] R. K. Kirkwood, et al. *Nat. Phys.* **14** (2017)

[9] V. M. Malkin, et al. *Phys. Rev. Lett.* (1999).

[10] J. Ren, et al. *Nat. Phys.* (2007).

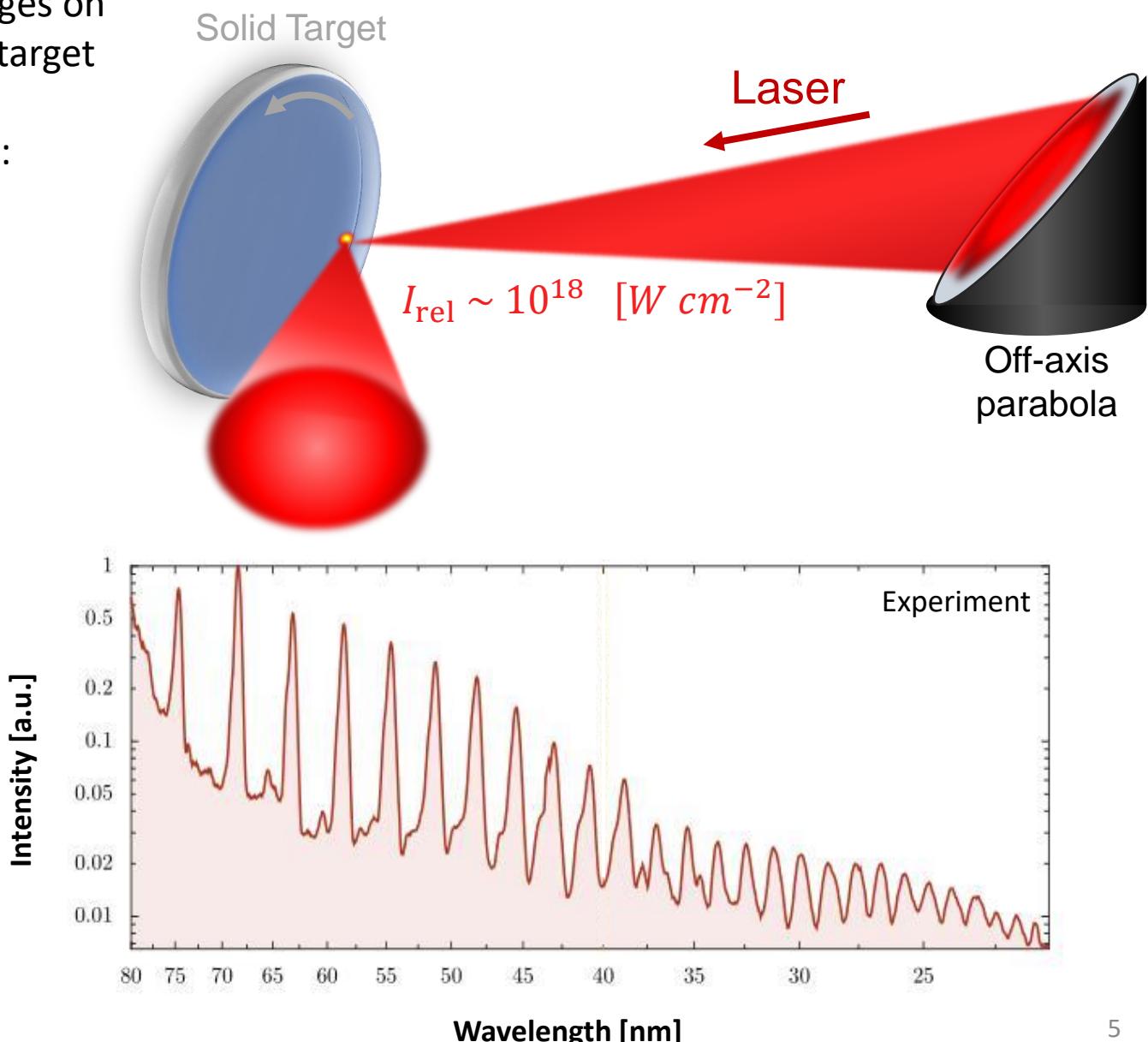
[11] M. R. Edwards, et al. *Phys. Rev. Lett.* (2022).

[12] M. R. Edwards et al. *Phys. Rev. Appl.* (2022).



Plasma mirrors

- Plasma mirrors are formed when intense light impinges on a solid target (e.g. glass), ionizing the surface of the target and yielding a high-density, opaque plasma
- They have several applications for high-power lasers:
 - Efficient reflection
 - [1] M. M. Murnane, et al. *Phys. Rev. Lett.* (1989).
 - [2] G. Doumy, et al. *Phys. Rev. E* (2004).
 - Non-linear temporal and spatial filtering
 - [1] H. C. Kapteyn, et al. *Opt. Lett.* (1991).
 - [2] R. Horlein, et al. *New J. Phys.* (2008).
 - [3] J. M. Mikhailova, et al. *Opt. Lett.* (2011).
 - Light intensification
 - [1] M. Nakatsutsumi, et al. *Opt. Lett.* (2010).
 - [2] H.-E. Tsai, et al. *Phys. Plasmas* (2017).
 - [3] R. Wilson, et al. *Phys. Plasmas* (2016).
 - Gratings and holograms
 - [1] S. Monchoce, et al. *Phys. Rev. Lett.* (2014).
 - [2] A. Leblanc, et al. *Nat. Phys.* (2017).
 - Harmonic generation
 - [1] P. Gibbon, *Phys. Rev. Lett.* (1996).
 - [2] B. Dromey, et al. *Nat. Phys.* (2006).
 - [3] J. M. Mikhailova, et al. *Phys. Rev. Lett.* (2012).
 - [4] M. R. Edwards and J. M. Mikhailova, *Sci. Rep.* (2020).





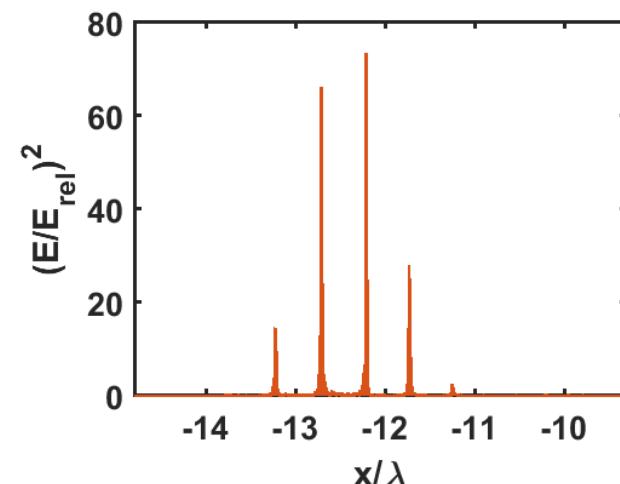
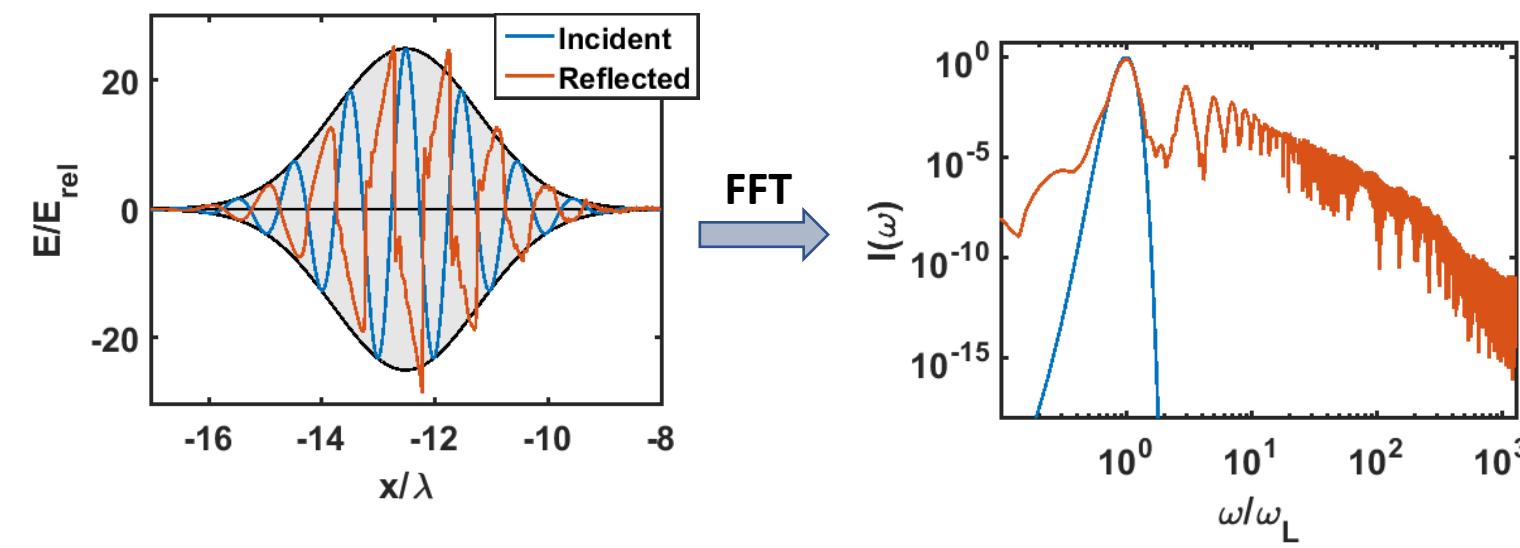
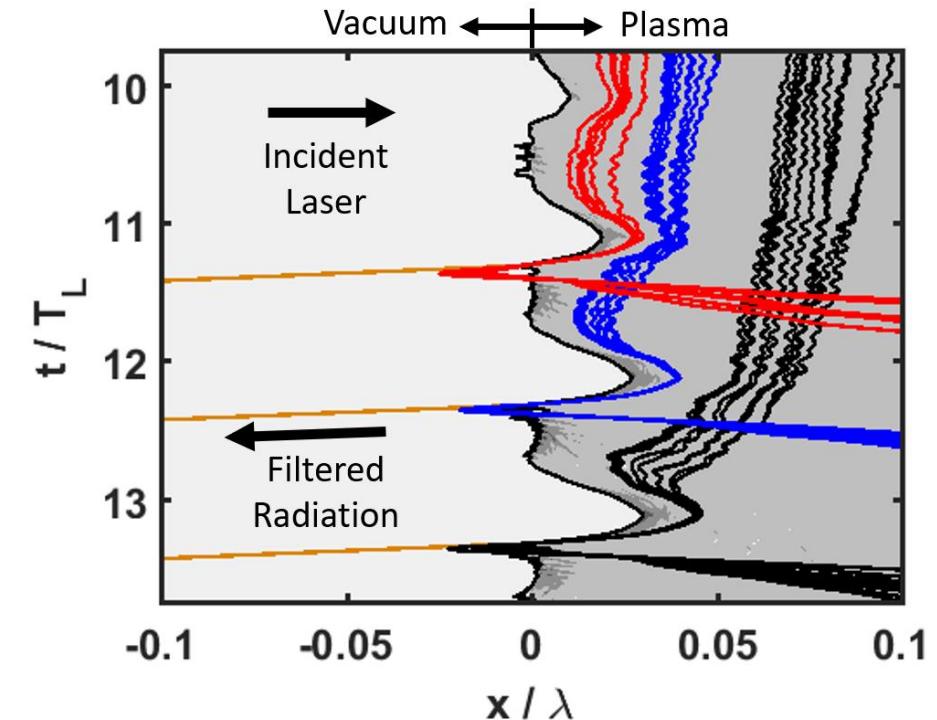
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Harmonic generation from plasma mirrors

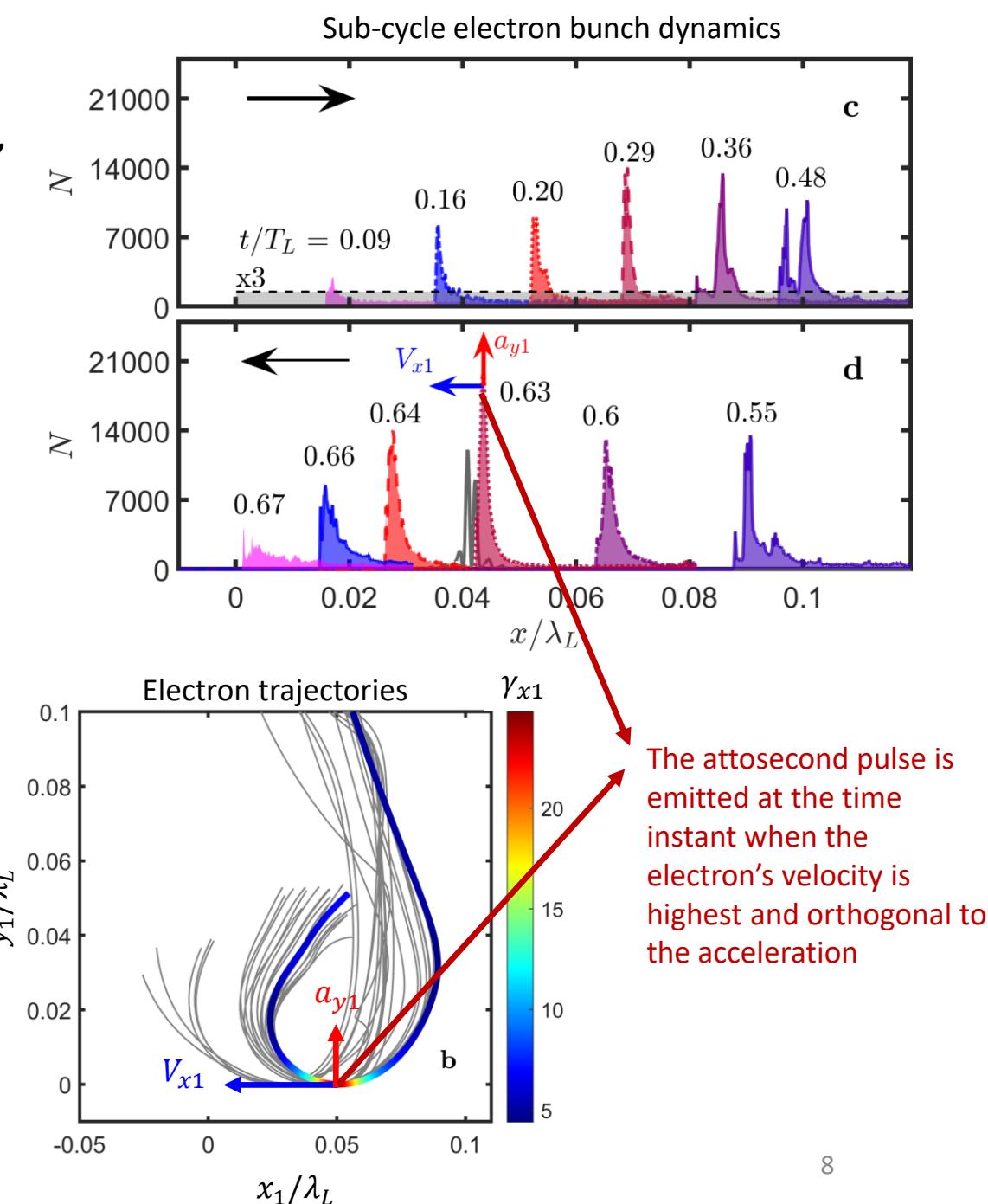
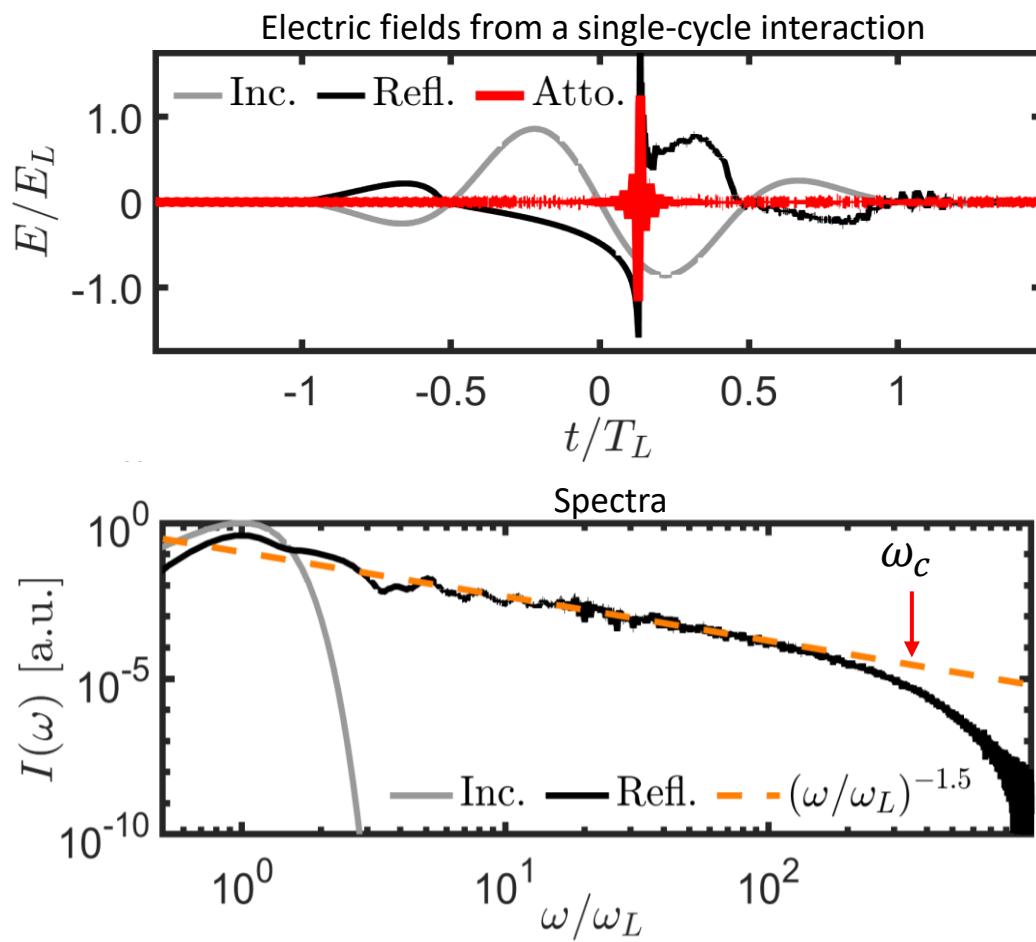
- Relativistic harmonic generation from plasma mirrors starts with a relativistically intense laser ($I_L > 10^{18} W/cm^2$) incident onto a fully ionized solid-density plasma target ($n_e > 10^{21} cm^{-3}$)
- The incident laser force and resulting plasma restoring force yield periodic oscillations of the electron number density
- Once per laser cycle, a dense bunch of electrons are formed and accelerated to highly relativistic speeds, eventually emitting an attosecond pulse





Sub-cycle electron bunch dynamics

- Harmonic generation from plasma mirrors is a sub-cycle process, where attosecond pulse emission occurs once per laser cycle.
- Harmonics arise from interference of periodically-spaced attosecond pulses in an attopulse train





Scaling of harmonic generation from plasma mirrors

- The efficiency of harmonic generation (η) is a function of the laser and plasma parameters and is optimized when the laser force ($\sim a_0$) and the plasma restoring force ($\sim N$) are close to balanced:

$$\eta = f(N, a_0, D, L, \tau_{fwhm}, \phi_{cep}, \theta, \omega_L)$$

Plasma density Laser intensity Target thickness Gradient scale length Pulse duration Carrier envelope phase Angle of incidence Laser frequency

- Simulations predict that the efficiency of harmonic generation scales with the dimensionless parameter a_0/N and is most efficient when the dimensionless parameter $a_0/N \approx 0.5$ [1], which requires laser intensities much greater than that available in state-of-the-art facilities
- Paths toward reaching optimal efficiency regime:
 - Increase laser intensity (i.e. build bigger lasers)
 - Decrease plasma density (e.g. foam targets or ultrathin foils)
 - Manipulate the frequency composition of the laser:

$$\eta \propto \left(\frac{a_0}{N}\right)^q = \left(\frac{q_e E_L}{m_e \omega_L c} \frac{m_e \omega_L^2}{4\pi q_e^2}\right)^q \propto (\omega_L)^q$$

- Relativistic harmonic generation is a frequency-dependent process that can be enhanced by adding higher-frequency components to the driving laser
- Our goal: Increase harmonic efficiency by combining the fundamental laser with its second harmonic to produce a two-color waveform

Definitions

Normalized laser amplitude

$$a_0 = \frac{q_e E_L}{m_e \omega_L c}$$

Normalized plasma density

$$N = \frac{n_e}{n_c} \quad n_c = \frac{m_e \omega_L^2}{4\pi q_e^2}$$

For $\lambda_L = 800\text{nm}$

$$n_c = 1.75 \times 10^{21} \text{cm}^{-3}$$

$$I_L = 2.14 \times 10^{18} \text{W/cm}^2 (a_0 = 1)$$



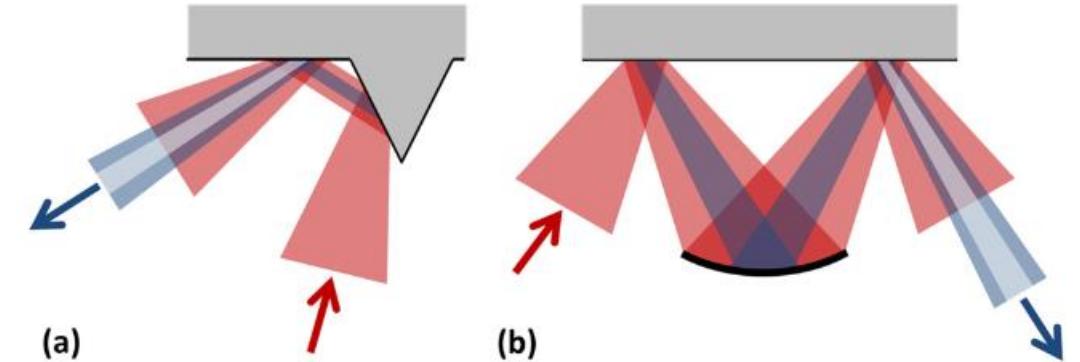
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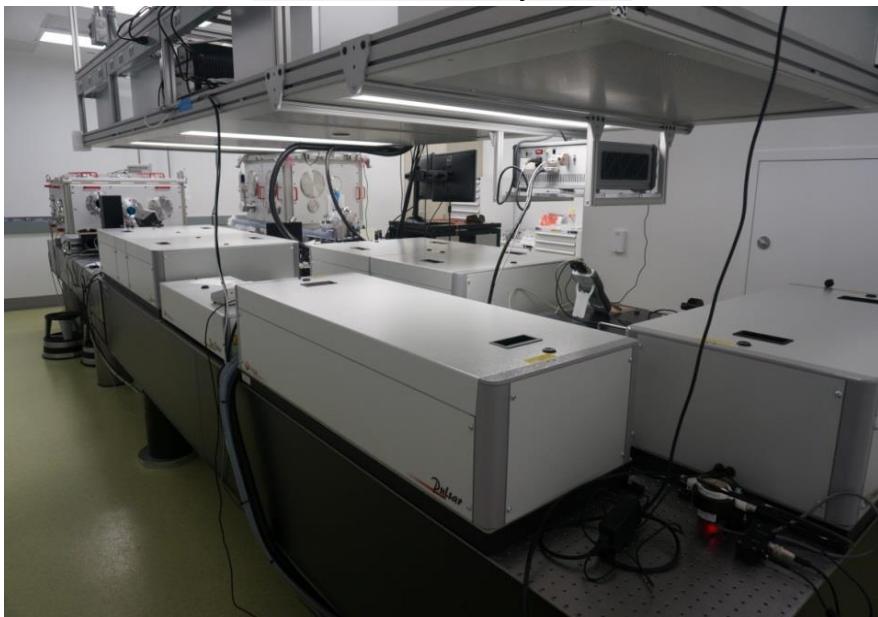
Cascaded plasma mirror configuration for enhanced harmonic generation

- Main idea: Use harmonics radiated from one plasma to synthesize a multi-color waveform which can then be used to efficiently generate harmonics from a second plasma mirror
- Experimental set-up uses three plasma mirrors (PM): one contrast cleaning PM and two relativistically-driven PMs that generate harmonics



M. R. Edwards and J. M. Mikhailova, *Phys. Rev. A* (2016).

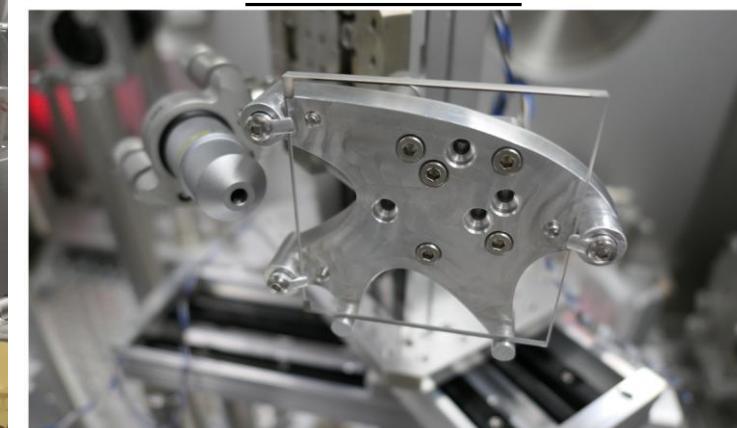
20TW Laser System



Experimental Chamber



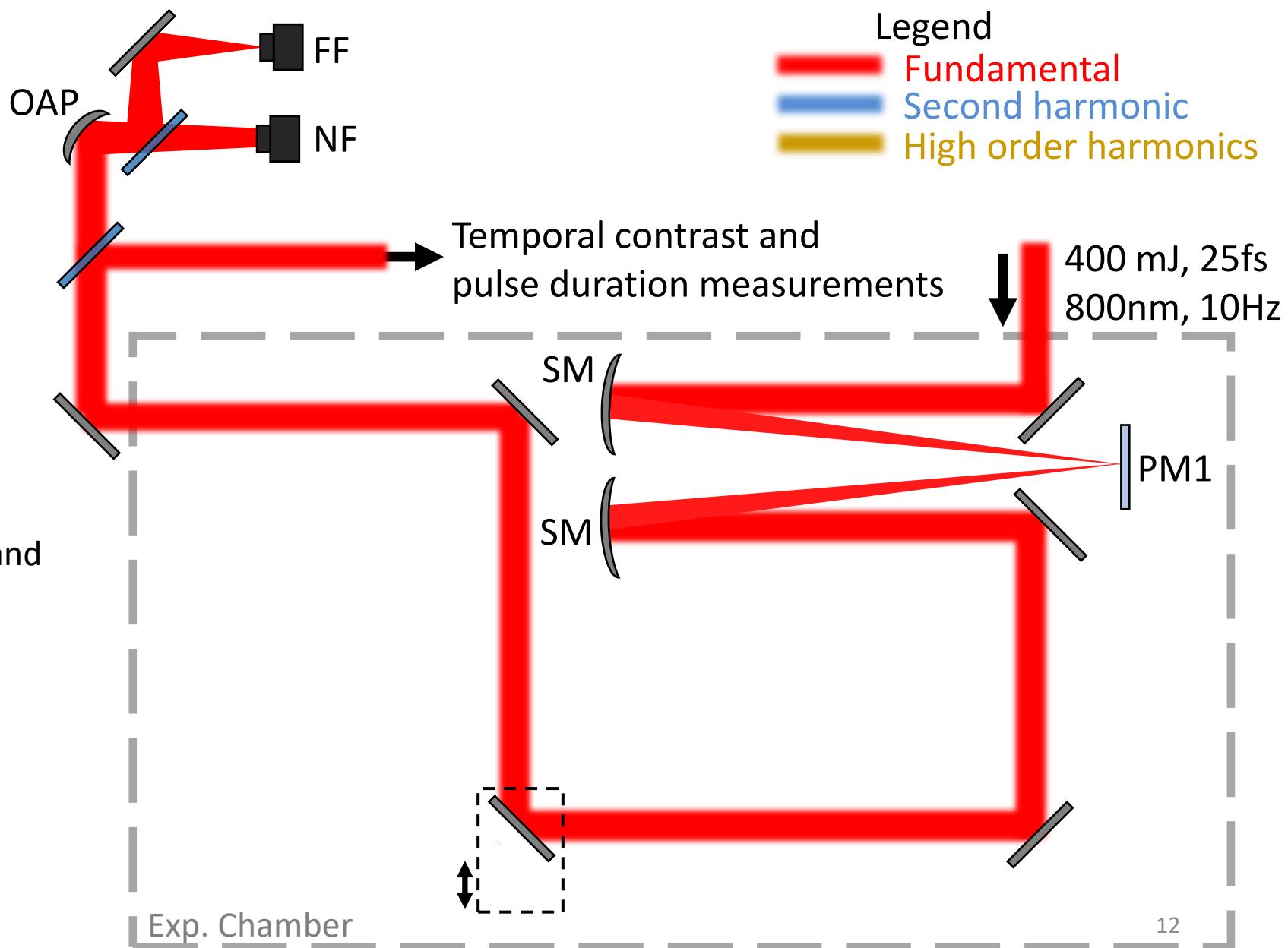
Plasma Mirror





Experimental set-up – PM1 only

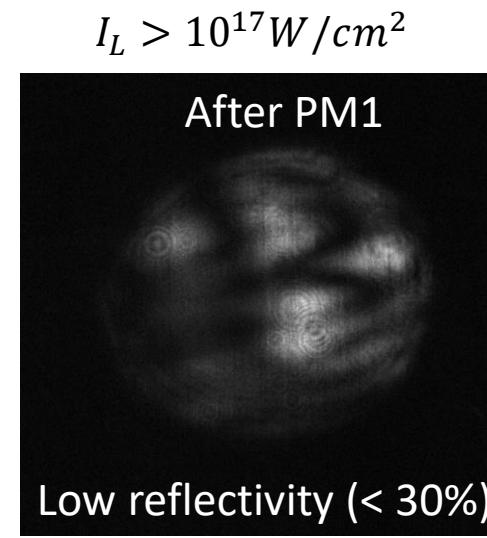
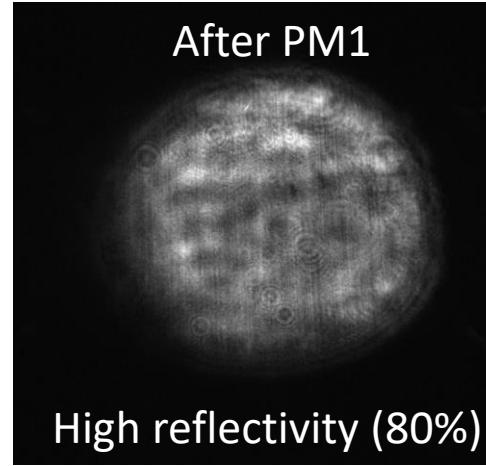
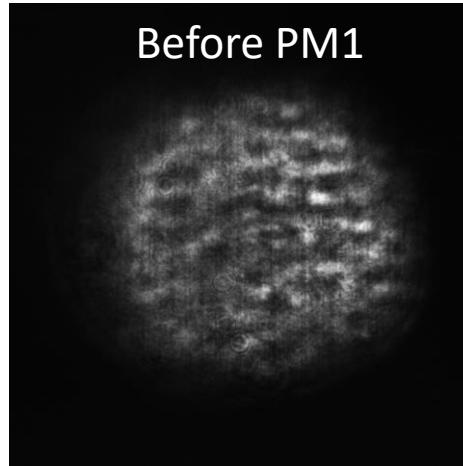
- The first plasma mirror (PM1) is used as a temporal contrast cleaner
- Pick-off optics inside the chamber allow for characterization of reflected beam's spatial profile, focusability, temporal contrast, and pulse duration
- Summary of PM1: 80% reflectivity and improves temporal contrast by two orders of magnitude improvement



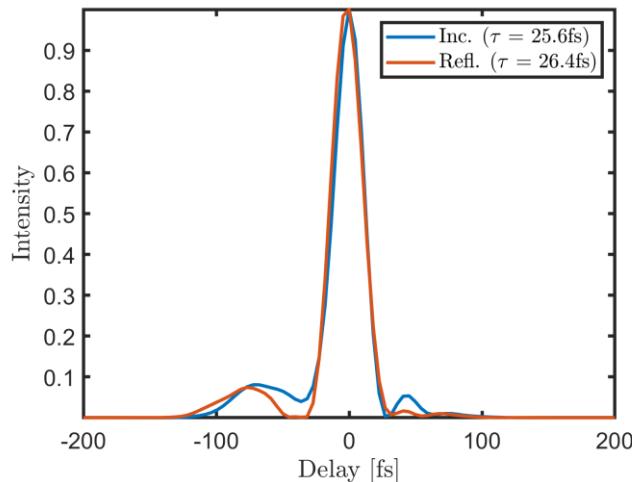


Key results from PM1: Contrast cleaning plasma mirror

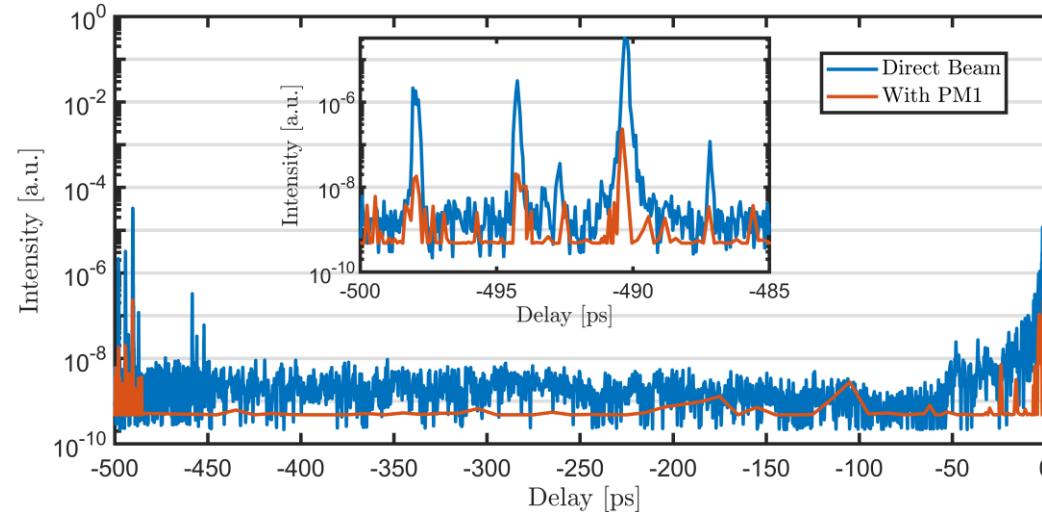
- Plasma mirrors operating at near-normal incidence have high reflectivity (up to 80%), depending on incident intensity



Temporal envelope is not affected



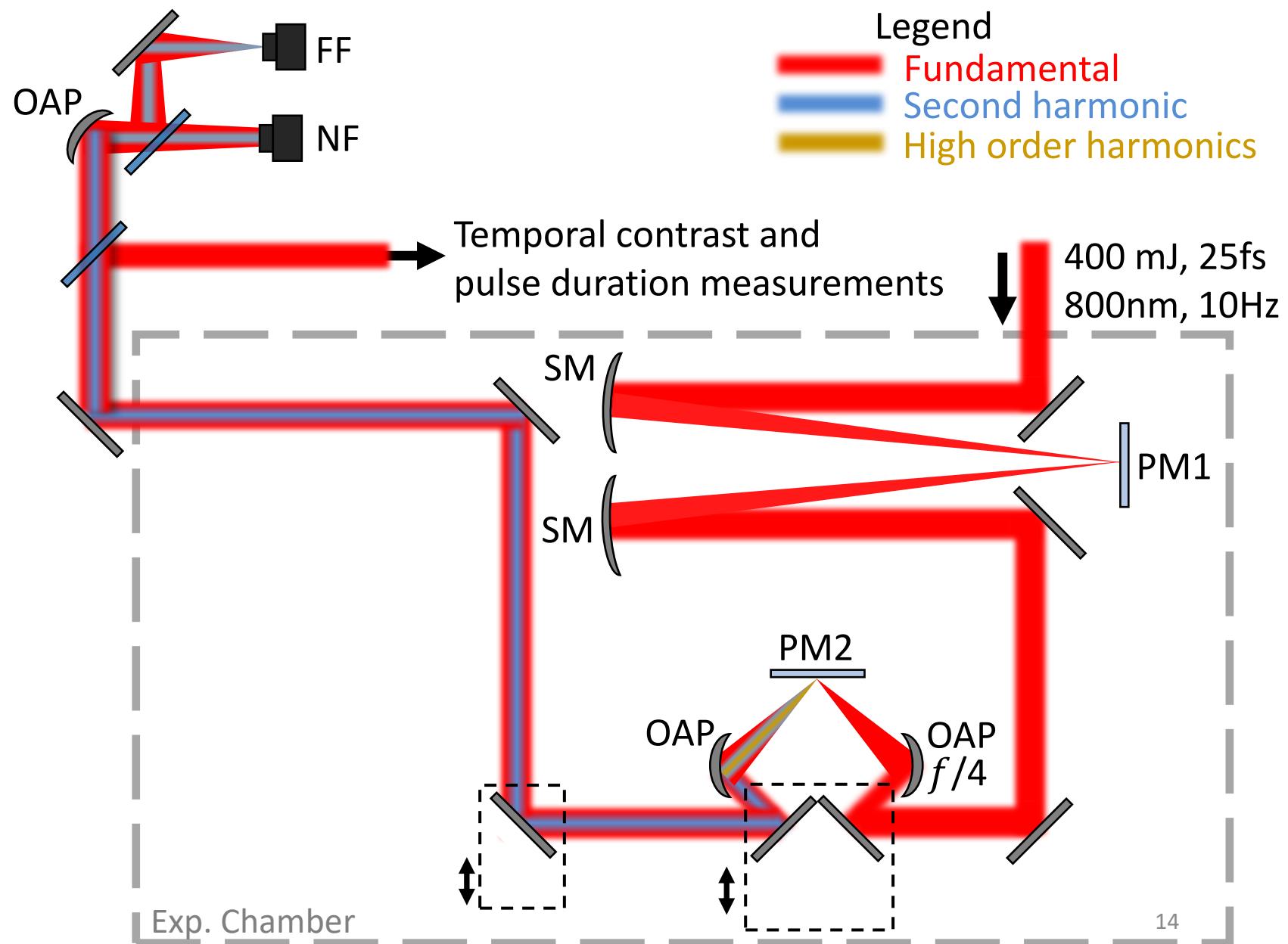
Temporal contrast enhanced by two orders of magnitude





Experimental set-up – PM1 and PM2

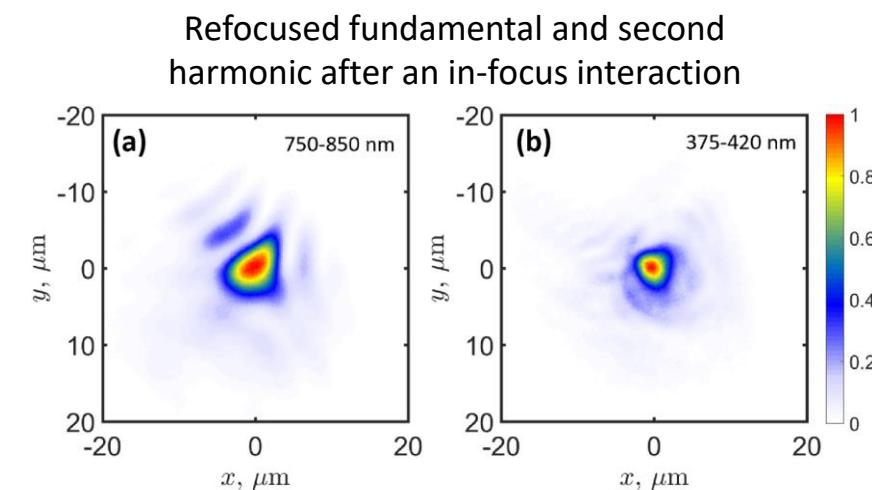
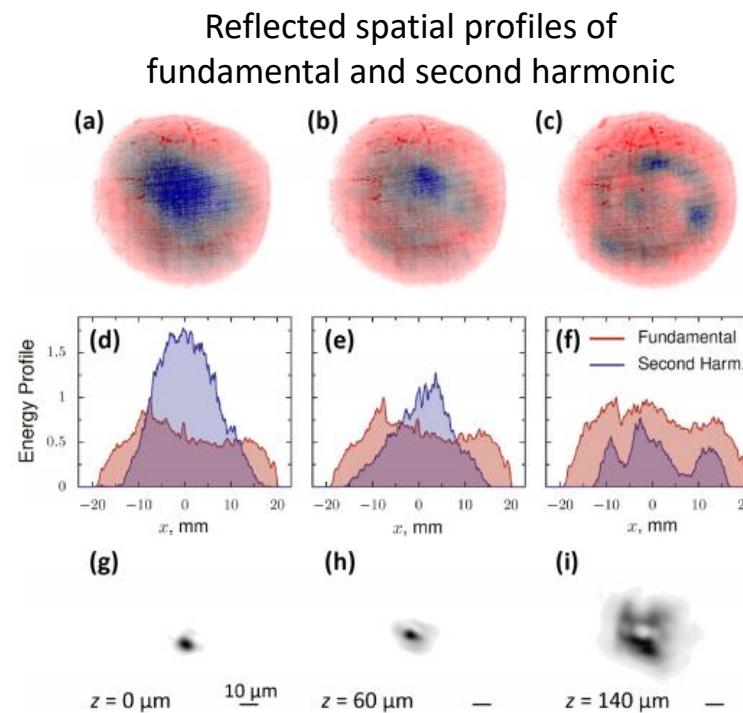
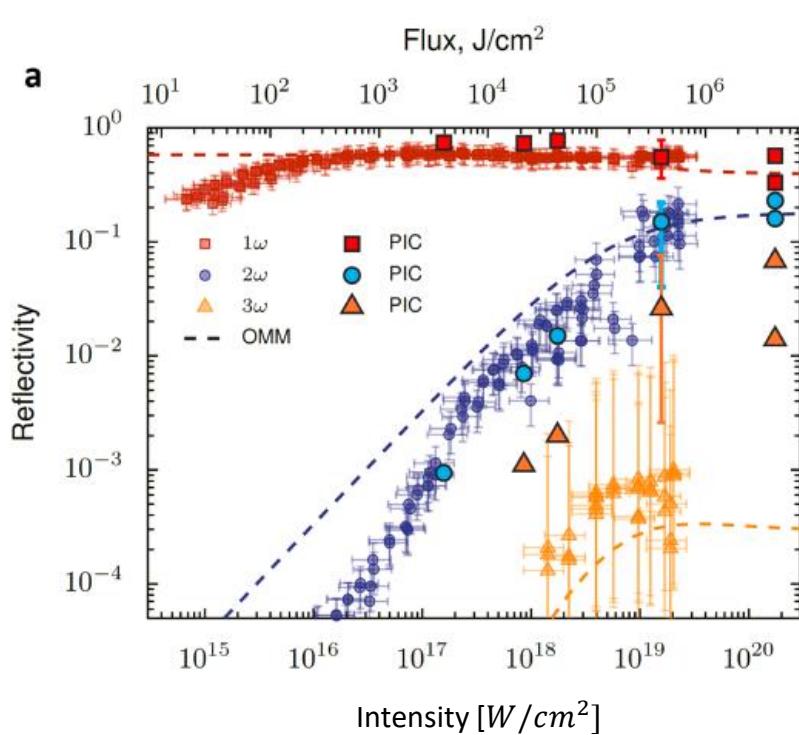
- The second plasma mirror (PM2) is used as a two-color waveform synthesizer
- In principle, we generate harmonic orders from the infrared to the xuv, but all orders greater than 2 are lost in the beam transport
- Pick-off optics inside the chamber allow for characterization of reflected beam's spatial profile, focusability, temporal contrast, and pulse duration





Key results from PM2 – Two-color waveform synthesizer

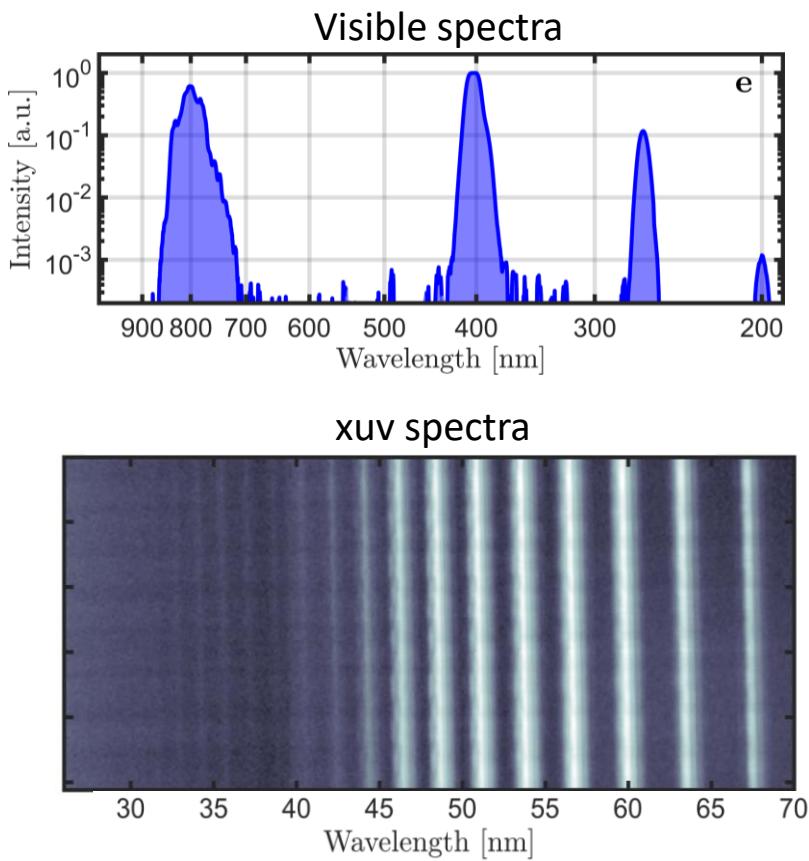
- Constructed a two-color terawatt beam, with up to 15% conversion efficiency into the second harmonic
- Both colors have good spatial quality and can be simultaneously focused to the same spot, provided that the plasma mirror is positioned within one Rayleigh range of the driving laser's focal spot



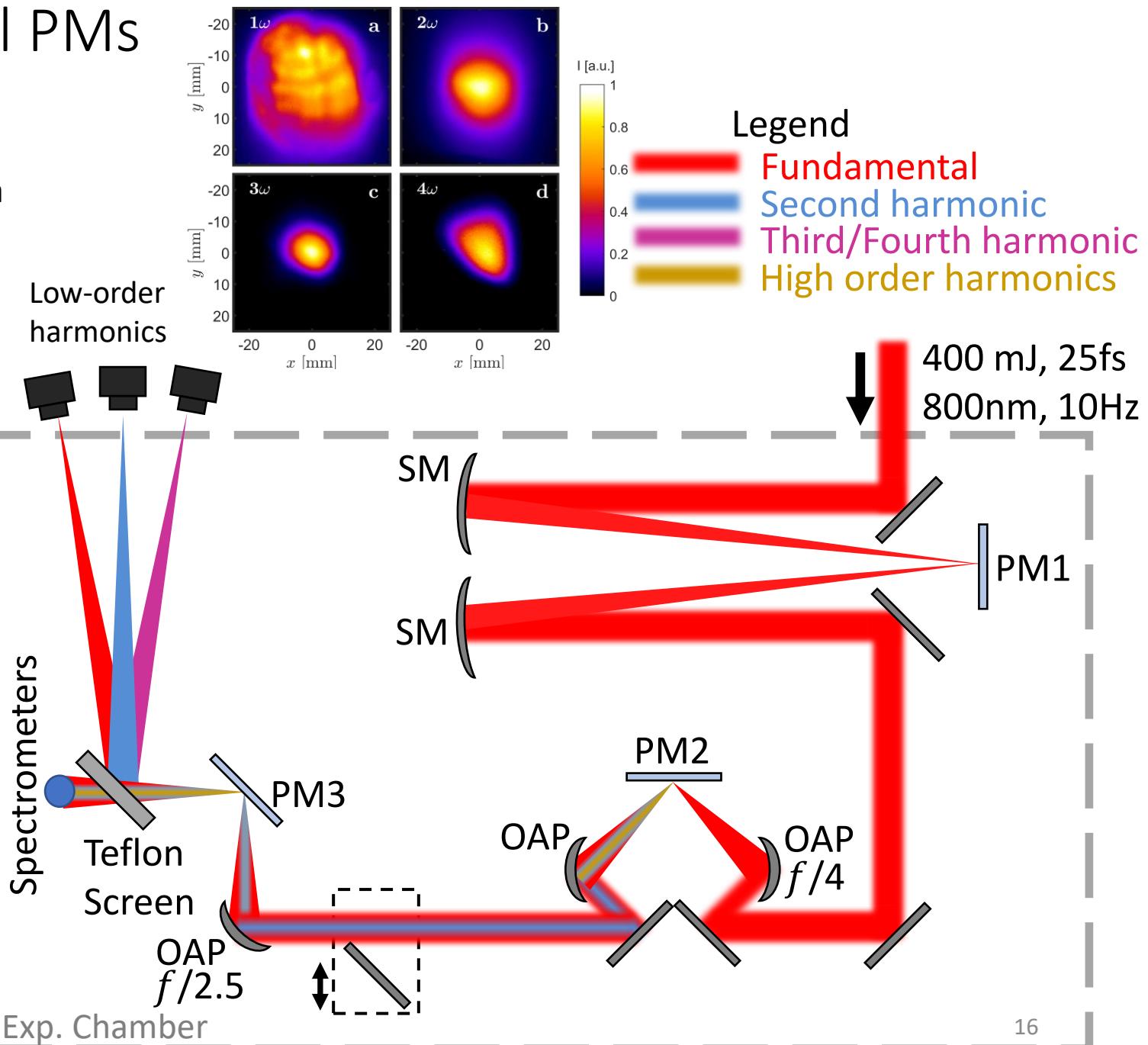


Experimental set-up – All PMs

- The third plasma mirror (PM3) is used to investigate harmonic generation from plasma mirrors driven by two-color waveforms



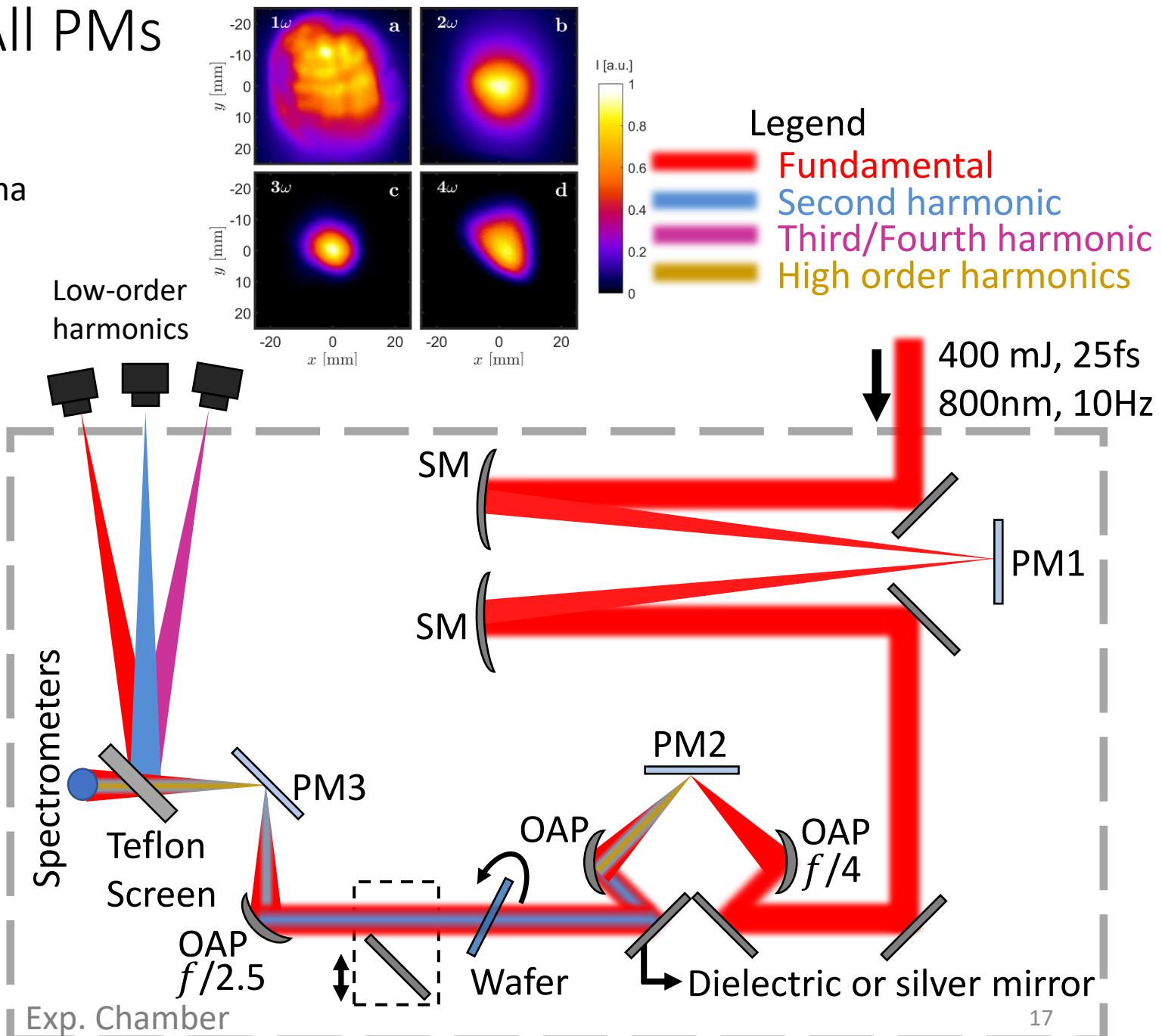
N. M. Fasano, et al. to be submitted (2023).





Experimental set-up – All PMs

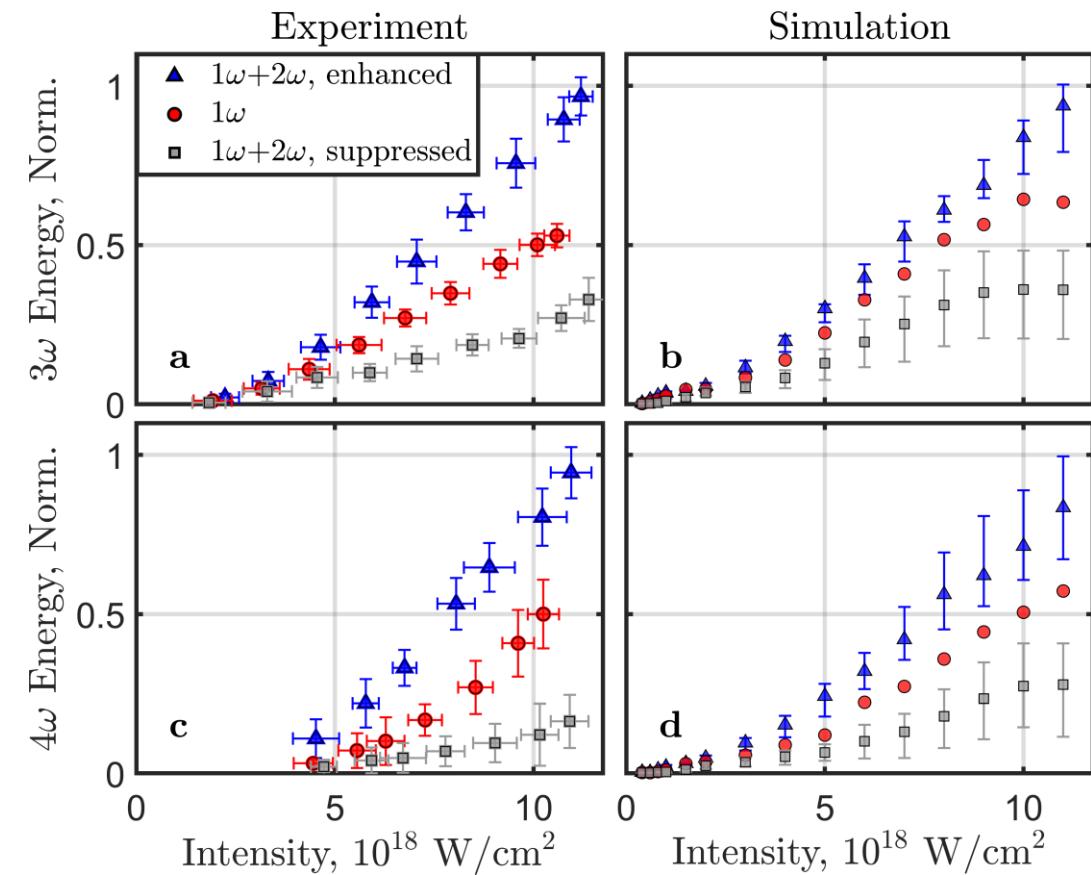
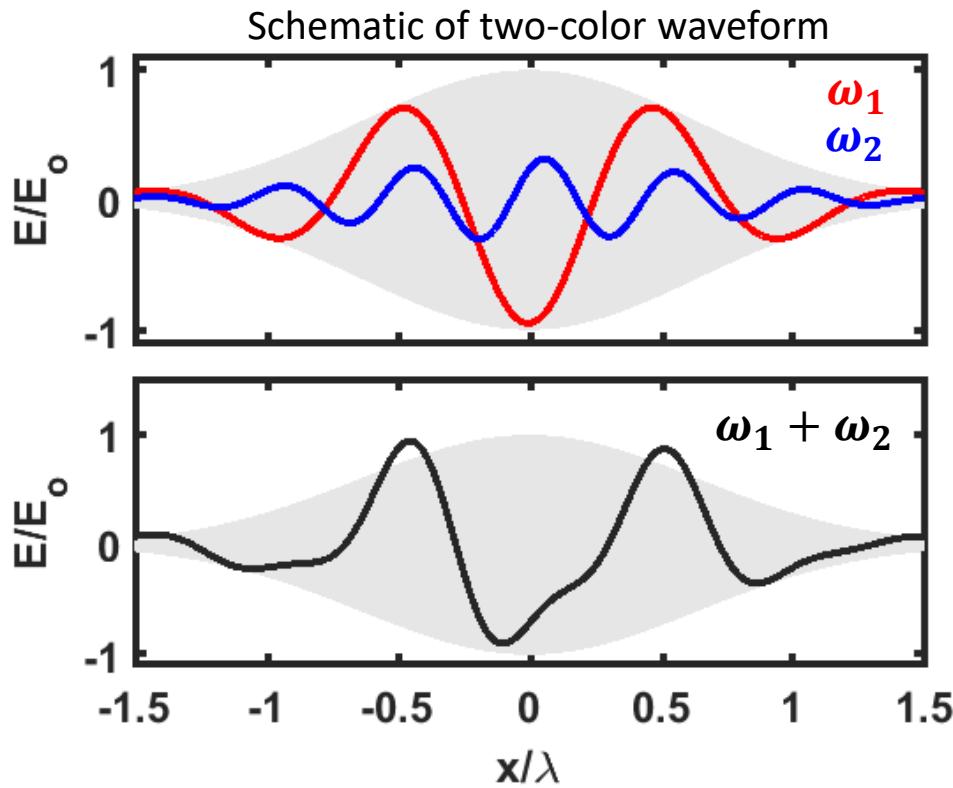
- The third plasma mirror (PM3) is used to investigate harmonic generation from plasma mirrors driven by two-color waveforms
- To compare one-color interactions with two-color interactions we replace a mirror after PM2 with either a dielectric mirror, which only reflects 800nm light, with a silver mirror, which reflects both 800nm and 400nm light.
- To control the waveform of the two-color beam, we insert a thin wafer ($100\mu m$ thick) into the beam path





Waveform-controlled engineering of harmonic generation

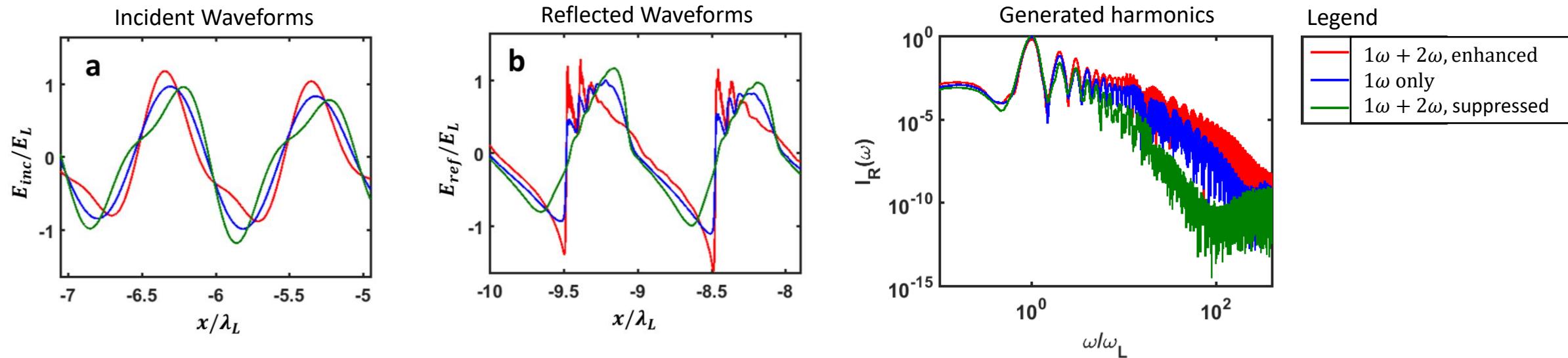
- A multi-color waveform can either suppress or enhance the harmonic generation process, depending on relative phase between the colors - $\Delta\phi_{12} = 2\phi_1 - \phi_2$
- We find an enhancement factor of 1.6 at an intensity of $1 \times 10^{19} W/cm^2$ when 10% of the laser's energy is in the second harmonic and the relative phase is appropriately tuned



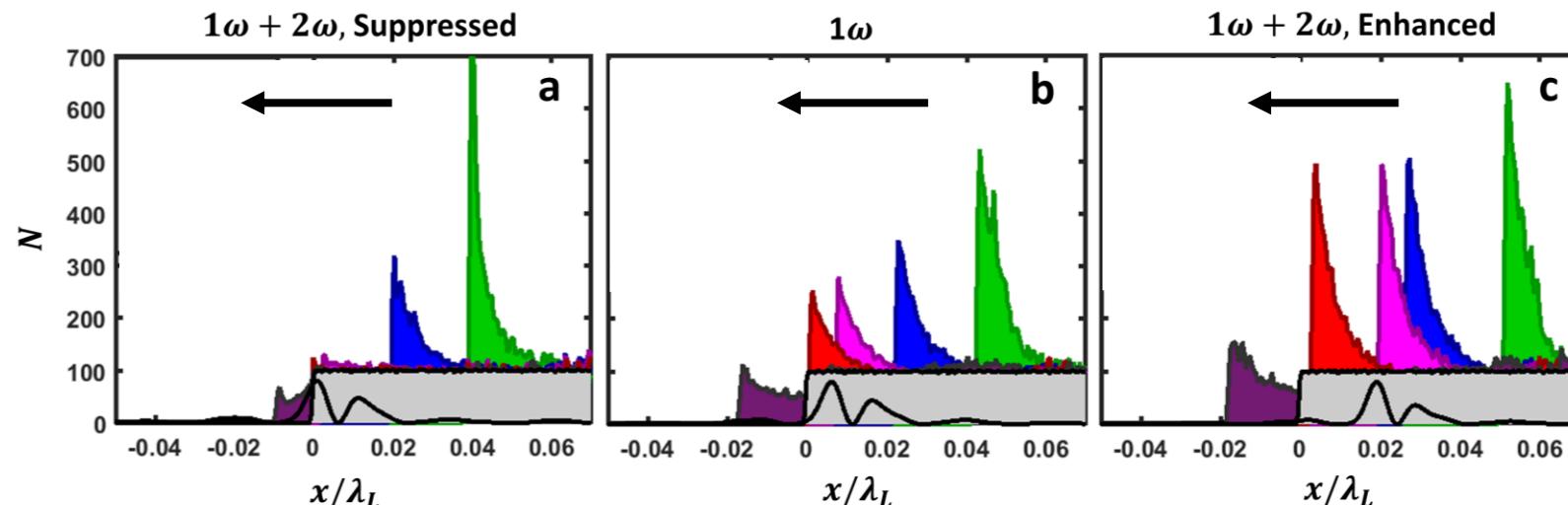


Waveform-controlled engineering of harmonic generation: electron bunch dynamics

- Adjusting the waveform of the driving laser provides fine-tuned control of the electron bunch dynamics at the attosecond timescale, yielding enhanced or suppressed harmonic efficiency



Attosecond electron bunch dynamics leading up to attosecond pulse emission





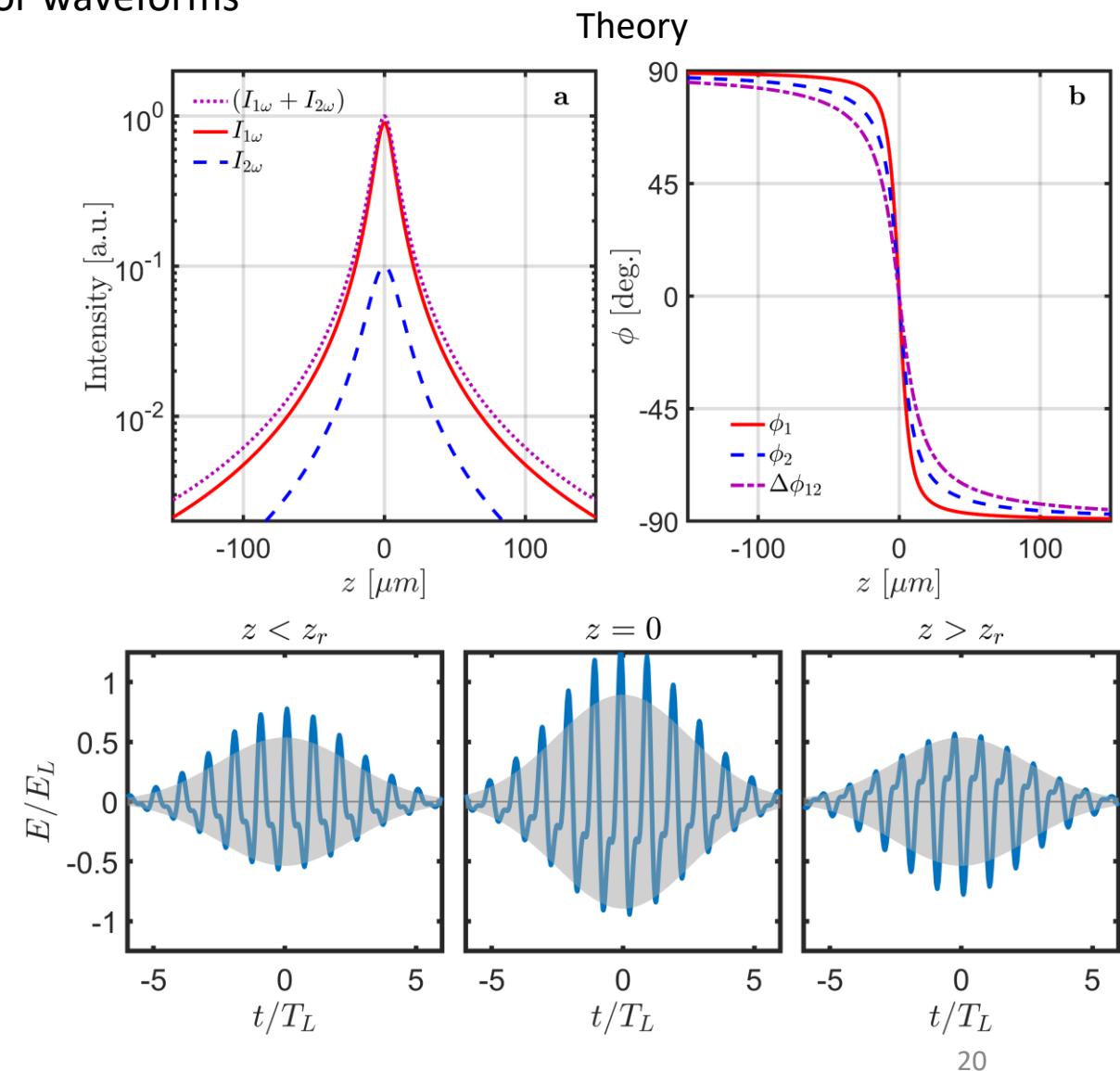
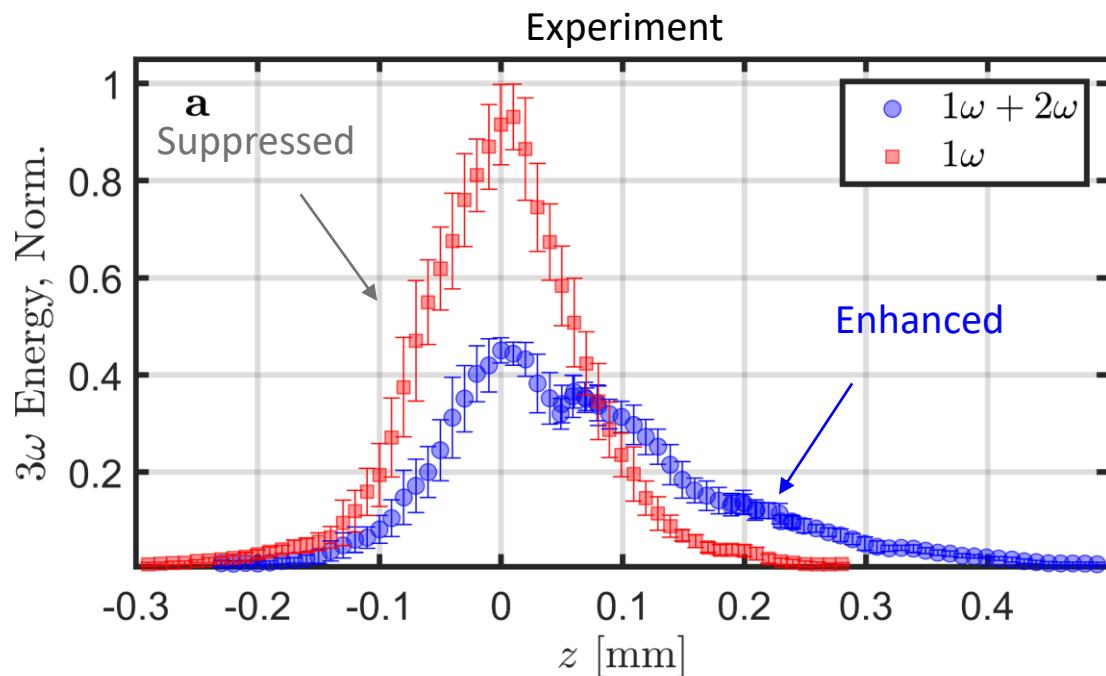
Effects of free space propagation on the two-color's waveform

- The waveform of the laser changes as it propagates in free space, which is an important consideration for single-color few-cycle and multi-color waveforms

Paraxial beam propagation equation:

$$E(\vec{r}, t) = E_L \frac{w_0}{w(z)} \exp\left[-\frac{r^2}{w(z)^2}\right] \exp\left[i \frac{k}{2} \frac{r^2}{R(z)}\right] \exp(i\phi_{Gouy}(z))$$

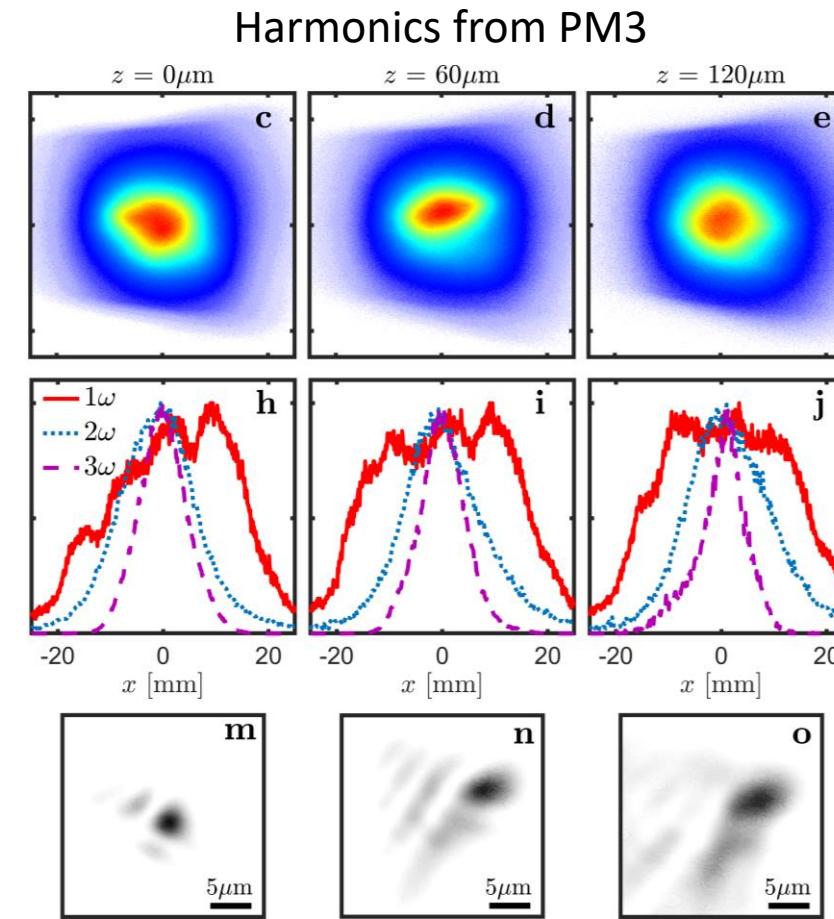
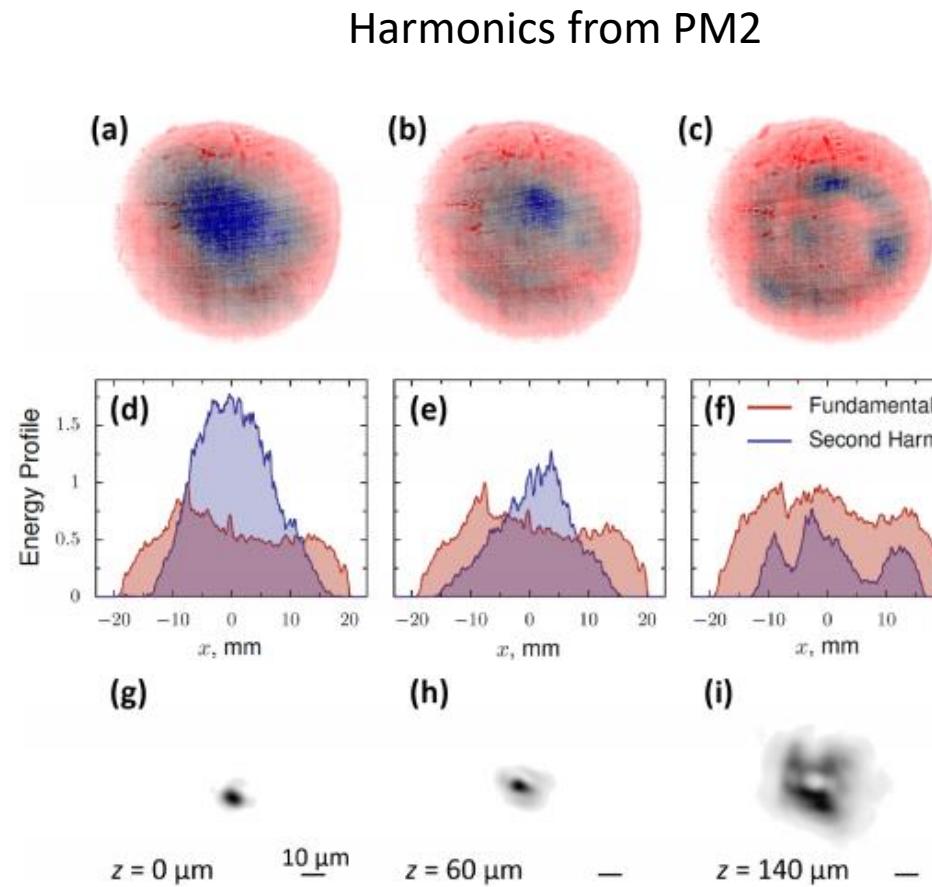
$$\text{Guoy Phase: } \phi_{Gouy}(z) = \arctan\left(-\frac{z}{z_r}\right)$$





Cascaded plasma mirror improves spatial quality of harmonics

- The spatial quality of radiated harmonics, specifically the second harmonic produced from interactions when the plasma mirror positioned outside the Rayleigh range, is substantially improved after a multi-pass plasma mirror configuration when compared to a single pass



M. R. Edwards, N. M. Fasano, et al. *Opt. Lett.* (2020).

N. M. Fasano, et al. to be submitted (2023).



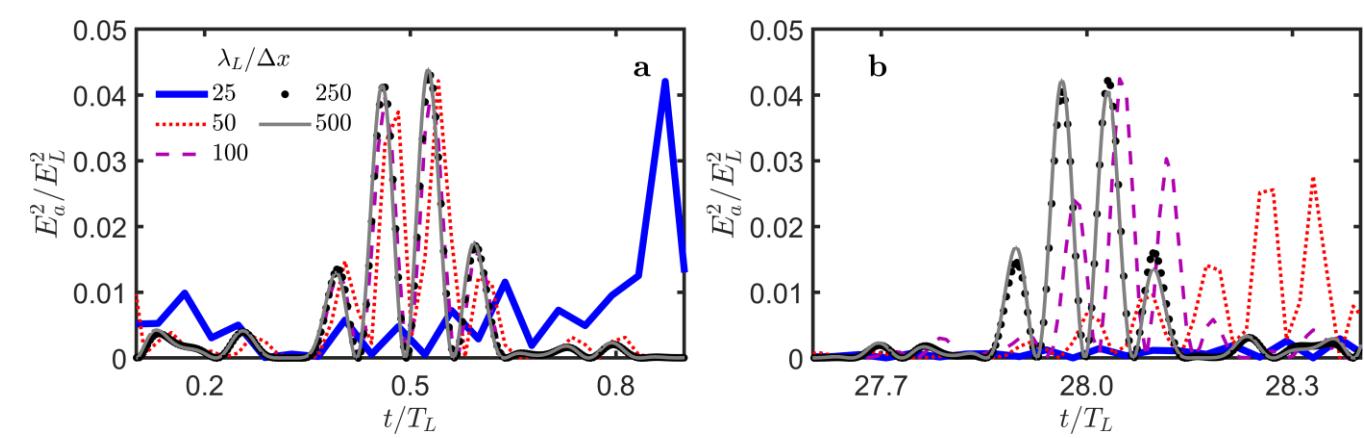
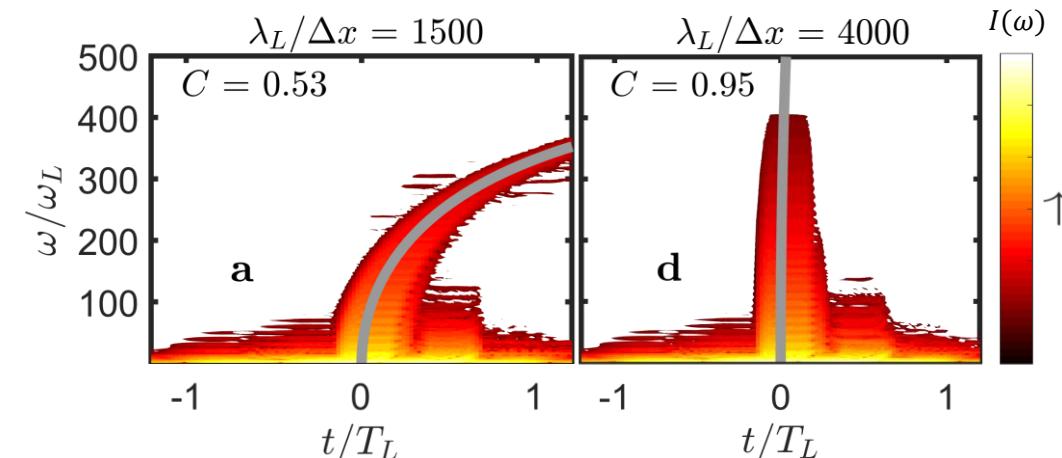
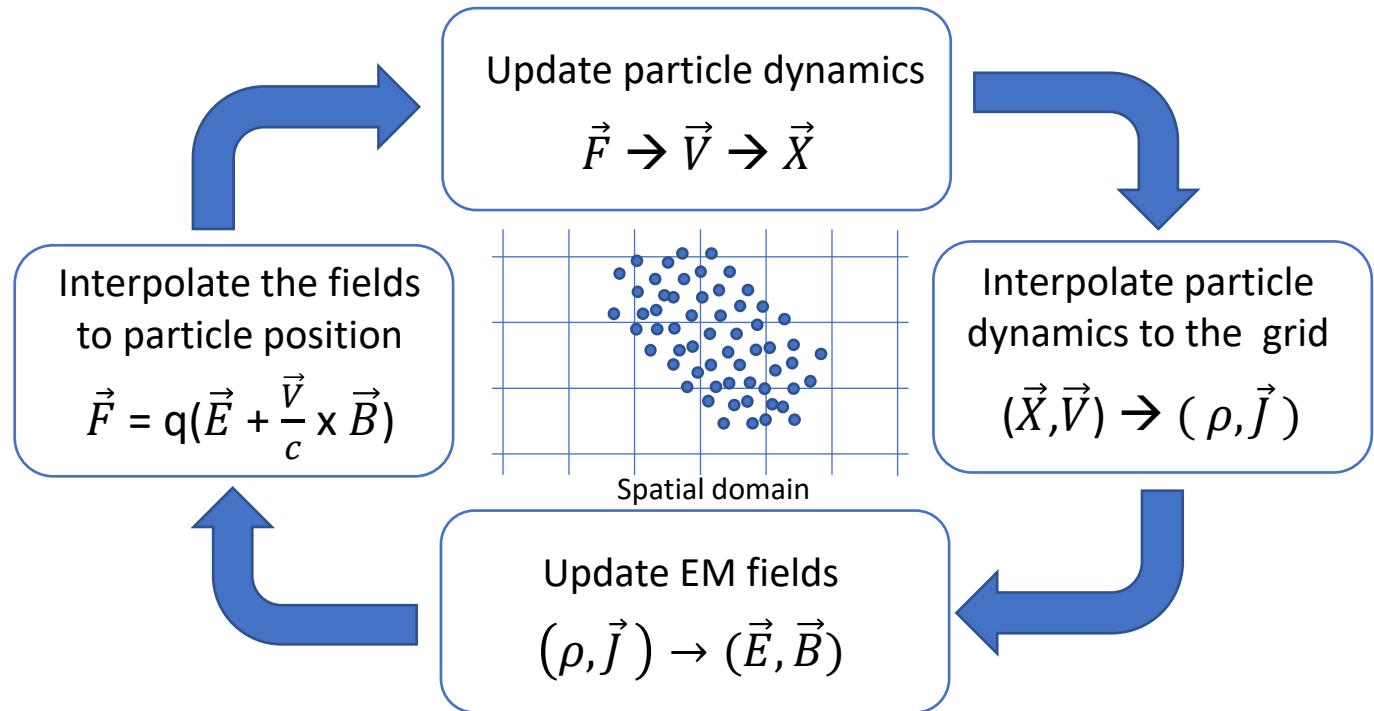
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Modeling of laser-plasma interactions: On density artifacts, collisions, and numerical dispersion in particle-in-cell simulations

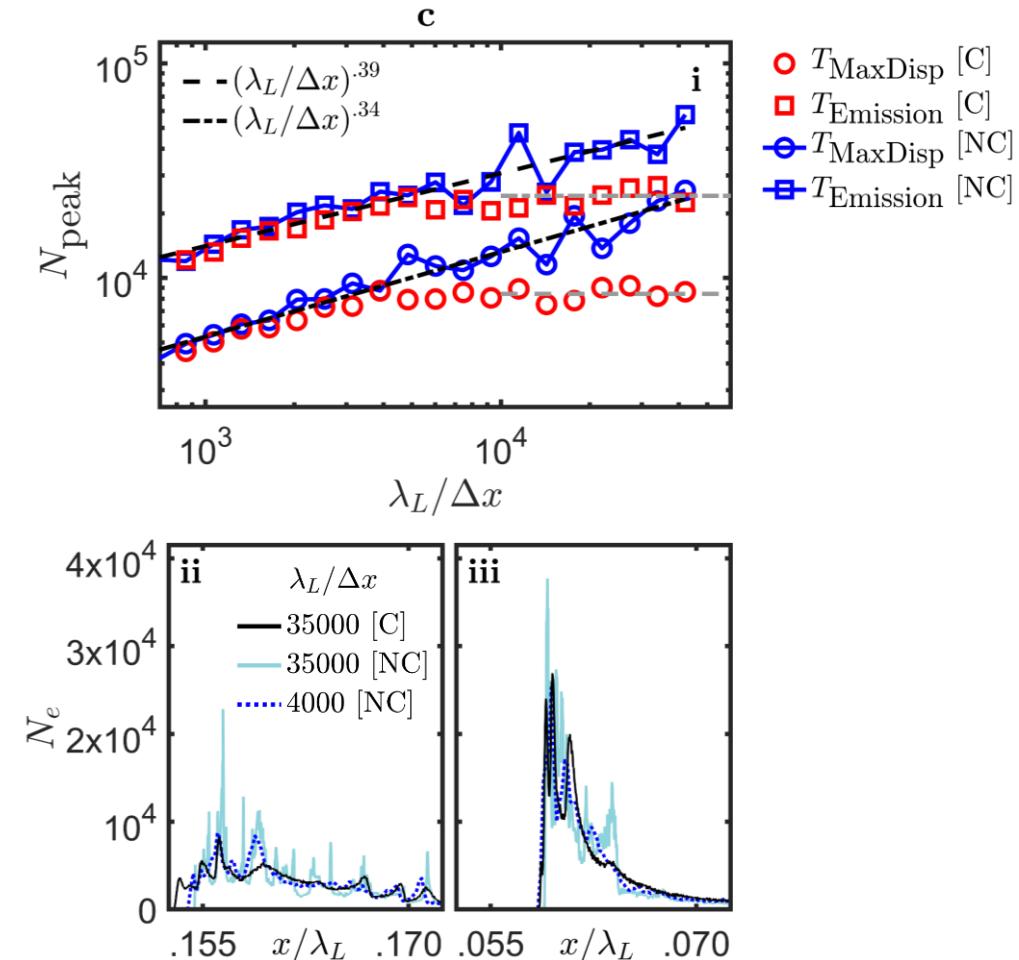
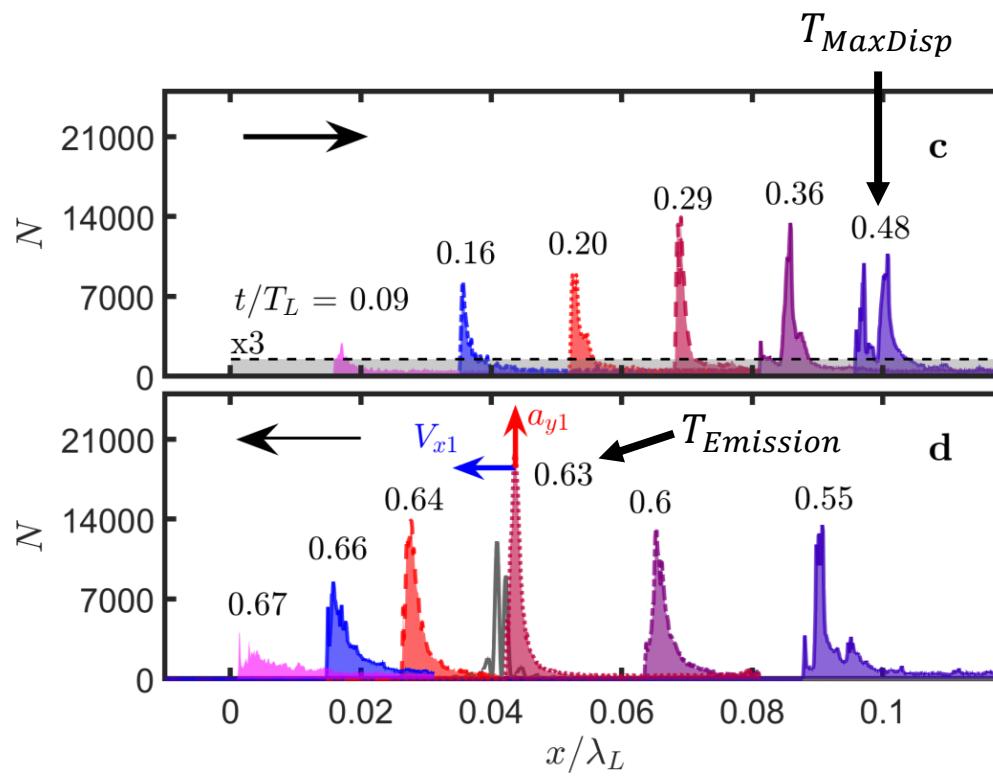
- PIC simulations have been the workhorse for modeling relativistic laser-plasma interactions
- Employs physical approximations and introduces numerical artifacts
 - Finite-difference time-domain algorithms lead to numerical dispersion
 - Finite-sized macroparticles smooth out short-range collisional interactions





Modeling of laser-plasma interactions: On density artifacts, collisions, and numerical dispersion in particle-in-cell simulations

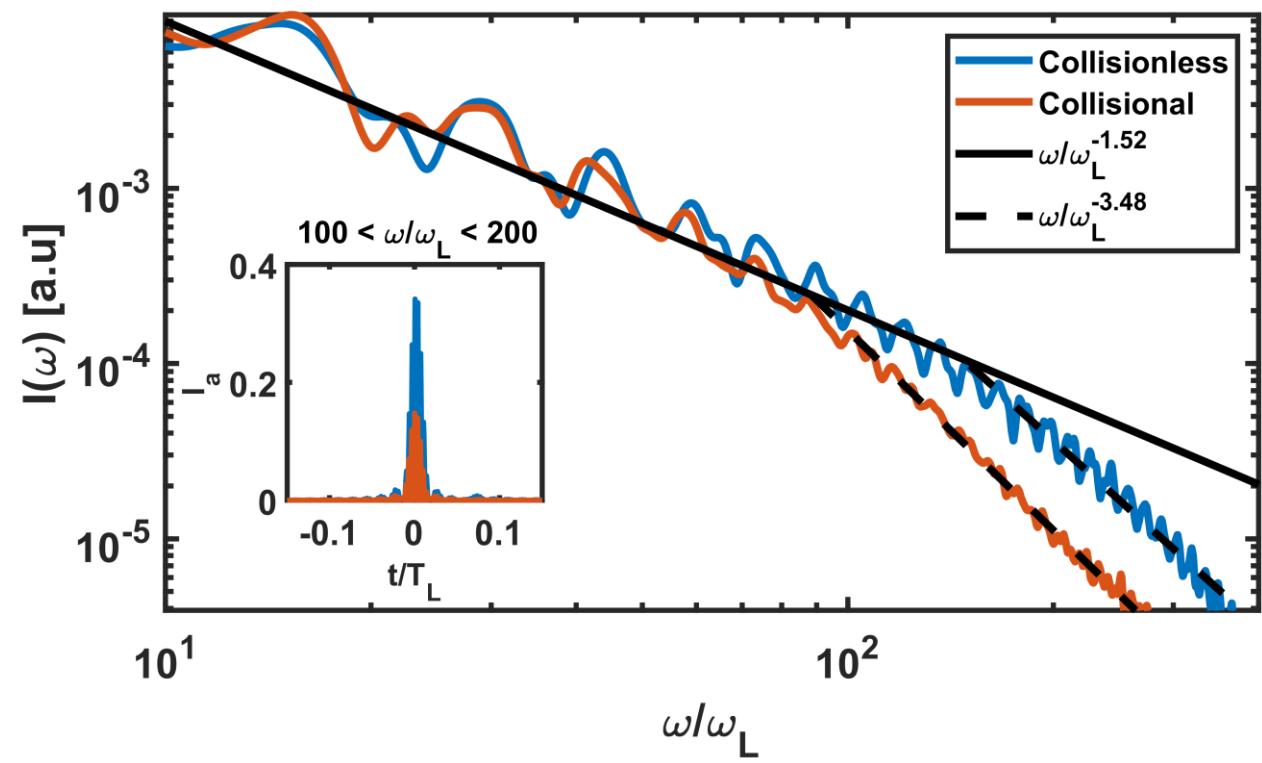
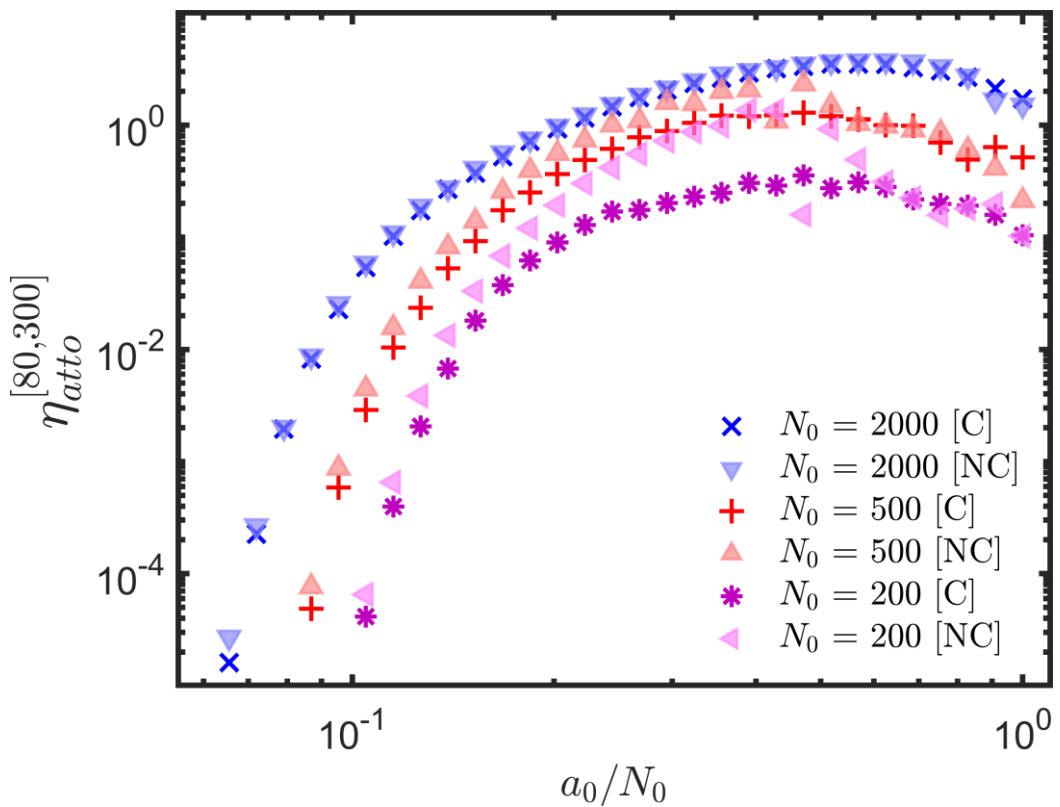
- In a collisionless PIC simulations, the peak density of the emitting electron bunch increases with the spatial resolution of the simulation grid.
- When collisions are added to the model, the peak electron density becomes independent of the spatial resolution.





Collisions reduce harmonic efficiency at short wavelengths

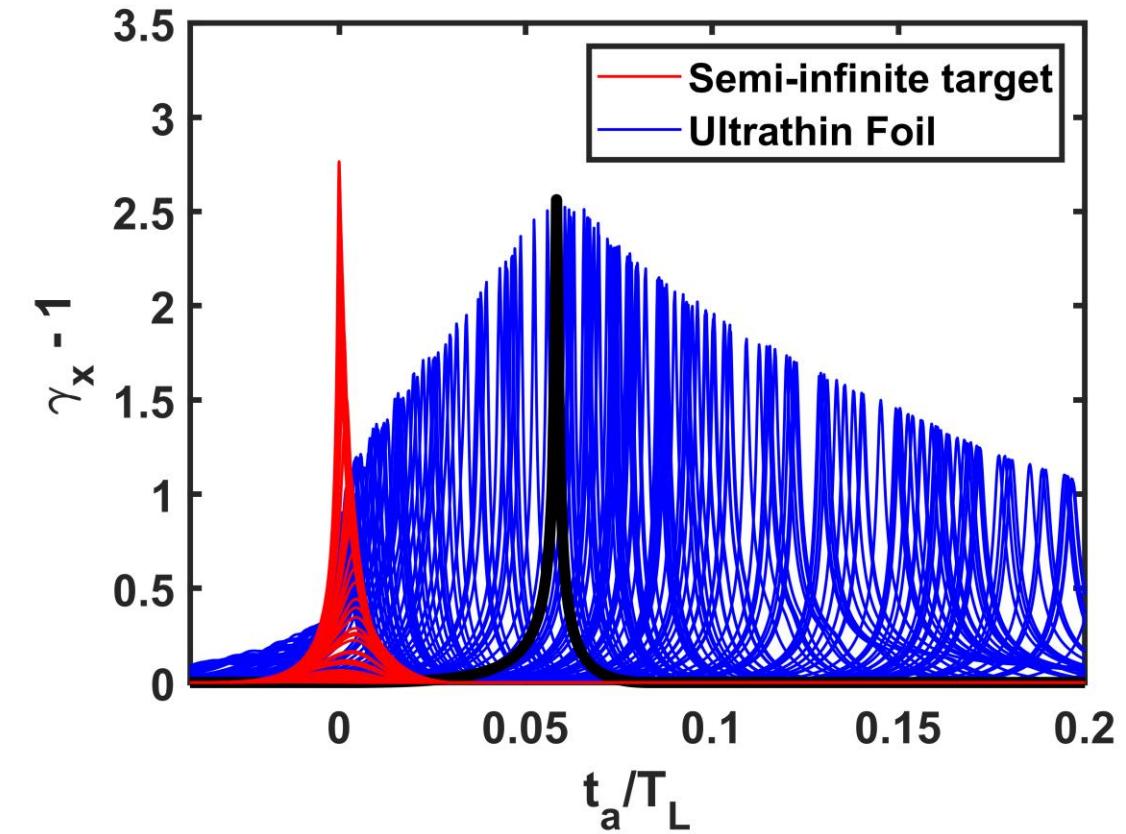
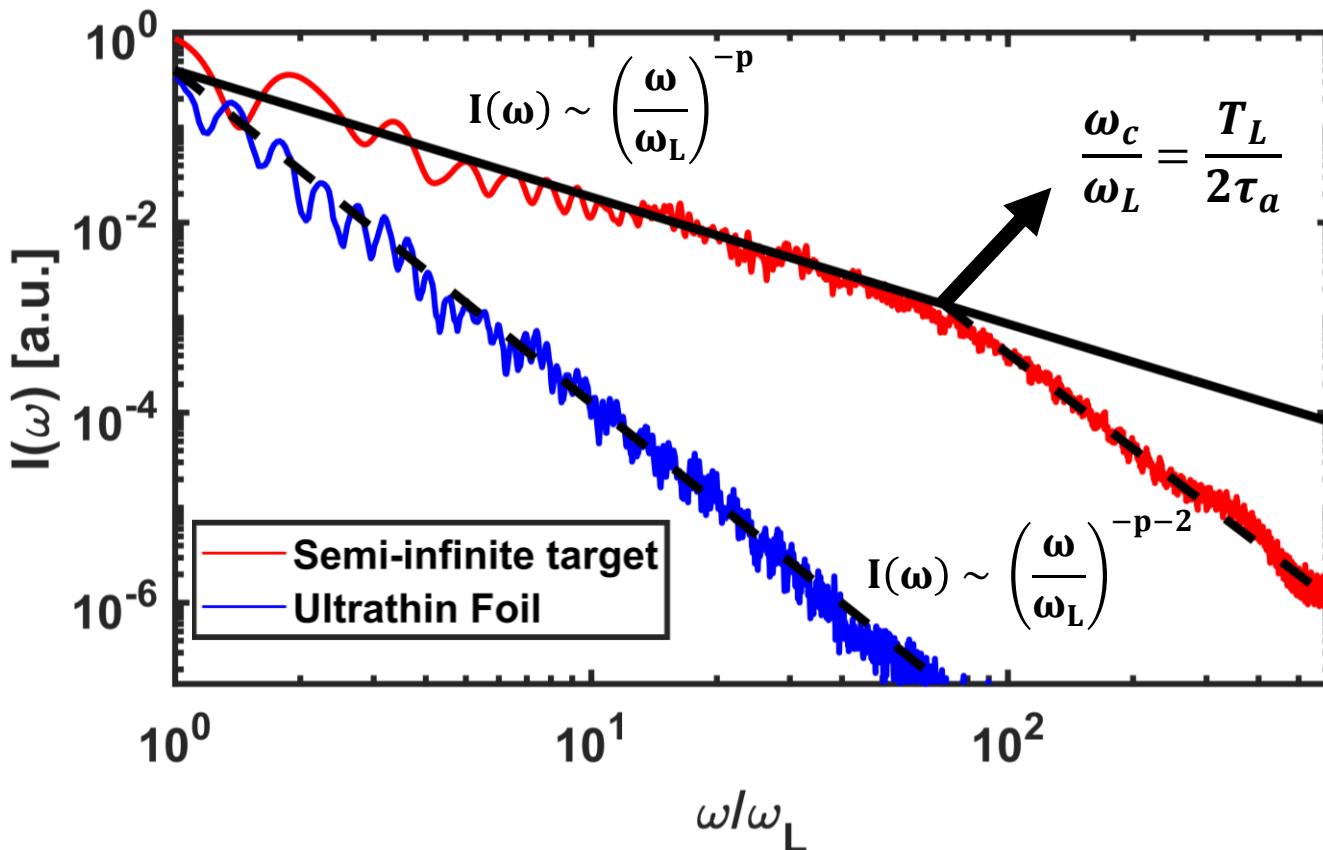
- Incorporating collisions into particle-in-cell simulations leads to an earlier spectral cut-off compared to the collisionless case.





Power-law scaling and the cut-off frequency

- Particle-in-cell simulations predict a power-law spectral scaling up until a cut-off frequency set by the finite duration of the emitting electron bunch [1-3]
- For ultrathin foils, the electron bunch width extends to several tenths of a laser period and dominates the spectral power law [4, 5]



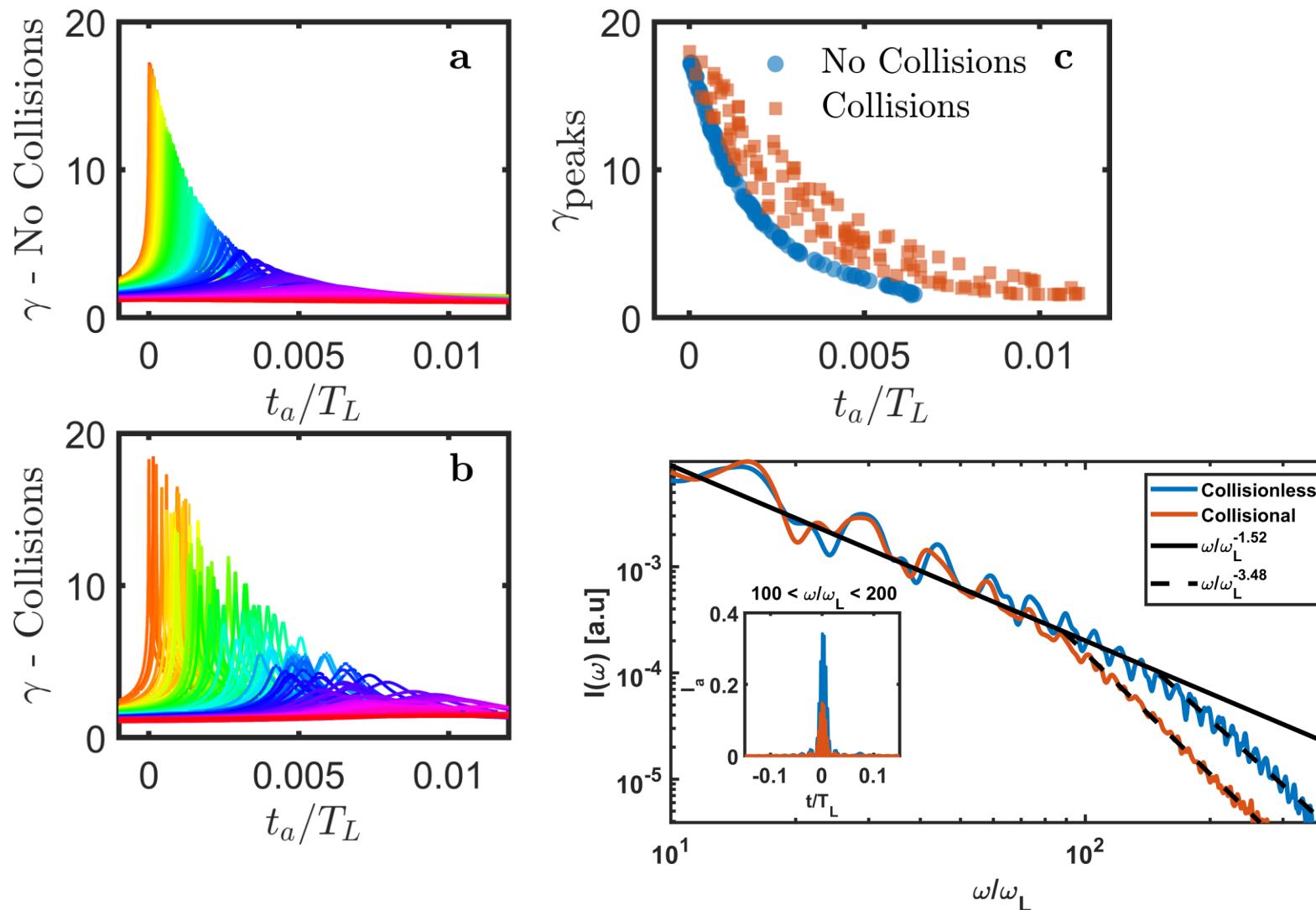
[1] D. an der Brügge et al. Phys. Plasmas, (2010).
[2] J. M. Mikhailova, et al. Phys. Rev. Lett. (2012).
[3] M. R. Edwards, et al., Sci. Rep. (2020).

[4] M. R. Edwards, N. M. Fasano, and J. M. Mikhailova, Phys. Rev. Lett. (2020).
[5] N. M. Fasano, M. R. Edwards, J. M. Mikhailova, CLEO (2020).



How collisions modify the emitting electron bunch

- Collisions result in a wider emitting electron bunch, resulting in an earlier frequency cut-off





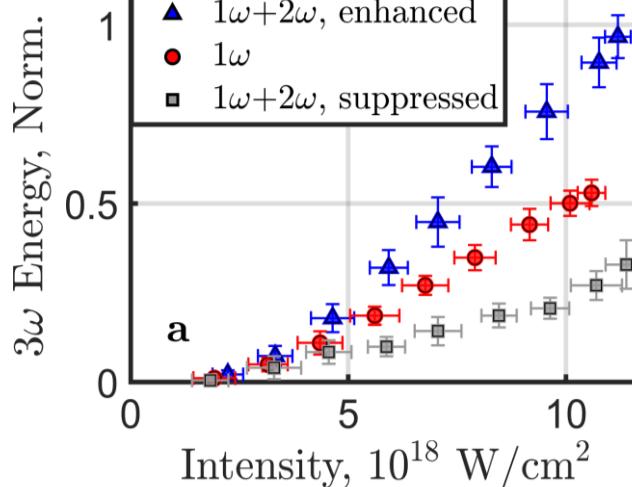
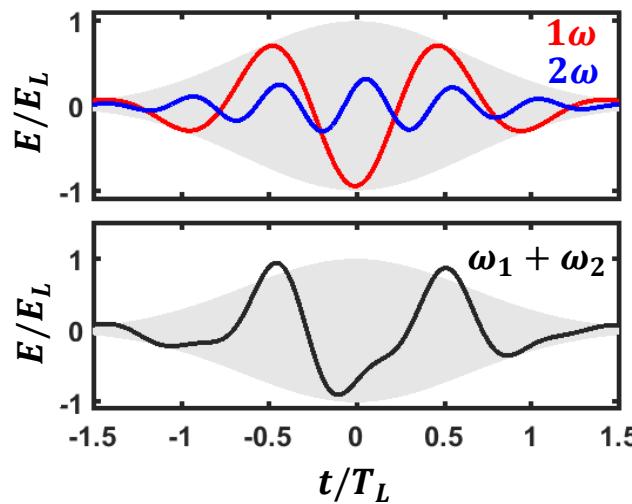
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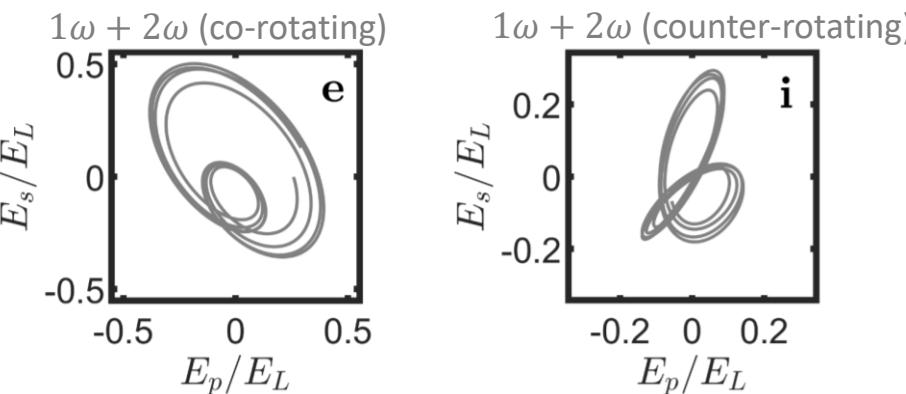
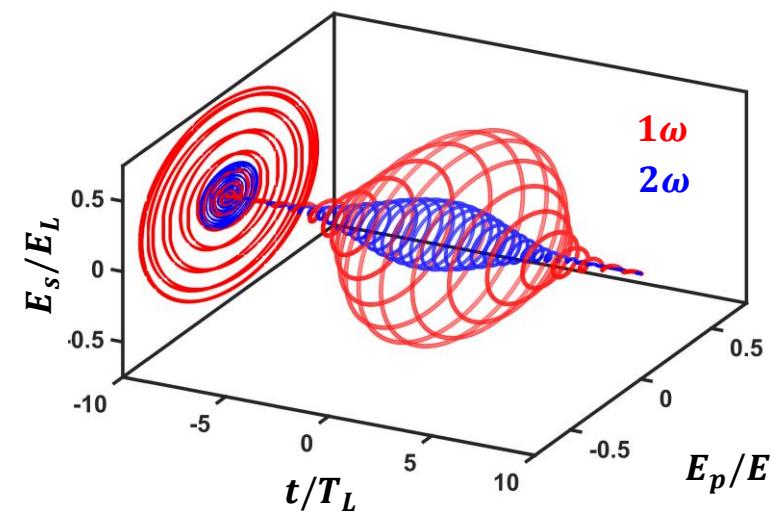
Generating structured light from plasma mirrors

Multi-color waveforms



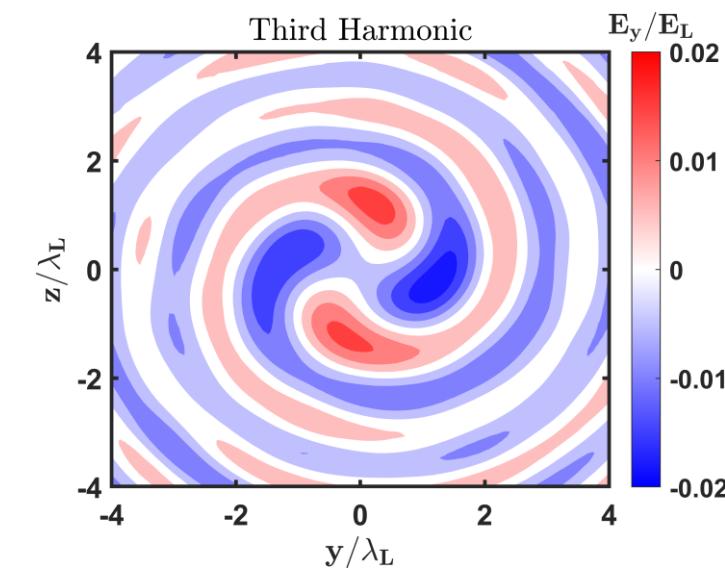
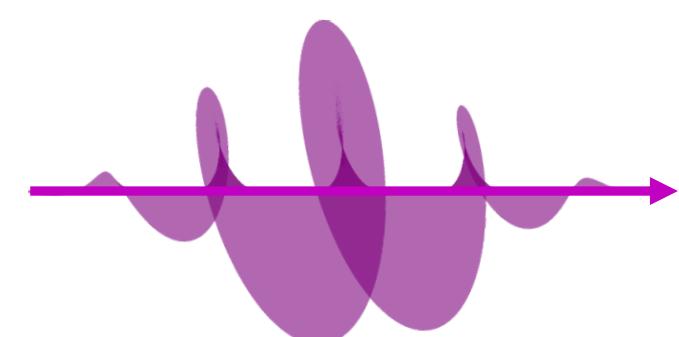
M. R. Edwards, et al. *Phys. Rev. Lett.* (2016).
M. R. Edwards, et al. *Opt. Lett.* (2014).
M. Yeung, et al. *Nat. Photon* (2017).

Elliptically polarized harmonics



R. Lichters, et al. *Phys. Plasmas* (1996).
Z.-Y. Chen, et al. *Nat. Commun.* (2016).
G. Ma, et al. *Opt. Express* (2016).
M. Blanco, et al. *Phys. Plasmas* (2018).

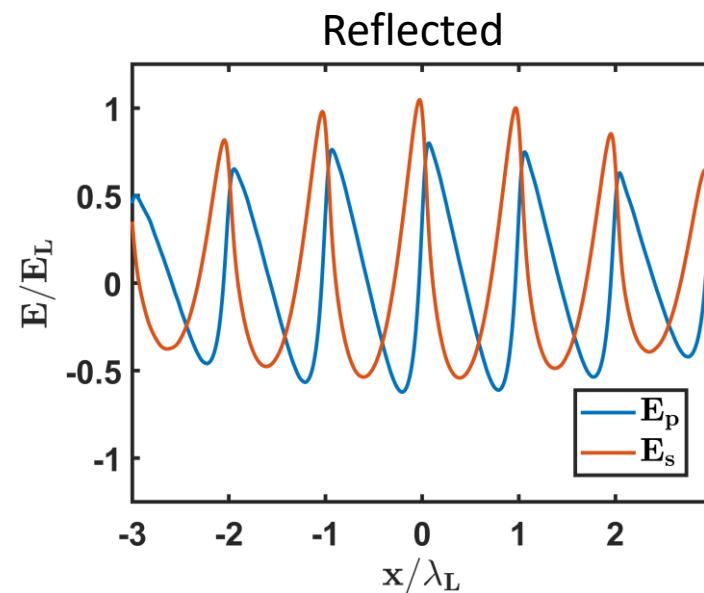
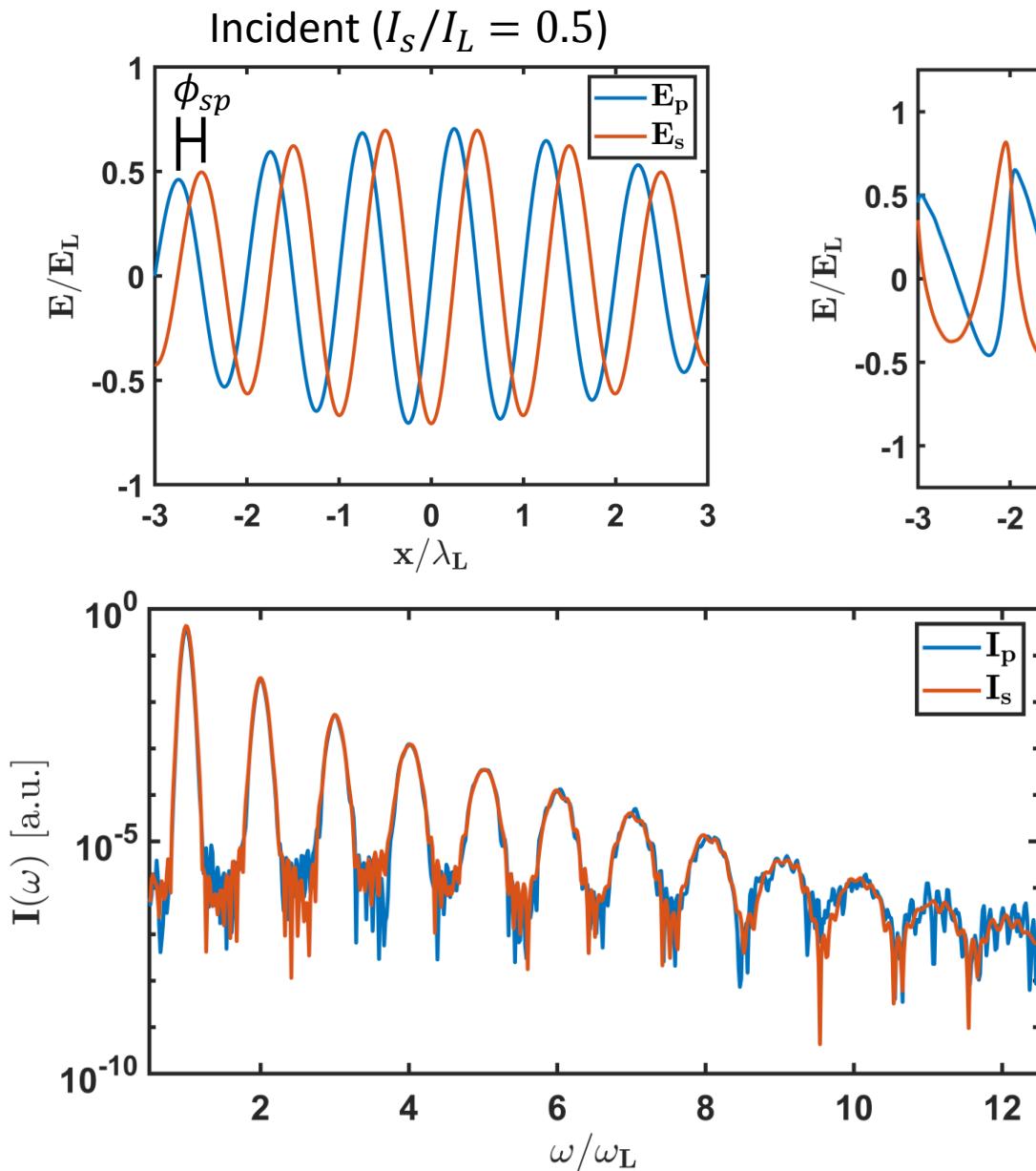
Vortex harmonics



N. M. Fasano and J. M. Mikhailova, *CLEO* (2020).
J. W. Wang, et al. *Nat. Commun.* (2019).
L. Yi, *Phys. Rev. Lett.* (2021).



Tuning the laser's ellipticity for circularly polarized (CP) harmonics



We can analyze individual harmonics by filtering the spectra in the range:
 $\omega_n = [n - 0.5, n + 0.5]\omega_L$

Simulation parameters

$\lambda_L = 800\text{nm}$
 $a_0 = 10$
 $\tau_{fwhm} = 8T_L$
 $W_0 = 4\lambda_L$
 $\theta = 45^\circ$
 $N = 400$
 $L = 0.05\lambda_L$

$\phi_{sp} = \pi/2$
 $I_s/I_L = 0.5$



Tuning the laser's ellipticity for circularly polarized (CP) harmonics

RH Circular polarization

Linear polarization

LH Circular polarization

P-polarized

Ellipticity

1

0

-0.5

-1

0

0.2

0.4

0.6

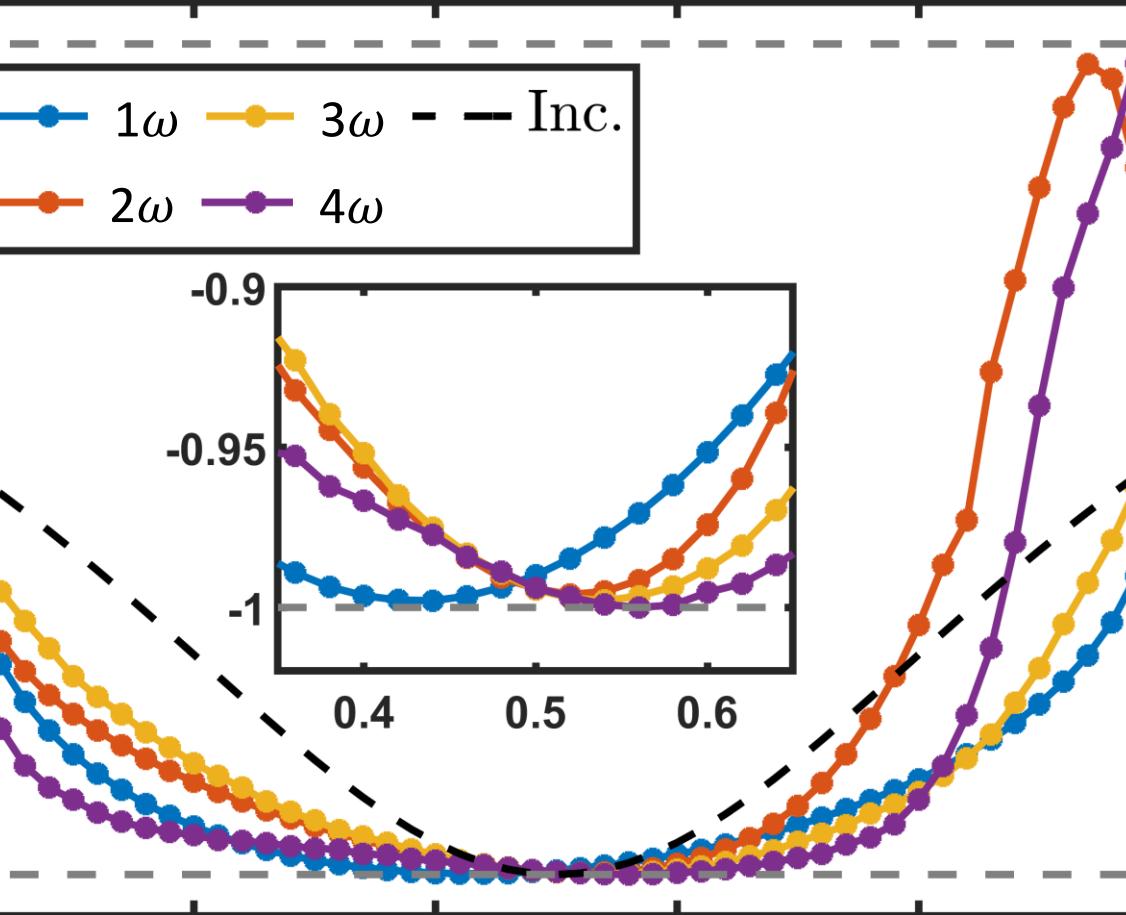
0.8

1

I_s/I_L

Circularly polarized

S-polarized



Simulation parameters

$$\lambda_L = 800\text{nm}$$

$$a_0 = 10$$

$$\tau_{fwhm} = 8T_L$$

$$W_0 = 4\lambda_L$$

$$\theta = 45^\circ$$

$$N = 400$$

$$L = 0.05\lambda_L$$

$$\phi_{sp} = \pi/2$$

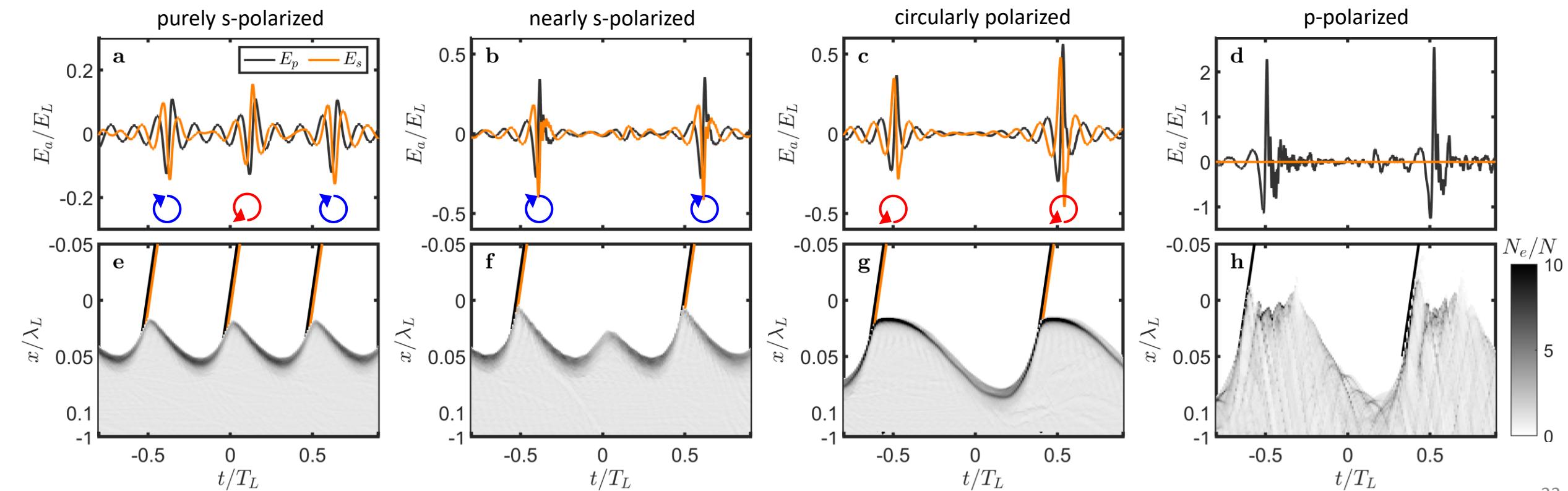
$$I_s/I_L = [0, 1]$$



Effects of polarization control on the electron density oscillations

- S-polarized interactions emit two CP attosecond pulses every laser period, where consecutive attosecond pulses in the train have opposite helicity
- Nearly s-polarized interactions emit only one CP attosecond pulse each laser period, since the component of the p-polarized electric field that acts normal to the plasma surface suppresses one of the two surface oscillations that took place for purely s-polarized interactions
- CP interactions also emit one CP attosecond pulse per every laser period, but the helicity of the attosecond pulse train has flipped compared to nearly s-polarized interactions

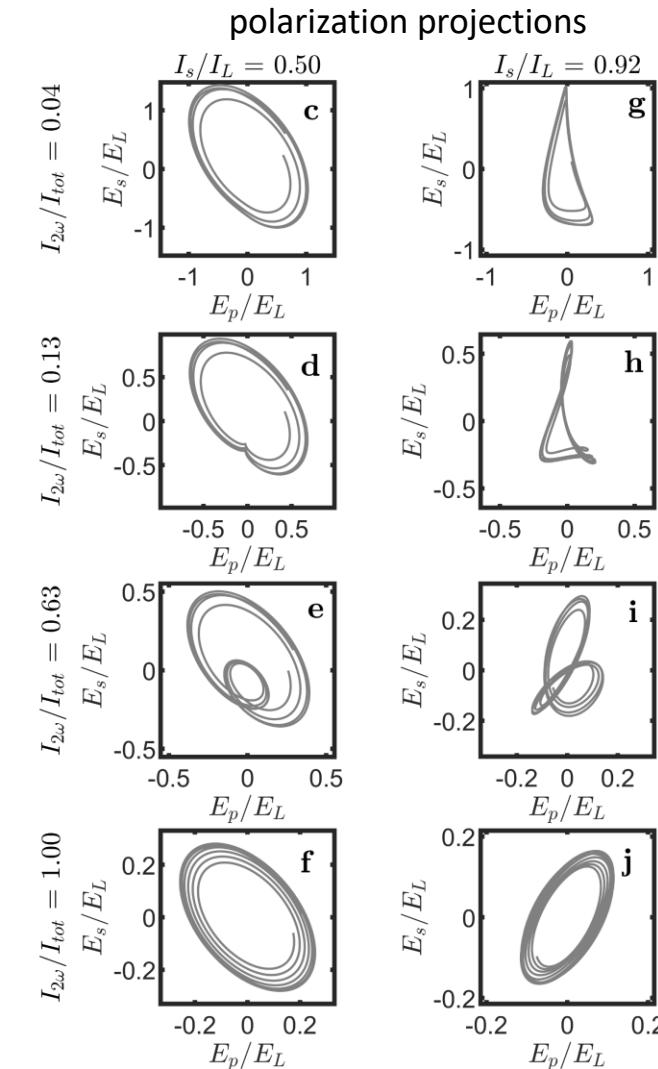
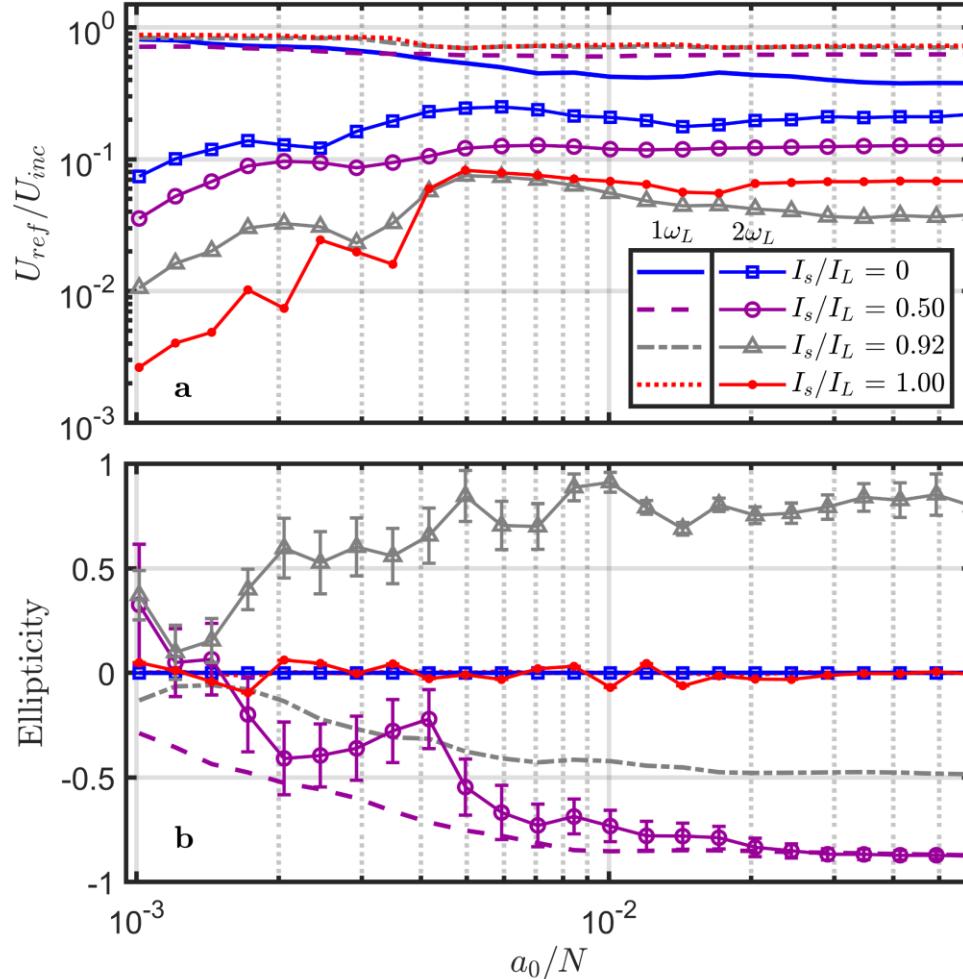
RH circularly polarized
 LH circularly polarized





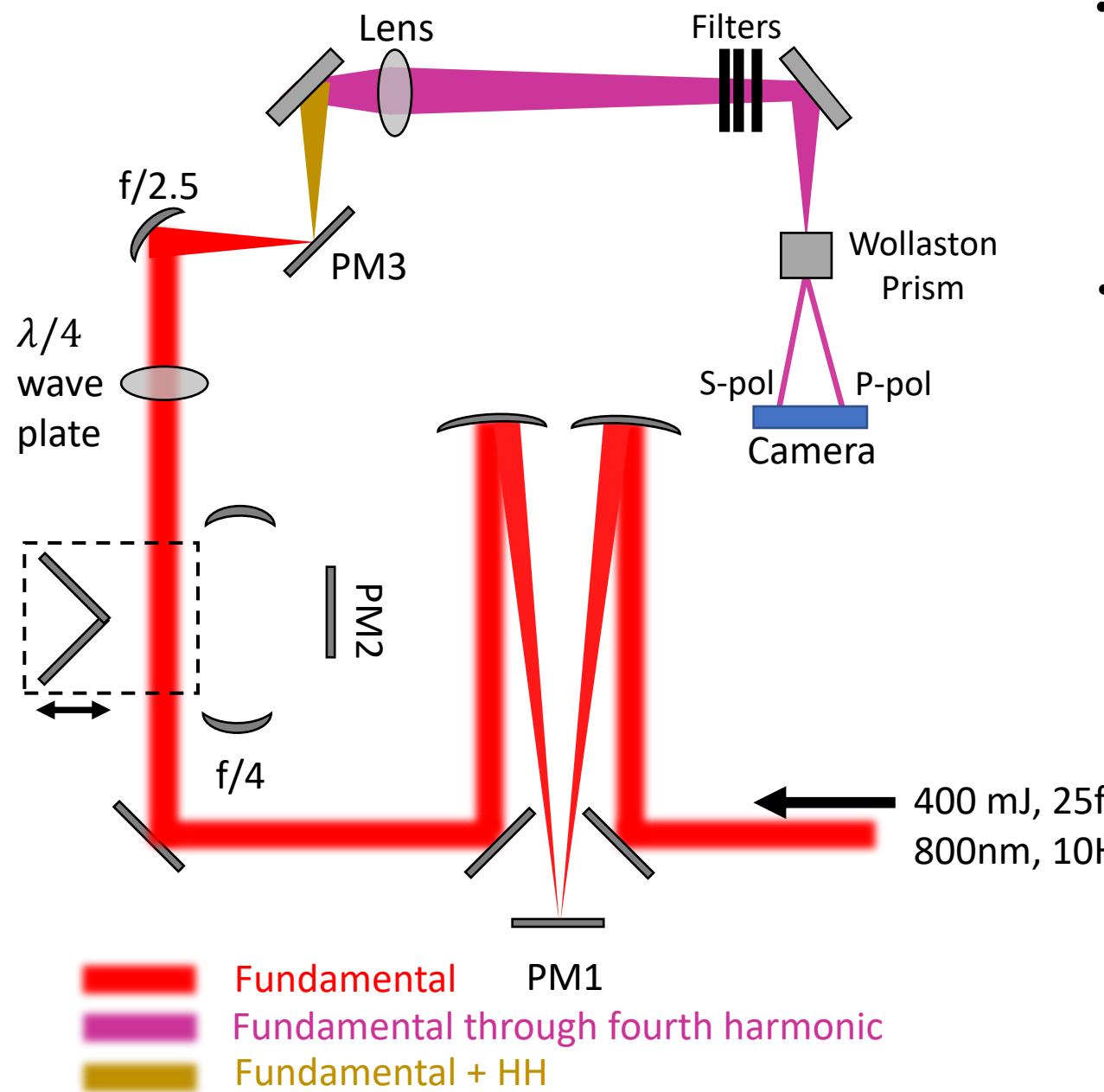
Two-color co- and counter-rotating terawatt laser beams

- Plasma mirrors provide a convenient way to generate high-power two-color co- and counter-rotating lasers with up to 11% and 4% conversion efficiencies, respectively, into the second harmonic

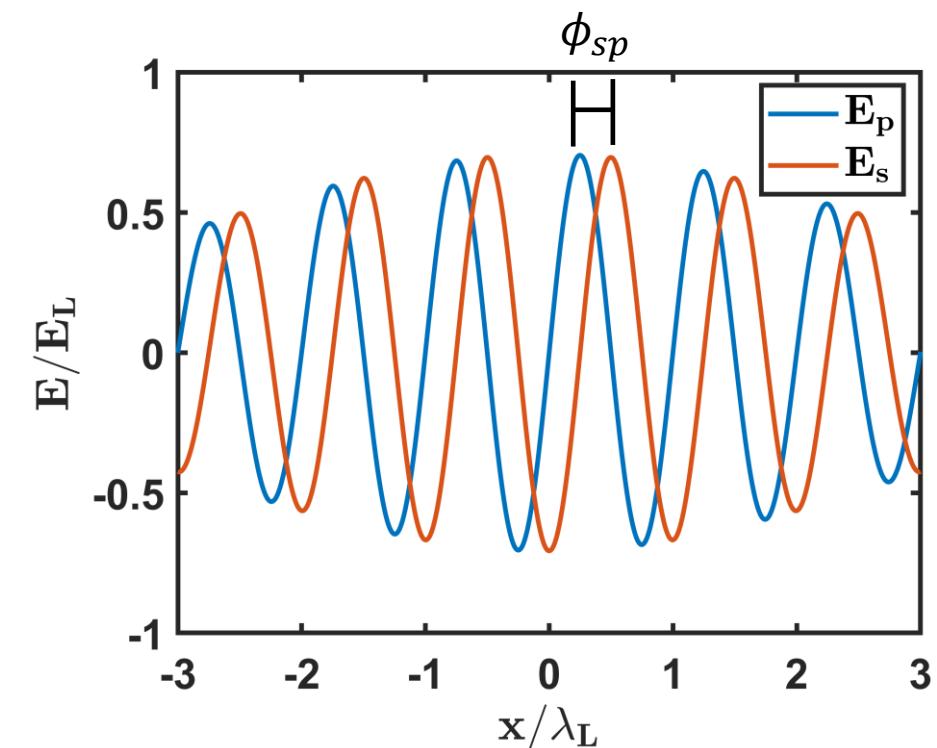




Next steps: detecting CP harmonics in experiments



- Investigating elliptically polarized driving lasers only requires small modifications to the original set-up
 - Quarter waveplate will convert LP driving laser to CP
 - Wollaston prism will allow for determining the complete polarization state of individual harmonics
- Fine tuning of the laser's polarization state will allow for selection of harmonic orders with circular polarization





Outline

- 1) Introduction to plasma optics and their applications
- 2) Relativistic harmonic generation from plasma mirrors: overview and scaling laws
- 3) Cascaded plasma mirrors for enhanced harmonic generation
- 4) Modeling harmonic generation from plasma mirrors with particle-in-cell simulation
- 5) Harmonic source with controlled polarization state
- 6) **Summary of significant results**



Summary of significant results

- 1) Cascaded plasma mirror configuration for enhanced harmonic generation
 - i. HHG from a plasma mirror is used to construct a two-color terawatt beam, with 15% conversion efficiency into the second harmonic.
 - ii. HHG driven with a multi-color waveform can either suppress or enhance the harmonic generation process, depending on relative phase between the colors.
 - iii. We find an enhancement factor of 1.6 at an intensity of $1 \times 10^{19} W/cm^2$ when 10% of the incident laser's energy is second harmonic
 - iv. The spatial quality of radiated harmonics is improved after a multi-pass plasma mirror configuration when compared to a single pass alone.
- 2) Modeling harmonic generation from plasma mirrors with particle-in-cell simulation: on density artifacts, collisions, and numerical dispersion
 - i. Incorporating collisions into particle-in-cell simulations removes artificial density spikes and leads to an earlier spectral cut-off compared to the collisionless case.
 - ii. Numerical dispersion from Yee algorithm can be suppressed for significant propagation lengths, so long as at least 10 cells per shortest wavelength of interest is used
- 3) Harmonic source with a controlled polarization state and spatial structure
 - i. Circularly polarized harmonics can be generated from plasma mirrors driven by circularly polarized lasers or by nearly s-polarized lasers, where a small fraction of energy (<5%) has been converted to the p-polarized component with a $\pi/2$ phase difference
 - ii. (see supplementary slides)



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ETOILES group members:

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Thesis committee and readers: Professor Michael Littman, Dr. Mikhail Shneider

FPO Examiners: Professor Nathaniel Fisch, Professor Aditya Sood

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Program of Plasma Science
and Technology



Thank you for your attention! Questions?



Citations

Papers under review or in preparation:

1. **N. M. Fasano**, M. R. Edwards, and J. M. Mikhailova, Electron bunch dynamics and emission in particle-in-cell simulations of relativistic laser-solid interactions: on density artifacts, collisions, and numerical dispersions, Under Review at Physics of Plasmas.

2. **N. M. Fasano** and J. M. Mikhailova, "Co- and Counter-Rotating Harmonics Radiated from Plasma Mirrors Driven by Elliptically Polarized Lasers", In preparation.

3. **N. M. Fasano**, M. R. Edwards, A. Giakas, A. Morozov, T. Bennett, and J. M. Mikhailova, "Harmonic Generation by Cascaded Plasma Mirrors", in preparation.

Peer-reviewed manuscripts:

1. M. R. Edwards, V. R. Munirov, A. Singh, **N. M. Fasano**, E. Kur, N. Lemos, J. M. Mikhailova, J. S. Wurtele, and P. Michel, "Holographic Plasma Lenses", Physical Review Letters 128, 065003 (2022).

2. M. R. Edwards, **N. M. Fasano**, T. Bennett, A. Griffith, N. Turley, B.M. O'Brien, and J. M. Mikhailova, "A multi-terawatt two-color beam for high-power field-controlled nonlinear optics", Optics Letters 45, 6542 (2020).

3. M. R. Edwards, **N. M. Fasano**, and J. M. Mikhailova, "Electron-nanobunch-width-dominated spectral power law for relativistic harmonic generation from ultrathin foils", Physical Review Letters 124, 185004 (2020).

Conferences:

1. **N. M. Fasano**, M. R. Edwards, A. Giakas, A. Morozov, T. Bennett, J. M. Mikhailova. Low-Order Harmonics Emitted from Relativistic Plasma Mirrors Driven by Two-Color and Elliptically Polarized Lasers, APS DPP meeting, Bulletin of the American Physical Society, 2021. (Oral)

2. M. R. Edwards, **N. M. Fasano**, N. Lemos, A. Singh, E. Kur, J. S. Wurtele, J. M. Mikhailova, P. Michel. High-Focusing High-Power Laser Pulses with Diffractive Plasma Lenses, APS DPP meeting, Bulletin of the American Physical Society, 2021.

3. **N. M. Fasano** and J. M. Mikhailova, High-Power Ultraviolet Vortex Beams Generated from a Relativistic Laser Interacting with an Ultrathin Foil, CLEO: Conference on Lasers and Electro-Optics, 2021. (Oral)

4. M. R. Edwards, **N. M. Fasano**, N. Lemos, A. Singh, V. Munirov, E. Kur, J. S. Wurtele, J. M. Mikhailova, P. Michel. Measuring the Optical Properties of Ionization Gratings in Air for Control of Femtosecond Lasers. CLEO: Conference on Lasers and Electro-Optics, 2021.

5. **N. M. Fasano**, M. R. Edwards, and J.M. Mikhailova, Particle-in-Cell Simulations of Harmonic Generation from Relativistic Plasma Mirrors: Effects of Collisions on the Emitting Electron Bunch Width. APS DPP meeting, Bulletin of the American Physical Society, 2020. (Oral)

6. **N. M. Fasano** and Julia M. Mikhailova, Numerical Modeling of Relativistic Harmonic Structure from Plasma Mirrors: Insights into Relativistic Plasma Dynamics, APS DPP meeting, Bulletin of the American Physical Society, 2020. (Poster)

7. M. R. Edwards, **N. M. Fasano**, N. Lemos, A. Singh, E. Kur, J. S. Wurtele, J. M. Mikhailova, P. Michel. High-Intensity Bragg Reflection of a Femtosecond Laser via Ionized Structures in Air, APS DPP meeting, Bulletin of the American Physical Society, 2020.

8. **N. M. Fasano**, M. R. Edwards, J. M. Mikhailova, Effects of Electron Bunch Width on the Efficiency of High-Order Harmonic Generation from Ultrathin Solid Targets, CLEO: Conference on Lasers and Electro-Optics 2020. (Poster)

9. M. R. Edwards, **N. M. Fasano**, E. Lepowsky, A. Giakas, T. Bennett, J. M. Mikhailova. Cascaded Plasma Mirrors for Enhanced Relativistic Harmonic Generation. CLEO: Conference on Lasers and Electro-Optics, 2020.

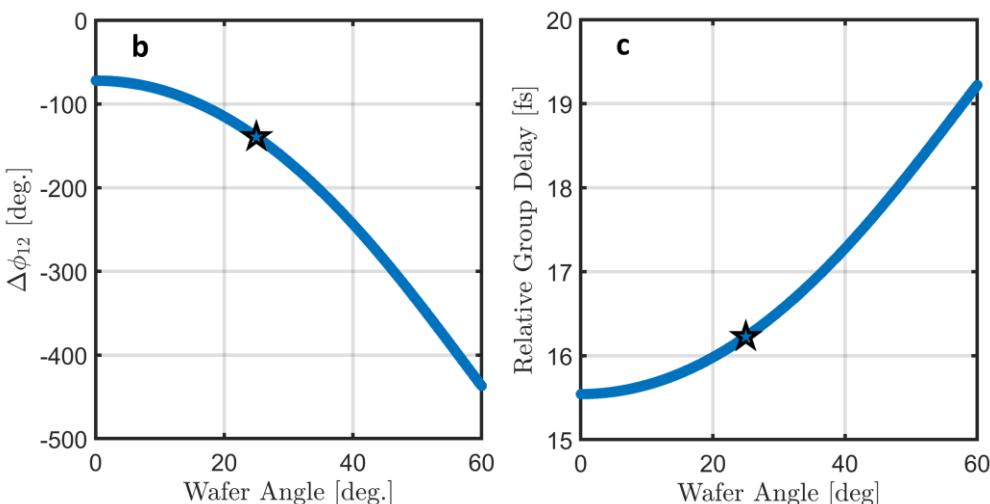
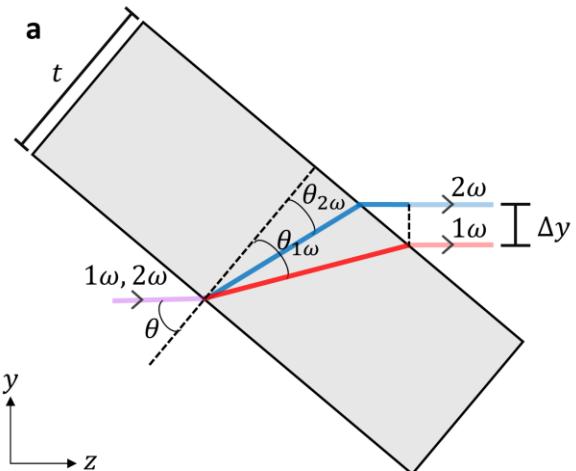
10. **N. M. Fasano**, M. R. Edwards, J.M. Mikhailova, Modeling the Formation of Nanometer-Scale High-Density Electron Bunches in Relativistic Laser-Solid Interaction: Effects of Numerical Resolution. Frontiers in Optics. Washington, DC. 2018. (Poster)

11. **N. M. Fasano**, M. R. Edwards, Julia M. Mikhailova, Sub-Optical-Cycle Dynamics of Electron Nano-Bunches in Relativistic Laser-Plasma Interactions: Insights From Numerical Simulations. 60th Annual Meeting of the APS Division of Plasma Physics. Portland, OR. 2018. (Poster)

12. M. R. Edwards, T. Bennett, A. Griffith, **N. M. Fasano**, B. O'Brien, N. Turley, and J. Mikhailova, Plasma Mirrors as Optical Components for Manipulation of Intense Light. Meeting of the APS Division of Plasma Physics. Portland, OR. 2018.



Effects of two-color laser after propagating through thin wafer



For fused silica with $\theta = 25^\circ$ and $t = 100\mu\text{m}$:

$$\begin{aligned}\Delta\phi_{12} &= -135^\circ \\ \Delta\tau_{12} &= 16\text{fs} \\ y_{12} &= 0.35\mu\text{m} \\ \tau_{out} - \tau_{inc} &< 0.1\text{fs}\end{aligned}$$

Phase difference

$$\Delta\phi_{12} = 2\phi_{1\omega} - \phi_{2\omega} = 2\pi t \left[\frac{2n_{1\omega}}{\lambda_{1\omega} \cos(\theta_{1\omega})} - \frac{n_{2\omega}}{\lambda_{2\omega} \cos(\theta_{2\omega})} - \frac{\sin(\theta)(\tan(\theta_{1\omega}) - \tan(\theta_{2\omega}))}{\lambda_{2\omega}} \right]$$

Group envelope difference

$$\Delta\tau_{12} = \tau_{2\omega} - \tau_{1\omega} = \frac{t}{c} \left[\frac{n_{2\omega} - \lambda_{2\omega} n'_{2\omega}}{\cos(\theta_{2\omega})} + \sin(\theta) (\tan(\theta_{1\omega}) - \tan(\theta_{2\omega})) - \frac{n_{1\omega} - \lambda_{1\omega} n'_{1\omega}}{\cos(\theta_{1\omega})} \right]$$

Spatial walk-off

$$\Delta y_{12} = y_{2\omega} - y_{1\omega} = t \cos(\theta) (\tan(\theta_{1\omega}) - \tan(\theta_{2\omega}))$$

Pulse broadening

$$\frac{\tau_{out}}{\tau_{inc}} = \sqrt{1 + \left(\frac{4 \ln(2) GL}{\tau_{inc}^2} \right)^2}$$



Stokes parameters for determining the polarization state of light

Definition of Stokes parameters

(Experiment)

$$S_0 = I_{90} + I_0 \quad = \langle E_1^2 \rangle + \langle E_2^2 \rangle$$

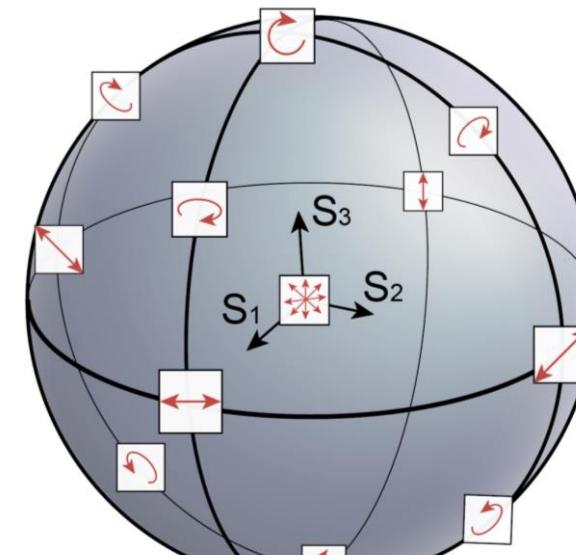
$$S_1 = I_{90} - I_0 \quad = \langle E_1^2 \rangle - \langle E_2^2 \rangle$$

$$S_2 = I_{45} - I_{135} \quad = 2 \langle E_1 \rangle \langle E_2 \rangle \cos(\delta)$$

$$S_3 = I_{RHP} - I_{LHP} = 2 \langle E_1 \rangle \langle E_2 \rangle \sin(\delta)$$

(Theory)

Poincare sphere



credit: wikipedia

For completely polarized light: $S_0^2 = S_1^2 + S_2^2 + S_3^2$

Some common stokes vectors

$$\mathbf{S} = \begin{bmatrix} S_0 \\ S_1 \\ S_2 \\ S_3 \end{bmatrix} \quad \begin{bmatrix} 1 \\ 1 \\ 0 \\ 0 \end{bmatrix}$$

$$\begin{bmatrix} 1 \\ 0 \\ 1 \\ 0 \end{bmatrix}$$

$$\begin{bmatrix} 1 \\ 0 \\ 0 \\ 1 \end{bmatrix}$$

$$\begin{bmatrix} 1 \\ 0 \\ \sqrt{2}/2 \\ \sqrt{2}/2 \end{bmatrix}$$

Linearly polarized
(vertical plane)

Linearly Polarized
(45° angle)

Circularly Polarized
(45° angle)

Elliptically Polarized
(45° angle)



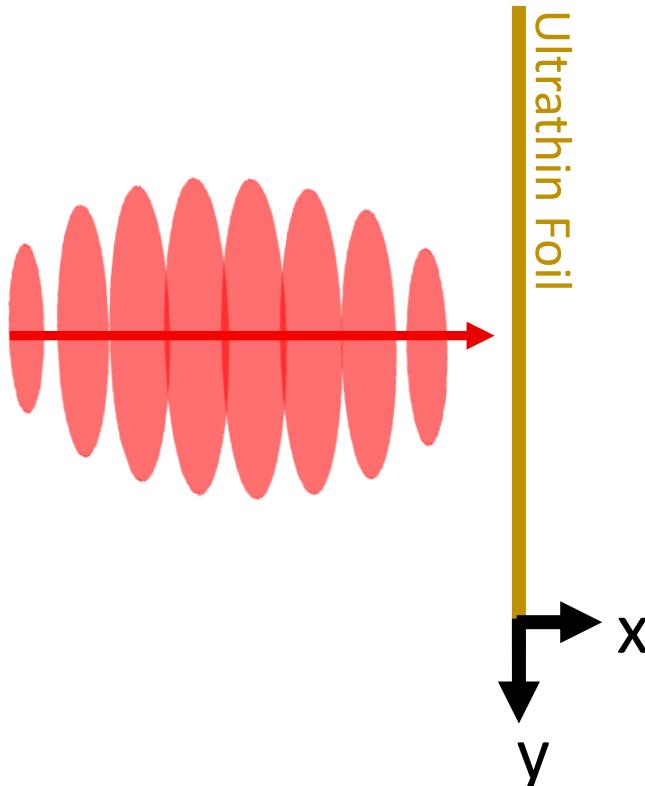
Vortex Beams

- Vortex beams, characterized by a twisted phase front and a null on-axis intensity, are useful for several applications such as optical communications [1], and optical manipulations [2].
 - [1] A.E. Willner et al. *Adv. Opt. Photonics* 7, 66 (2015).
 - [2] M. Padgett and R. Bowman, *Nat. Photon.* 5, 343–348 (2011).
- Schemes have been proposed to realize intense, short wavelength vortex beams:
 - High harmonic generation from plasma mirrors driven by vortex beams [3]
 - [3] A. Denoeud, L. Chopineau, A. Leblanc, and F. Quéré, *Phys. Rev. Lett.* 118, 033902 (2017).
 - Solid density plasma mirrors [4], or micro-apertured targets [5], driven by circularly polarized beams at normal incidence
 - [4] J. W. Wang, M. Zepf, and S. G. Rykovanov, *Nat. Commun.* 10, 5554 (2019).
 - [5] L. Yi, *Phys. Rev. Lett.* 126, 134801 (2021).
- In this work, we use particle-in-cell simulations to demonstrate vortex beam harmonic generation from circularly polarized laser interacting with an ultrathin foil



Ultrathin Foils for Harmonic Generation

$$\eta = f(\mathbf{N}, a_o, \mathbf{D}, L, \tau_{fwhm}, \phi_{cep}, \theta, w_o)$$



Normalized
Plasma Density

$$N = \frac{n_e}{n_c} = n_e \left(\frac{4\pi e^2}{m_e \omega_L^2} \right)$$

Normalized
Laser Amplitude

$$a_o = \frac{e E_L}{m_e \omega_L c}$$

Normalized
Target Thickness

$$D = \frac{d}{\lambda_L}$$

- Harmonic generation from laser-solid interactions is most efficient when the laser and plasma forces are close to balanced [1, 2].
- In the ultrathin foil regime, adjusting the **plasma thickness** adjusts the **effective plasma density (ND)** [3, 4].

$$\eta = f(\mathbf{ND}/a_o, L, \tau_{fwhm}, \phi_{cep}, \theta, w_o)$$

[1] M.R. Edwards, J.M. Mikhailova, Phys. Rev. Lett. 117, 125001 (2016).

[2] M.R. Edwards, J.M. Mikhailova, Sci. Rep. 10, 5154 (2020).

[3] Yu. M. Mikhailova, V. T. Platonenko, and S. G. Rykov, JETP Lett. 81, 571 (2005).

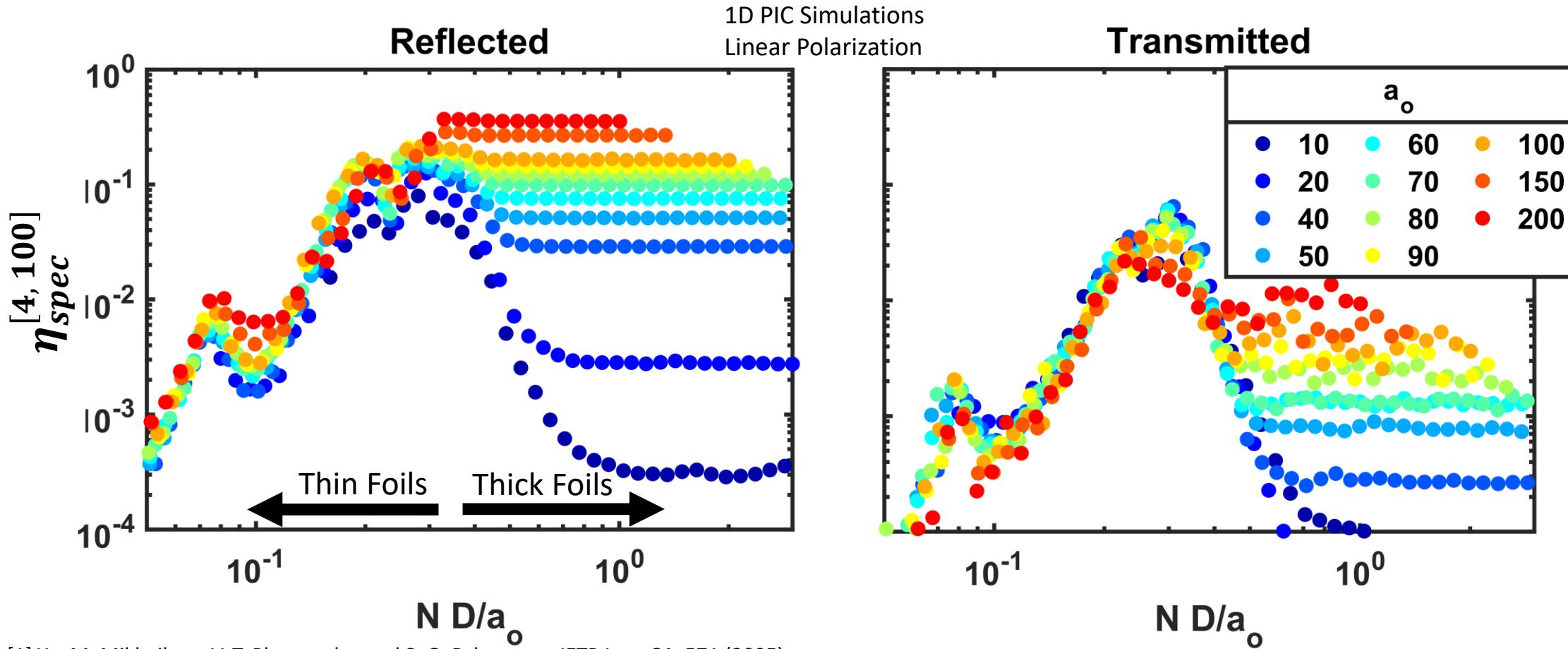
[4] M. R. Edwards, N. M. Fasano, and J. M. Mikhailova, Phys. Rev. Lett. 124, 185004 (2020).



Ultrathin Foils for Harmonic Generation driven by LP laser

- Previously, ultrathin foils have been demonstrated to enhance harmonic generation efficiency for **linearly polarized** driving lasers [1-3].

$$f(\mathbf{N}, a_o, d, \lambda, \phi_{cep}, \theta, \tau_{fwhm}, L) = f(ND/a_o, \phi_{cep}, \theta, \tau_{fwhm}, L)$$



[1] Yu. M. Mikhailova, V. T. Platonenko, and S. G. Rykovyanov, JETP Lett. **81**, 571 (2005).

[2] J. M. Mikhailova, M. V. Fedorov, N. Karpowicz, et al. Phys. Rev. Lett. **109**, 245005 (2012).

[3] M. R. Edwards, N. M. Fasano, and J. M. Mikhailova, Phys. Rev. Lett. (2020).

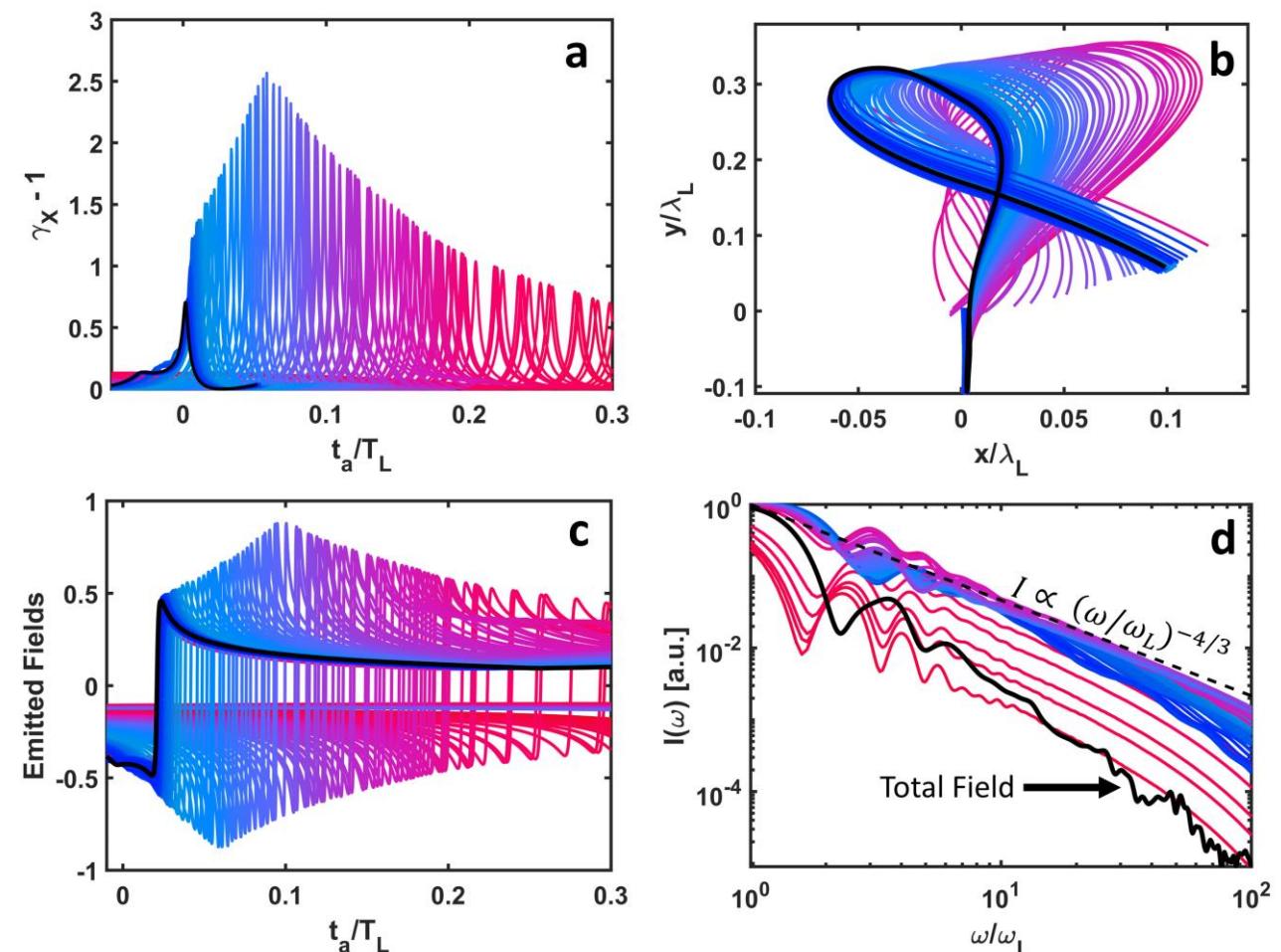


Ultrathin Foils for Harmonic Generation driven by LP laser

- With detailed knowledge of the particle trajectories, it is possible to reconstruct the emitted fields of individual electrons:

$$E_y = \frac{2\pi\sigma V_y}{c - V_x \operatorname{sgn}(x - x_0)}$$

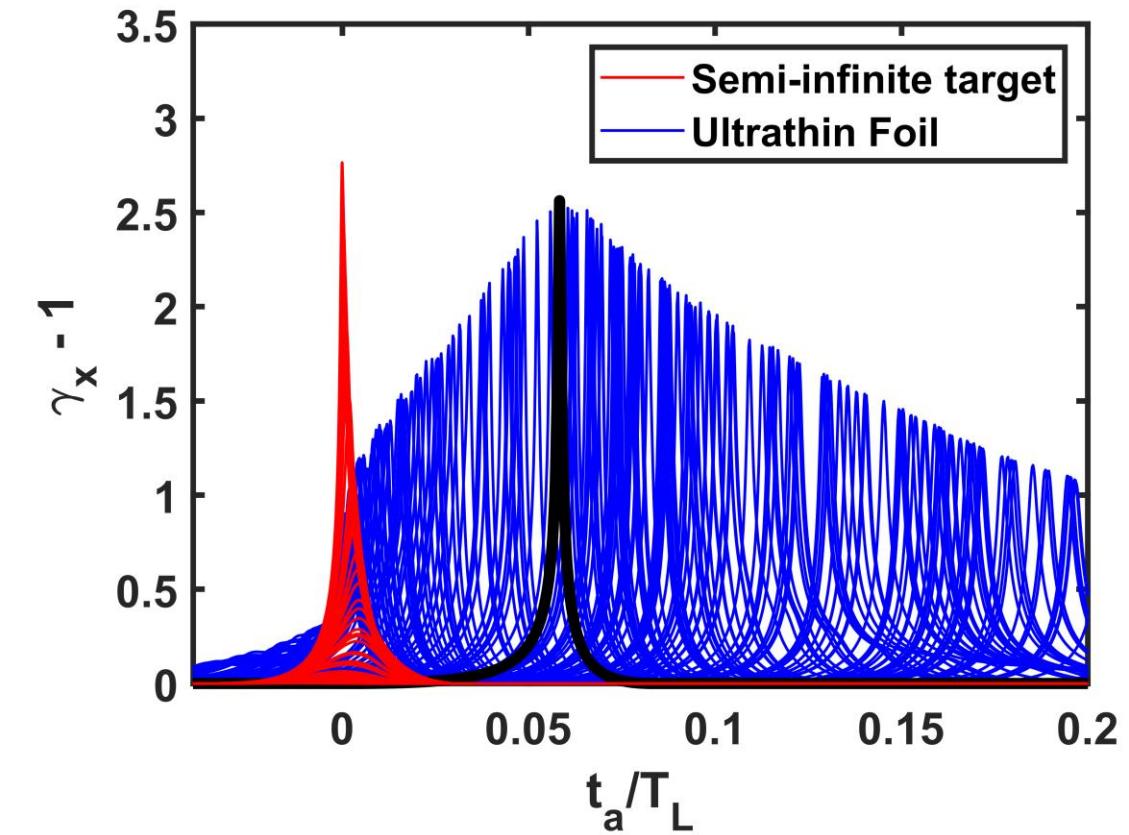
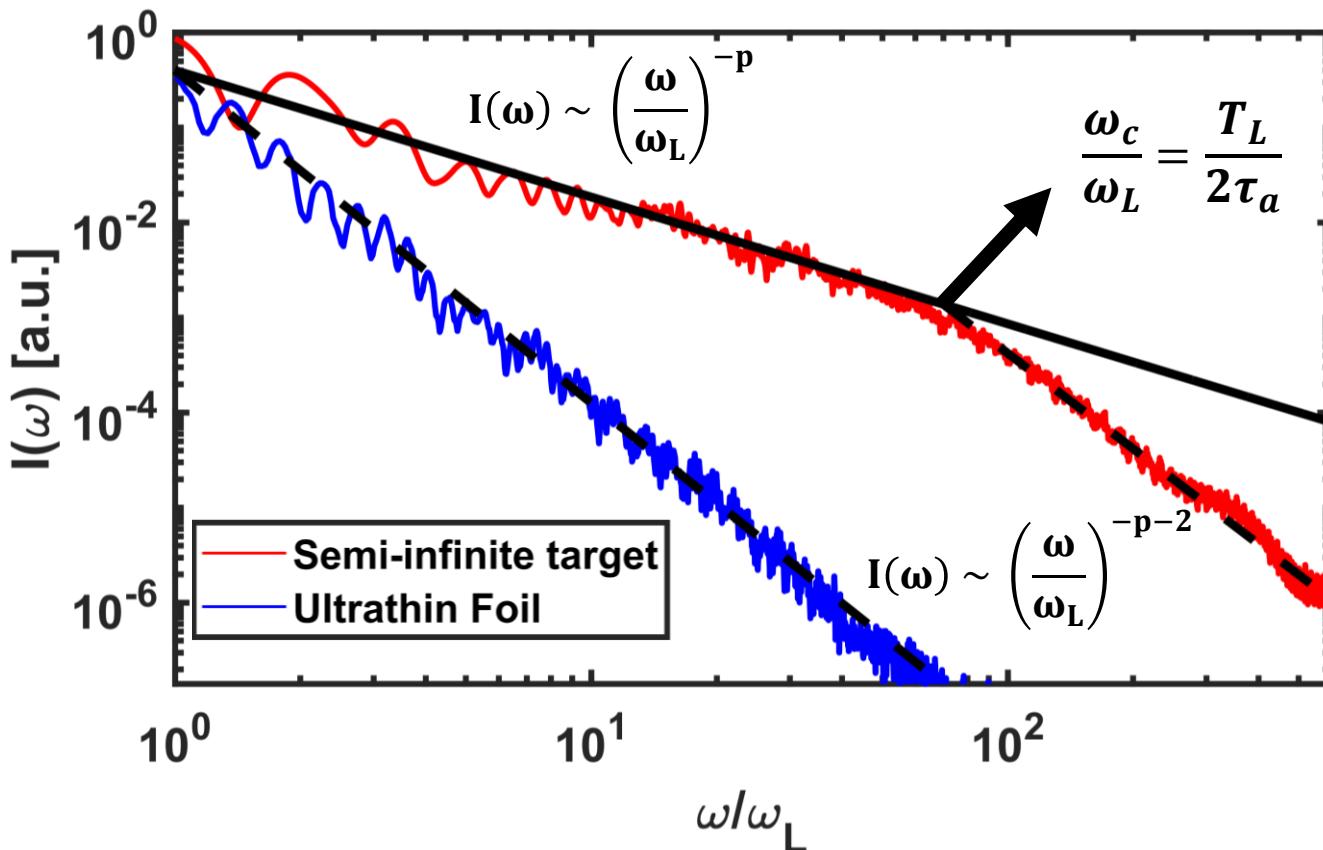
- Most individual electrons have a spectral scaling in agreement with the Coherent Synchrotron emission (CSE) model which predicts a power-law spectral scaling with an exponent of $-4/3$.
- However, the final spectrum obtained after superposition of all the emitting electrons is generally much steeper with a $-10/3$ power-law spectral scaling





Power-law scaling and the cut-off frequency

- Particle-in-cell simulations predict a power-law spectral scaling up until a cut-off frequency set by the finite duration of the emitting electron bunch [1-3]
- For ultrathin foils, the electron bunch width extends to several tenths of a laser period and dominates the spectral power law [4, 5]



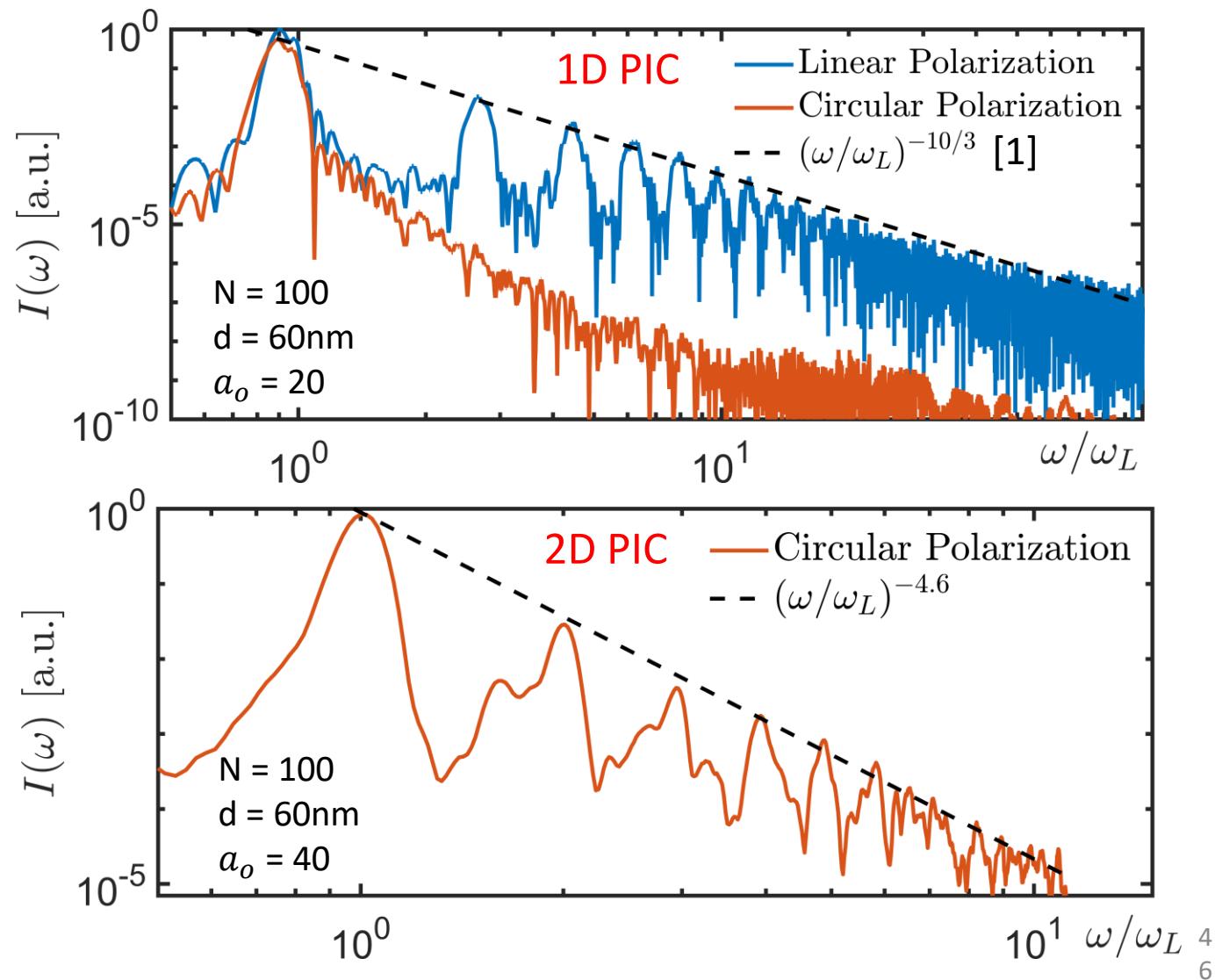
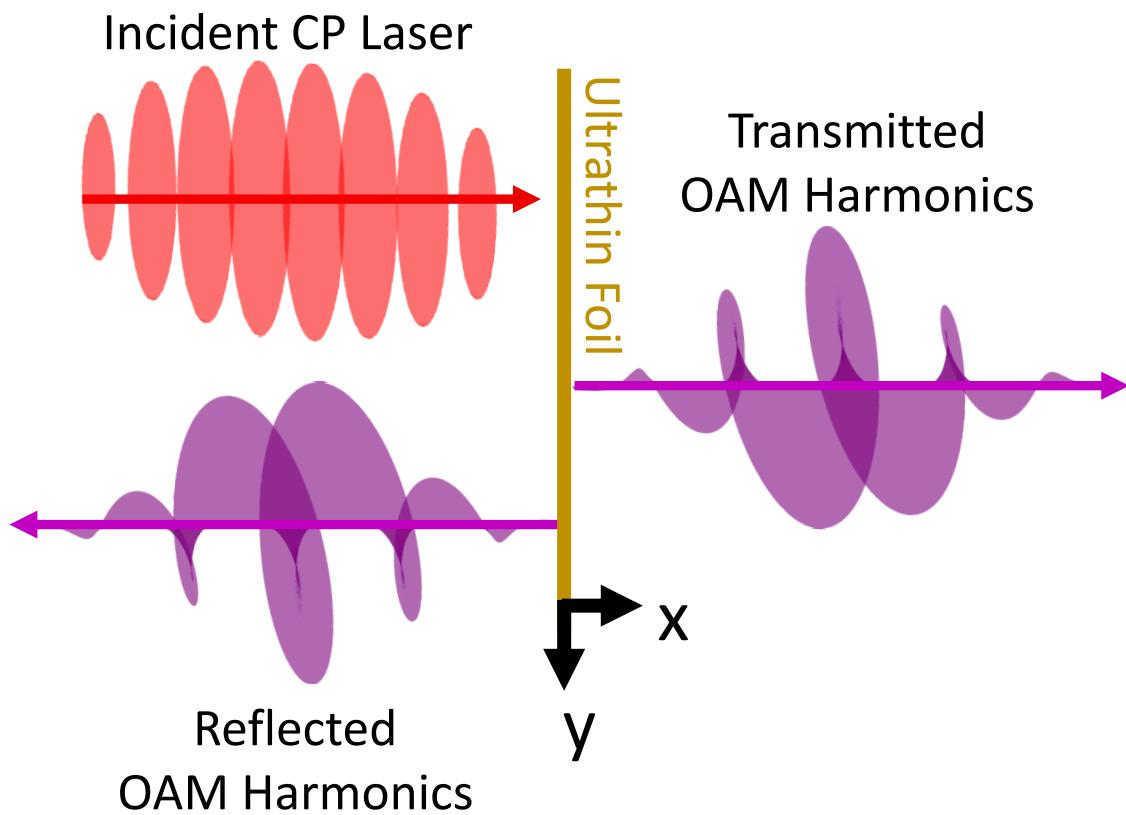
[1] D. an der Brügge et al. Phys. Plasmas, (2010).
[2] J. M. Mikhailova, et al. Phys. Rev. Lett. (2012).
[3] M. R. Edwards, et al., Sci. Rep. (2020).

[4] M. R. Edwards, N. M. Fasano, and J. M. Mikhailova, Phys. Rev. Lett. (2020).
[5] N. M. Fasano, M. R. Edwards, J. M. Mikhailova, CLEO (2020).



Ultrathin Foils Driven by Circularly Polarized (CP) Lasers

- For a 1D geometry, there is no harmonic generation for normally incident CP lasers since there is no fast oscillation of the plasma surface.



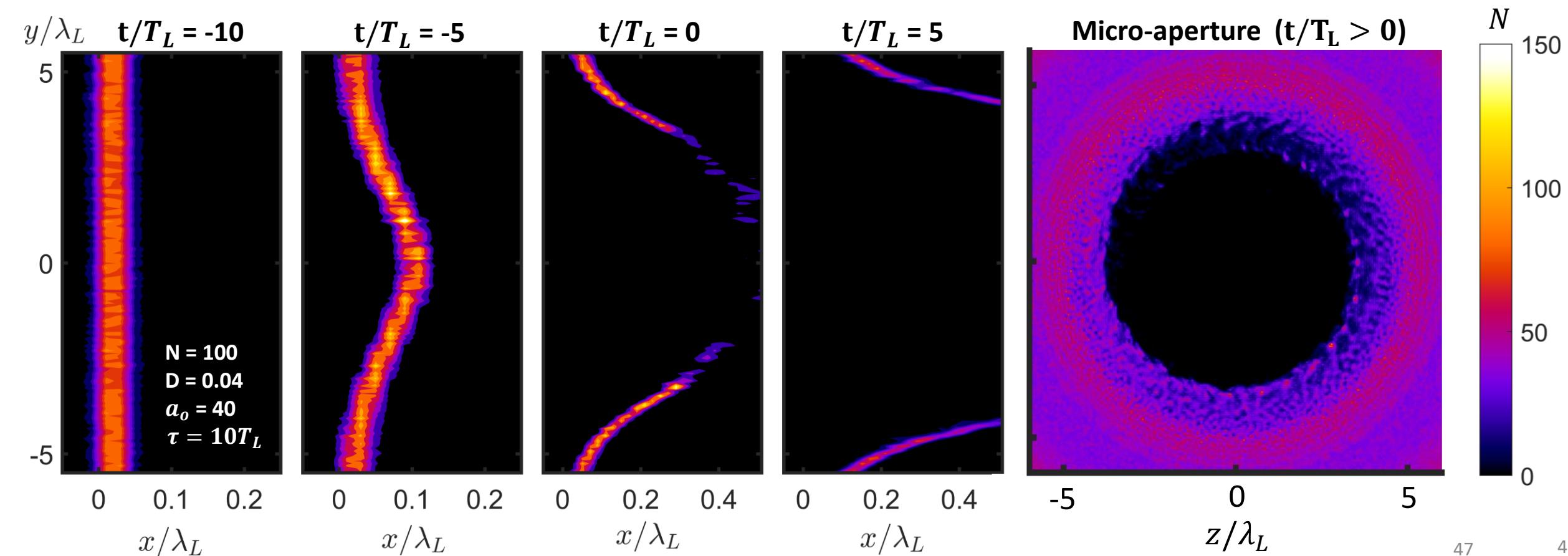


Plasma Dynamics from CP Driven Ultrathin Foils I

- 3D particle-in-cell simulations reveal the importance of transverse effects to shape the plasma target and allow for harmonic generation.
- The laser's pressure leads to plasma **surface denting** [1], and eventually drills a **micron-sized aperture** [2].

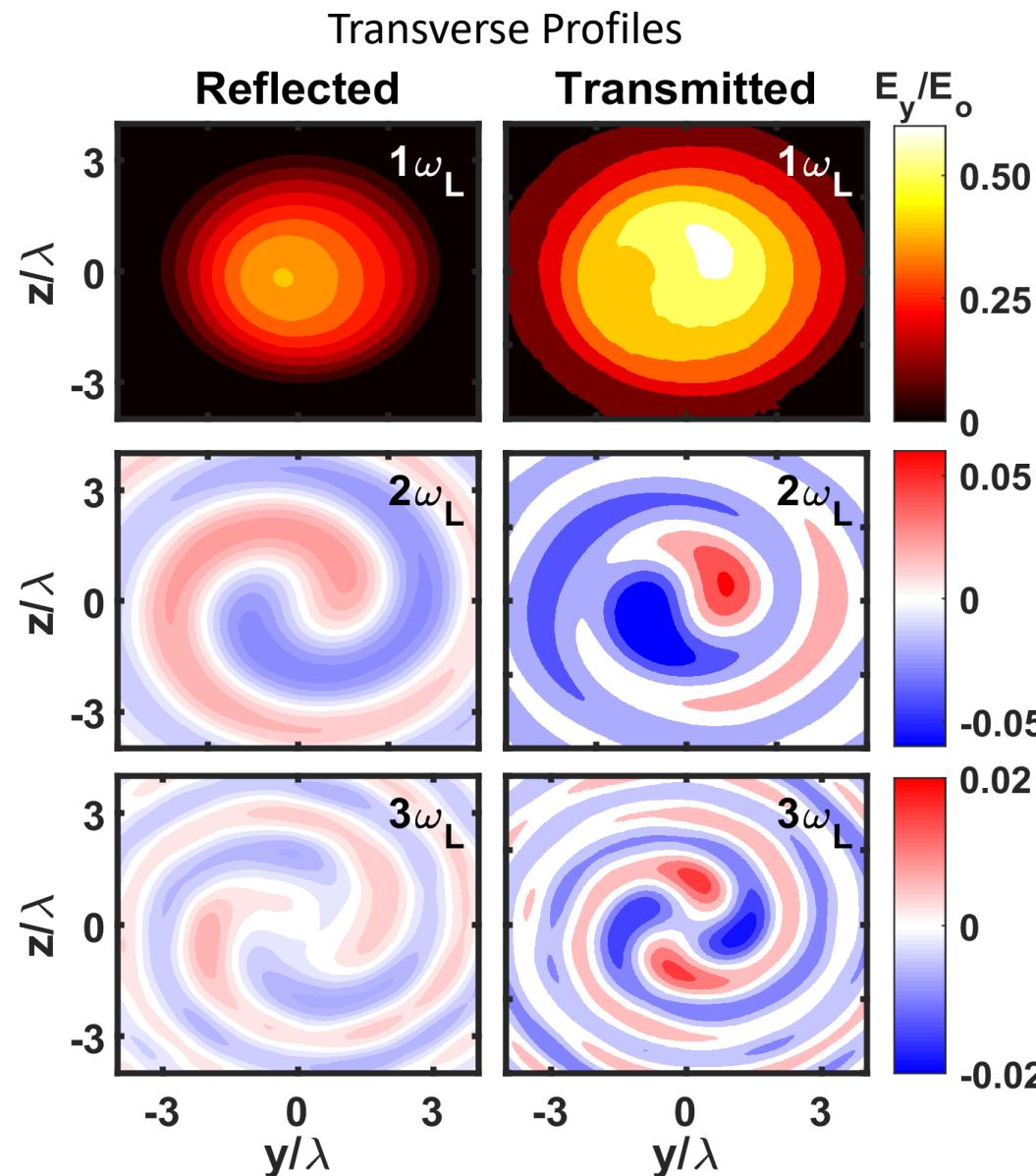
[1] J. W. Wang, M. Zepf, and S. G. Rykovanov, Nat. Commun. 10, 5554 (2019).

[2] L. Yi, Phys. Rev. Lett. 126, 134801 (2021).

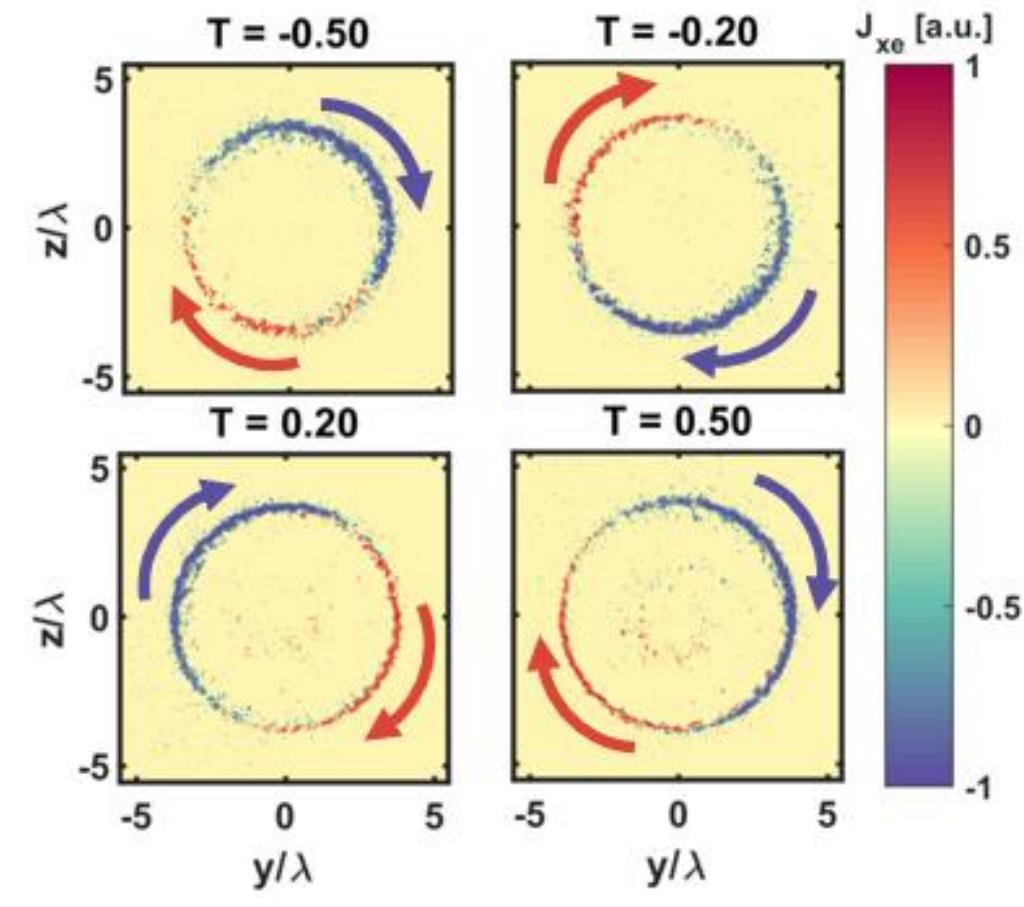




Conversion of spin angular momentum to orbital angular momentum Facilitated by Electron Currents

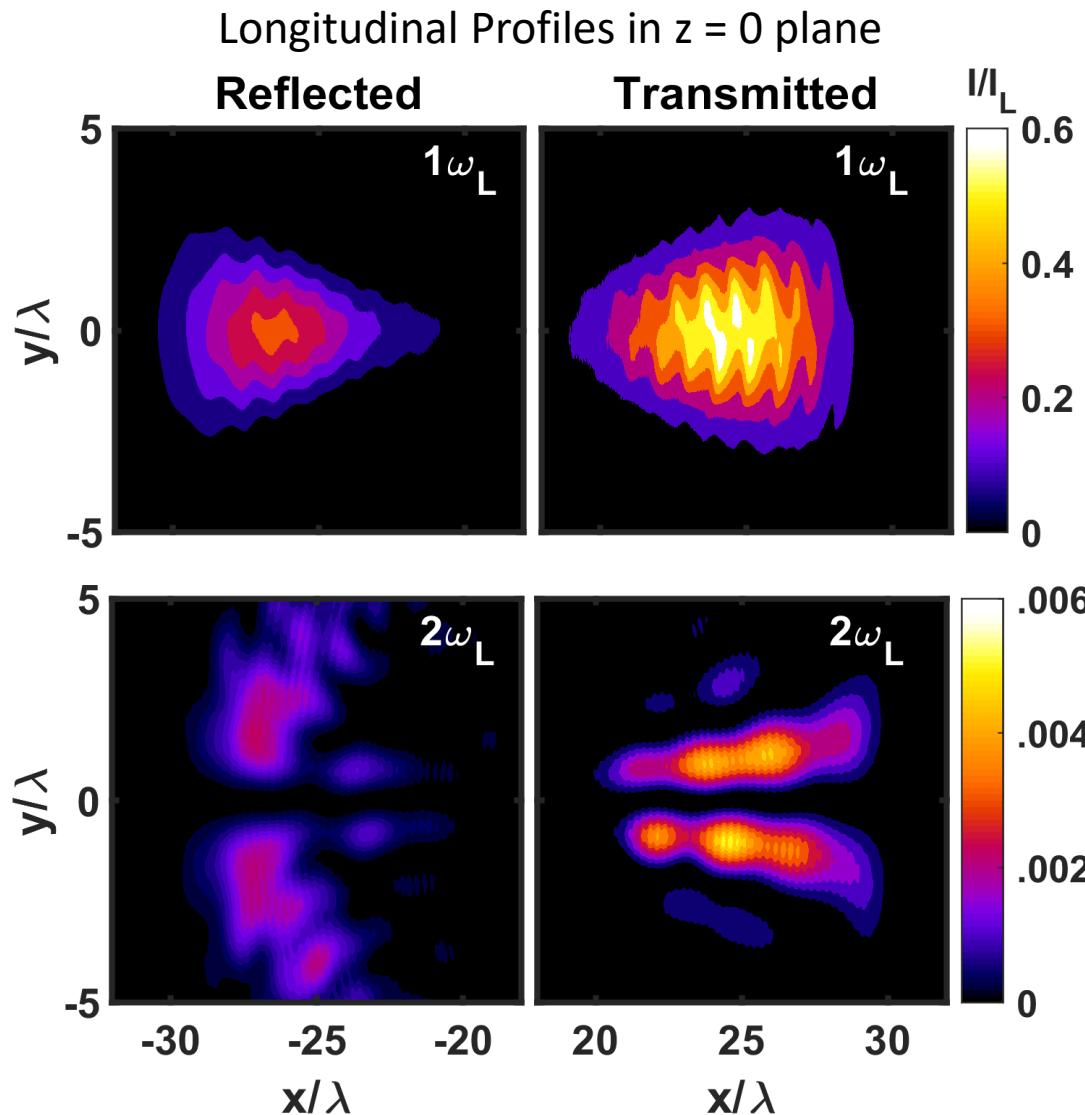


- Generation of vortex wavefronts is related to the angularly-dependent longitudinal currents that rotate with time $\exp(-il\phi)$





High-Order Spatial Structure in the Generated Harmonics



- Fundamental beam still has a Gaussian profile while the generated harmonics contain higher-order transverse structure
- Vortex beams are characterized with a **null on-axis intensity and spiral phase fronts.**

$$E(\rho, \phi, x) \sim \rho \exp(-il\phi)$$

l = topological charge

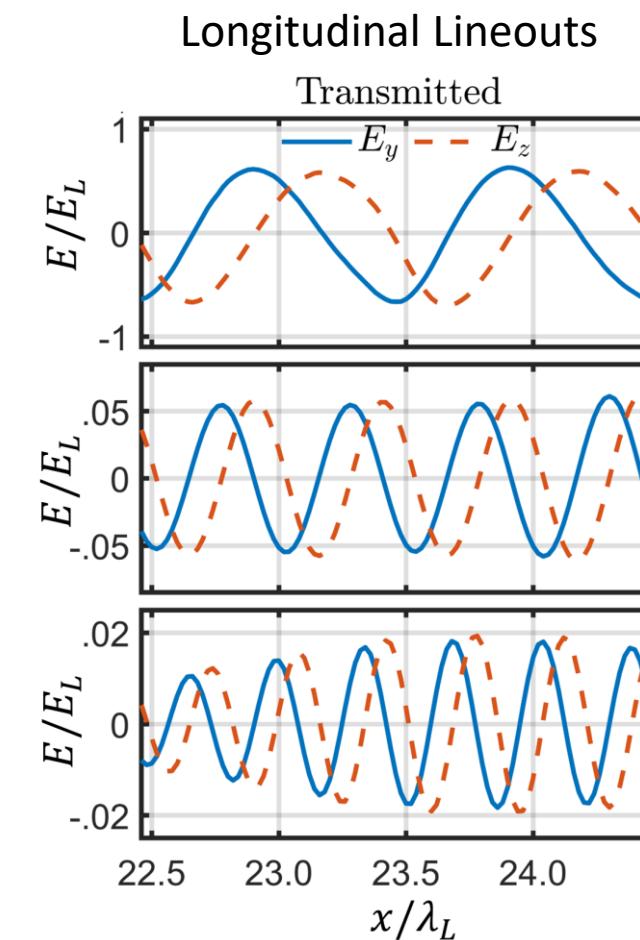
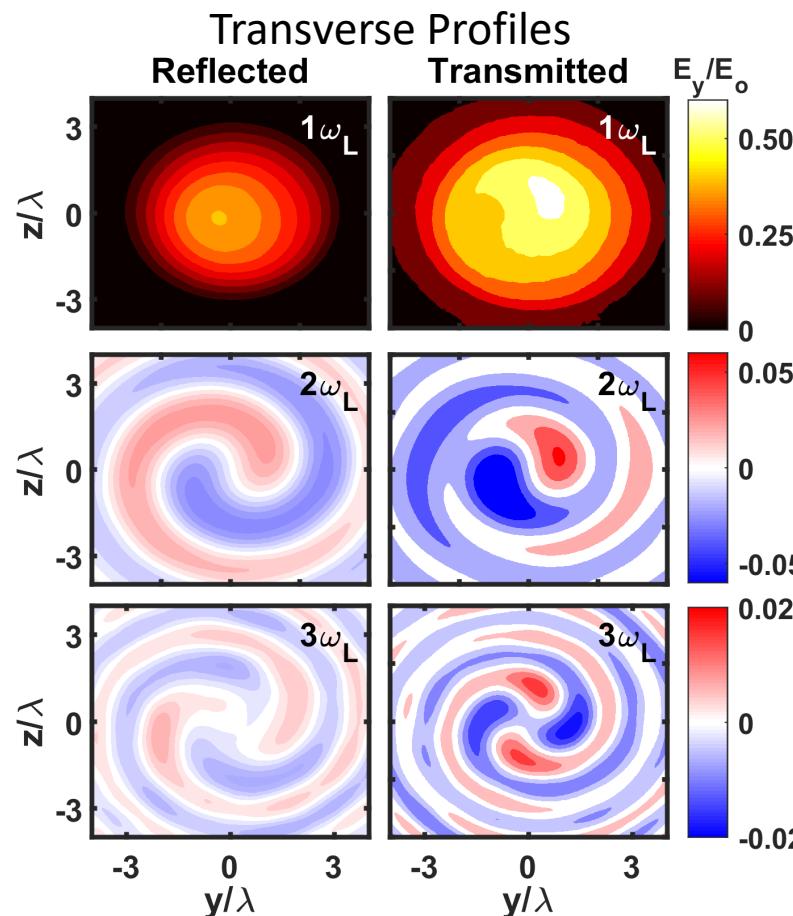
ϕ = azimuthal angle

$$\rho^2 = y^2 + z^2$$



Helical Phase Fronts in the Generated Harmonics

- The transmitted and reflected fundamental remains Gaussian in the transverse plane
- The n^{th} order harmonic has a topological charge of $l = (n-1)$ $\exp(-il\phi)$
- All three harmonics remain circularly polarized

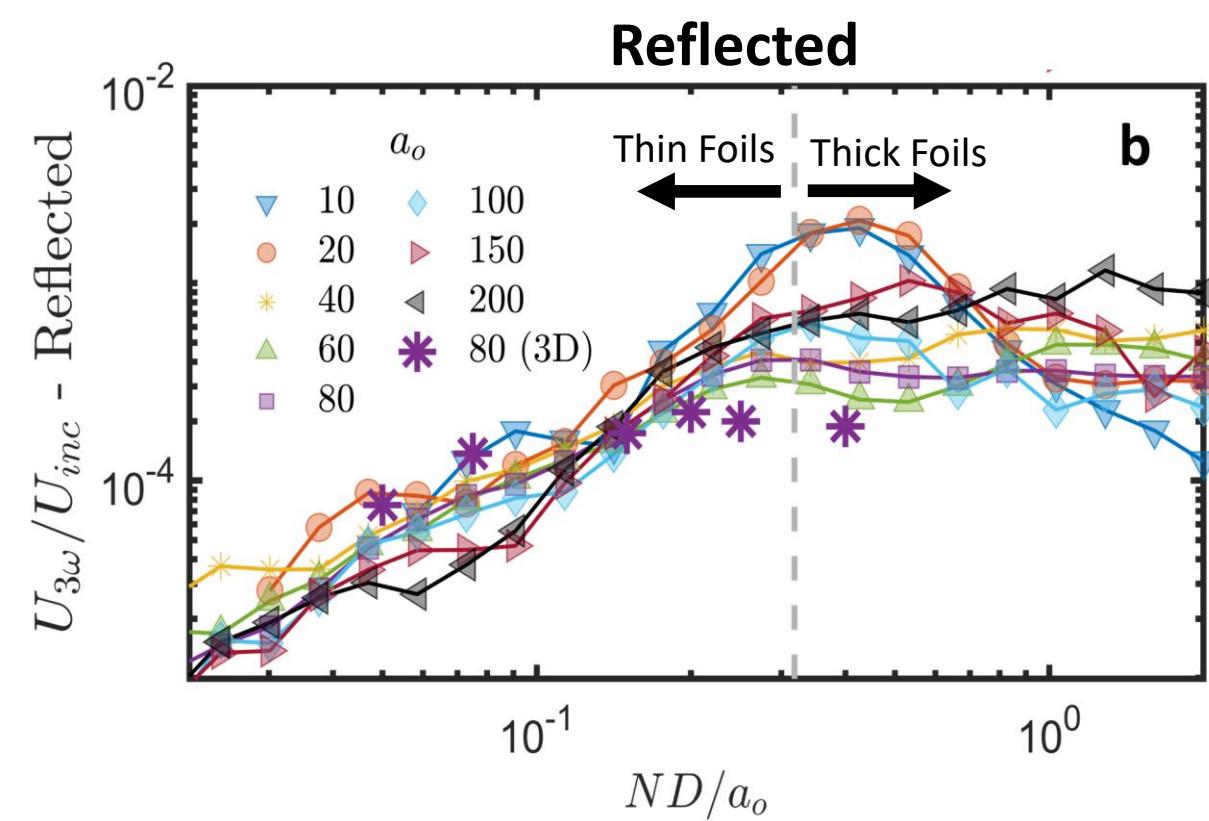
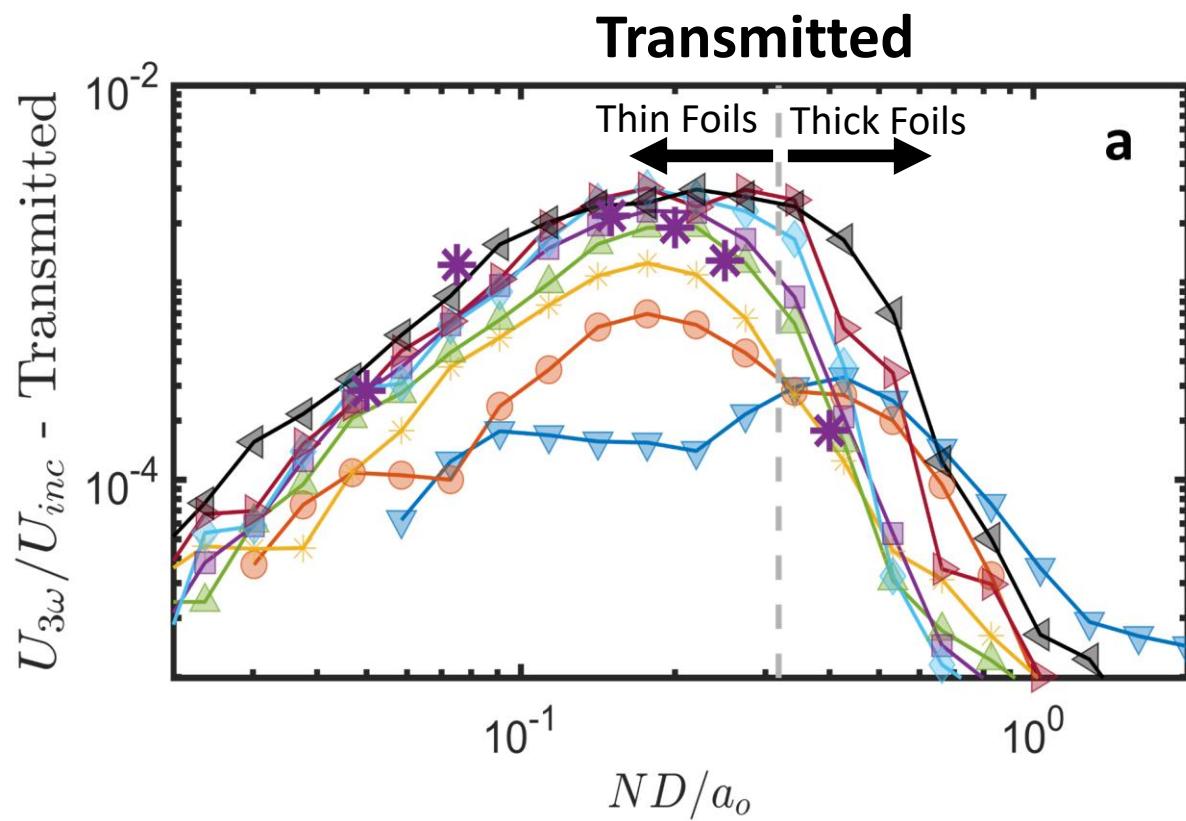




Dependence on the Laser and Plasma Parameters (ND/a_0)

- Two-dimensional geometry allows for transverse effects but at a lower computational cost permitting higher resolution and finer parameter scans.

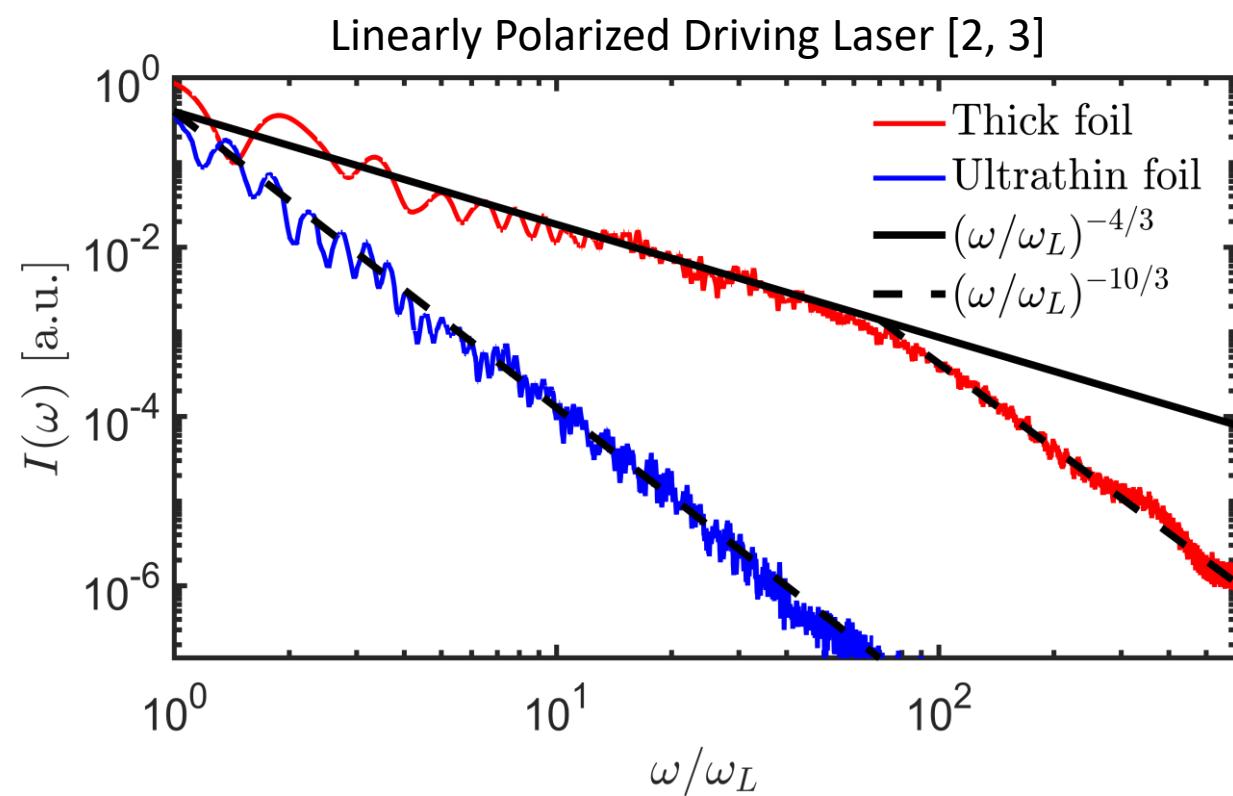
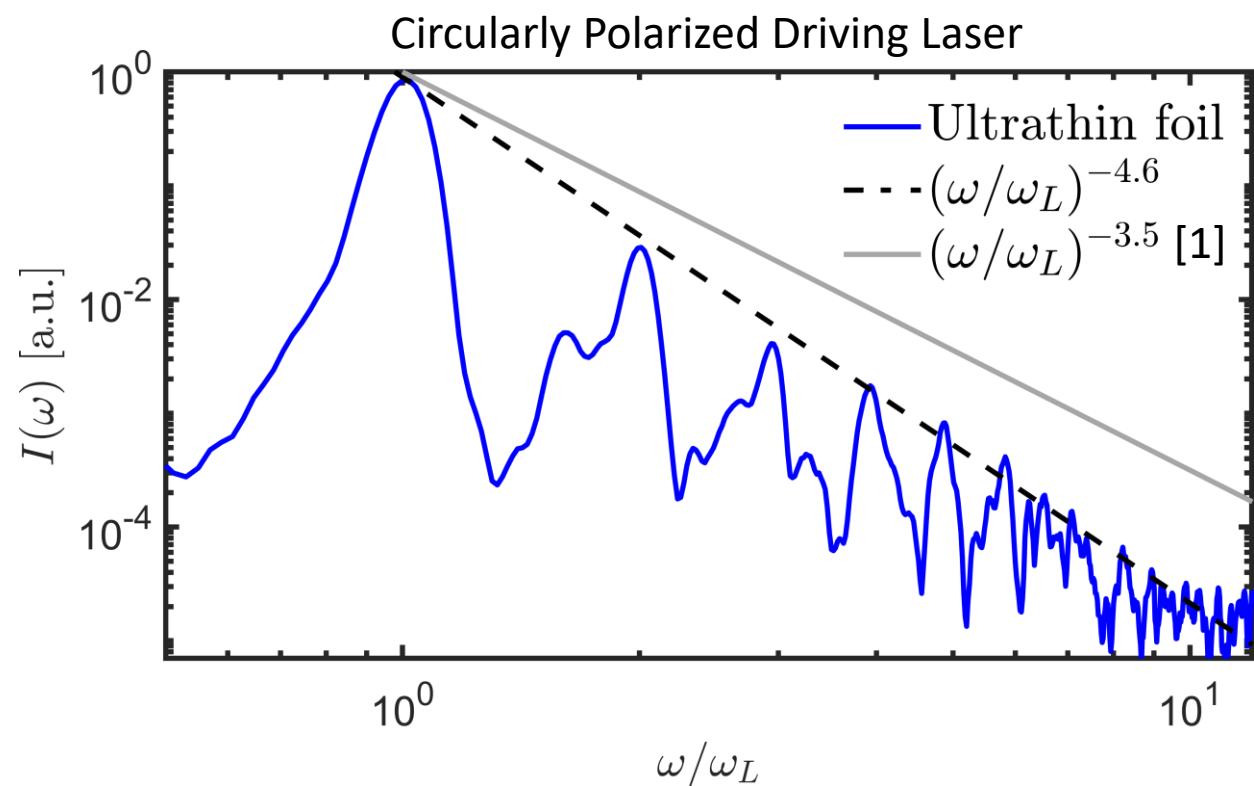
$N = 100$
 $D = 0.005 - 1.5$
 $d = 4 - 1,200\text{nm}$





Spectral Power-Law Scaling

- Ultrathin foils driven by circularly polarized lasers at normal incidence provide comparative power-law exponents to that of micro-apertures driven by circularly polarized light and ultrathin foils driven by linearly polarized light.



[1] L. Yi, Phys. Rev. Lett. 126, 134801 (2021). [micro-apertured target]

[2] M. R. Edwards, N. M. Fasano, and J. M. Mikhailova, Phys. Rev. Lett. (2020).

[3] M. R. Edwards, J.M. Mikhailova, Sci Rep **10**, 5154 (2020).



Summary

- Transverse effects such as plasma surface denting and micro-aperture formation allows for the generation of harmonics when an ultrathin foil is irradiated with a circularly polarized laser at normal incidences.
- The laser-plasma dynamics facilitates the generation of vortex harmonics which possess orbital angular momentum.
- Tuning of laser and plasma parameters allows for this geometry to reach comparable efficiencies as for linearly polarized driven ultrathin foils and circularly polarized driven micro-apertured foils.

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Program of Plasma Science
and Technology PRINCETON
UNIVERSITY



Thank you for your attention!

Laguerre-Gauss Beam Definition

- Written in cylindrical coordinates with the x-axis being the propagation direction

$$E_p^L(\rho, \phi, x) = \sqrt{\frac{2p!}{\pi(|L| + p)!}} \frac{w_o}{w(x)} \left(\frac{\rho\sqrt{2}}{w(x)} \right)^{|L|} \exp\left(-\frac{\rho^2}{w^2(x)}\right) L_p^{|L|}(\Gamma) \exp\left(-i\left(\frac{k\rho^2}{2R(x)} + L\phi - N\psi(x)\right)\right)$$

$$\rho^2 = y^2 + z^2$$

$$w(x) = w_o \sqrt{1 + \left(\frac{x}{x_R}\right)^2}$$

$$\psi(x) = \text{atan}\left(\frac{x_R}{x}\right)$$

$$N = |L| + 2p + 1$$

$$\phi = \text{atan}\left(\frac{z}{y}\right)$$

$$R(x) = x \left(1 + \left(\frac{x_R}{x}\right)^2\right)$$

$$x_R = \frac{\pi w_o^2}{\lambda}$$

$$\Gamma = \frac{2\rho^2}{w^2(x)}$$

First Few Generalized Laguerre Polynomials

$$L_p^0 = 1$$

$$L_p^1 = -\Gamma + (p + 1)$$

$$L_p^2 = \frac{1}{2}\Gamma^2 - \Gamma(p + 2) + \left(\frac{1}{2}p^2 + \frac{3}{2}p + 1\right)$$

$$L_p^3 = -\frac{\Gamma^3}{6} + \Gamma^2 \left(\frac{P}{2} + \frac{3}{2}\right) - \Gamma \left(\frac{P^2}{2} + \frac{5P}{2} + 3\right) + \left(\frac{P^3}{6} + P^2 + \frac{11P}{6} + 1\right)$$

References:

1. A. E. Siegman, Lasers (1986).
2. R. Aboushelbaya, K. Glize, A. F. Savin, et al. Physics of Plasmas **27**, 053107 (2020).
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