



Laboratory for
Ultra Fast Optical Science (UFOs)



Strong THz generation from air plasma using a long wavelength laser

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& **APPLIED PHYSICS**



physics
University of Maryland

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Project goals

- ❑ Study of THz/microwave emission from 10 μm filamentation
 - Investigate THz/microwave generation mechanisms (single-color, two-color, 10.3 μm + 10.6 μm mixing schemes)
 - High-power THz/microwave generation

- ❑ Development of THz/microwave detection schemes
 - THz/microwave characterization (energy, spectrum, polarization,...)
 - Single-shot THz/microwave spectroscopy

- ❑ Characterization of CO₂ laser produced air filaments
 - THz/microwave radiation spectral analysis
 - Plasma density measurement with a B-dot probe
 - Time-resolved THz spectroscopy with a femtosecond laser

Outline

- ❑ Strong THz field generation
 - Two-color laser mixing

- ❑ THz/harmonics generation at long wavelengths
 - 1% laser-to-THz conversion efficiency
 - Coherent control of ionization, THz, and harmonics

- ❑ Future experiments & simulation
 - mJ-level THz generation
 - Accurate phase-dependent measurements
 - UPPE simulation

- ❑ Summary

Strong THz field generation:

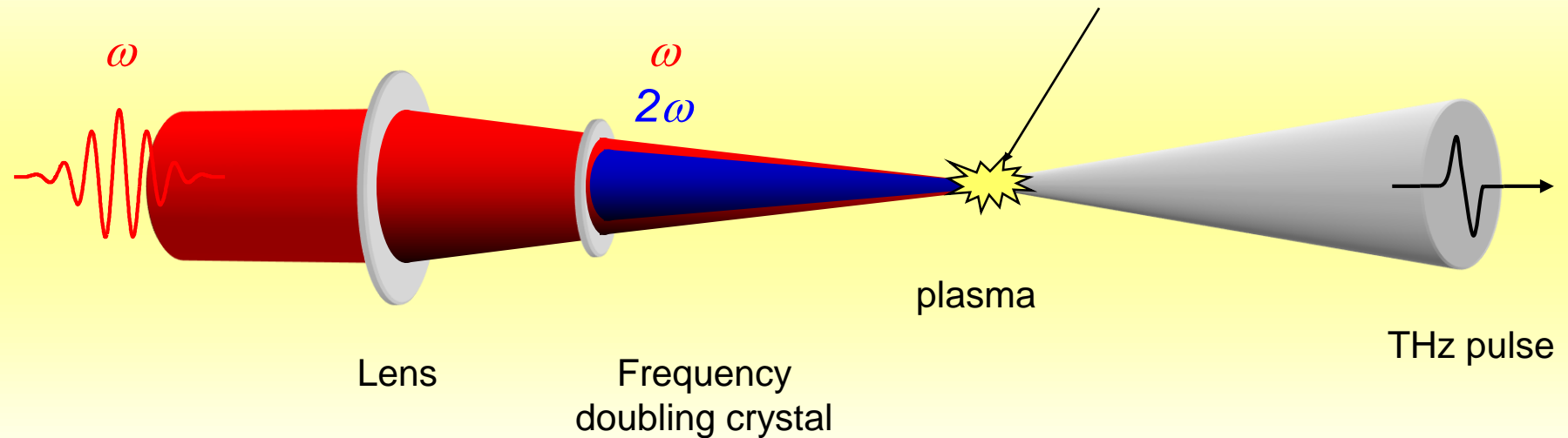
Two-color laser mixing

THz generation via two-color laser mixing:

Air

Four-wave mixing

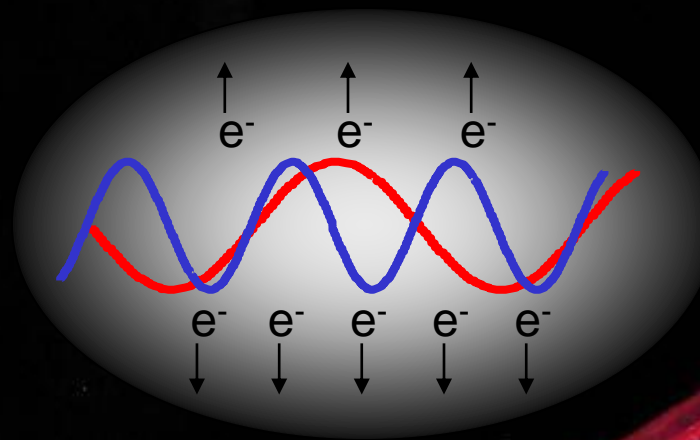
$$\omega_{\text{THz}} = \omega + \omega - 2\omega \quad ?$$



$$\mathbf{P}(t) = \varepsilon_0 \left(\chi^{(1)} \mathbf{E}(t) + \chi^{(2)} \mathbf{E}^2(t) + \chi^{(3)} \mathbf{E}^3(t) + \dots \right)$$

THz generation mechanism:

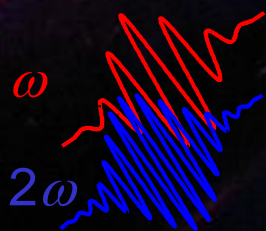
Plasma current model*



Directional quasi-DC current

THz

Current surge
→ THz generation



BBO crystal

ω

*K. Y. Kim *et al.*, Nature Photonics **2**, 605 (2008).

K. Y. Kim *et al.*, Optics and Photonics News **19**, 49 (2008).

Plasma current model (semiclassical model):

Laser field

$$E_L(t) = \underbrace{E_1 \cos(\omega t)}_{\omega \text{ field}} + \underbrace{E_2 \cos[2\omega t + \theta]}_{2\omega \text{ field}}$$

θ : relative phase

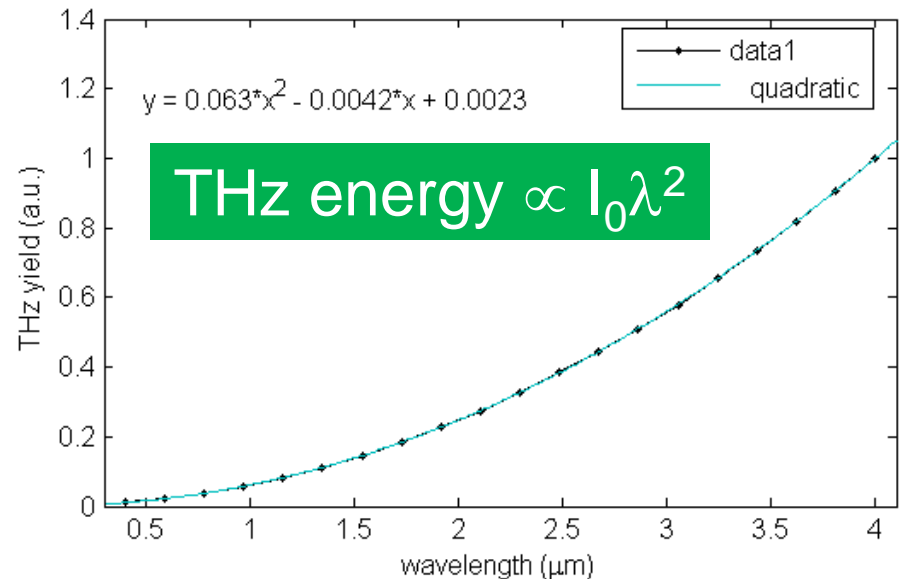
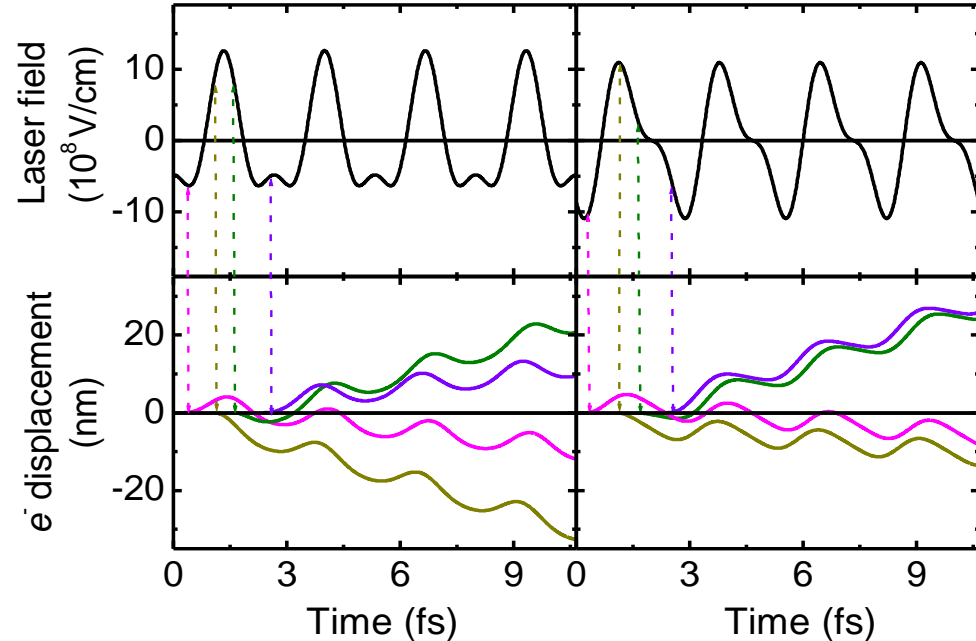
Electron drift velocity

$$v_d = \frac{eE_1}{m_e \omega} \sin \phi + \frac{eE_2}{2m_e \omega} \sin(2\phi + \theta)$$

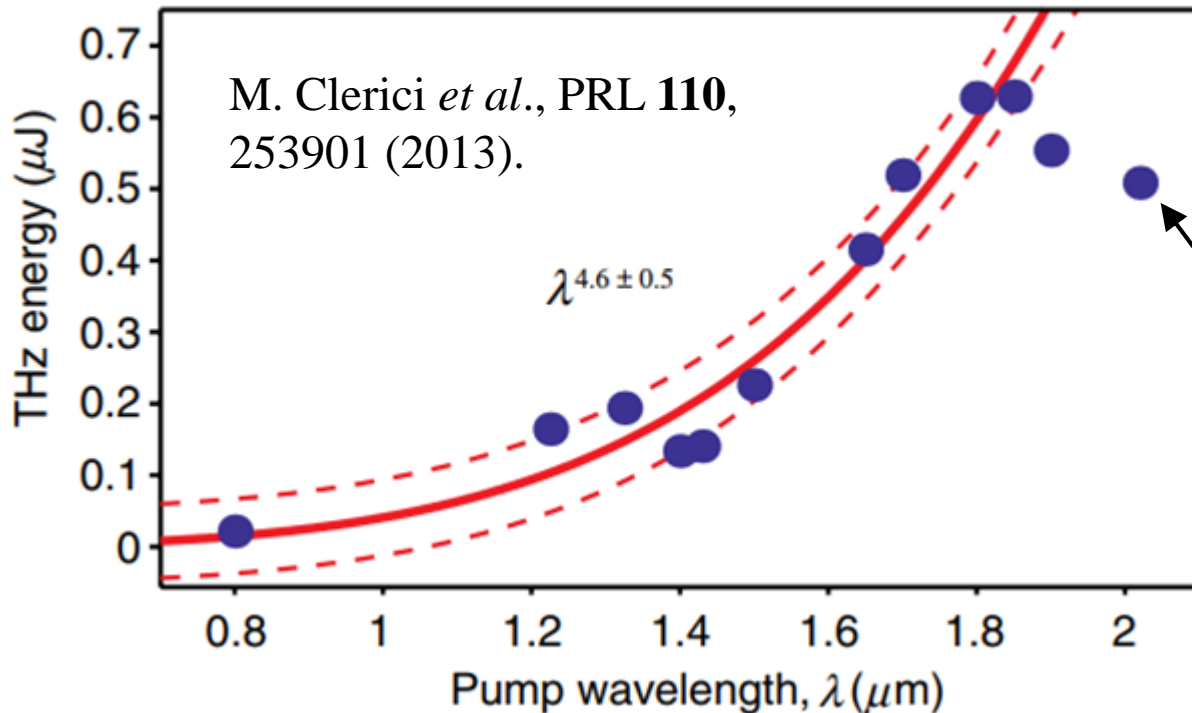
electron freed at ϕ

$\theta = 0$

$\theta = \pi/2$



Wavelength scaling with two-color mixing



Due to laser peak intensity drop

THz energy scaling:

Need more studies!

Plasma current ($\sim \lambda^2$)

Plasma length & radius ($\sim \lambda$)

Peak intensity ($\sim \lambda^{-2}$)

Longitudinal current $J_z^{(2)}$ ($\sim \lambda^4$)

Transverse current $J_x^{(3)}$ ($\sim \lambda^6$)

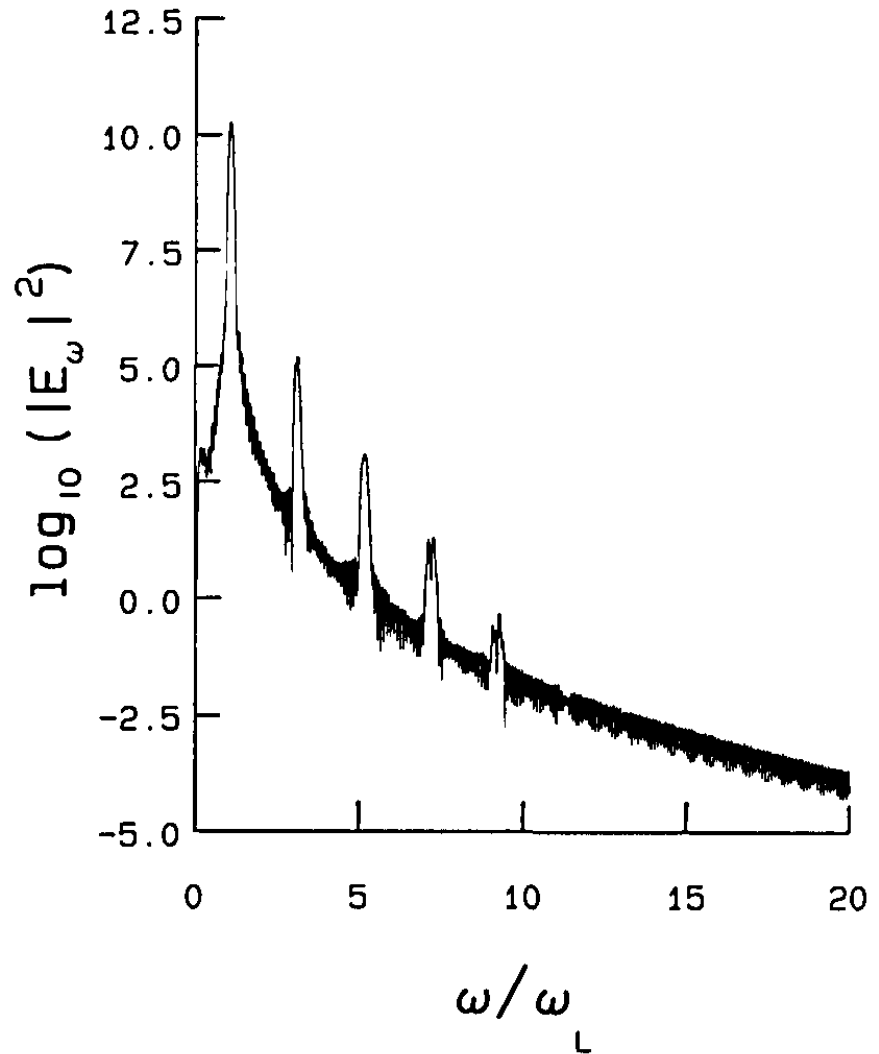
Brunel radiation*

- Brunel proposed harmonic generation due to plasma effects in a gas undergoing ionization
- Bound-free transition (plasma current) produces harmonics
- THz = 0th order Brunel radiation
- Explains lower-order harmonics

(N. H. Burnett *et al.*, PRA **51**, R3418 (1995))

- Higher-order harmonics due to recollisions

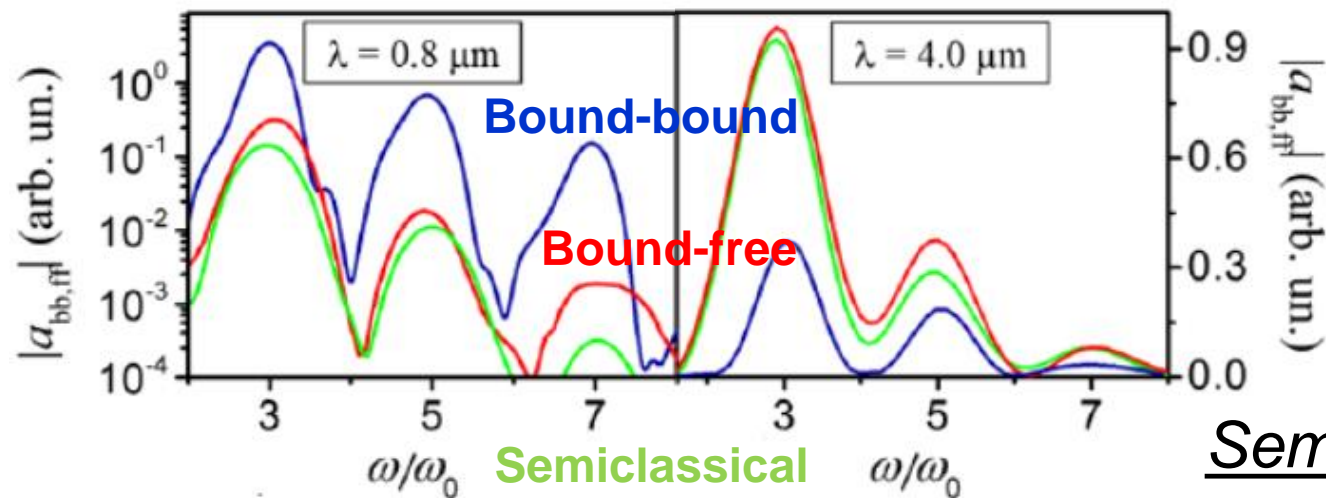
(P. B. Corkum, PRL **71**, 1994 (1993))



*F. Brunel J. Opt. Soc. Am. B **7**, 521 (1990)

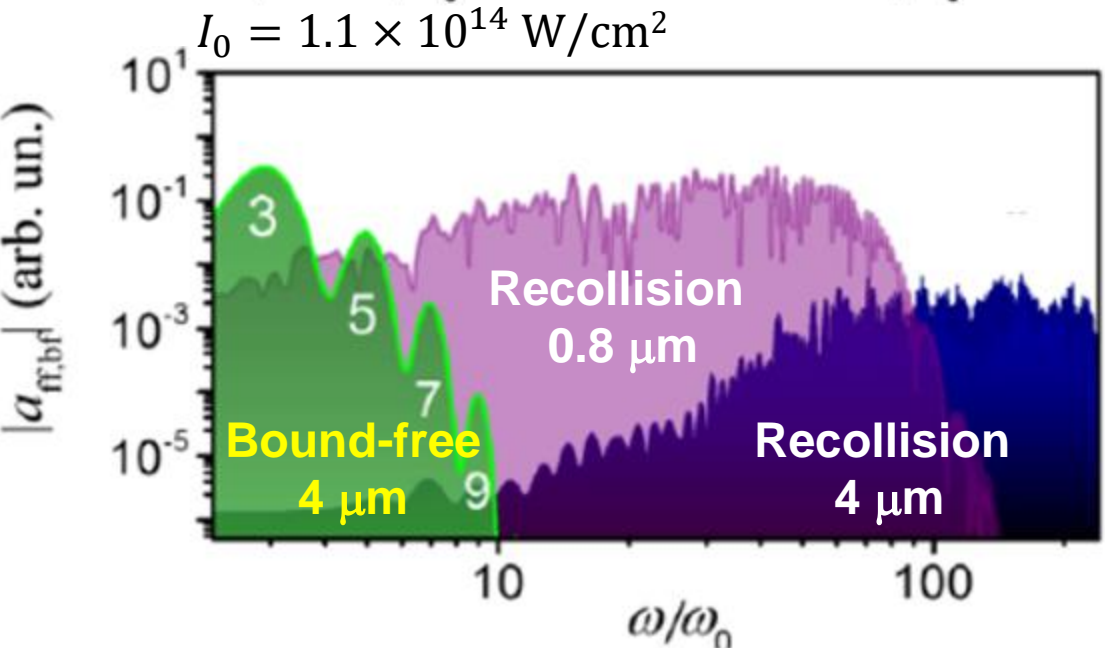
Contributions from bound vs free electrons

Time-dependent Schrödinger equation (TDSE)



At long wavelengths

Bound-free
 \downarrow
Bound-bound



Semiclassical plasma current (Brunel radiation)
dominant

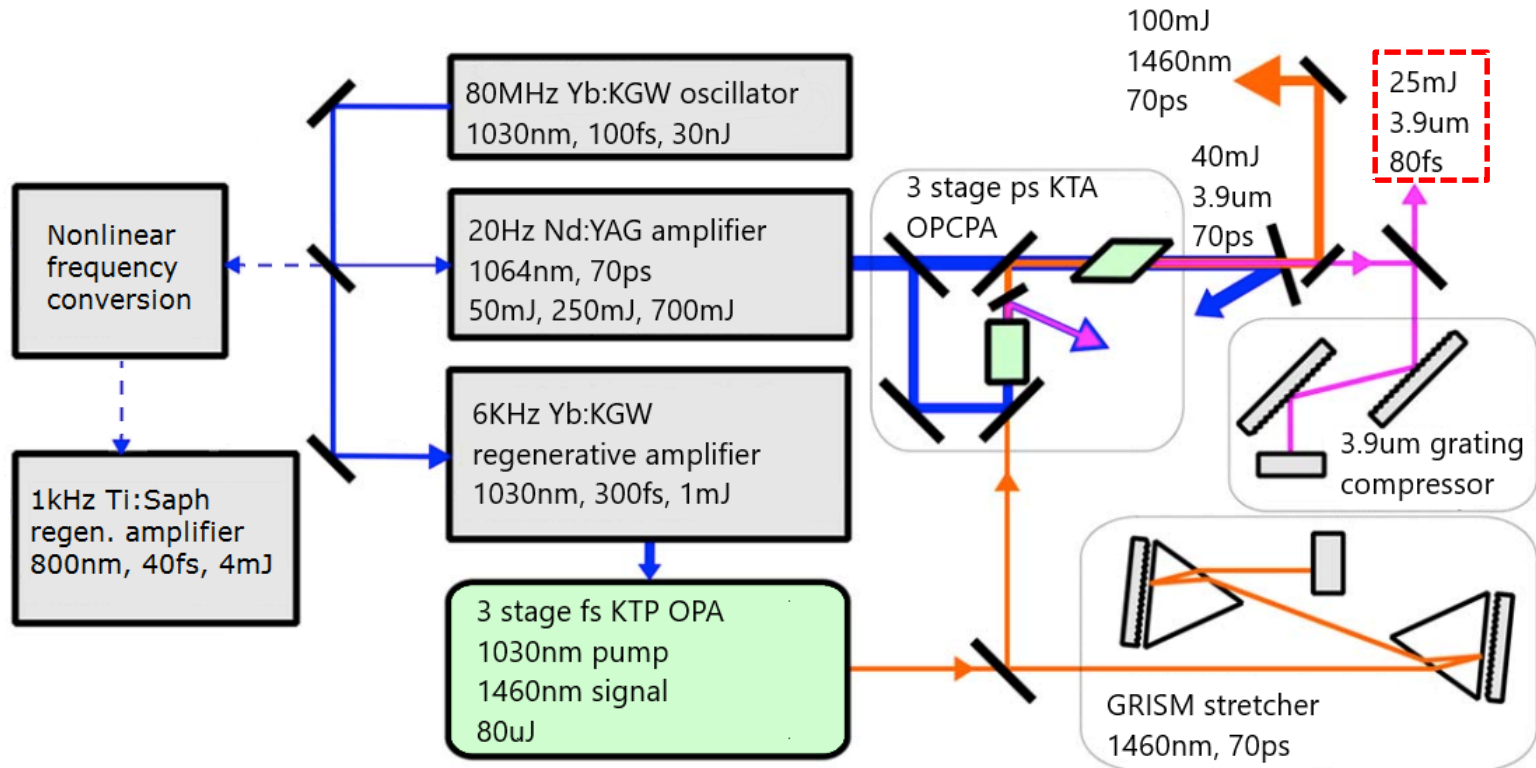
Bound-free
 \downarrow
Free-bound (Recollision)

THz & harmonics generation:

Experiment

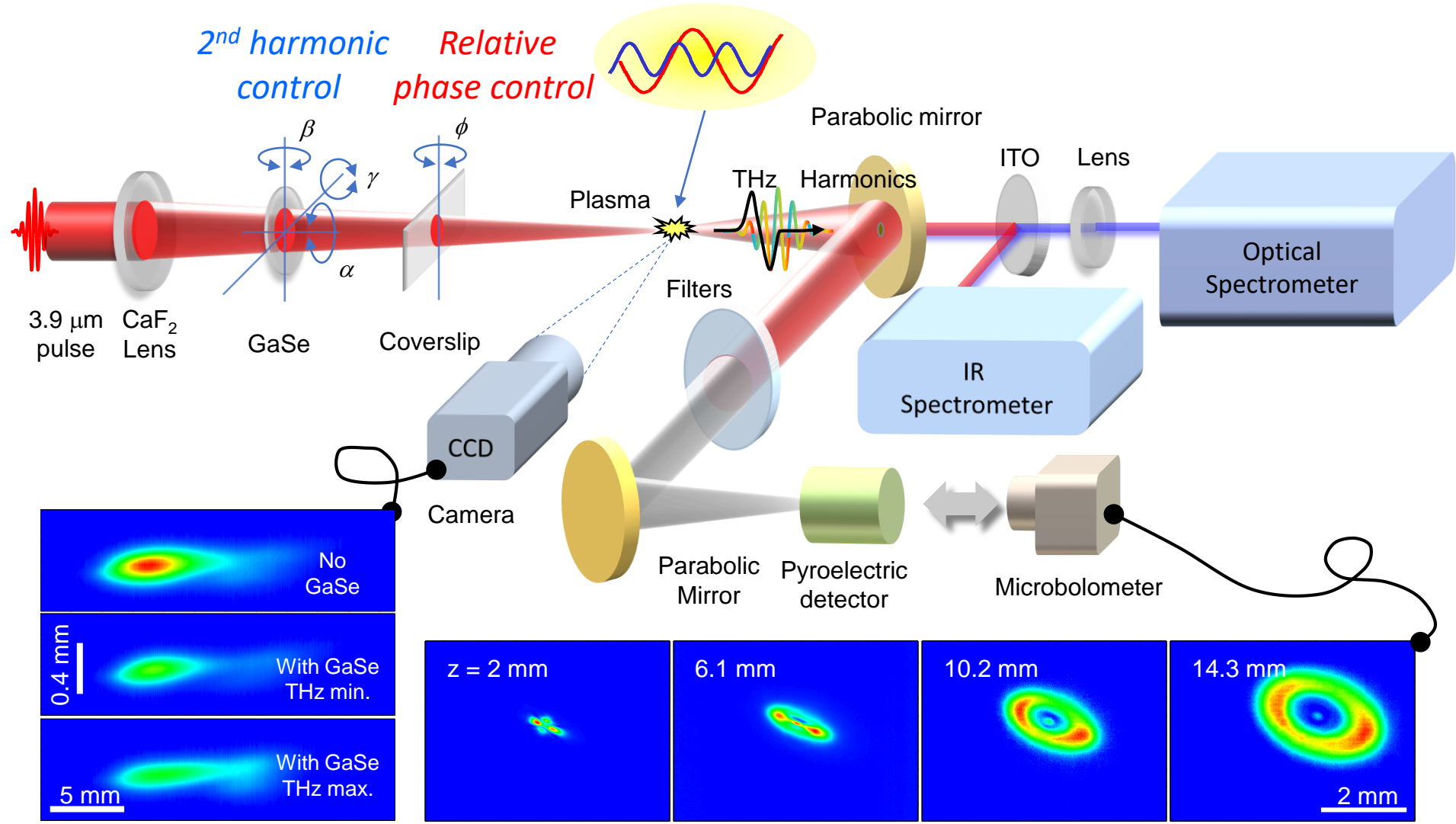
OPCPA laser at UMD ($\lambda = 3.9 \mu\text{m}$)

Optical Parametric Chirped-pulse Amplification (OPCPA)



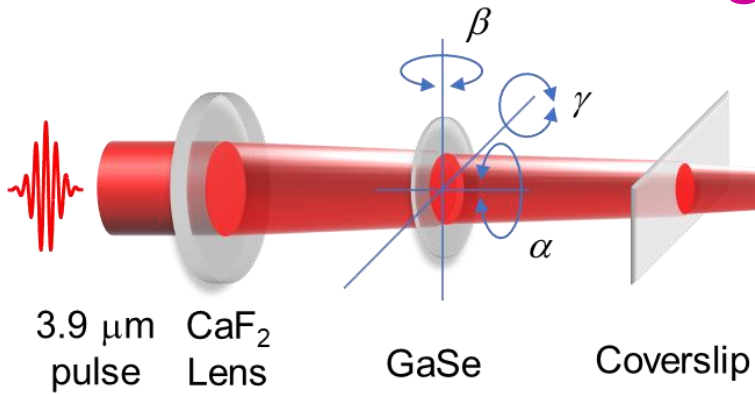
- High harmonic generation: T. Popmintchev et al., *Science* **336**, 1287 (2012).
- Mid-IR filamentation in air: A. V. Mitrofanov et al., *Sci. Rep.* **5**, 8368 (2015).
- Plasma wakefield acceleration: D. Woodbury et al., *Opt. Lett.* **43**, 1131 (2018).

Experimental setup

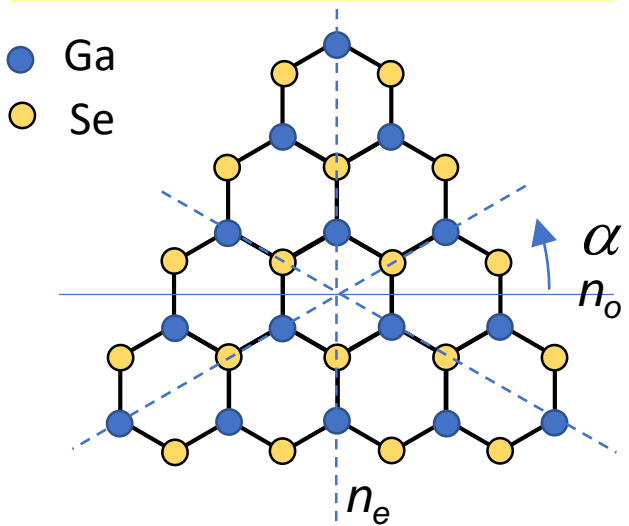


- Measured phase dependent THz, harmonics, and plasma fluorescence

Second harmonic generation in GaSe

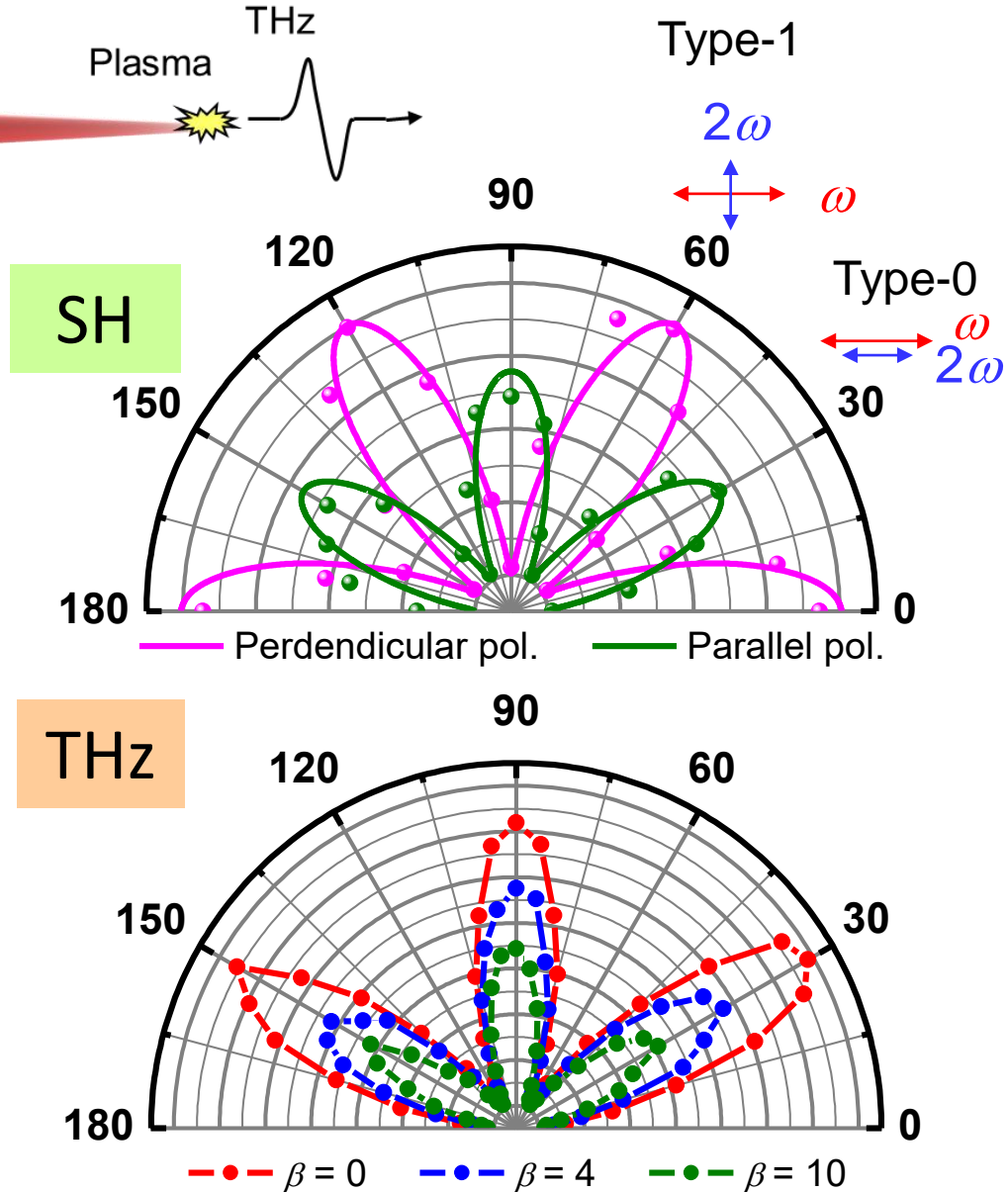


GaSe (layered material)

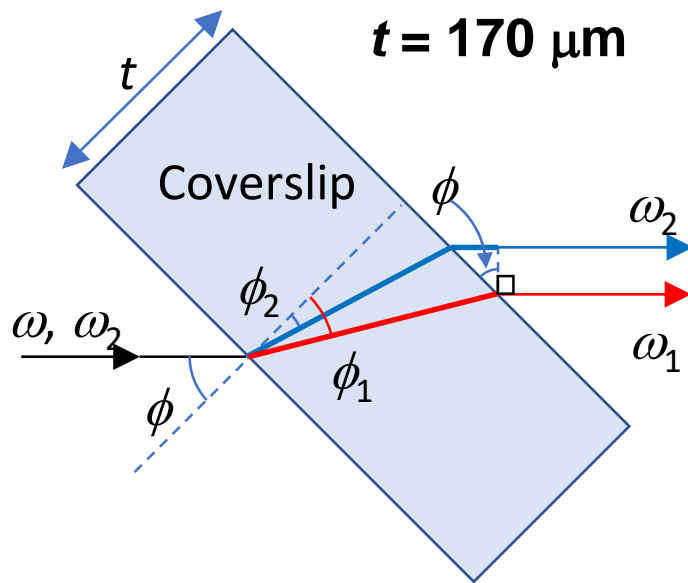


Six-fold rotational symmetry

Max. THz generation from colinear and ω and 2ω



Relative phase θ control



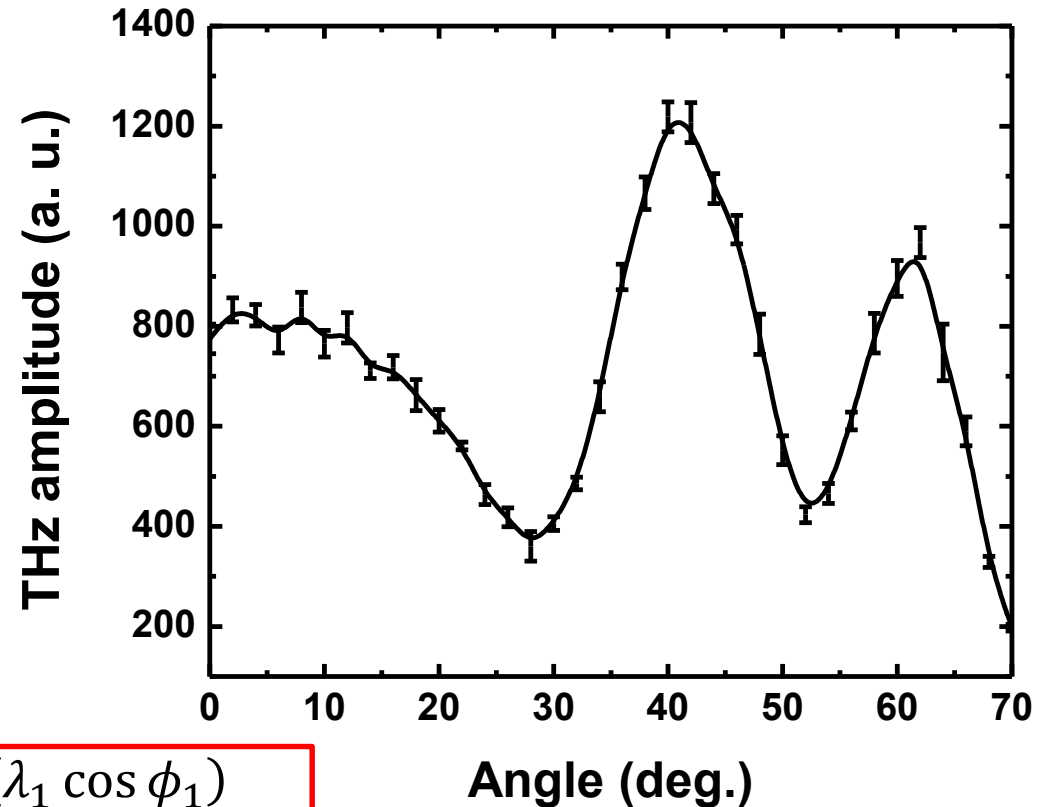
$$\phi_1 = \sin^{-1}(\sin \phi / n_1)$$

$$\phi_2 = \sin^{-1}(\sin \phi / n_2)$$

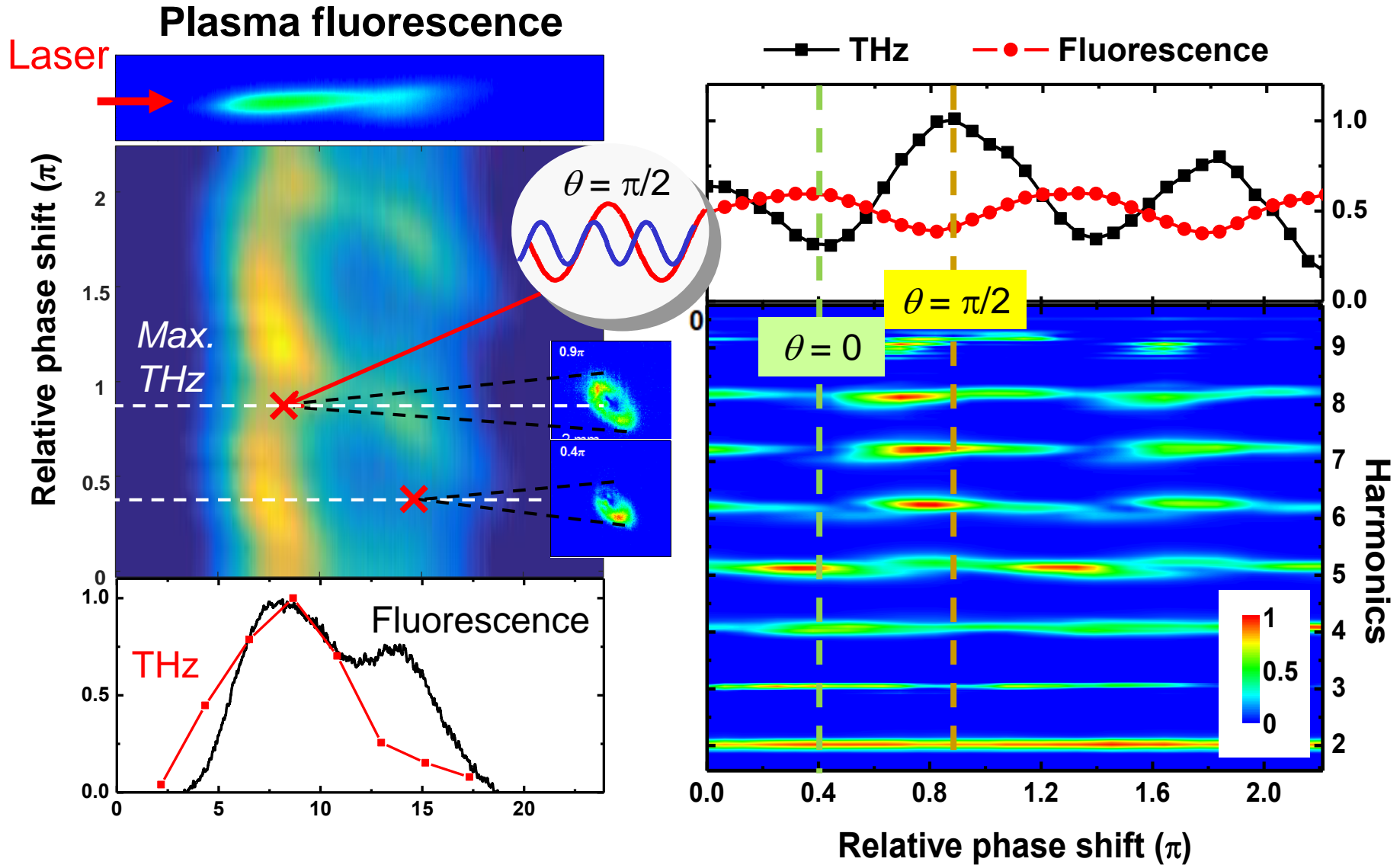
$$\theta = 2\pi t \left\{ n_2 / (\lambda_2 \cos \phi_2) - 2n_1 / (\lambda_1 \cos \phi_1) + \sin \phi (\tan \phi_1 - \tan \phi_2) / \lambda_2 \right\}$$

- Laser energy loss $\approx 10\%$ at $\phi = 0^\circ$
- Group velocity walk-off ≈ 20 fs at $\phi = 42^\circ$
- Transverse beam separation $\approx 1 \mu\text{m}$ at $\phi = 42^\circ$

THz yield vs coverslip angle ϕ



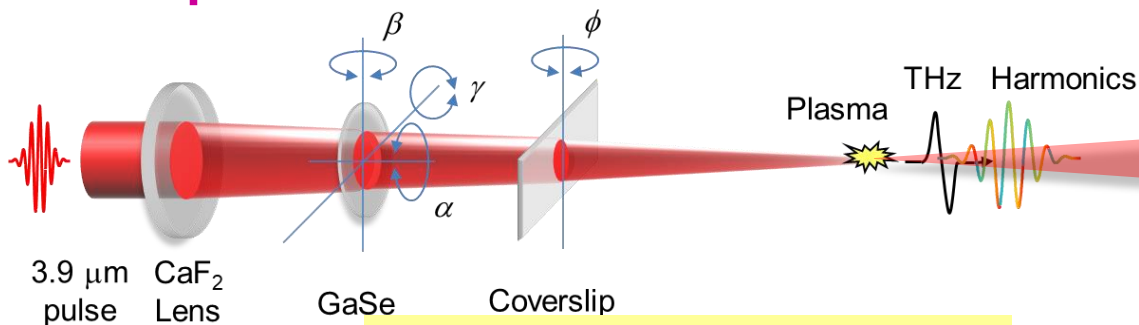
Phase dependent ionization, THz, & harmonics



At $\theta = \pi/2$, less ionization but more THz expected from plasma current model

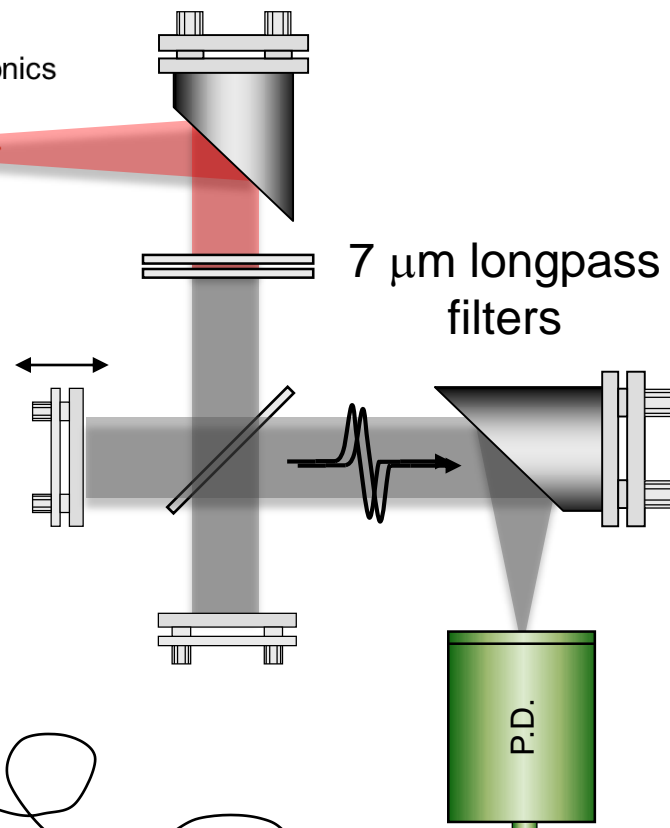
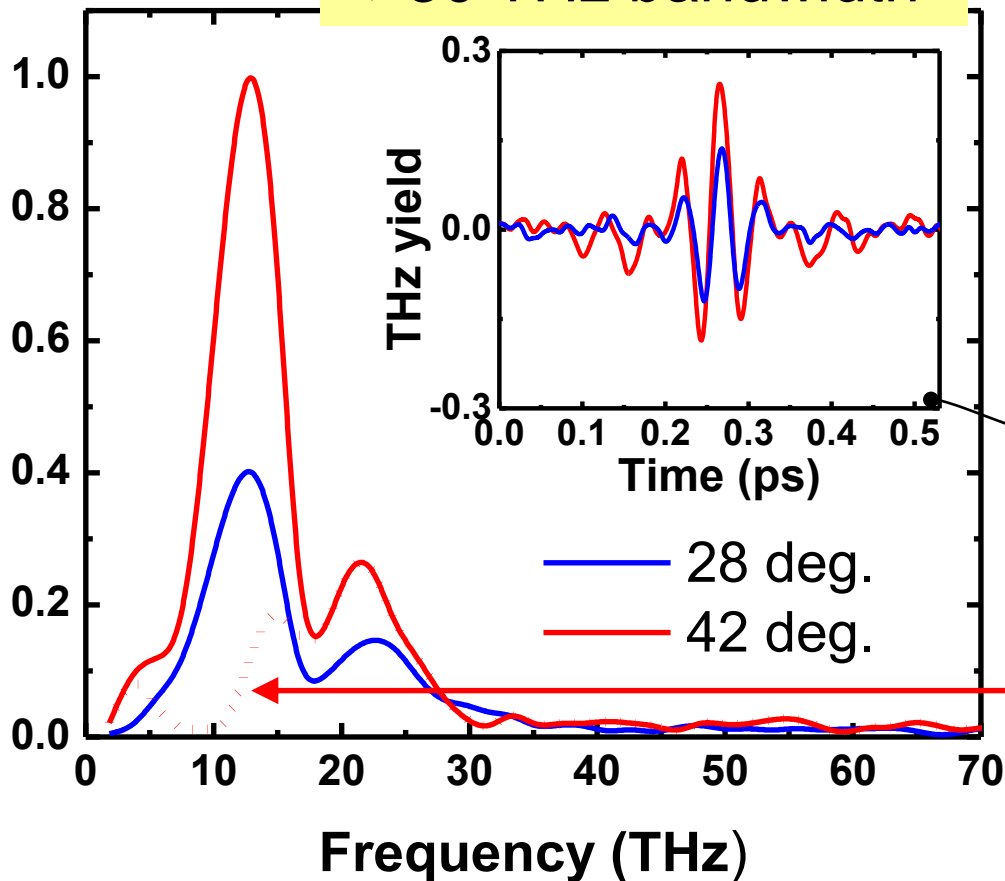
THz spectrum

Fourier-transform infrared (FTIR) spectrometer



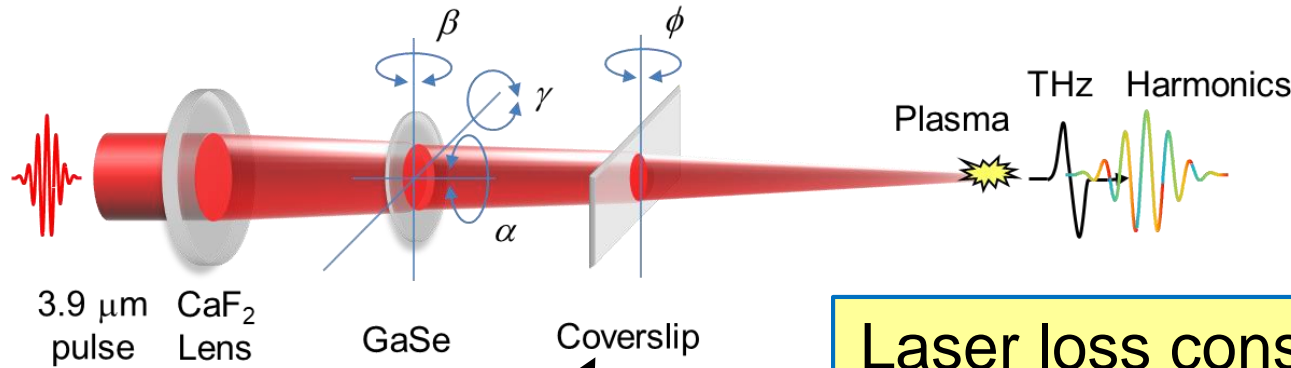
>30 THz bandwidth

Spectral power (arb. unit)



Spectrum distorted by 7 μm longpass filters used to block laser beams.

Laser-to-THz conversion efficiency

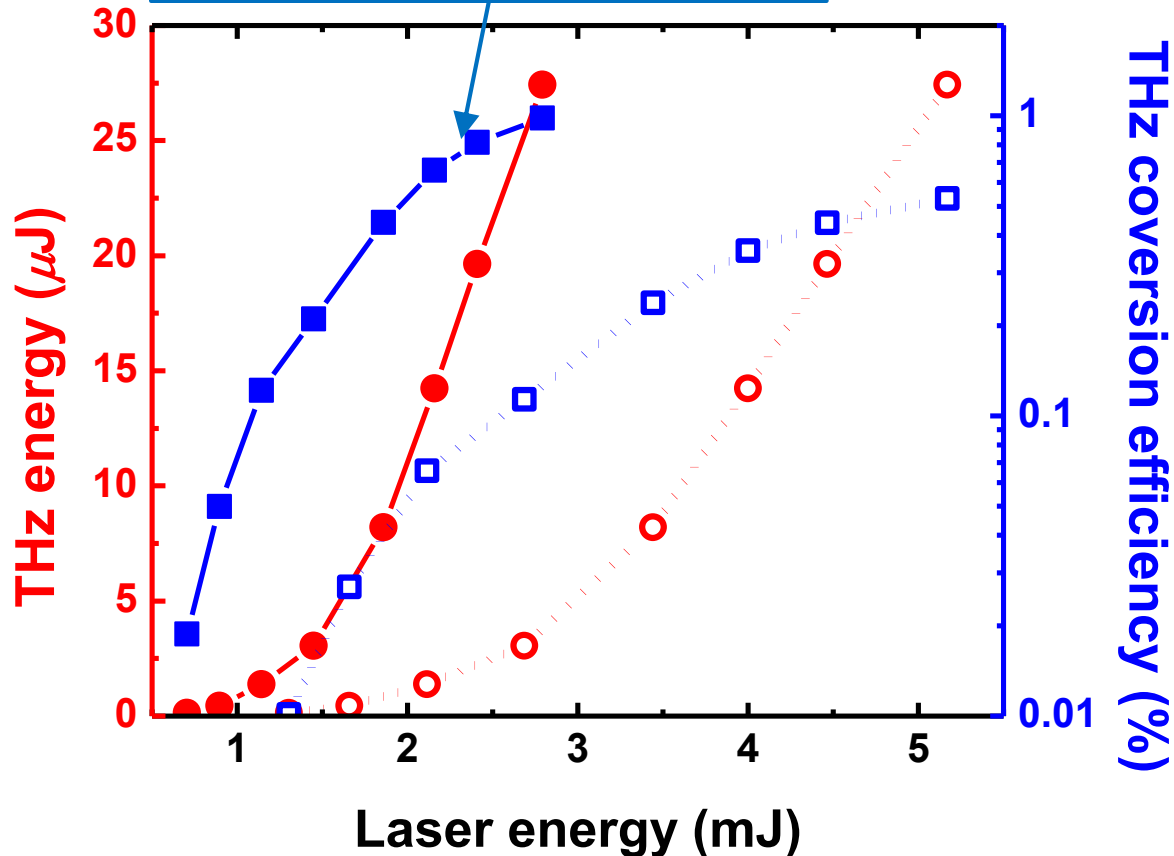


30%
Fresnel loss

10%
loss

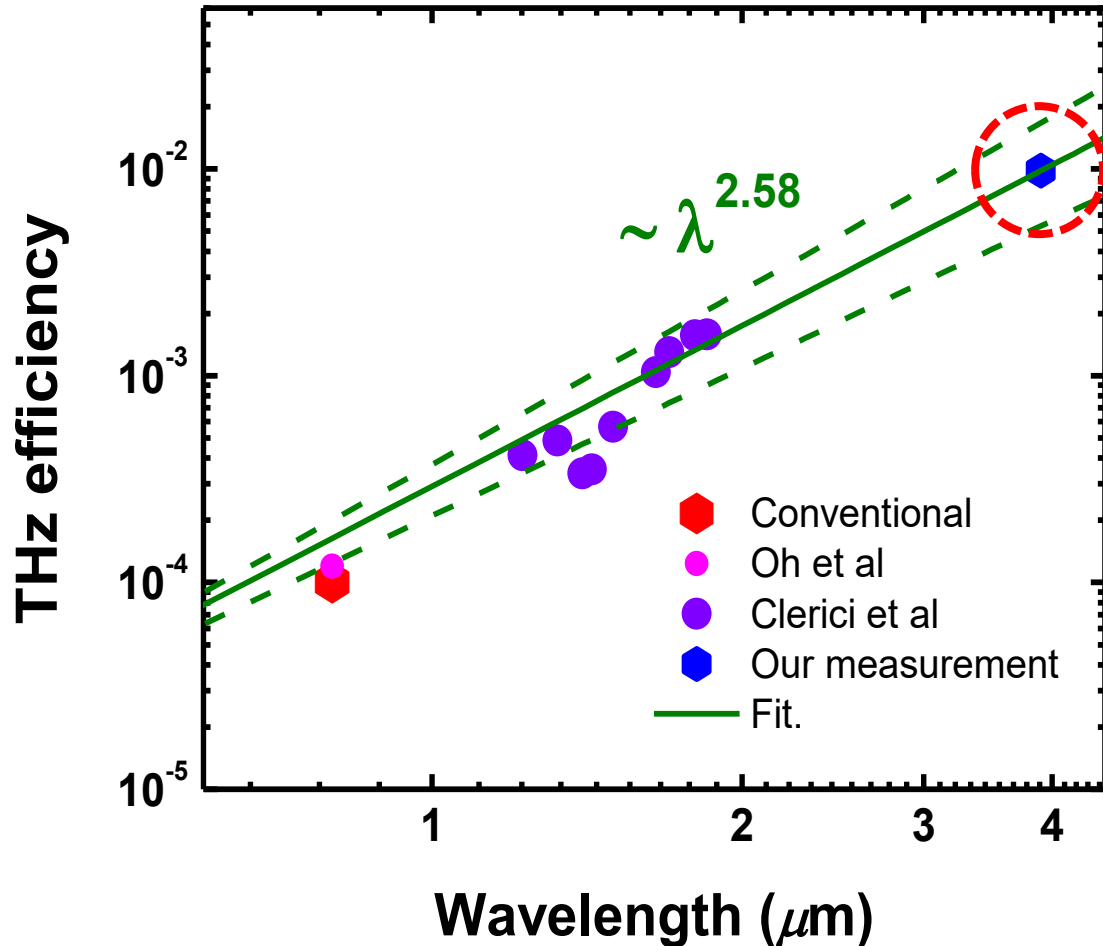
- Max. 1% efficiency with $I_{2\omega}/I_{\omega} = 0.02$
- Higher efficiency expected with greater $I_{2\omega}/I_{\omega}$

Laser loss considered



Wavelength scaling

THz generation via two-color mixing



- Better than λ^2
- Surprisingly high THz energy expected at 10 μm

Coherent control of broad EM waves

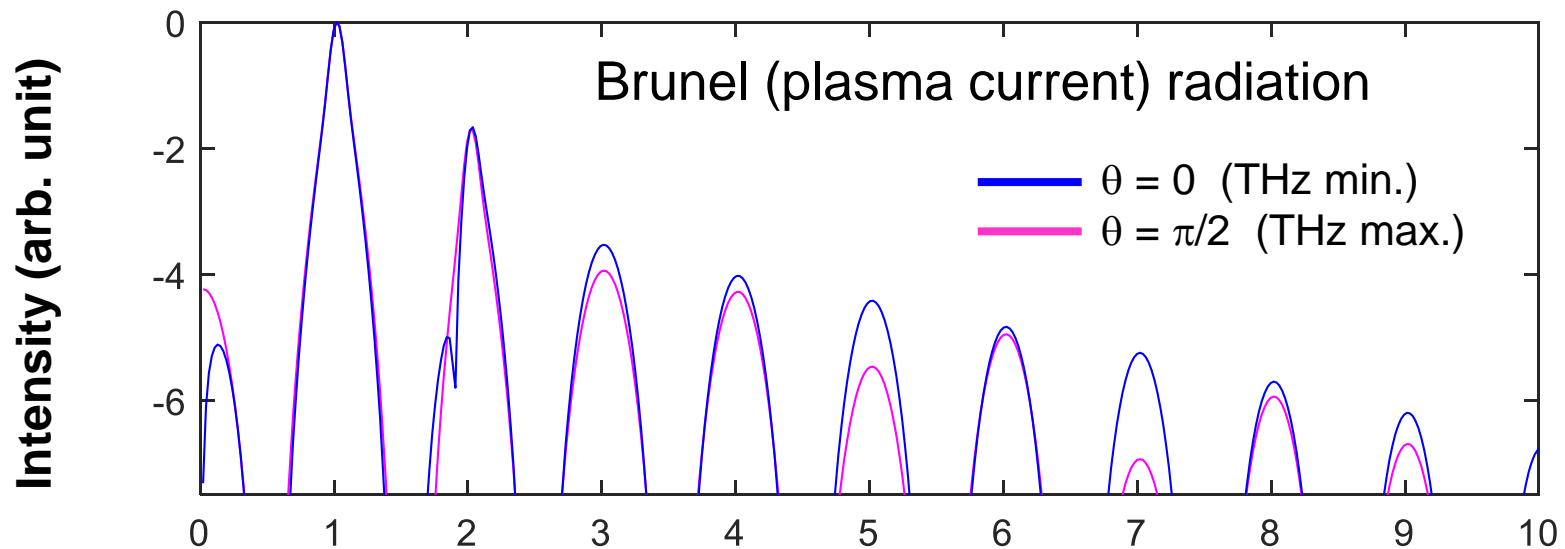
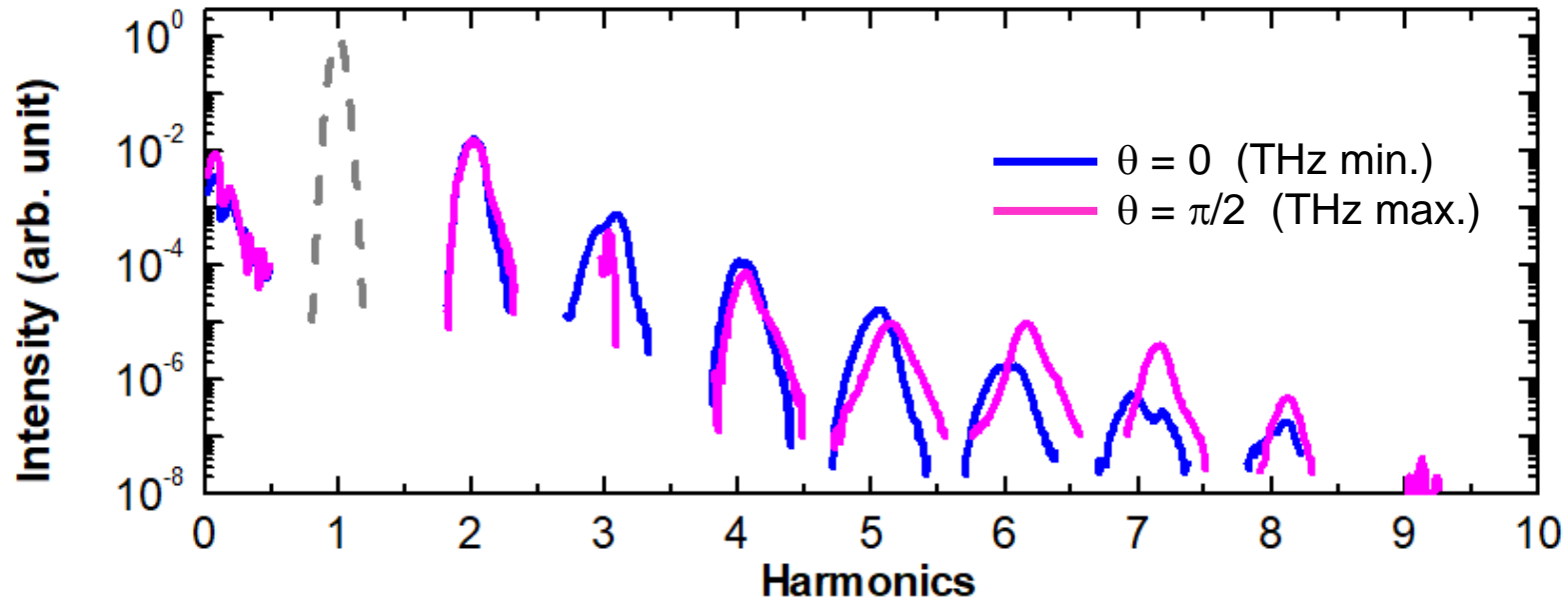
RF/ μ -wave

THz

IR

Visible

UV



Future Experiments & Simulation:

UPPE simulation

Unidirectional pulse propagation equation (UPPE)*

$$\frac{\partial \tilde{E}(z, \omega, k)}{\partial z} = ik_z \tilde{E}(z, \omega, k) + \frac{i\omega^2}{2\varepsilon_0 c^2 k_z} \tilde{P}_{NL}(z, \omega, k) - \frac{\omega}{2\varepsilon_0 c^2 k_z} \tilde{J}(z, \omega, k)$$

Dispersion

Nonlinear polarization

Plasma current

$$k_z = k_z(\omega, k) = \sqrt{\omega^2 \varepsilon(\omega) / c^2 - k^2}$$

$$\tilde{P}_{NL} = \varepsilon_0 (\chi^{(3)} E^3 + \chi^{(5)} E^5 + \dots)$$

$$\tilde{J} = \frac{e^2 (v_e + i\omega)}{m_e (v_e^2 + \omega^2)} \tilde{\rho} E$$

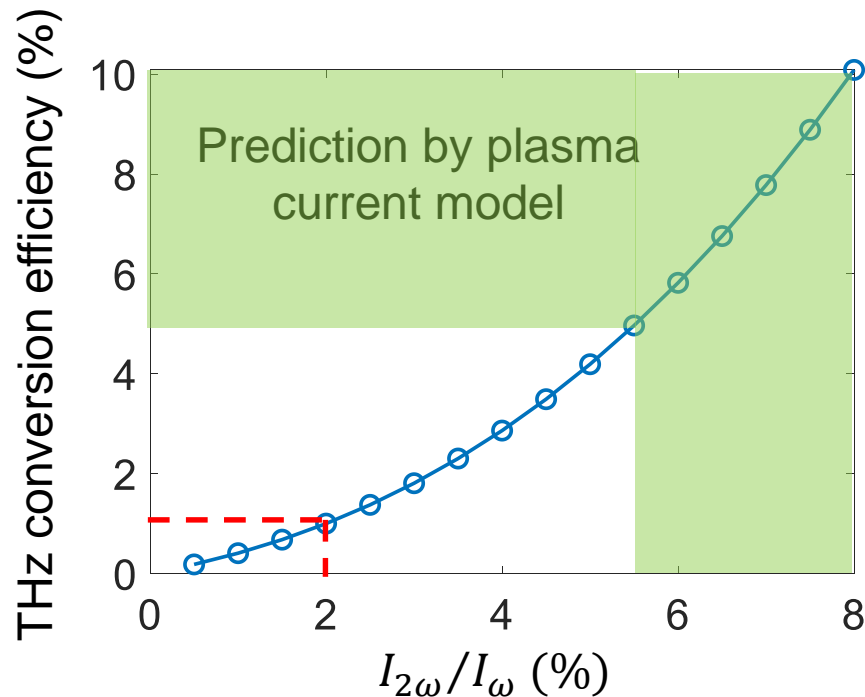
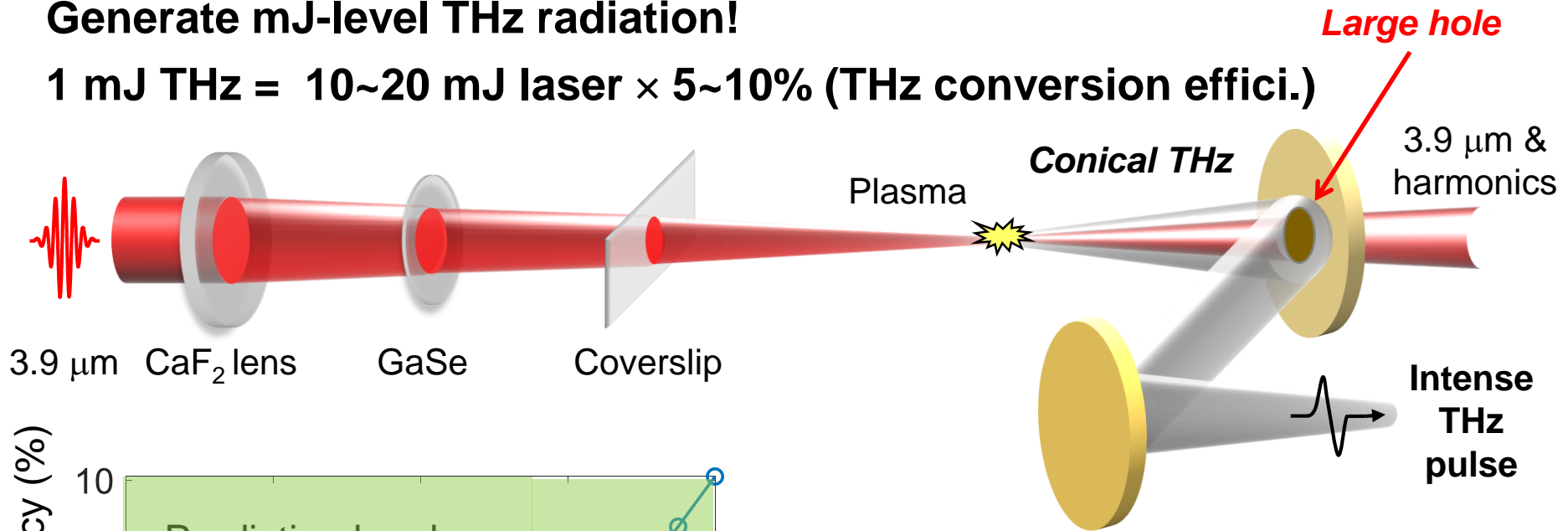
- Carrier based, no envelope approximations used
- Capture phase-dependent plasma, THz, harmonics generation with propagation

*M. Kolesik and J. V. Moloney, PRE **70**, 036604 (2004)

Next experiment with 3.9 μm laser (1)

Generate mJ-level THz radiation!

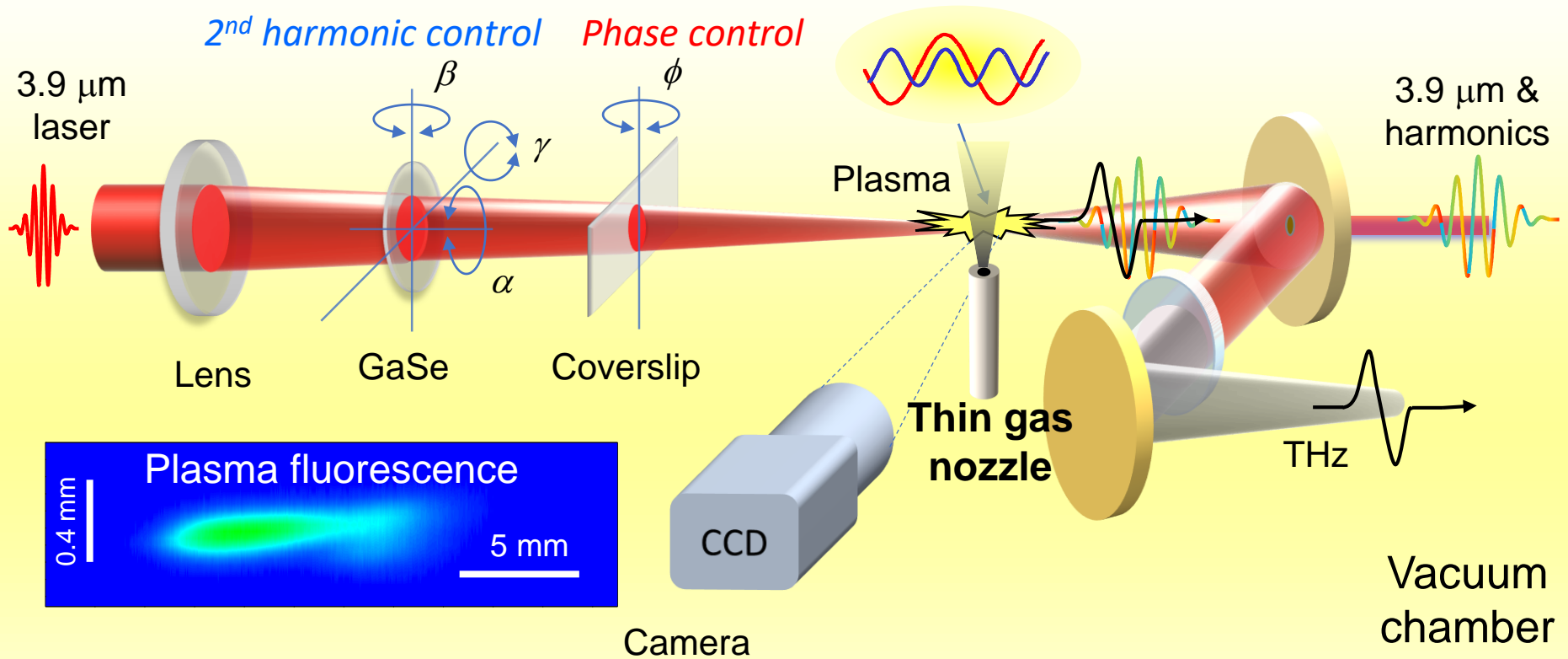
1 mJ THz = 10~20 mJ laser \times 5~10% (THz conversion effici.)



- **More 2nd harmonic energy**—use a more efficient SHG crystal
- **THz/laser separation**—use a parabolic mirror with a large hole (no filter necessary).

Next experiment with 3.9 μm laser (2)

Short plasma generation for accurate phase measurements



- Phase integrated effects occur by a long plasma created in air (due to plasma dispersion and Gouy phase shift)
- A short plasma will allow accurate measurements of phase θ dependent plasma, THz, and harmonic generation

Summary:

□ THz generation at long wavelengths:

- Observed 1% THz conversion efficiency with two-color laser mixing in air
- Generated broadband EM waves from microwave to UV
- Studied coherent control of ionization, THz, and harmonic generation

□ Near future experiments & simulation:

- Generate mJ level THz radiation with 5~10% efficiency
- Use a thin nozzle to localize phase-dependent effects in plasma, THz, and harmonic generation
- UPPE simulation for propagation effects

□ Anticipate more exciting results over the next 4 years!