Coherent control

with precision optical sources

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September 27, 2023

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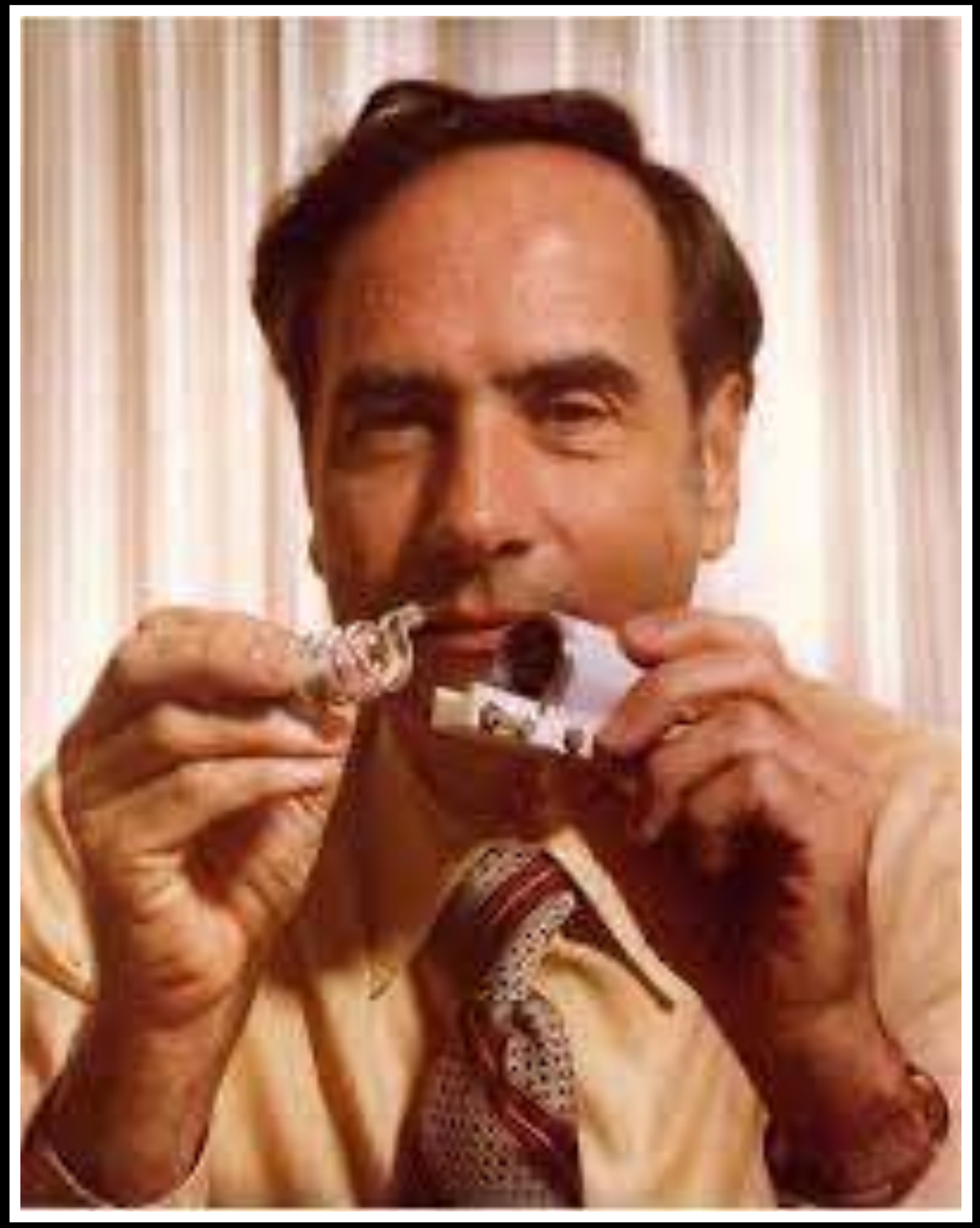
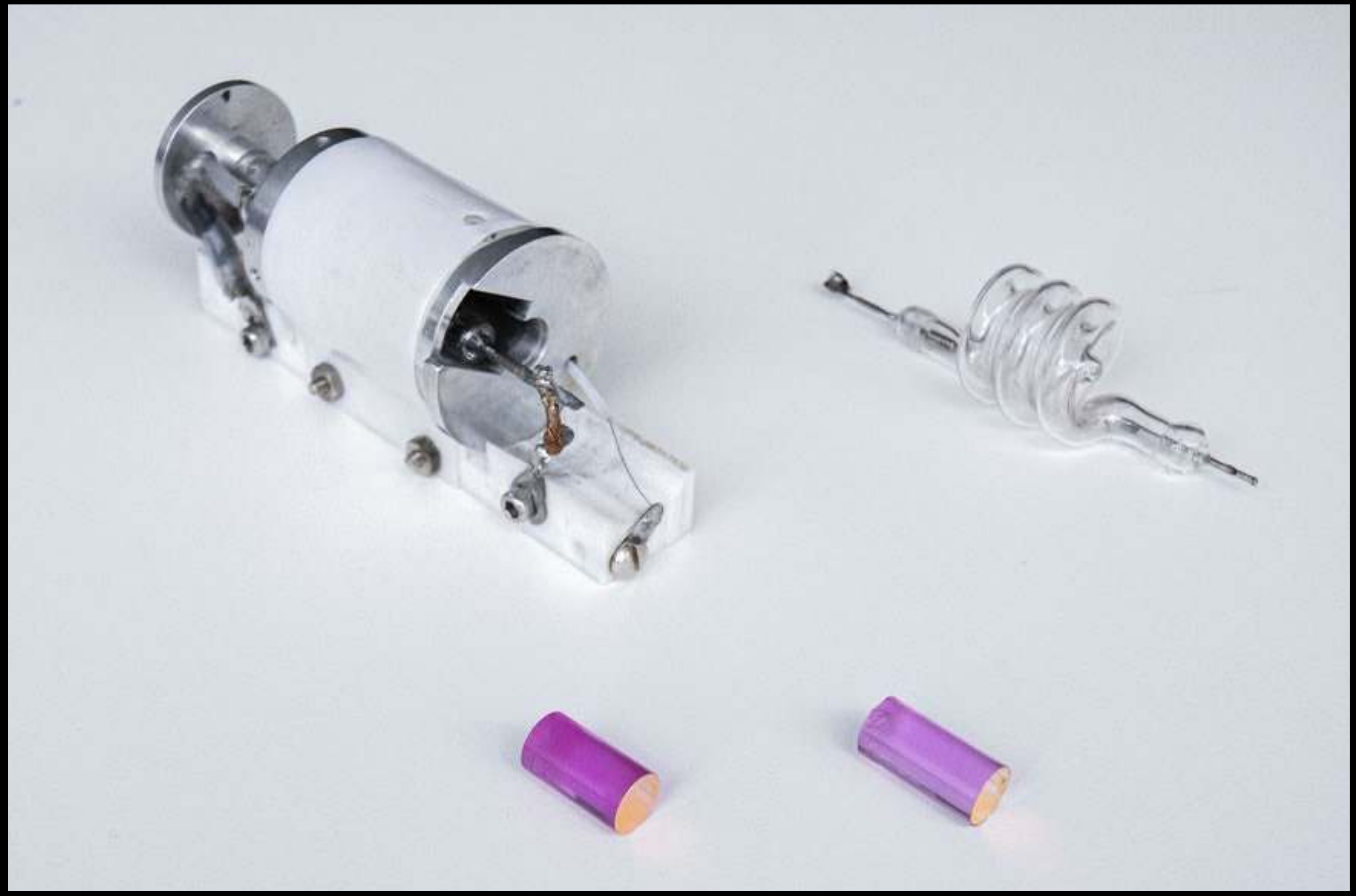
SFU MPQ NRC·CNRC uOttawa JASLab

- Introduction to coherent control
- Some early developments related to coherent control in solids
- Perturbative coherent control with structured light
    - Magnetic field transients
- Strong-field coherent control with single-cycle pulses
    - PHz-bandwidth waveform sampling

- Since 2021, I've been an Assistant Professor at Simon Fraser University
- Located in Metro Vancouver, BC, Canada
- Founded in 1965
- ~30k students

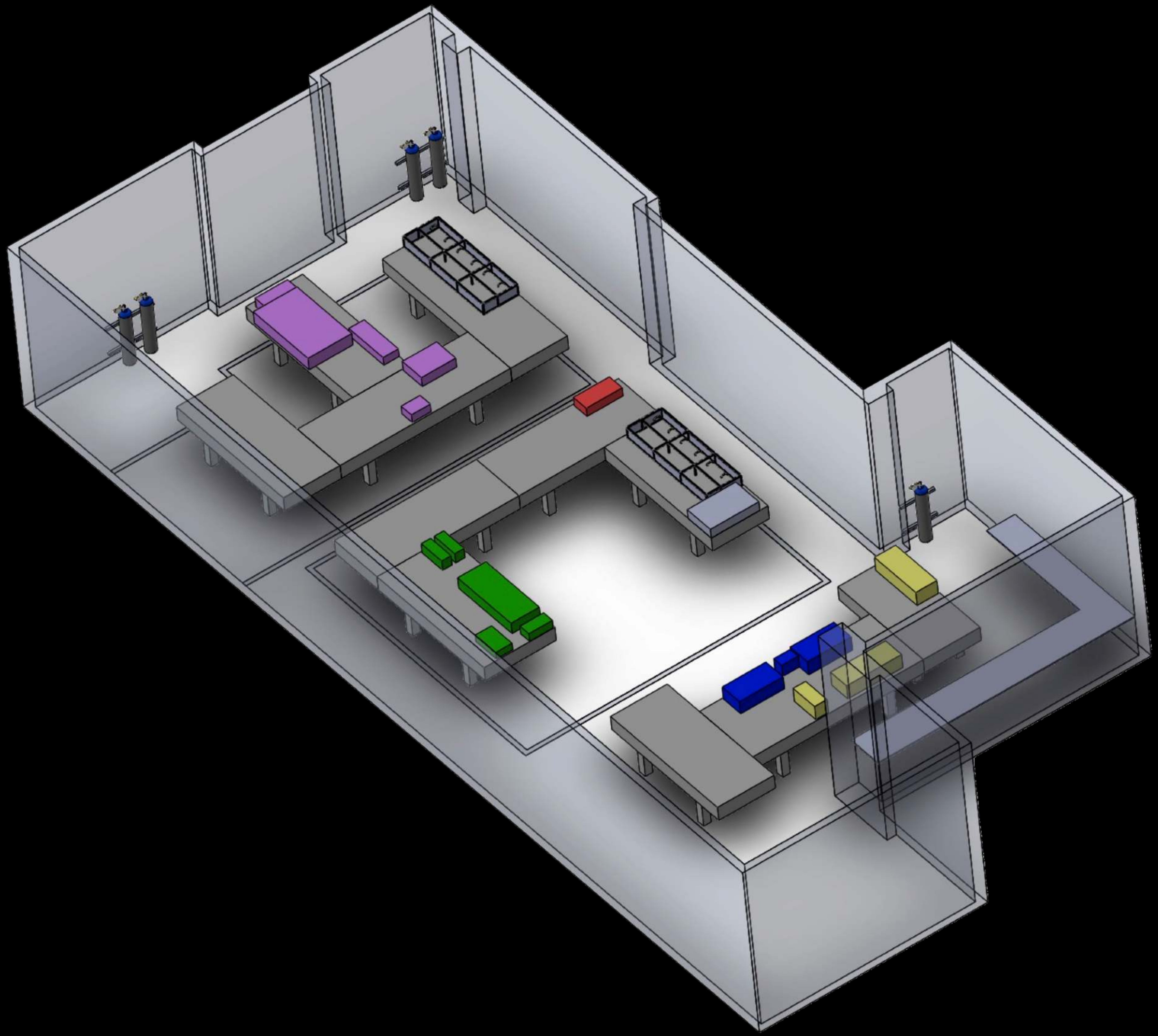
Simon Fraser University





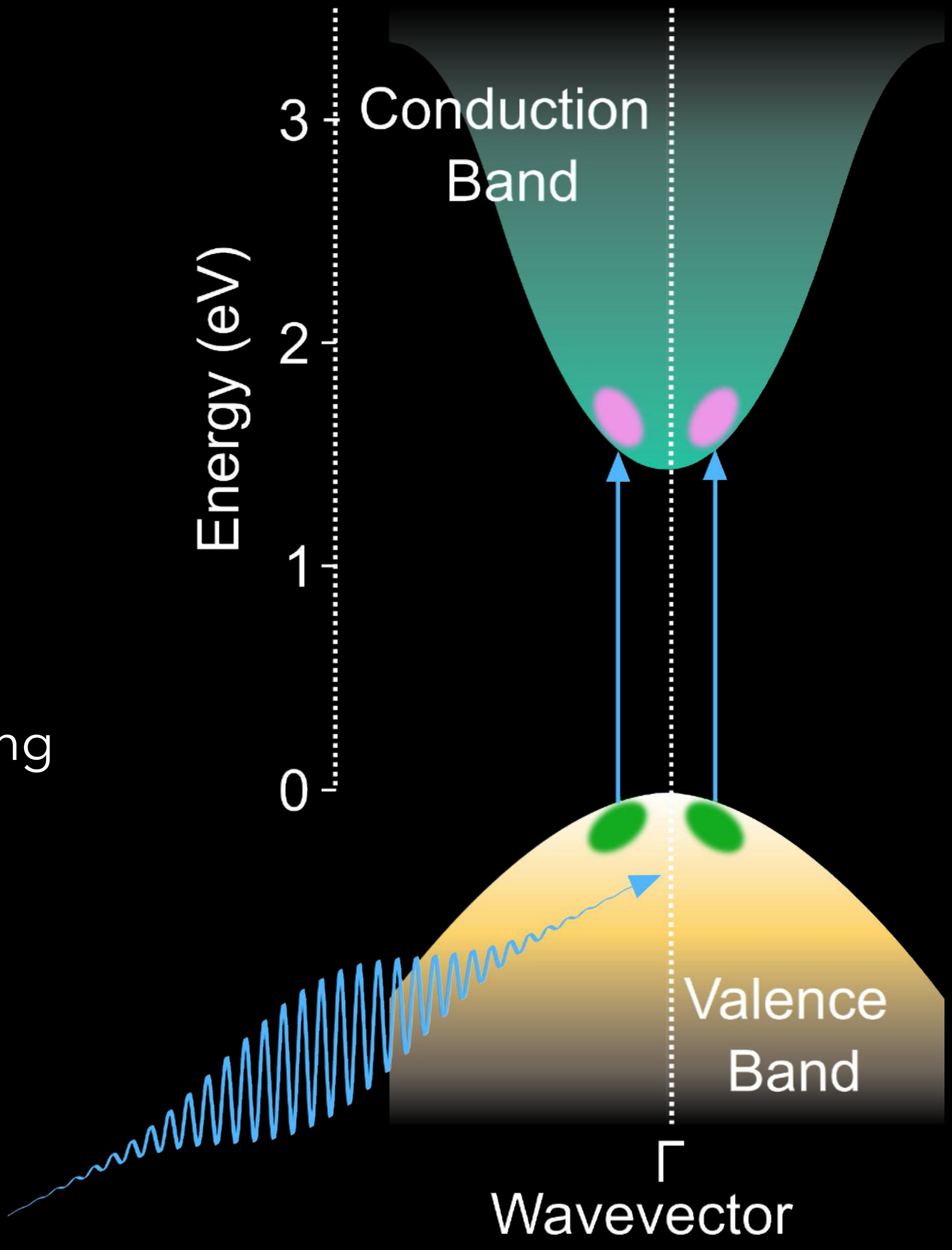
- Theodore Maiman spent his later years in Vancouver and was an Adjunct Professor at SFU
- His laser spent some time in our department and is currently on loan to the Max Planck Institute of Quantum Optics

our link to Theodore Maiman

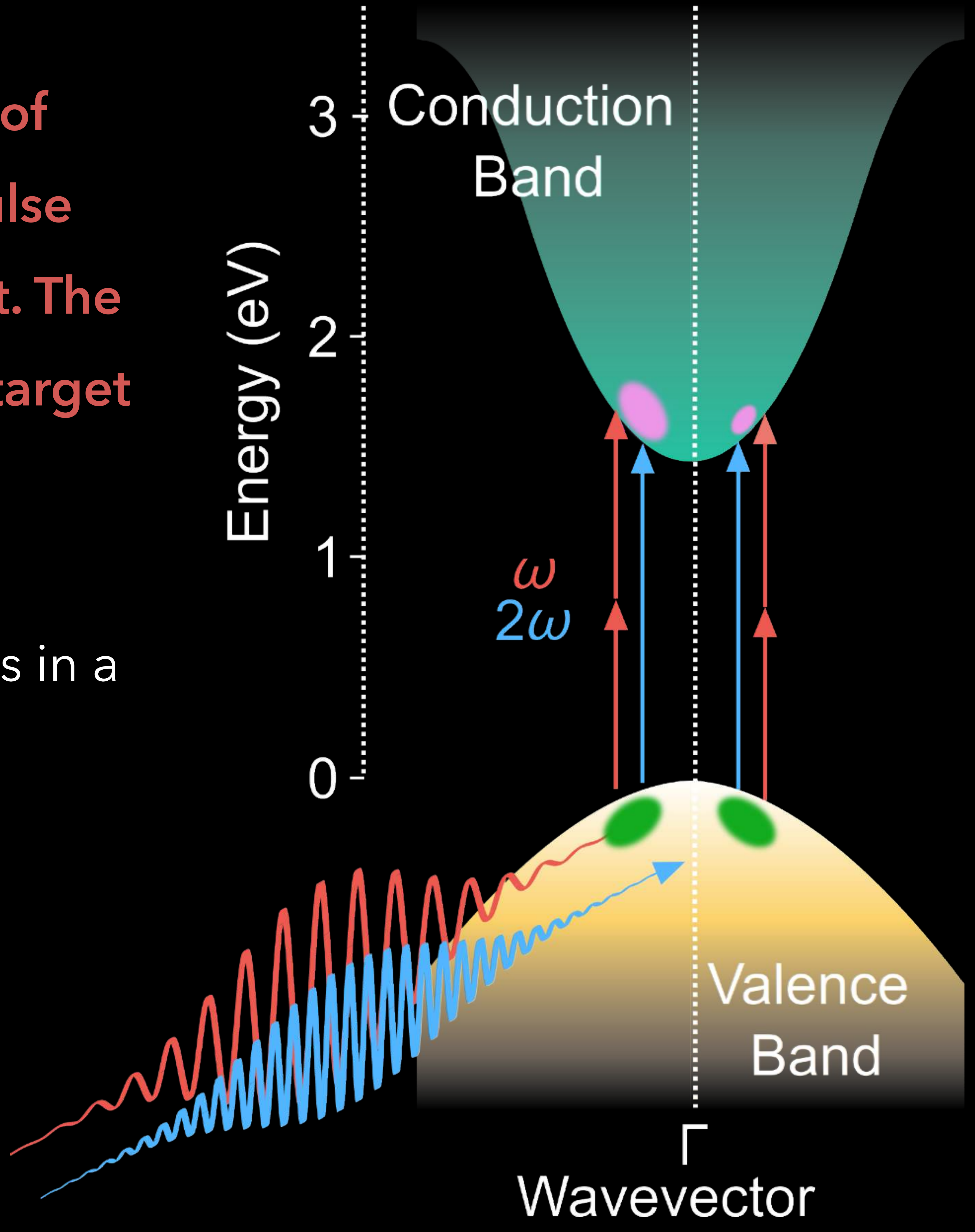


- We are in the process of developing our lab infrastructure in a >200 m^2 facility featuring vibrational isolation and air conditioning
- Our current research efforts are focused on:
  - Generation and field-resolved measurement of single-cycle structured light pulses
  - Quantum control in emerging materials and devices, with an emphasis on magnetic systems
- We are looking for a postdoc to join us!

- Steering and recording fast electronic processes is a central pursuit in ultrafast optics
    - **Example:** exciting electron-hole pairs in a semiconductor and recording their subsequent thermalization and recombination
    - Ultrafast spectroscopy has revealed that electron dephasing in solids occurs on 10 - 100 fs timescales



- Coherent control or quantum control requires an extra degree of precision. The outcome of the interaction between the laser pulse and the target system is sensitive to the phase of the laser light. The laser pulse imparts coherence of some sort to electrons in the target system that persists for some time after the laser pulse has left
    - **Examples:**
        - controlling momentum asymmetry of charge carriers in a semiconductor
        - sorting electron spins in momentum space
        - controlling molecular interactions
        - creating exotic molecular states

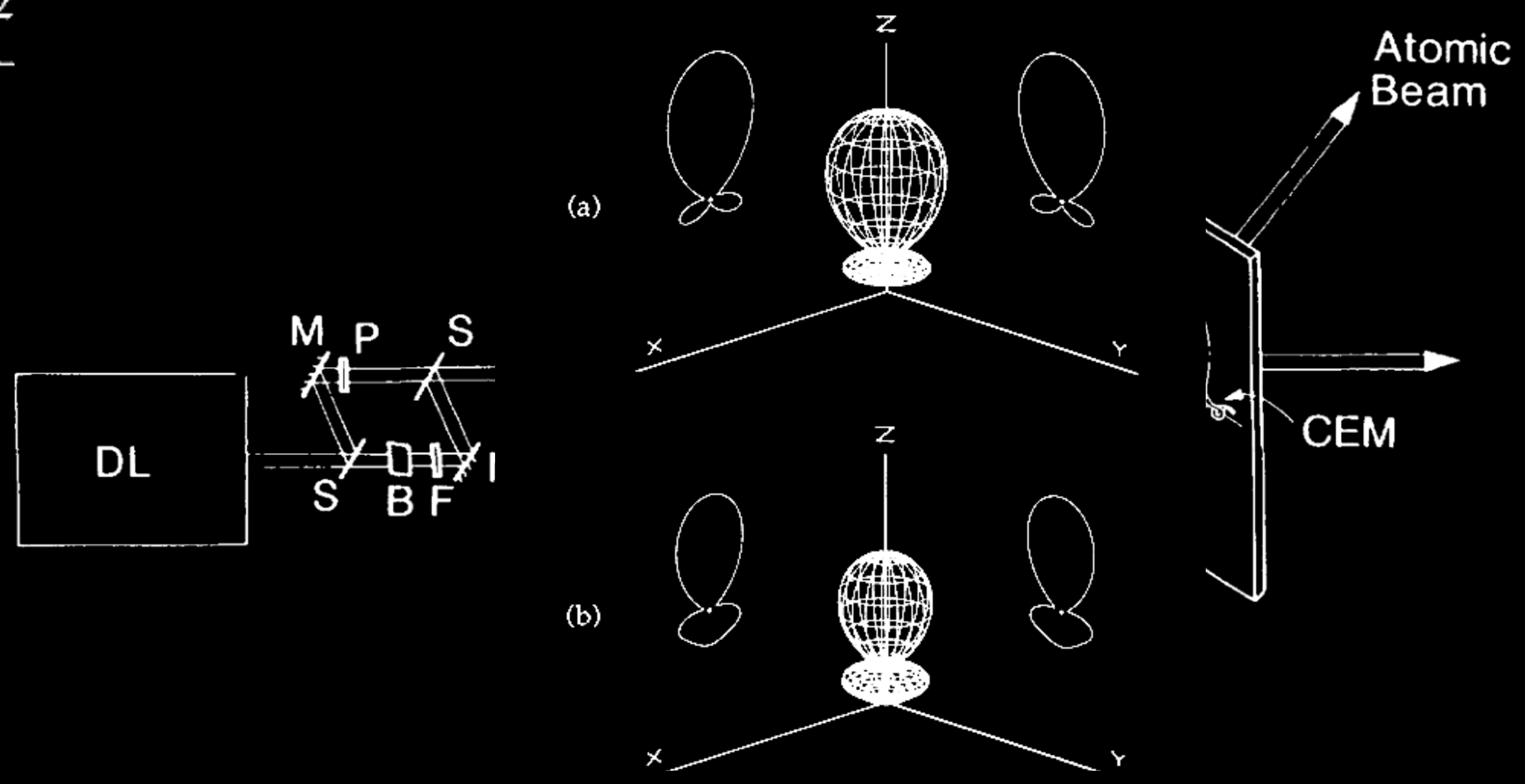


VOLUME 69, NUMBER 16 PHYSICAL REVIEW LETTERS 19 OCTOBER 1992

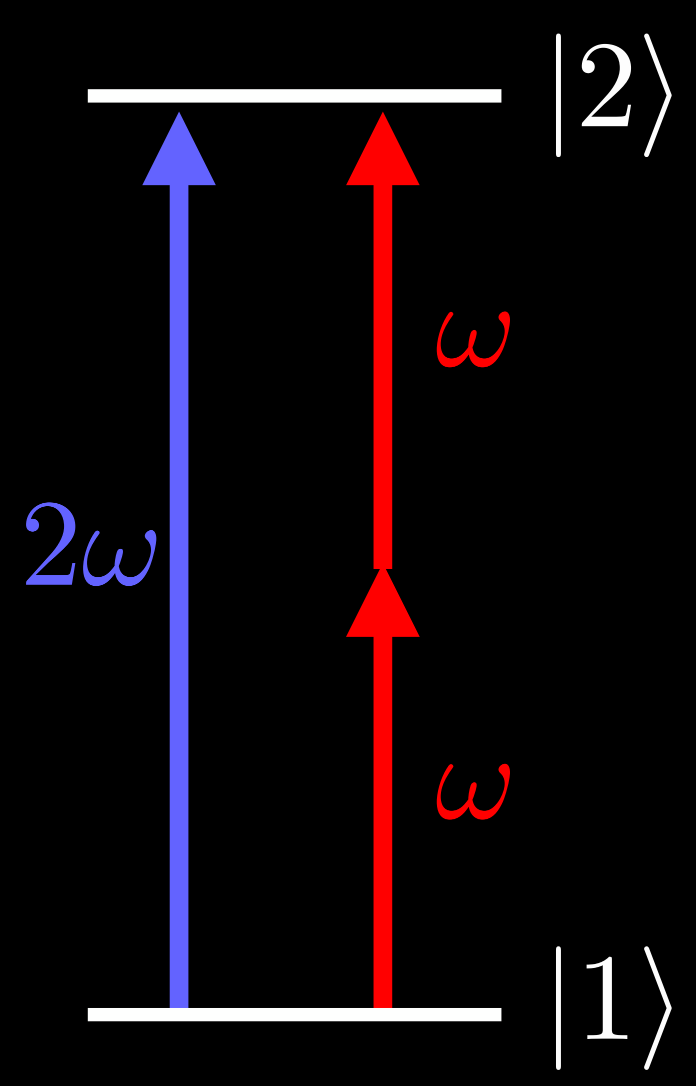
Asymmetric Photoelectron Angular Distributions from Interfering Photoionization Processes

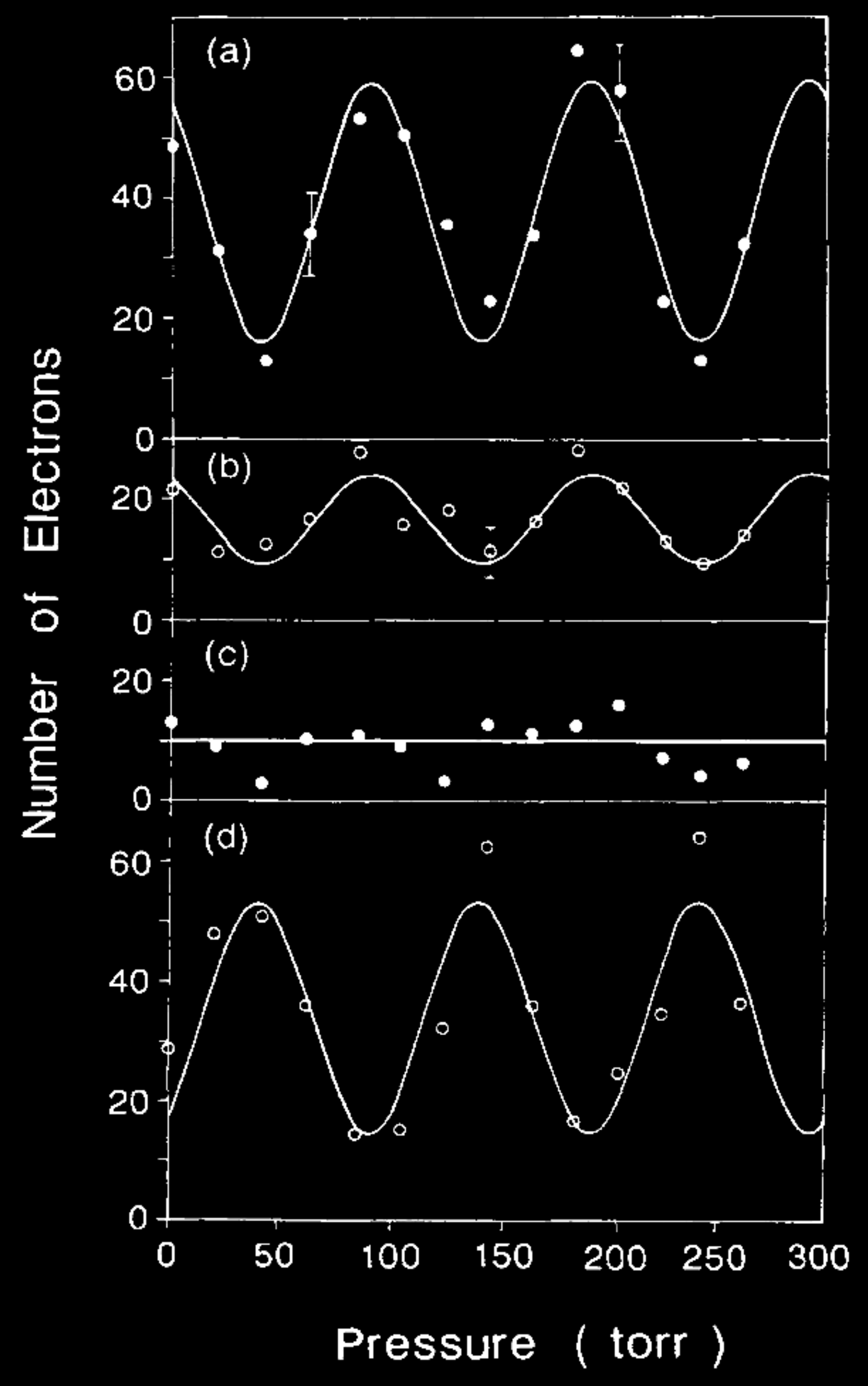
Yi-Yian Yin, Ce Chen, and D. S. Elliott
School of Electrical Engineering, Purdue University, West Lafayette, Indiana 47907-1285

A. V. Smith
Sandia National Laboratories, Albuquerque, New Mexico 87185



- **Target system: atomic rubidium (I_p = 4.18 eV)**
  - 280 nm: single-photon ionization
  - 560 nm: two-photon ionization
  - Interference between even- and odd-parity wave functions depends sinusoidally on the relative phase between the two colors of light
  - Asymmetry in photoelectron momentum is controlled



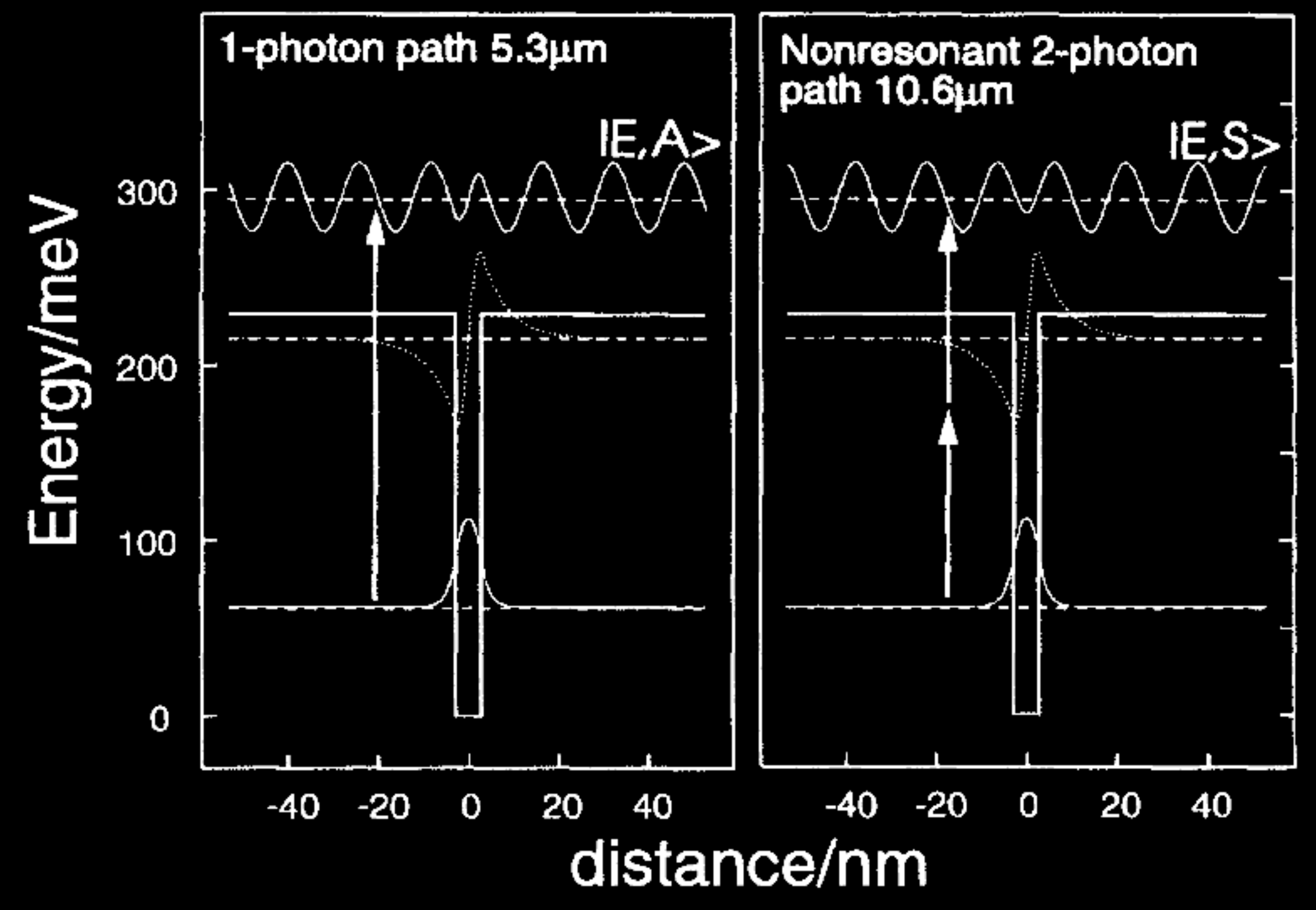


VOLUME 74, NUMBER 18 PHYSICAL REVIEW LETTERS 1 MAY 1995

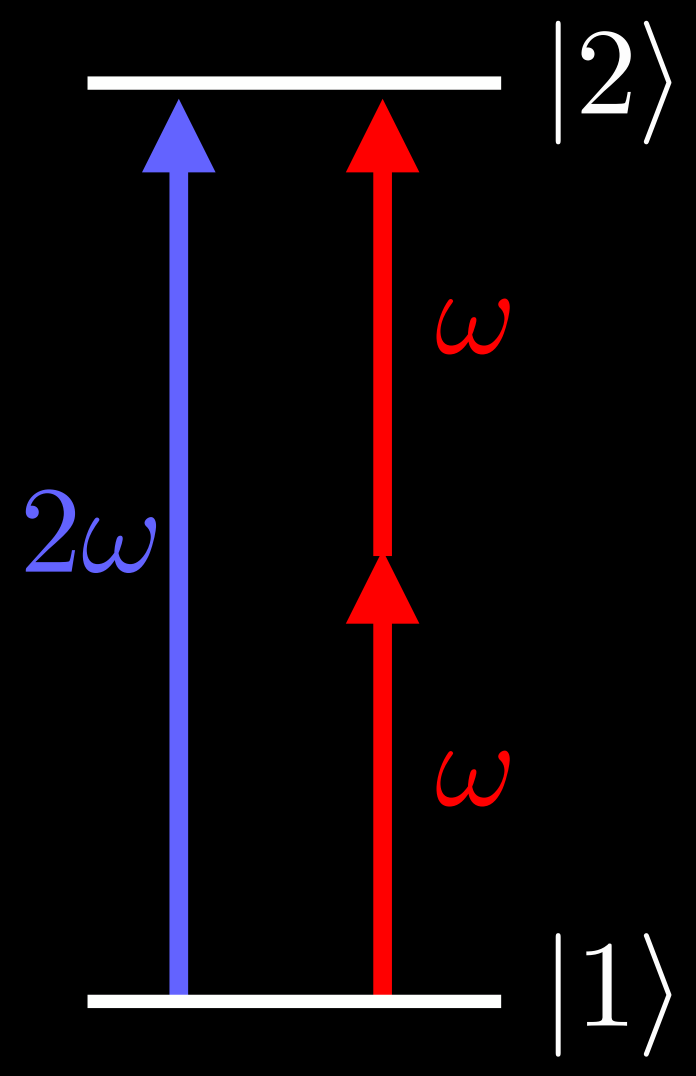
Phase-Controlled Currents in Semiconductors

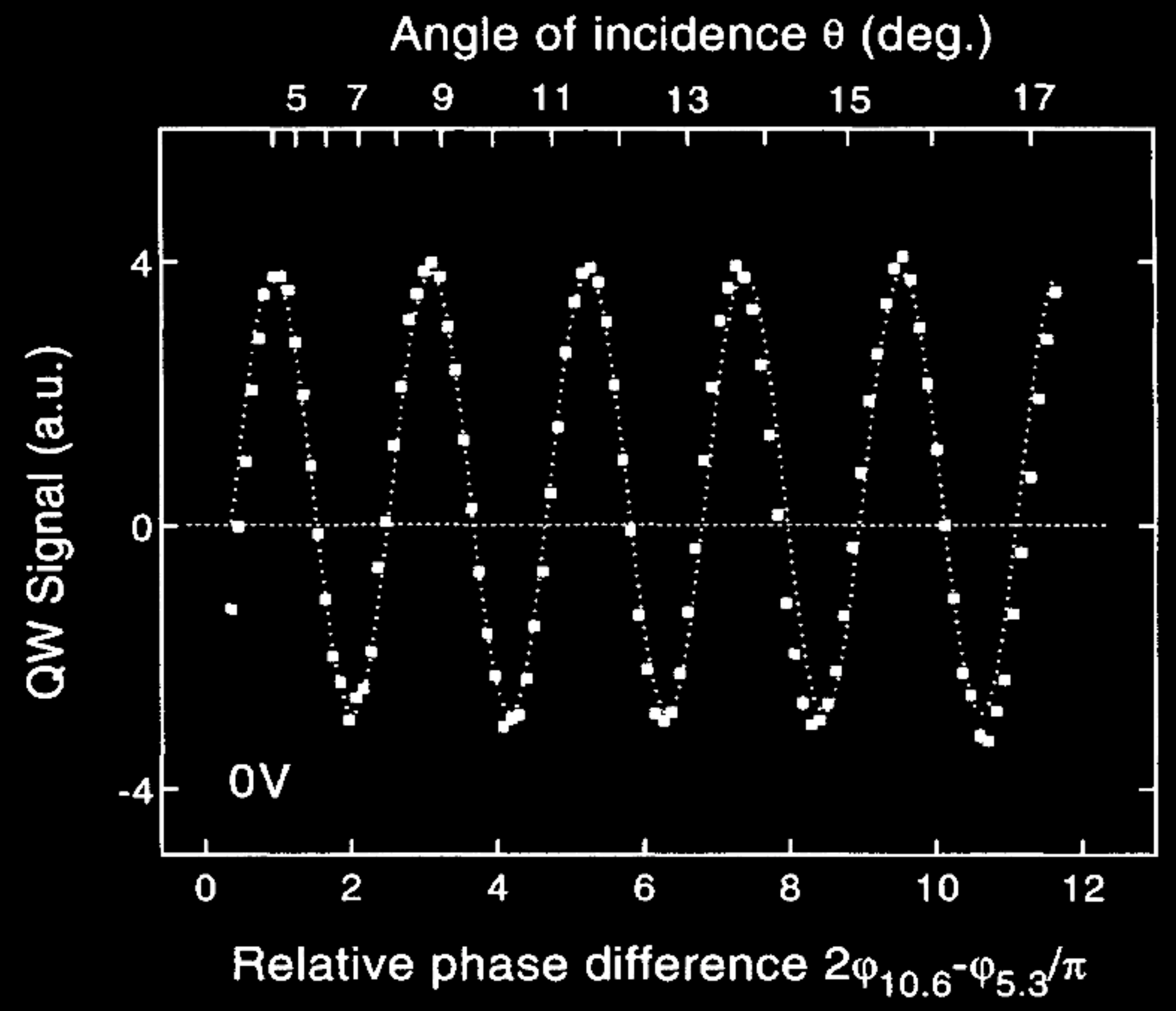
E. Dupont* and P. B. Corkum
Steacie Institute for Molecular Sciences, National Research Council, Ottawa, Ontario, Canada K1A 0R6

H. C. Liu, M. Buchanan, and Z. R. Wasilewski
Institute for Microstructural Sciences, National Research Council, Ottawa, Ontario, Canada K1A 0R6



- **GaAs/AlGaAs quantum wells (@ 82 K) irradiated by 10.6-μm + 5.3-μm pulses**
- Quantum interference currents detected with integrated electrodes
    - Sinusoidal dependence on relative phase



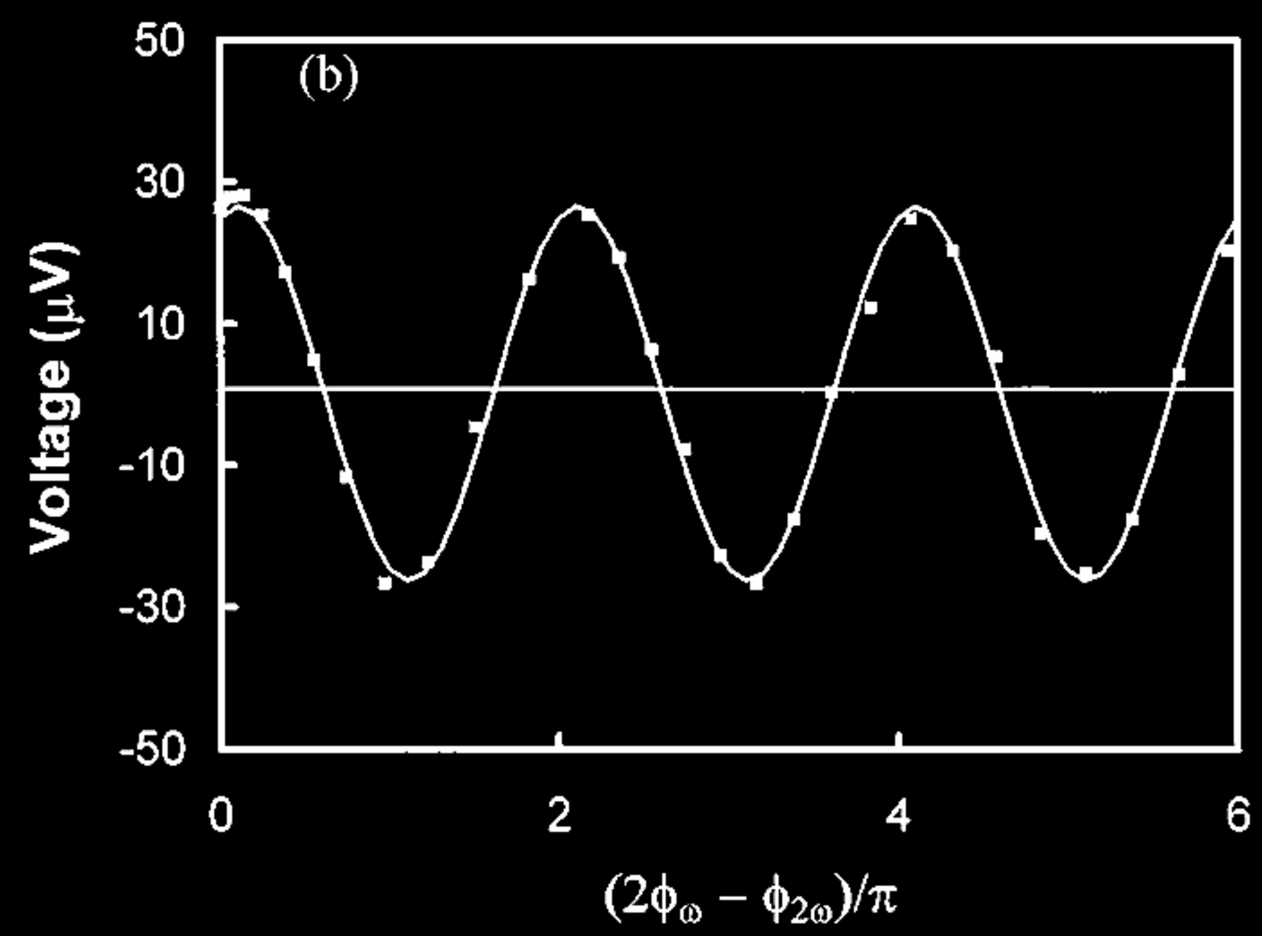


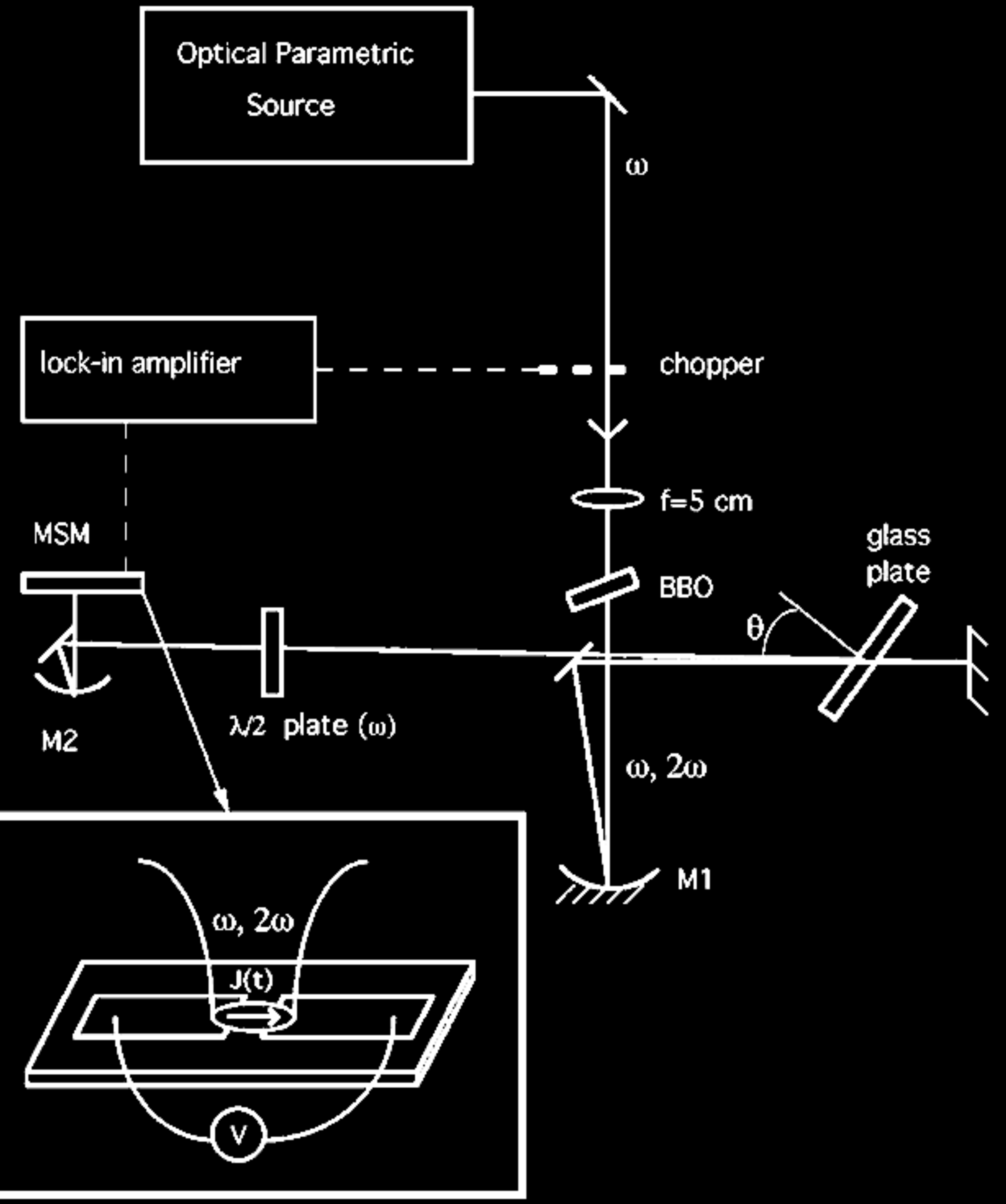
VOLUME 78, NUMBER 2 PHYSICAL REVIEW LETTERS 13 JANUARY 1997

Observation of Coherently Controlled Photocurrent in Unbiased, Bulk GaAs

A. Haché, Y. Kostoulas, R. Atanasov, J. L. P. Hughes, J. E. Sipe, and H. M. van Driel
Department of Physics, University of Toronto and Ontario Laser and Lightwave Research Centre, Toronto, Canada, M5S 1A7

- The previous measurements involved continuum states
- Subsequent experiments extended this to electrons excited from the valence band into the conduction band of a conventional semiconductor (low-temperature-grown GaAs)



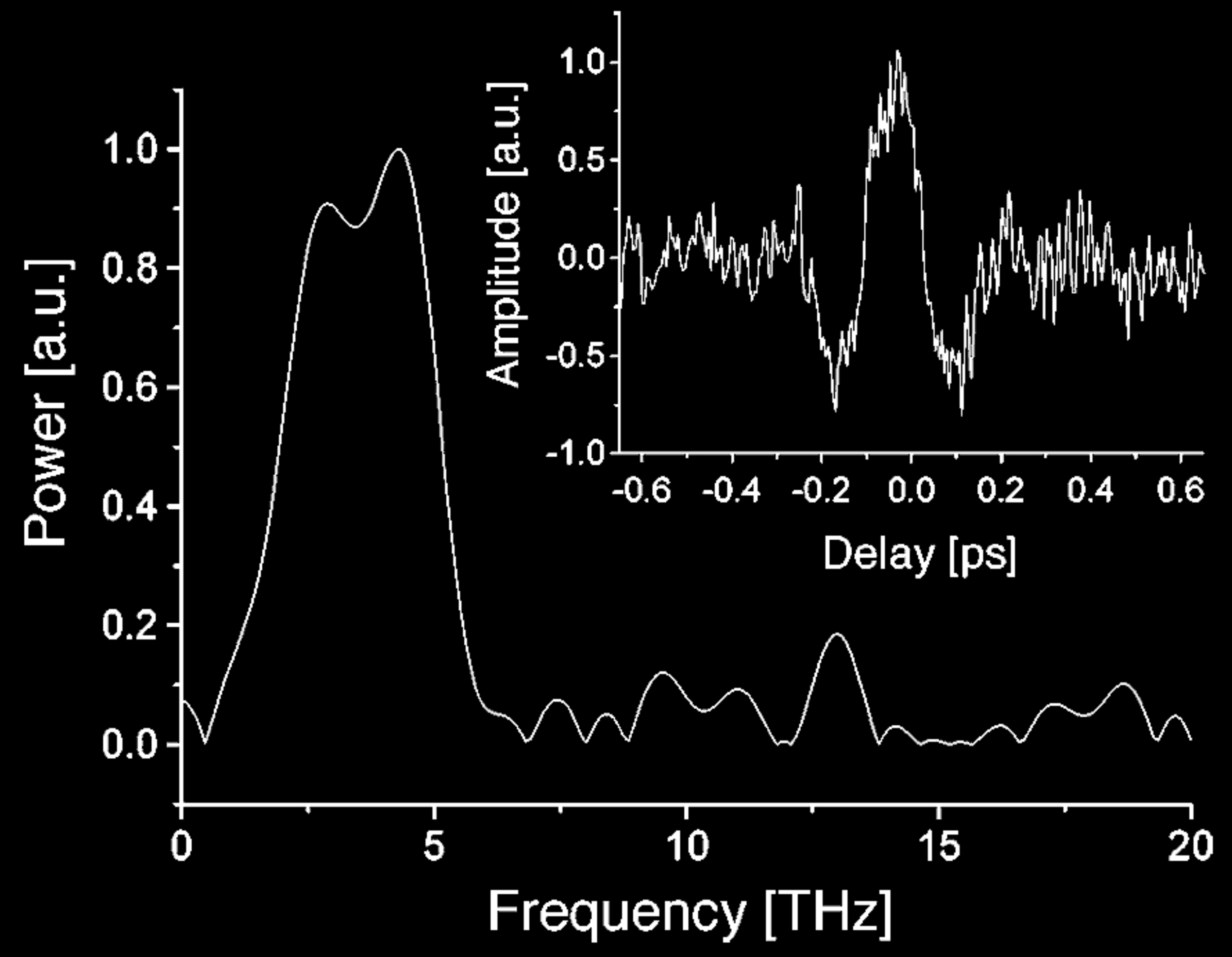


and to conventional semiconductors

APPLIED PHYSICS LETTERS VOLUME 75, NUMBER 25 20 DECEMBER 1999

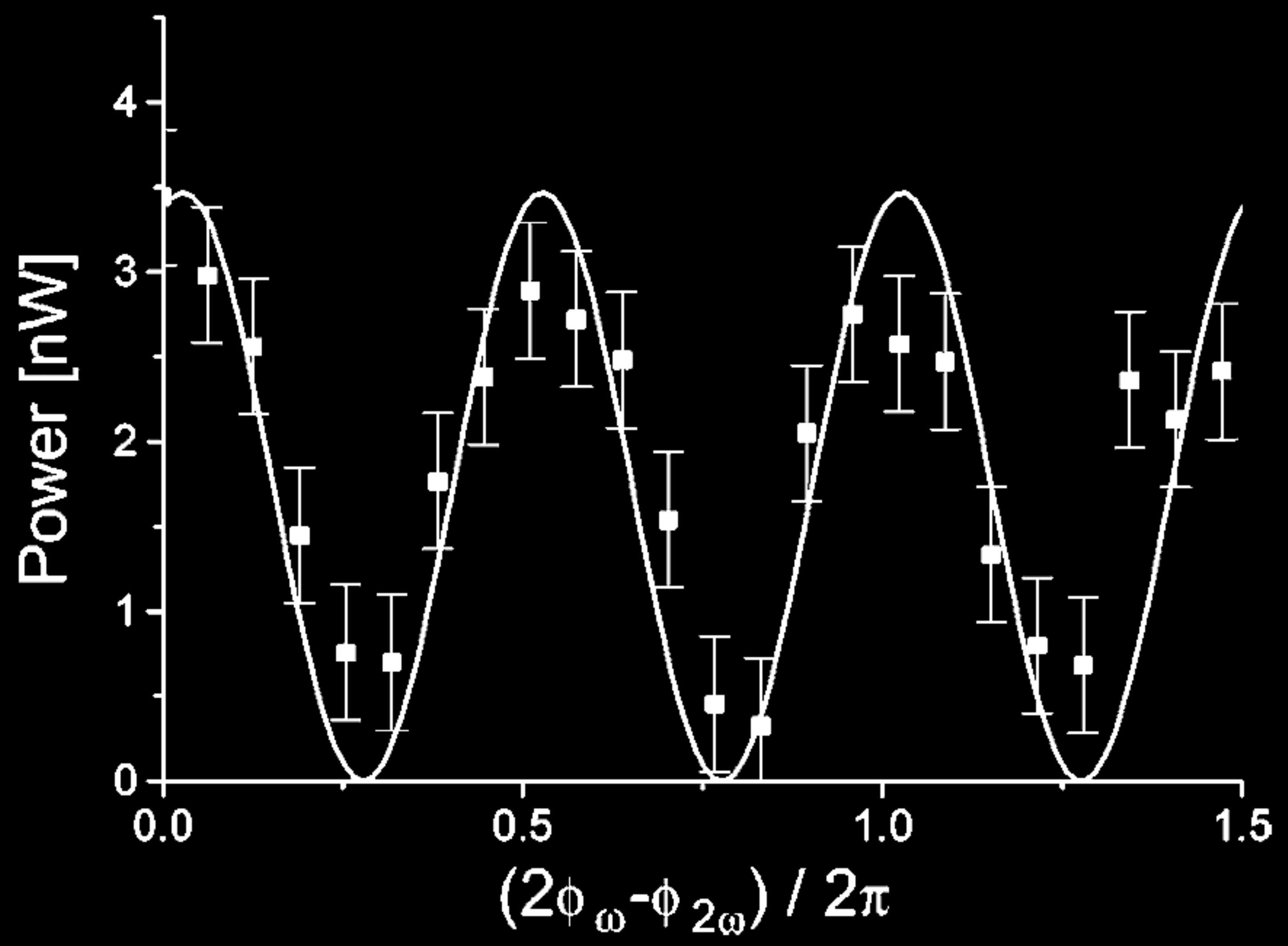
THz emission from coherently controlled photocurrents in GaAs

D. Côté[a] and J. M. Fraser
Department of Physics, University of Toronto, Toronto, Ontario, Canada M5S 1A7
M. DeCamp and P. H. Bucksbaum
Department of Physics, University of Michigan, Ann Arbor, Michigan 07974-2070
H. M. van Driel
Department of Physics, University of Toronto, Toronto, Ontario, Canada M5S 1A7



- ✦ Current is turned on within the timescale of the laser pulse duration
- ✦ It relaxes on a ~100 fs timescale
- ✦ This current transient radiates a broadband terahertz pulse
- ✦ The waveform of this pulse can be controlled using the relative phase
- ✦ The equilibrium charge carriers have sufficient density in many materials to shield this THz!

$$\frac{\partial \mathbf{J}_{e,h}}{\partial t} = \dot{\mathbf{j}}^{I}_{e,h} - \frac{\mathbf{J}_{e,h}}{\tau_{e,h}} + \frac{N_{e,h}e^2}{m^*_{e,h}}\mathbf{E}_{sc}$$

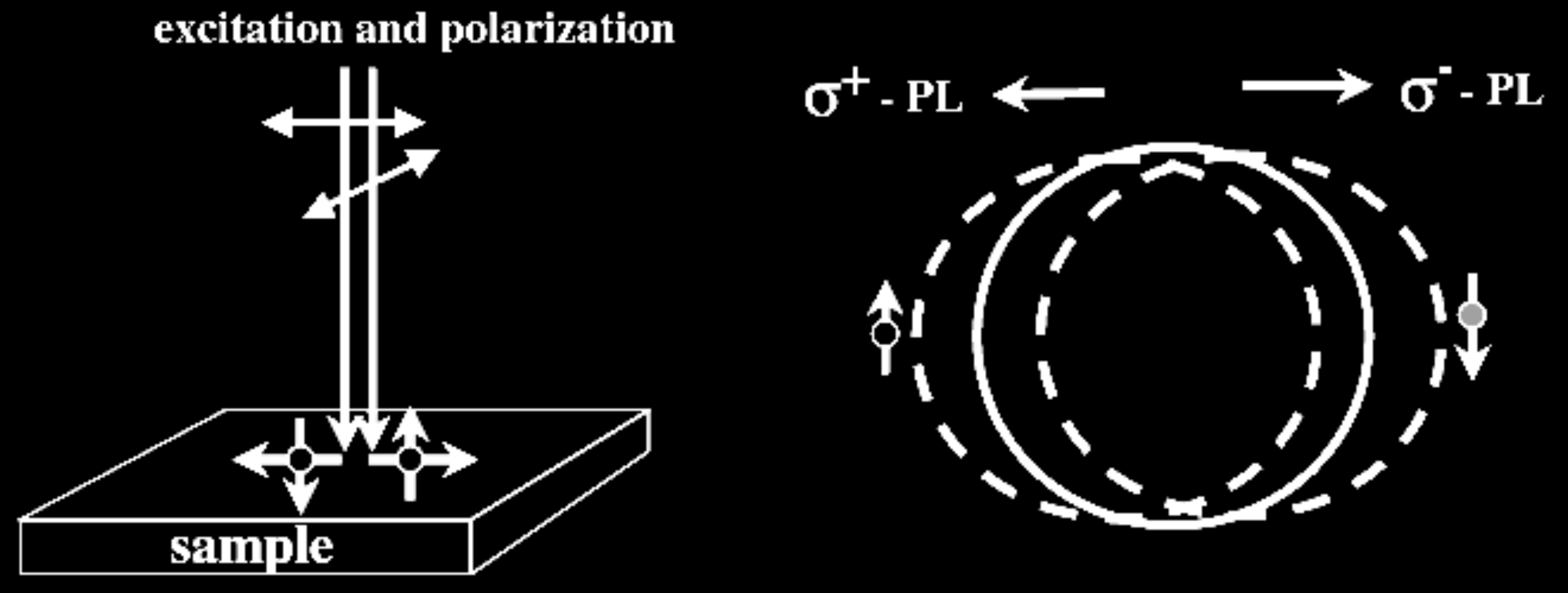


Current dynamics and THz radiation

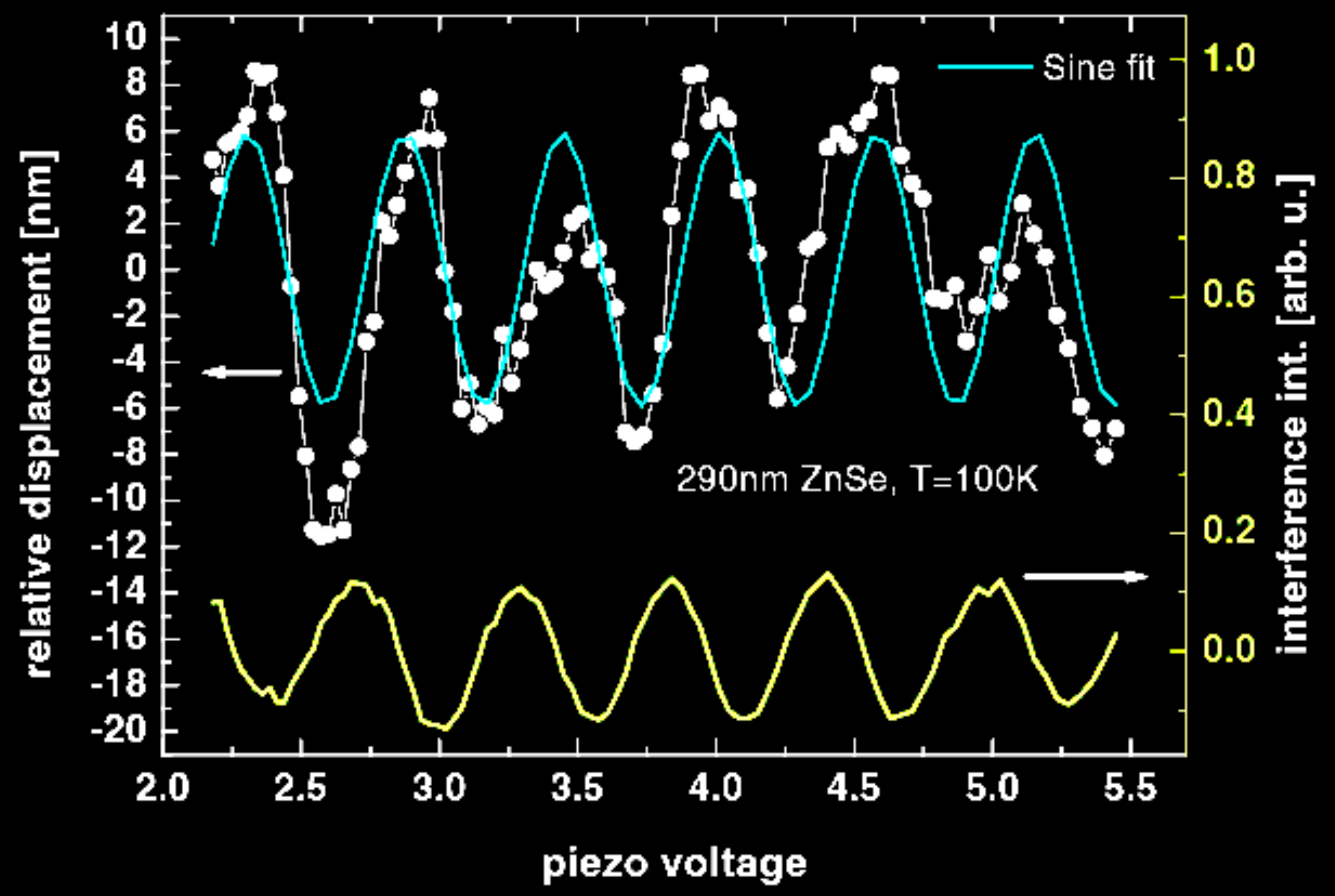
VOLUME 90, NUMBER 21 PHYSICAL REVIEW LETTERS week ending 30 MAY 2003

Direct Observation of Optically Injected Spin-Polarized Currents in Semiconductors

J. Hübner and W. W. Rühle
Department of Physics and Materials Science Center, University of Marburg, Renthof 5, 35032 Marburg, Germany
M. Klude and D. Hommel
Institute of Solid State Physics, Semiconductor Epitaxy, University of Bremen, P.O. Box 330440, 28334 Bremen, Germany
R. D. R. Bhat, J. E. Sipe, and H. M. van Driel
Department of Physics, University of Toronto, 60 St. George Street, Toronto, Ontario M5S 1A7, Canada



- Beyond charge currents, the possibility to control spin currents has also been explored using both cross-polarized bichromatic fields and bichromatic fields with circular polarization
- Detection: Spatial separation of circularly-polarized photoluminescence with opposite handedness



- **Some of the key results:**
  - Quantum pathway interference was observed in solids
  - It was used to control charge currents, spin currents, and THz waveforms
- **As laser and optical technology advance, new degrees of freedom for coherent control become available.**
- **Today I will discuss:**
  - **Perturbative coherent control using bichromatic structured light pulses**
    - I will show that structure is transferred from light to currents and place a particular emphasis on driving ring currents for the purpose of introducing magnetic fields within femtoseconds
  - **Strong-field coherent control in large bandgap solids using single-cycle pulses**
    - I will show how this can be harnessed for the purpose of PHz-bandwidth electric field waveform sampling

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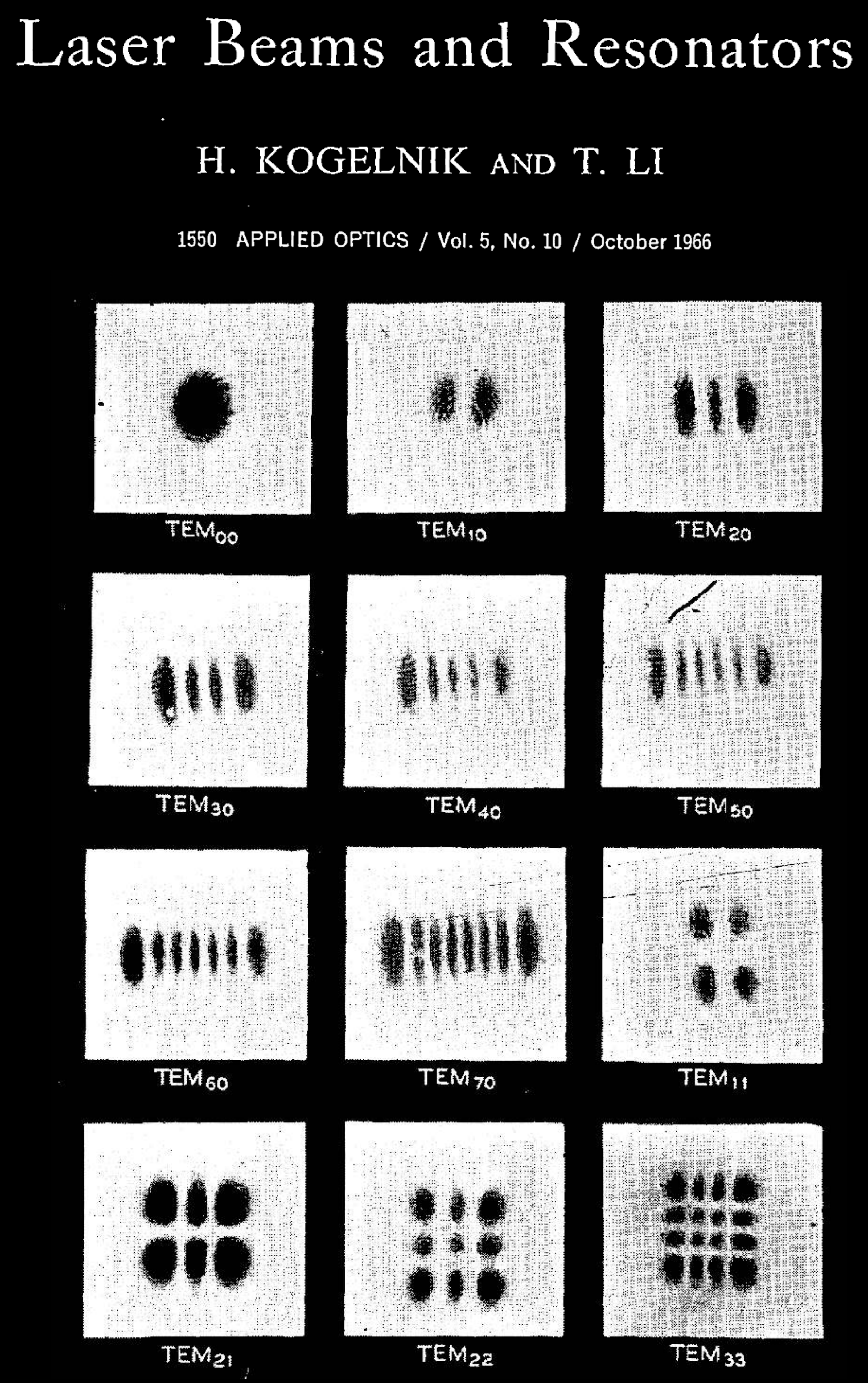
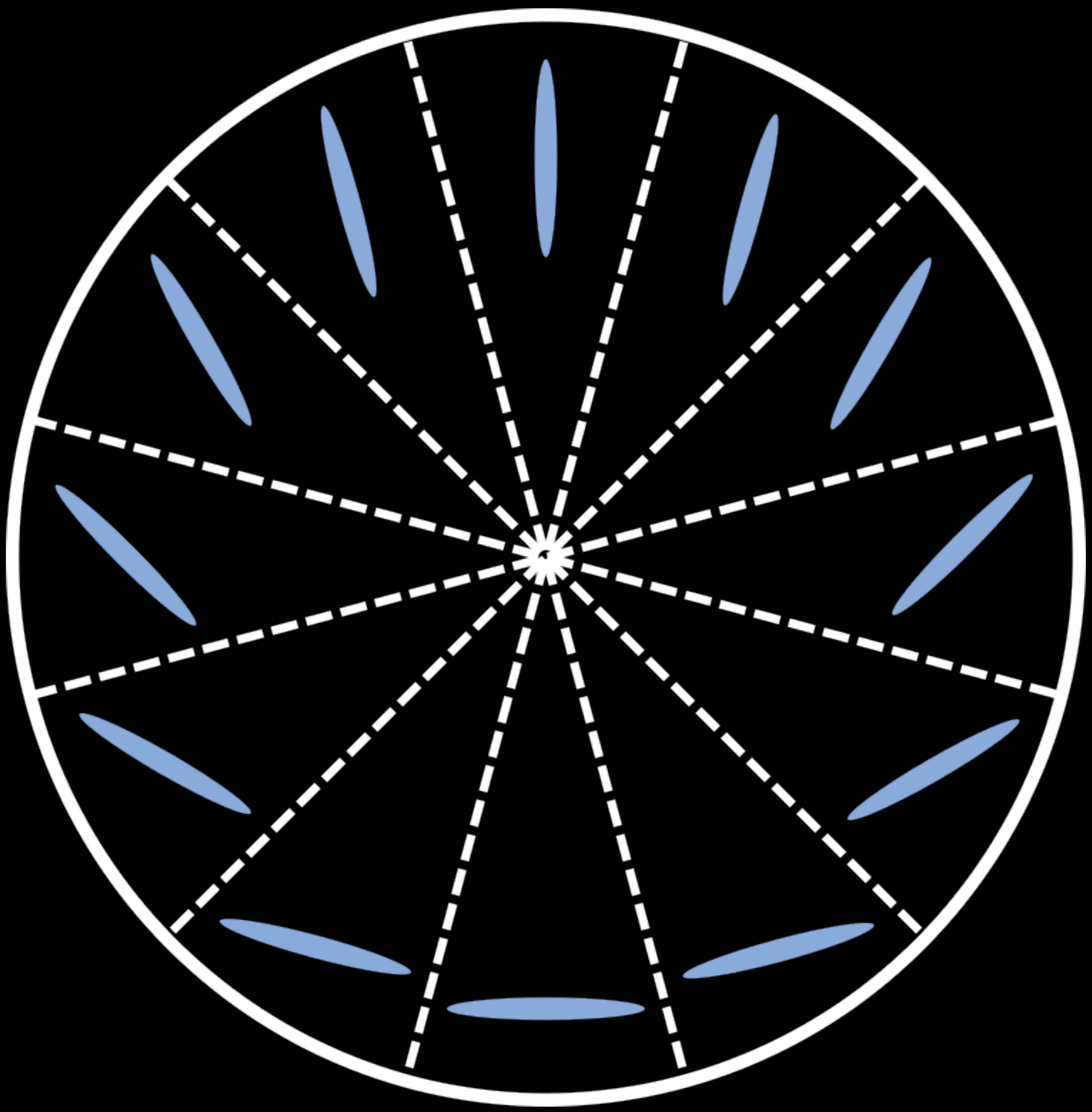
MPQ

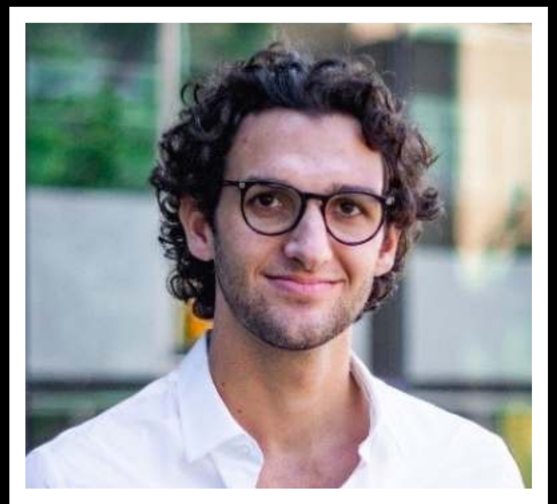
- ✦ Structured light beams have tailored spatial properties.
  - ✦ We can consider higher-order Hermite-Gaussian or Laguerre-Gaussian modes to be structured light
- ✦ **Most often, structured light beams are generated using a component that modifies the amplitude, direction, and/or phase of the incident optical fields in a pixel-by-pixel manner in the transverse plane.**
- ✦ **Examples of these components are:**
  - ✦ Spatial light modulators
  - ✦ q-plates, s-plates
  - ✦ Metasurfaces





- We can think of a q-plate as being a pixelated half-wave plate.
- It rotates the polarization of each pixel of the incident light by a predetermined, fixed angle.
- Depending on the polarization of incident light, the transmitted light can be imparted with radial polarization, azimuthal polarization, or orbital angular momentum.

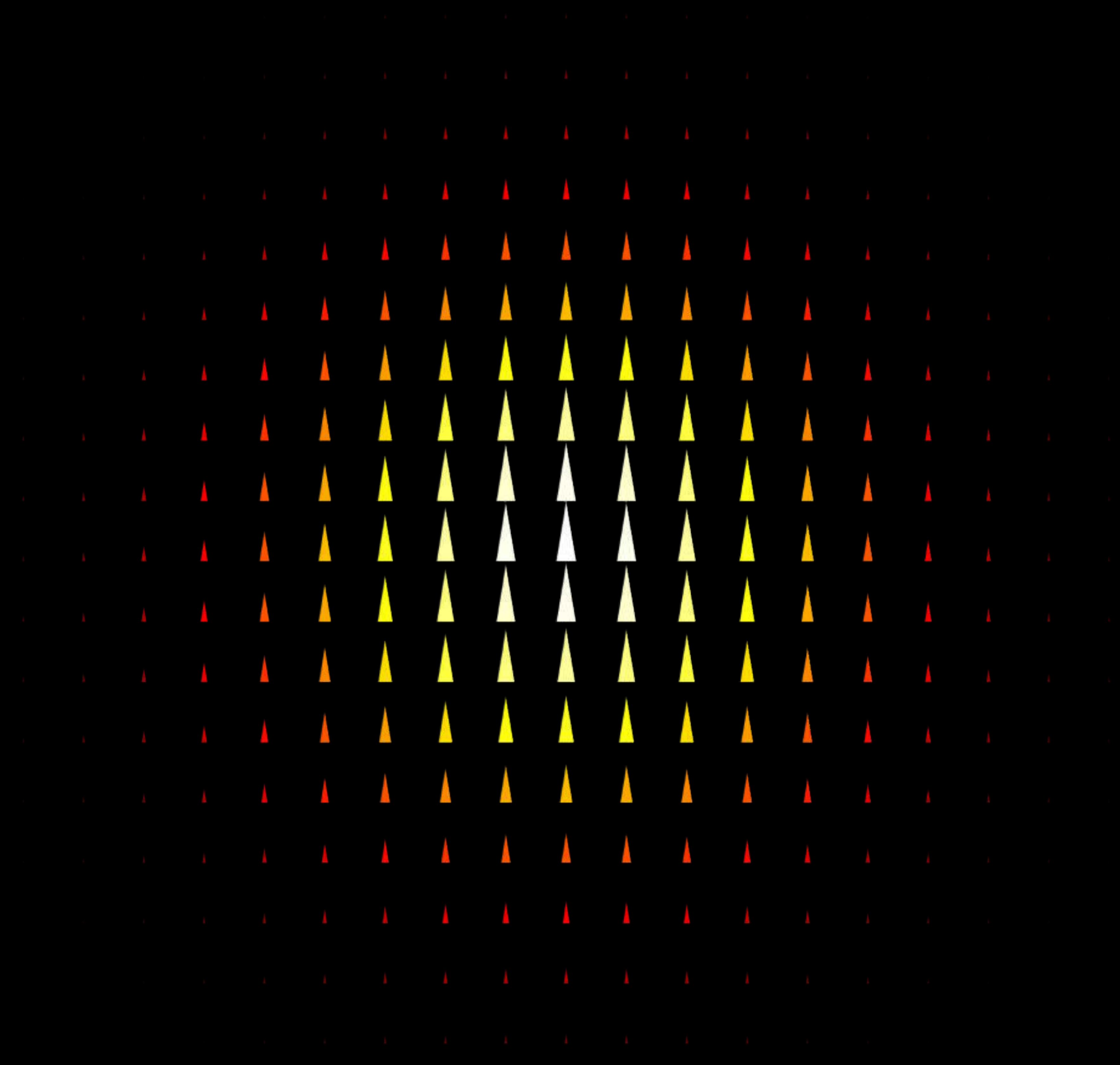
Prof. E. Karimi



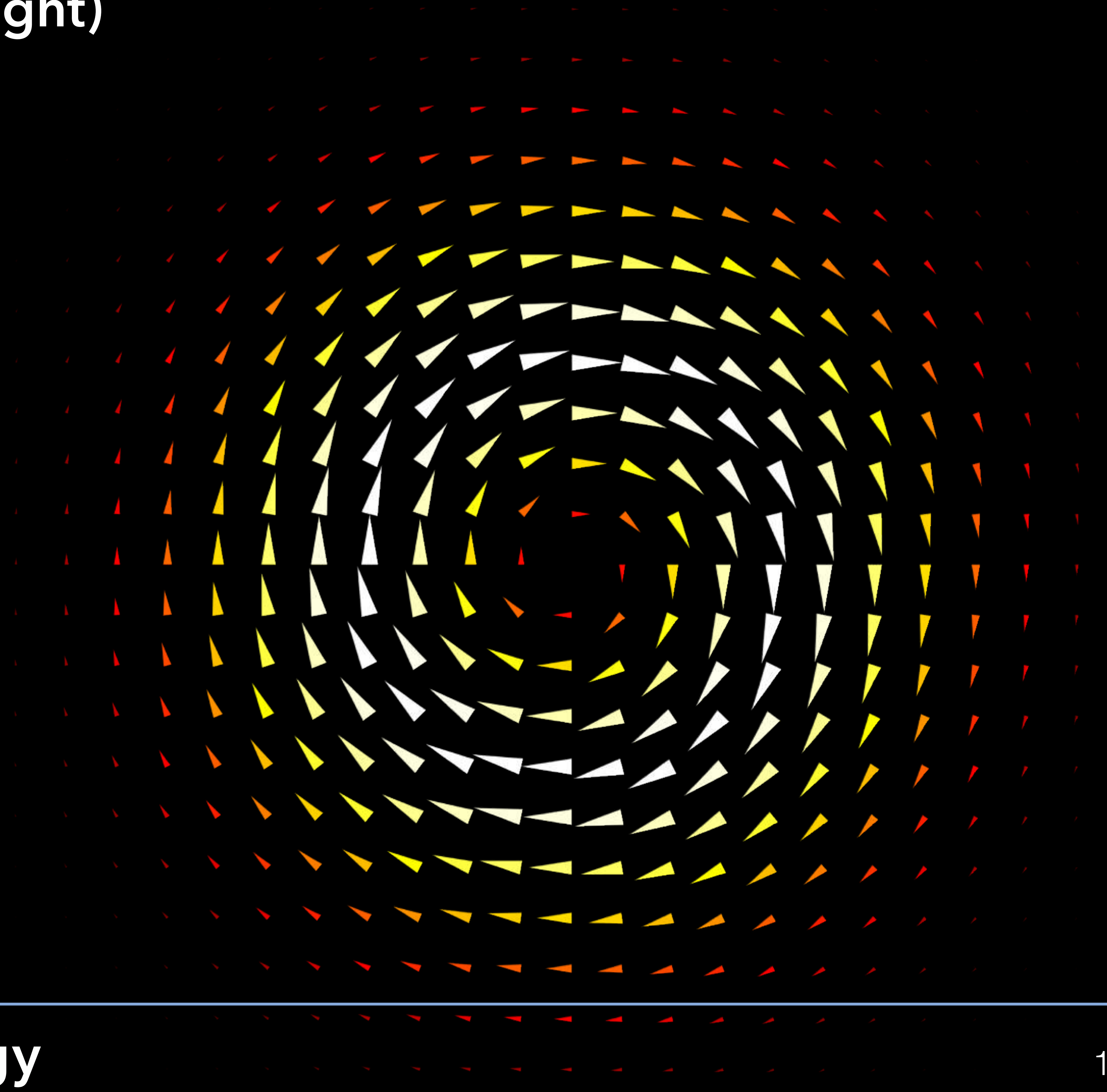
F. Hufnagel

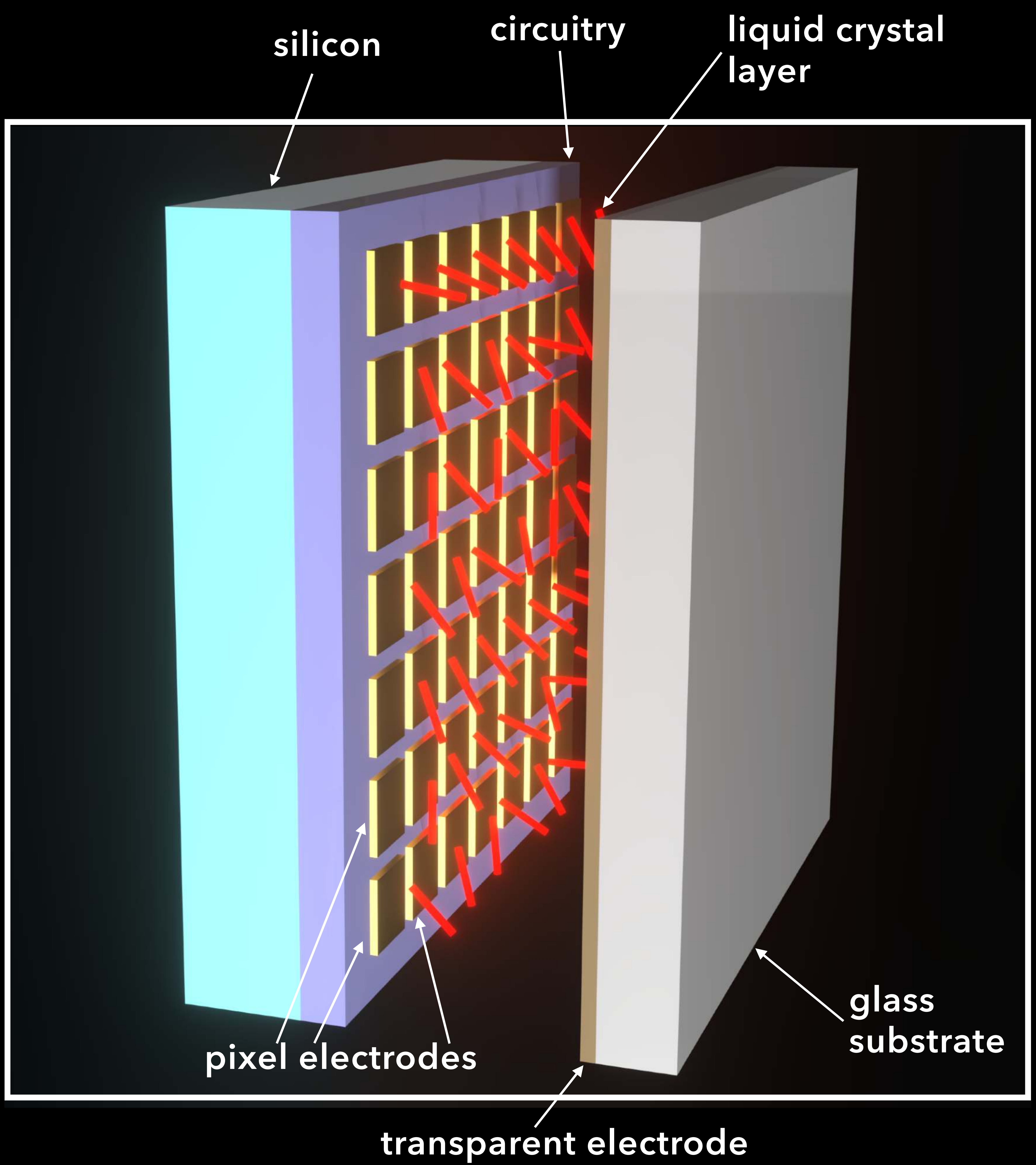
A. D'Errico

- The figures below show the electric field vectors of a laser beam. The local field amplitude is encoded onto the size and color of the triangles.
- If a Gaussian laser beam (left) is transmitted through a q-plate, it can be transformed into a cylindrical vector beam with azimuthal polarization (right)

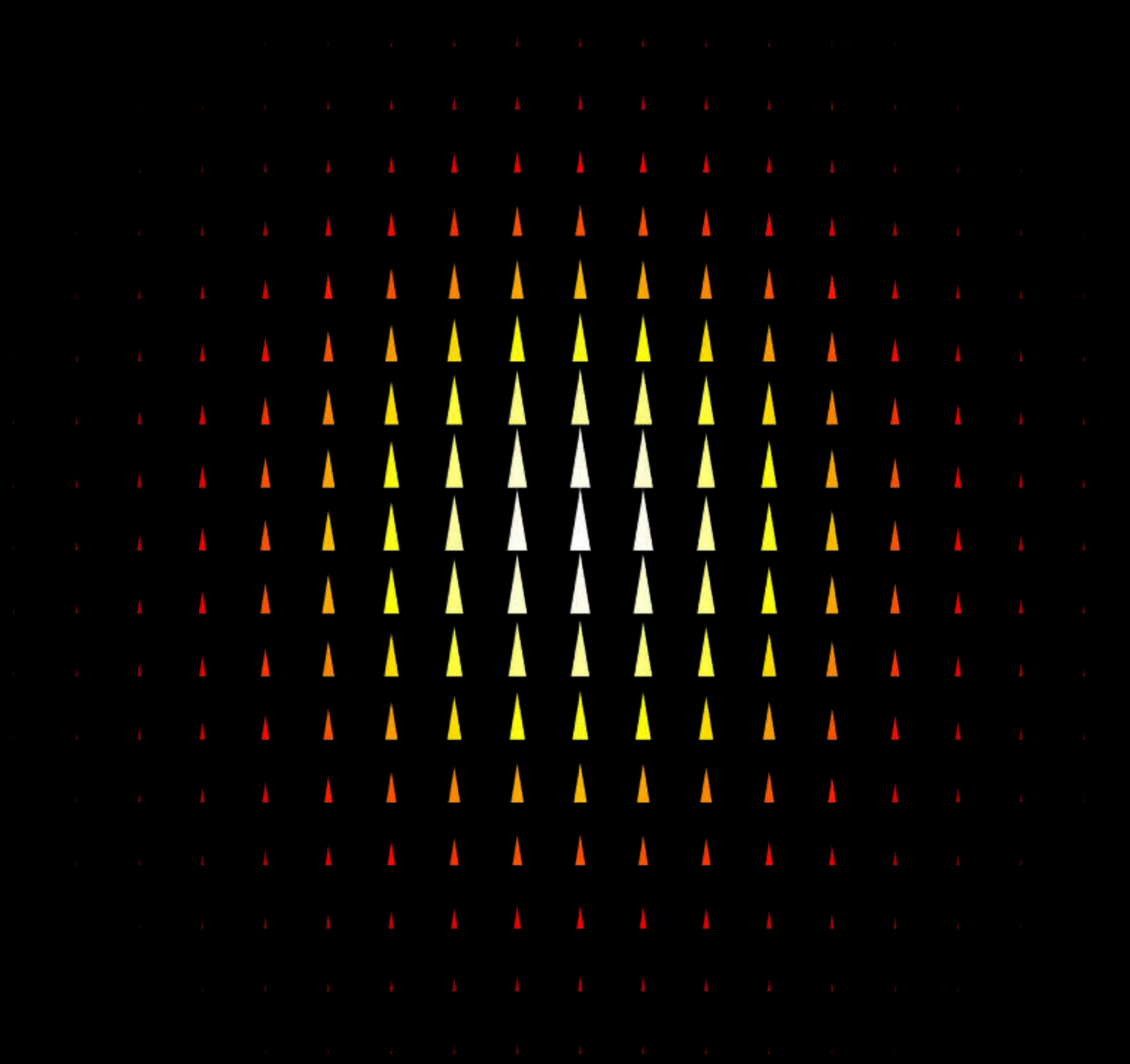


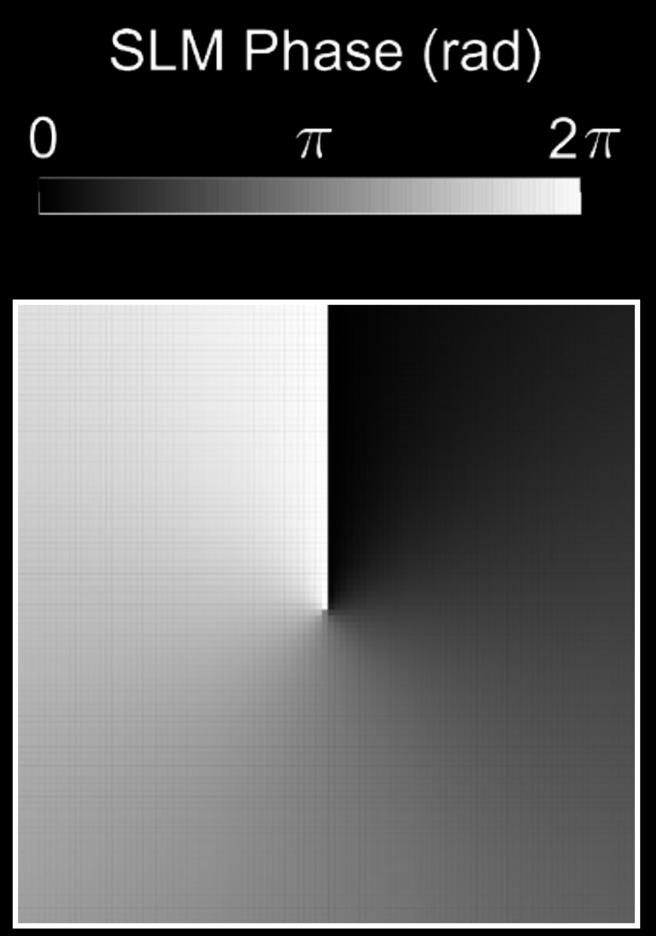
q-plate →





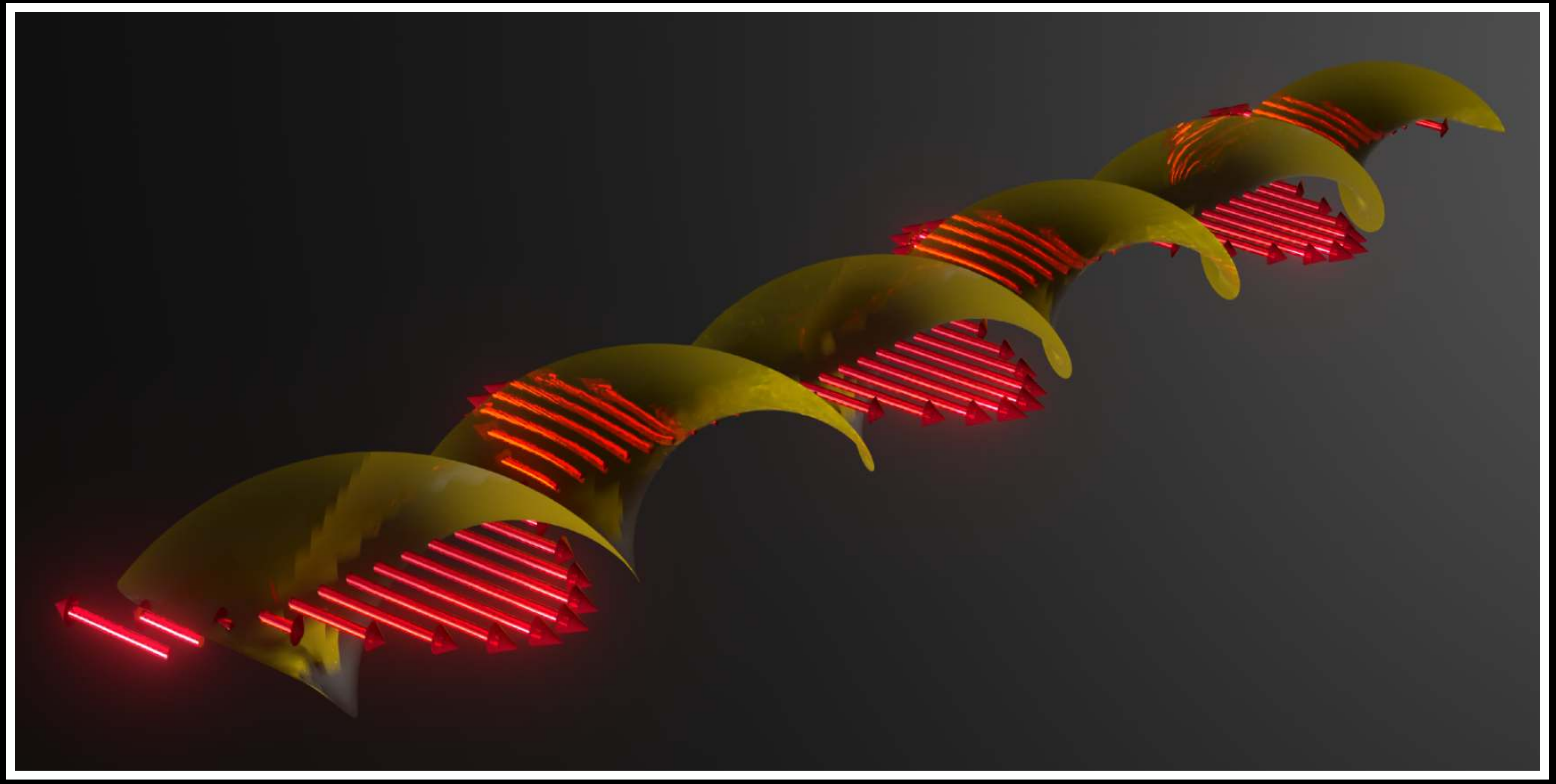
- Liquid crystal on silicon architecture
- The incident beam goes through the front window and reflects from the electrode array
- Electrode array enables local tilting of liquid crystal molecules, controlling the local refractive index
- The phase front of the reflected beam is sculpted by the SLM pattern
- This is a programmable megapixel array, so there is vast flexibility



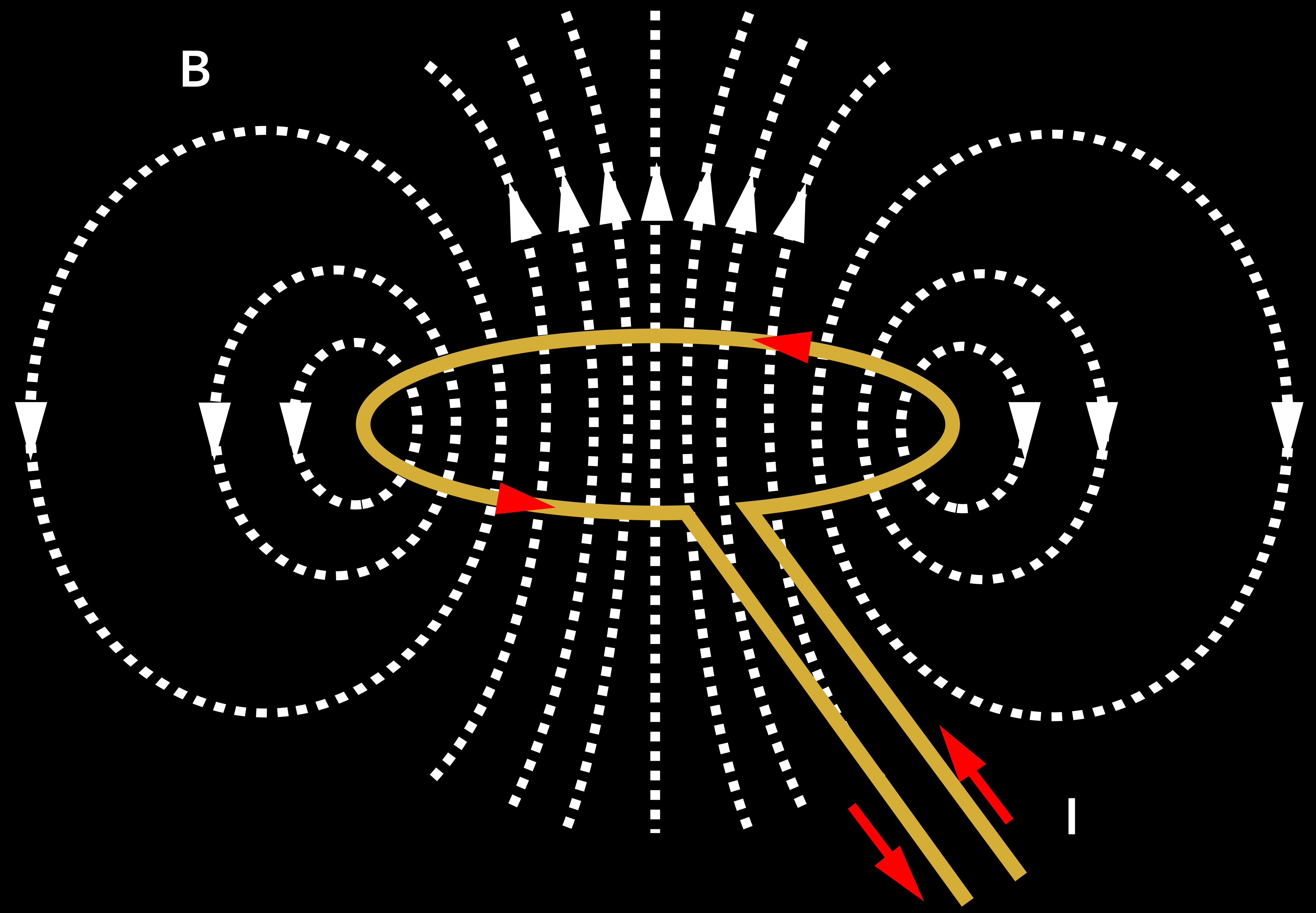


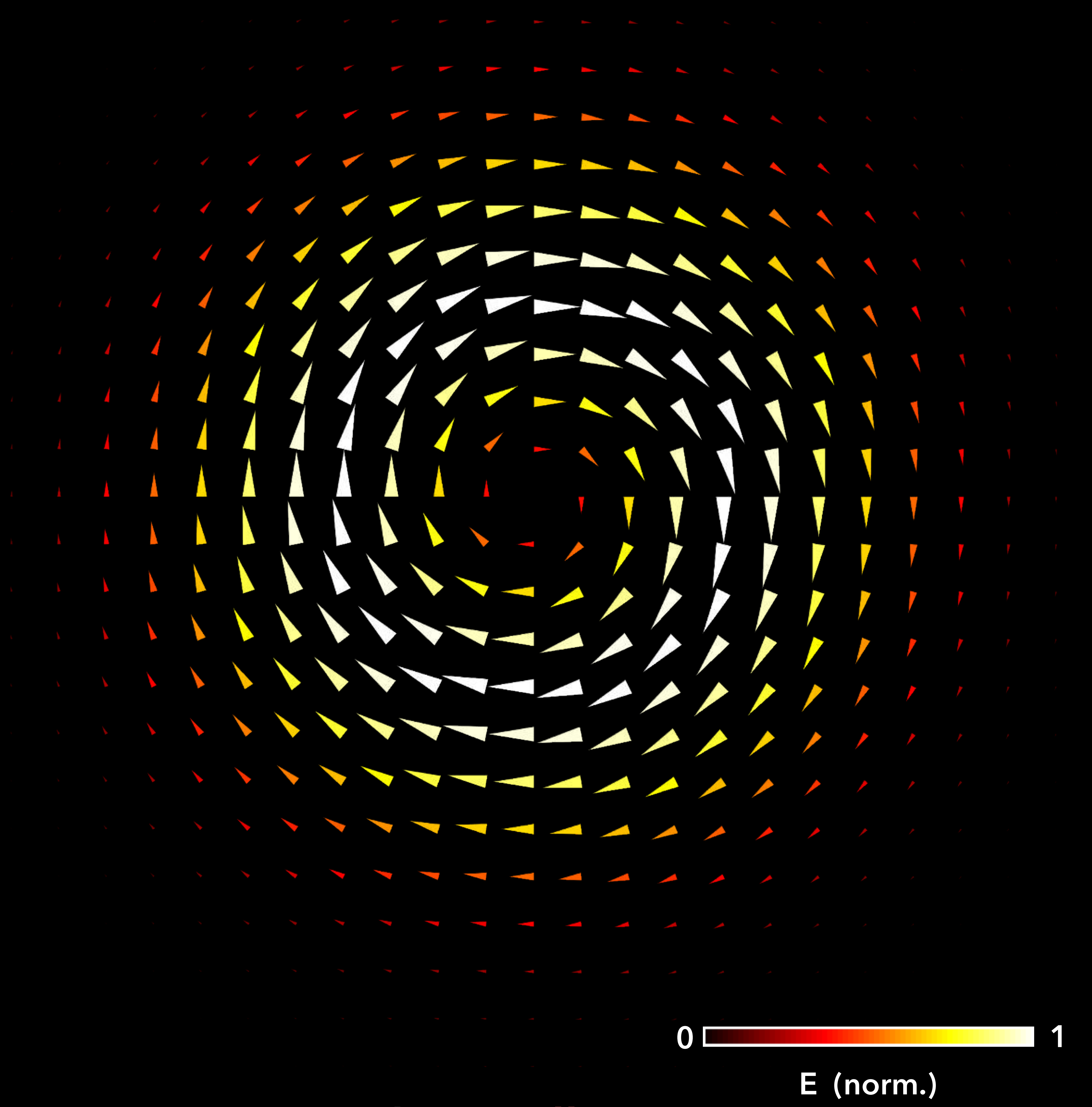
SLM

- Applying the phase pattern shown here introduces one unit of orbital angular momentum to the laser beam



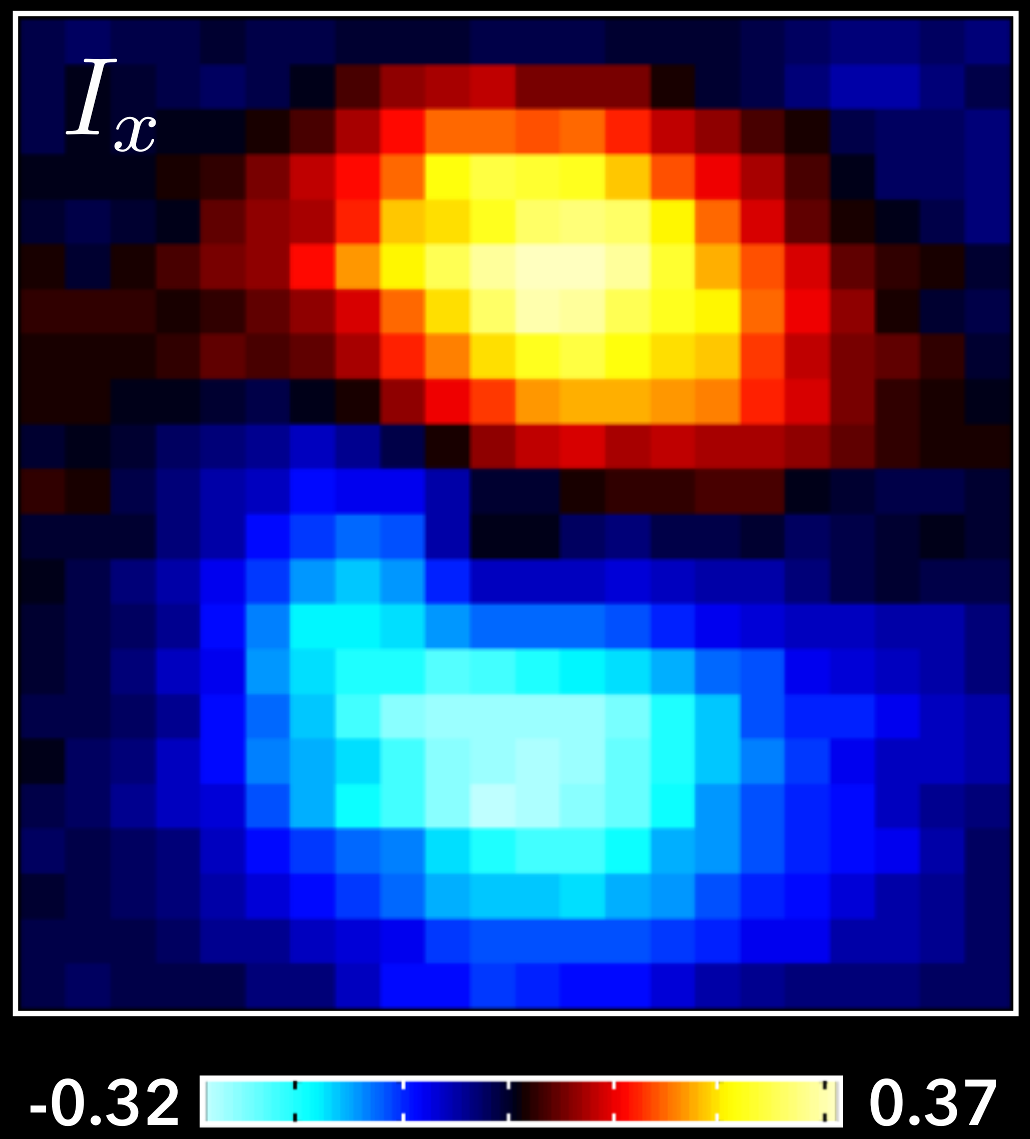
coherent control using structured light

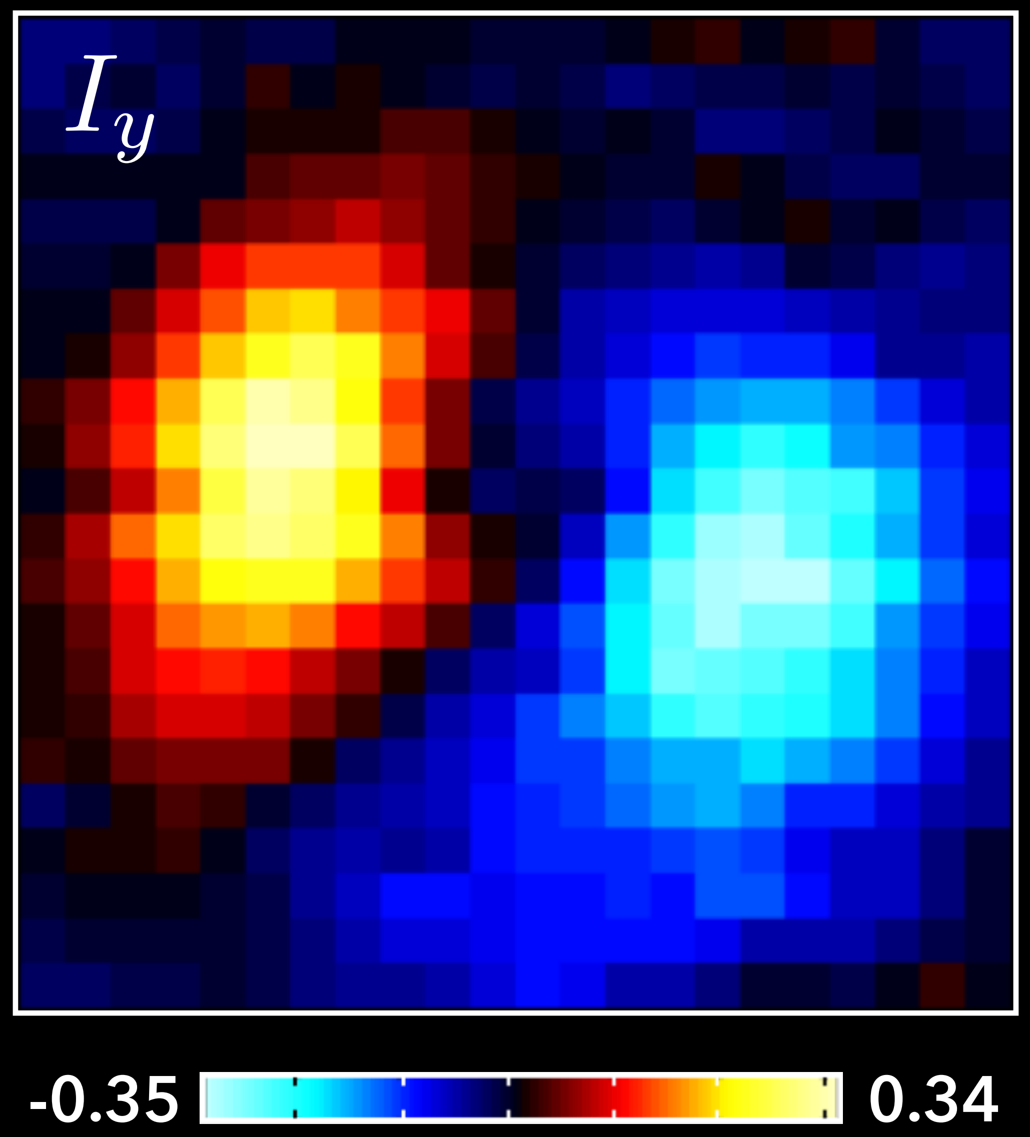




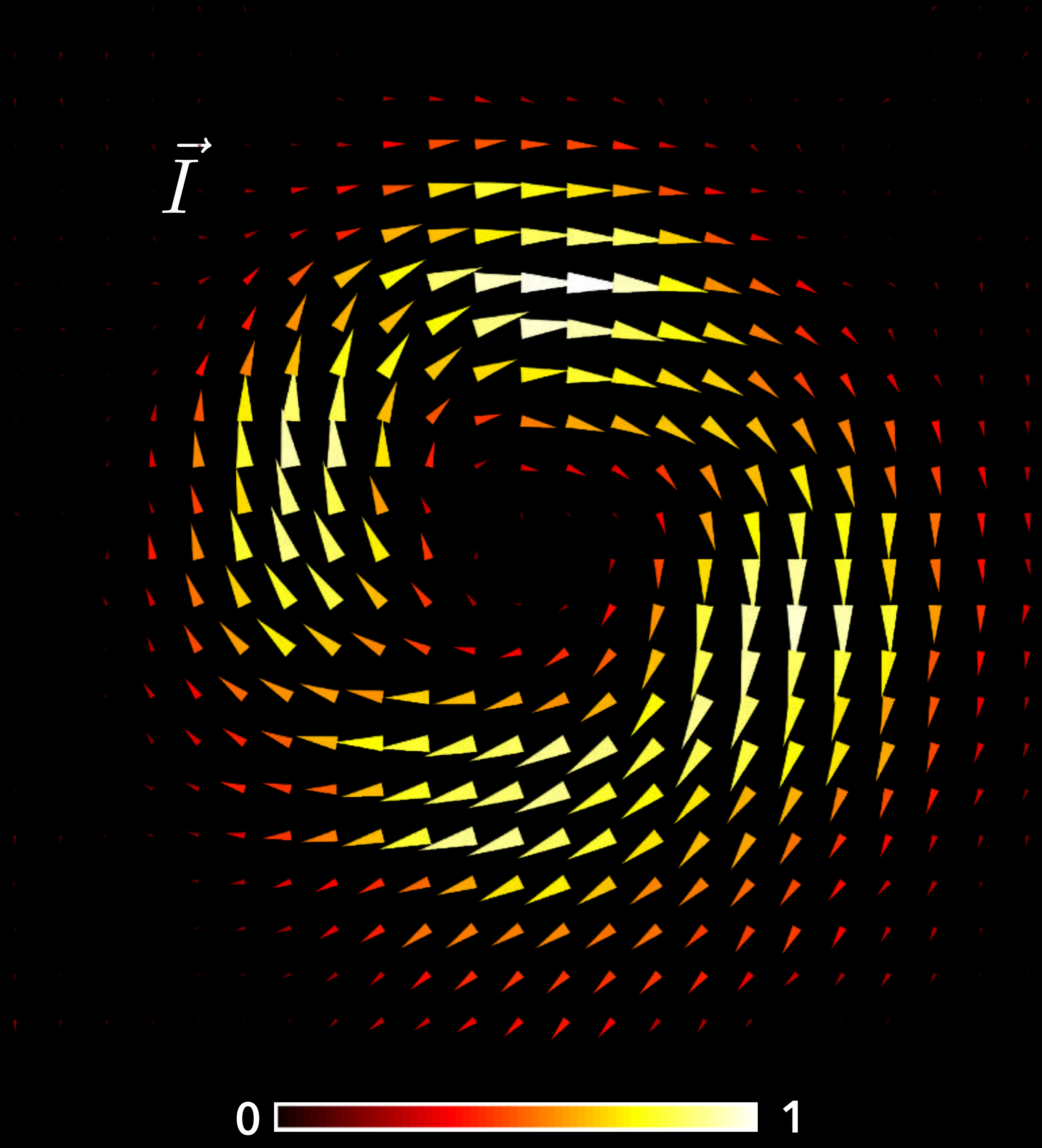
- ✦ We are particularly interested in generating ring currents and spatially-tailored THz pulses

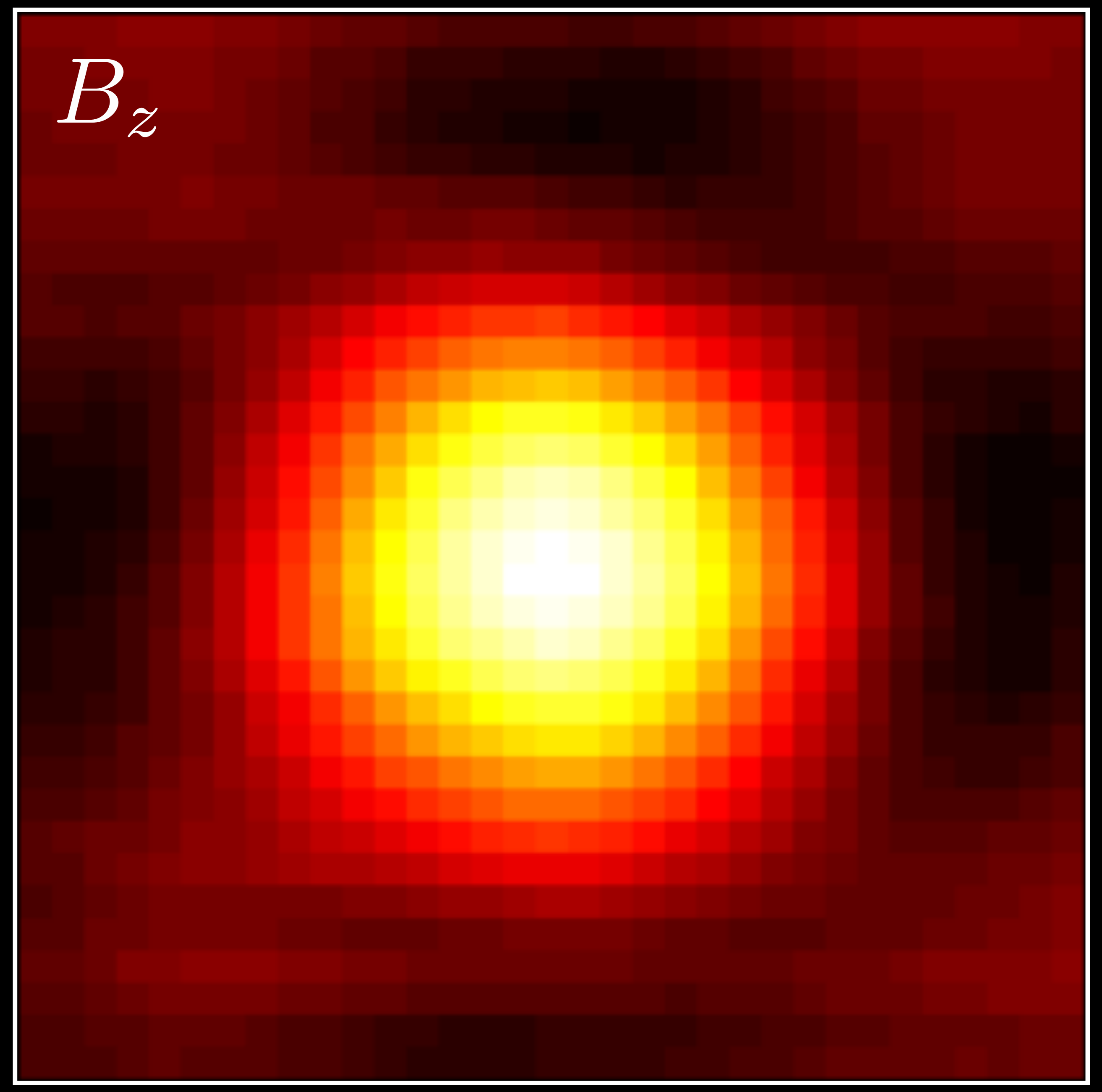

Detection
f = 0.5 m
BBO
Lock-in
reference
DM
2ω
ω
DM
WGP
QP
HWP
M
M
M
M
QP
WGP
HWP
$\Delta\varphi_{\omega,2\omega}$
Vector beam
generation
$\lambda_\omega = 1480\text{nm}$
$w_0 = 185\mu\text{m}$
$\tau_p = 42\text{fs}$
$E_p = 1.5\mu\text{J}$
$I_{0,\omega} = 6.7\ \text{GW}/\text{cm}^2$
$\lambda_{2\omega} = 740\text{nm}$
$w_0 = 185\mu\text{m}$
$\tau_p = 60\text{fs}$
$E_p \approx 25\text{nJ}$
$I_{0,2\omega} = 77.5\ \text{MW}/\text{cm}^2$
Counts (norm.)
Wavelength (nm)
1350 1400 1450 1500 1550 1600 1650
680 700 720 740 760 780 800 820
0 0.2 0.4 0.6 0.8 1

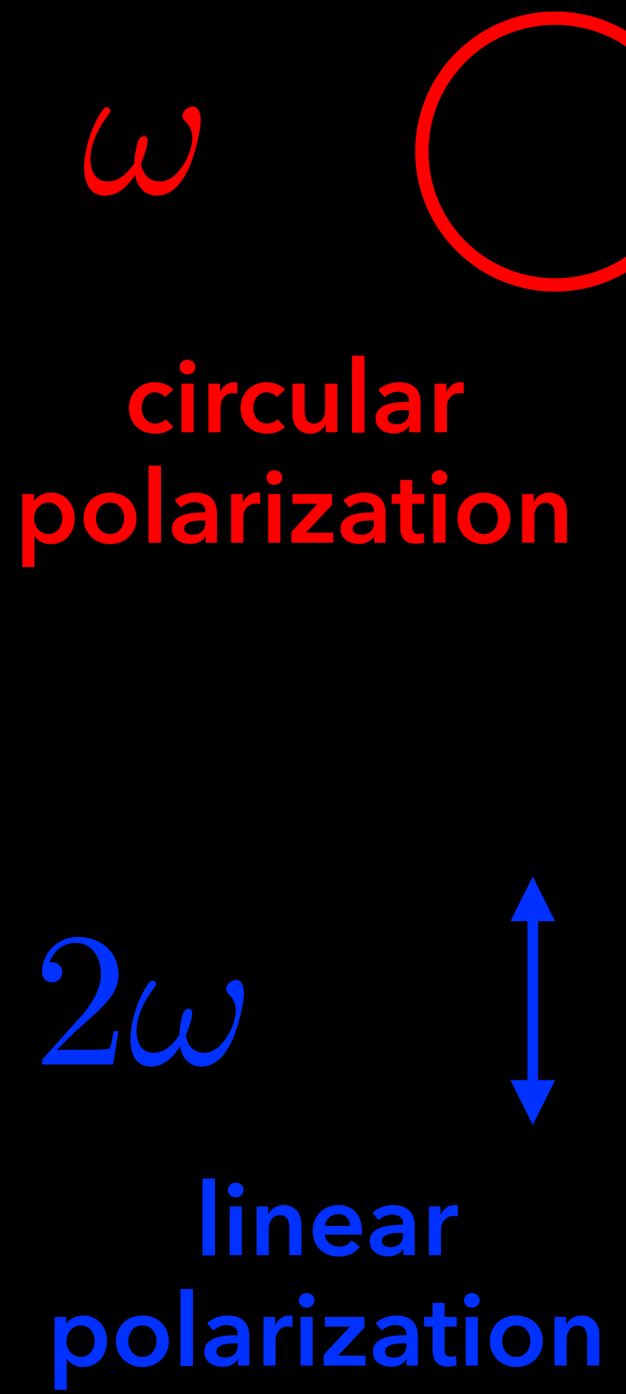


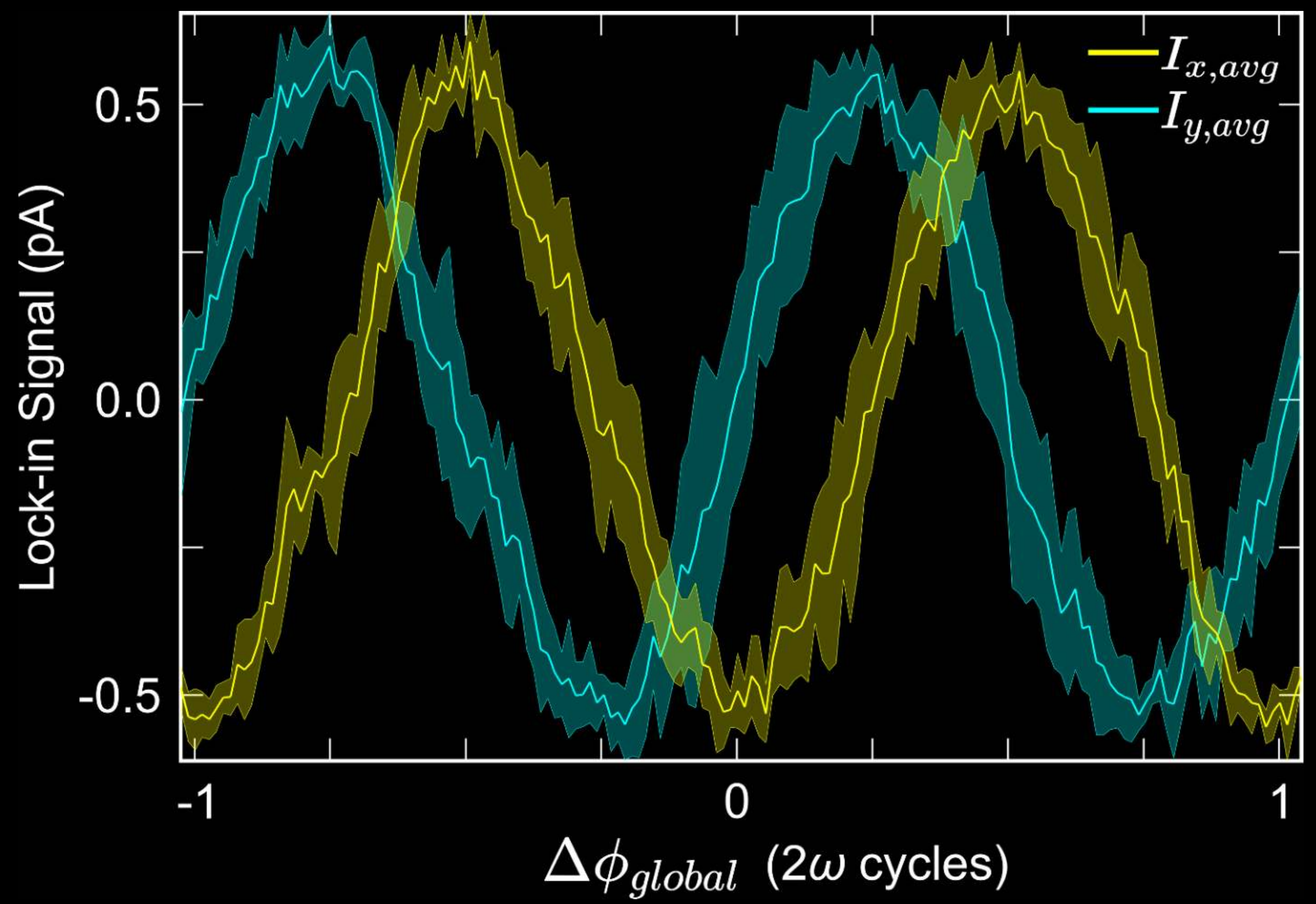


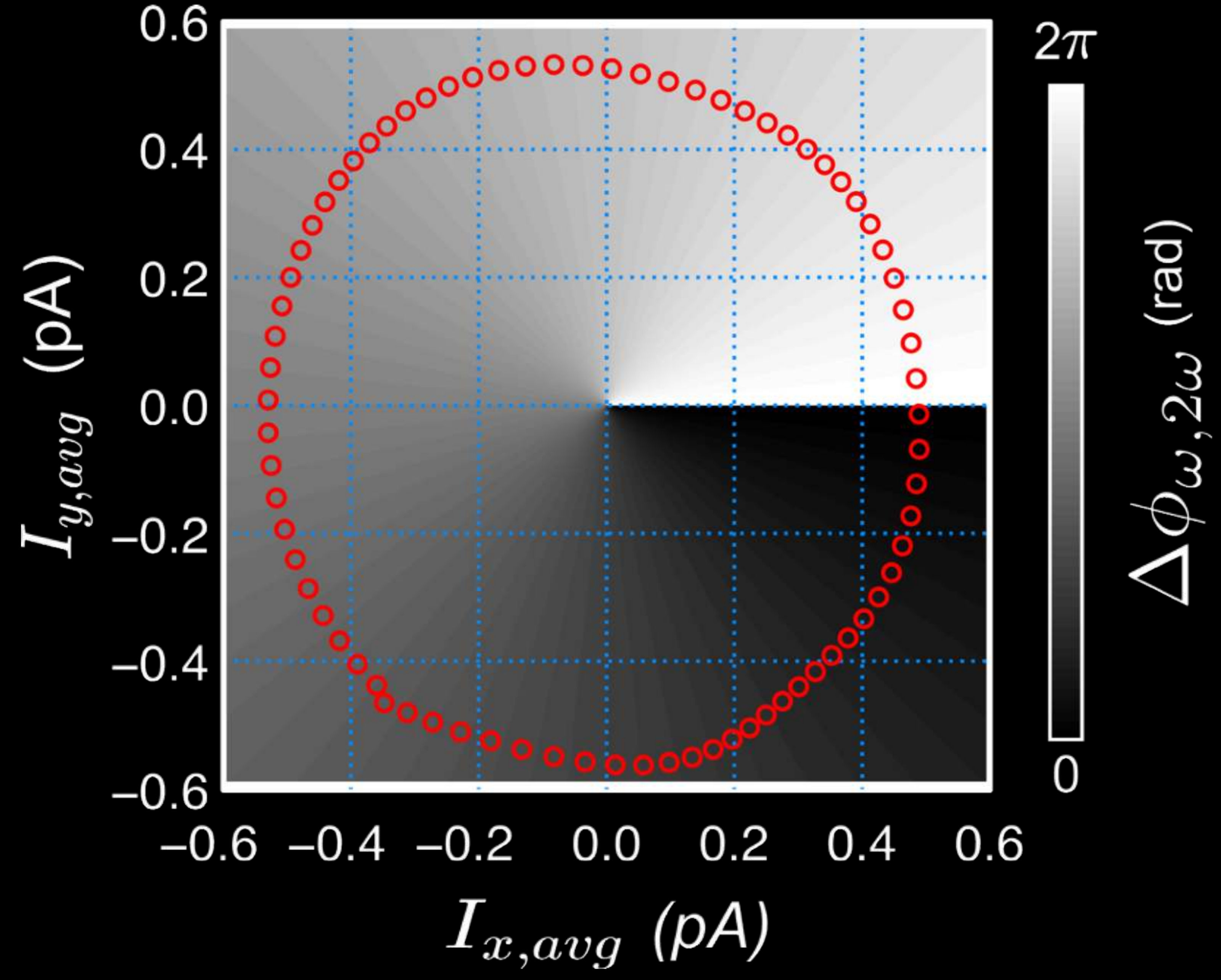
- Indpendent raster scans for the *x*- and *y*-component of current
- Retarded magnetic potentials are used to calculate magnetic field snapshots
  - 50-fs rise-time magnetic field





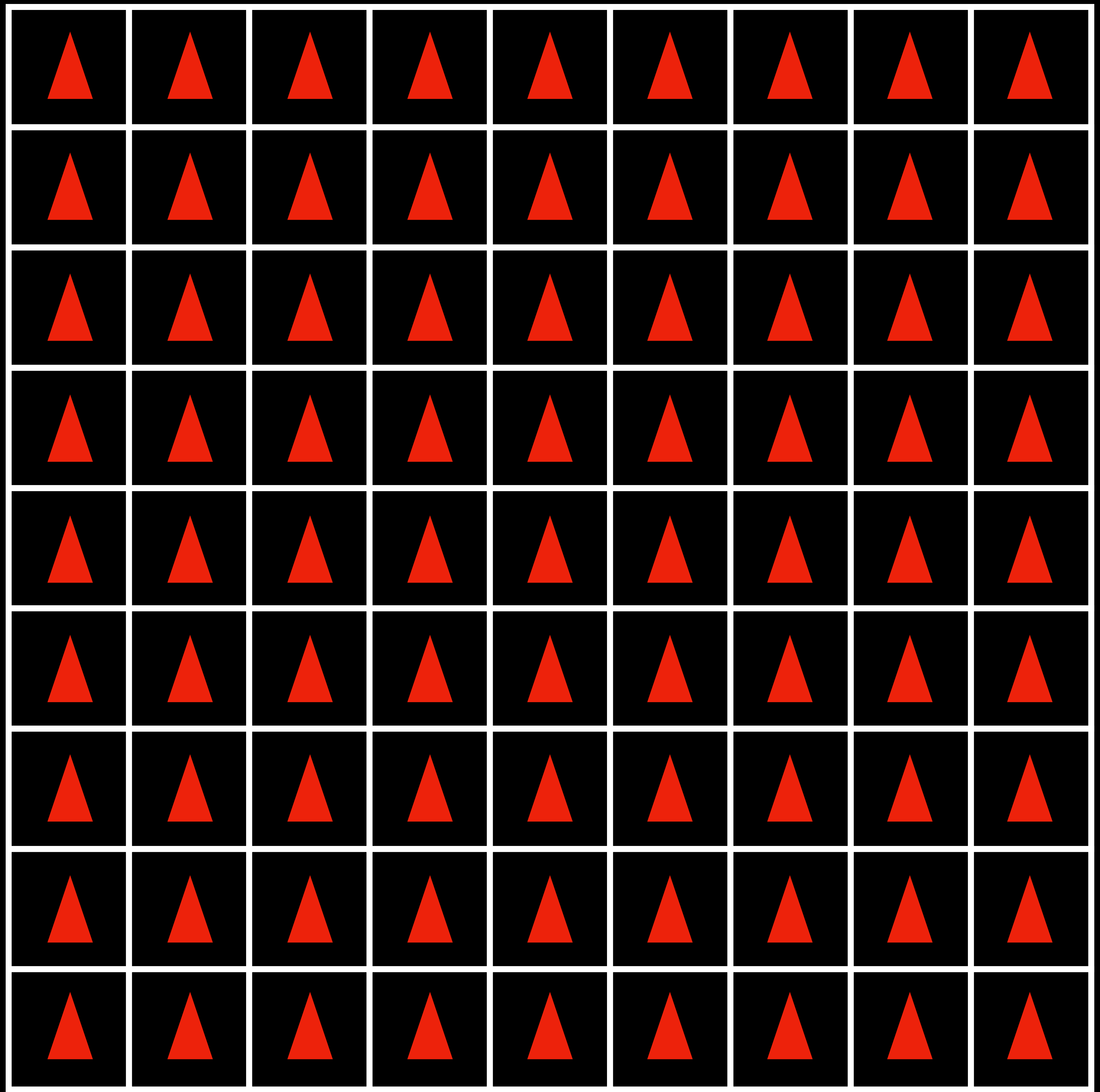


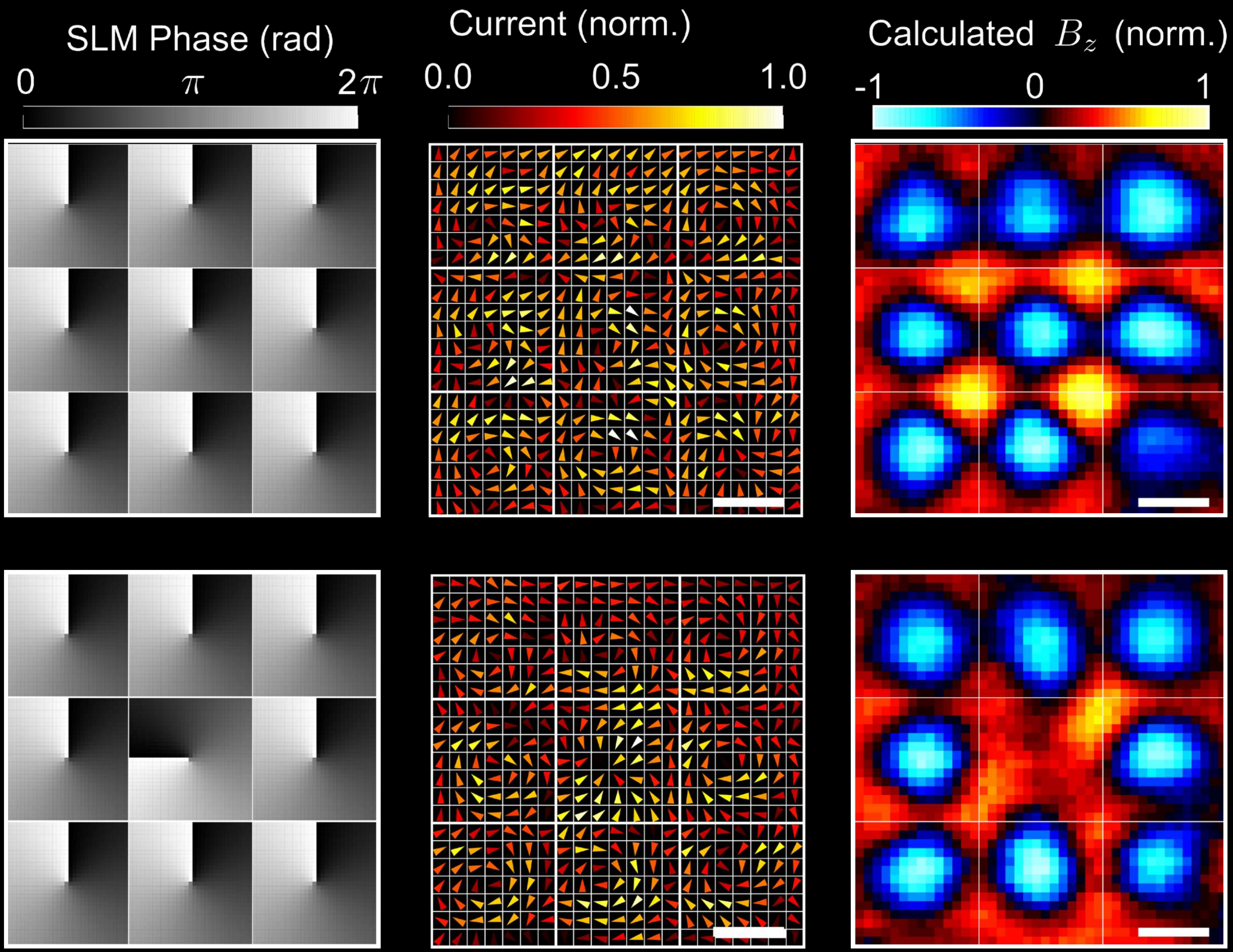




- Circular polarization can be used to steer the current direction
- *x*- and *y*-current components oscillate 90 deg. out of phase
- **Plotting parametrically, we see that current can be directed in any transverse direction**
- **If we can control the phase of pixels within our beam, we can create arbitrary optoelectronic circuits!**
  - **Programmable, pixelated phase control is achieved with an SLM**

Jana et al., *Nature Photon.* **15**, 622 (2021)

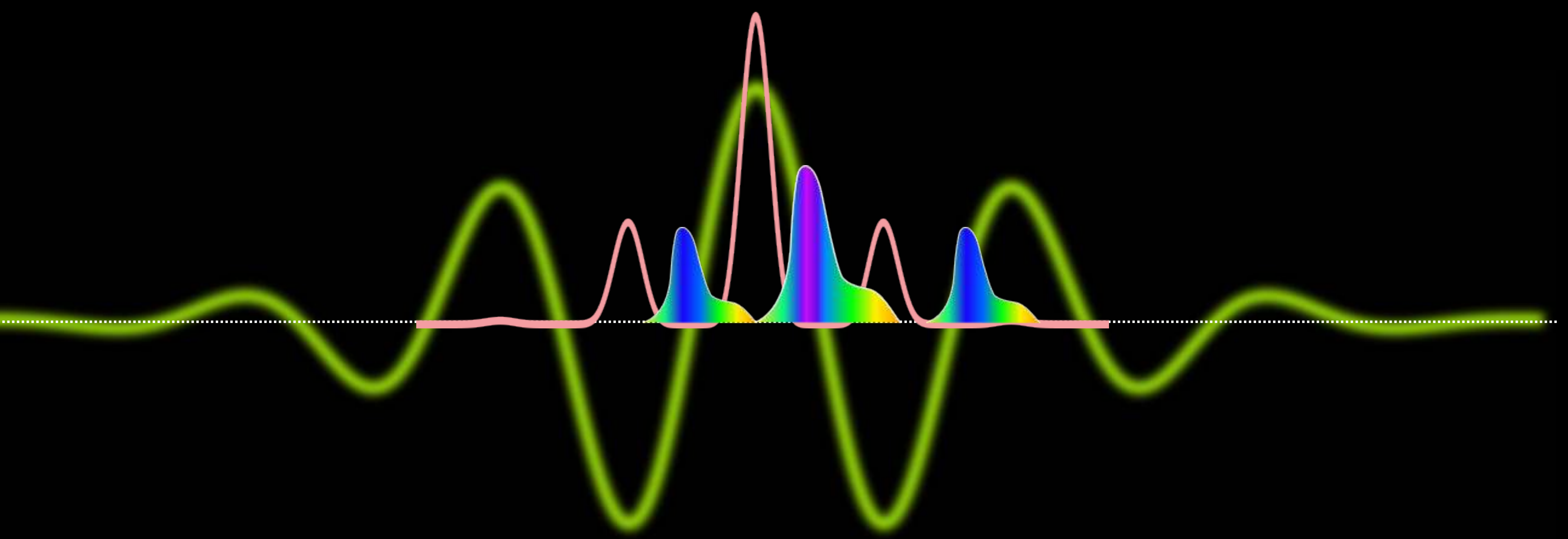


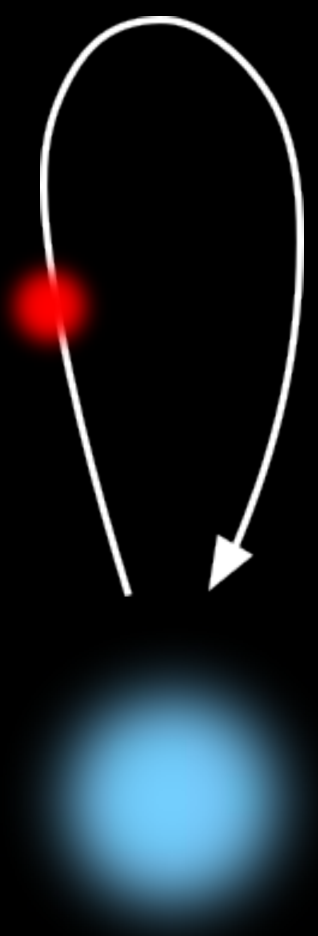


Jana et al., *Nature Photon.* **15**, 622 (2021)

magnetic field lattices

- Controlling the spatial structure of currents opens up new tools for studying magnetization dynamics, among other things
- We can also consider the temporal structure of our laser pulses
- Over the last 25 years, the attosecond community has devoted considerable effort to nonlinear compression of ~mJ-scale laser pulses
    - These are used to drive high-harmonic generation and, subsequently, to obtain isolated attosecond-duration XUV bursts

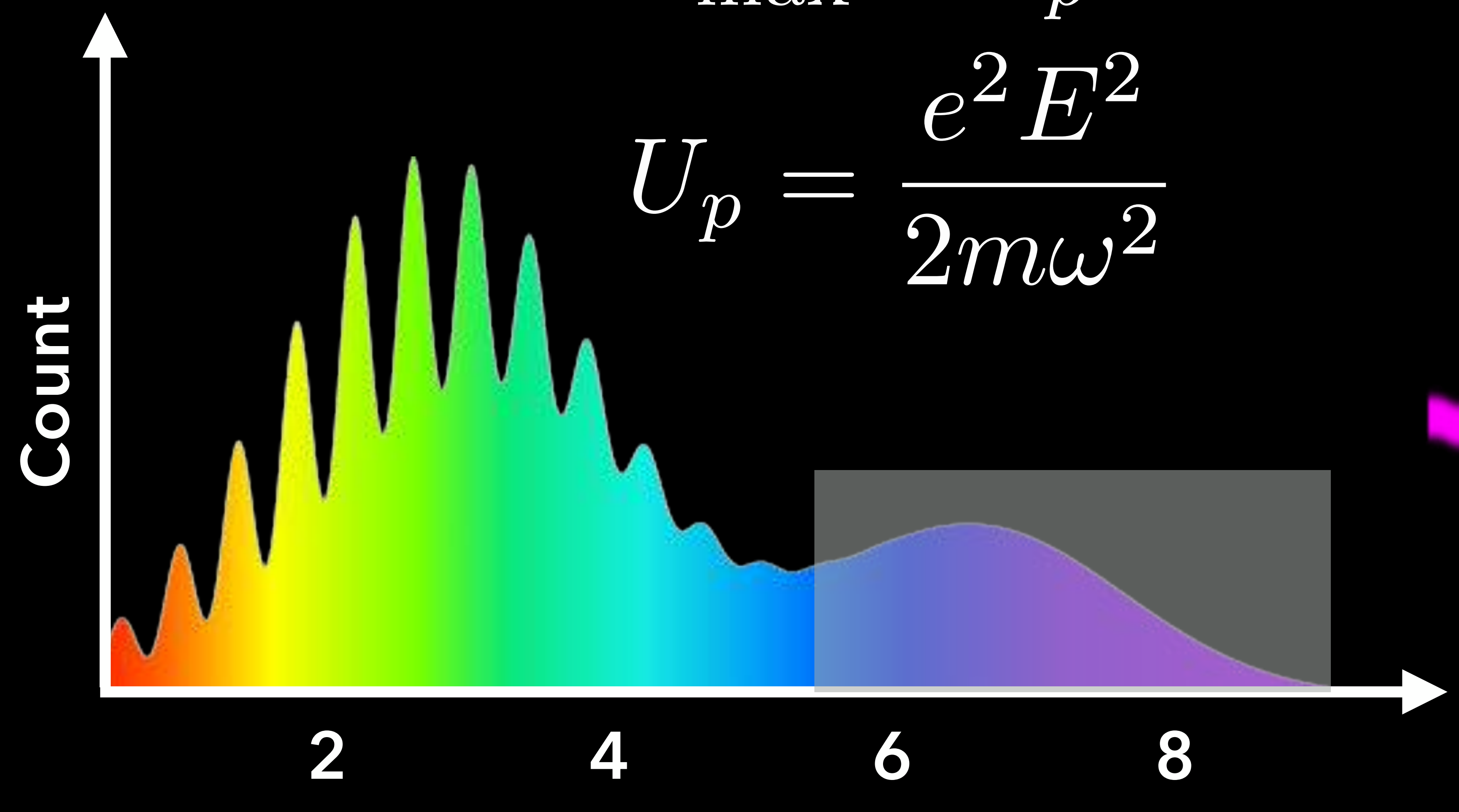




Paul Corkum
University of Ottawa

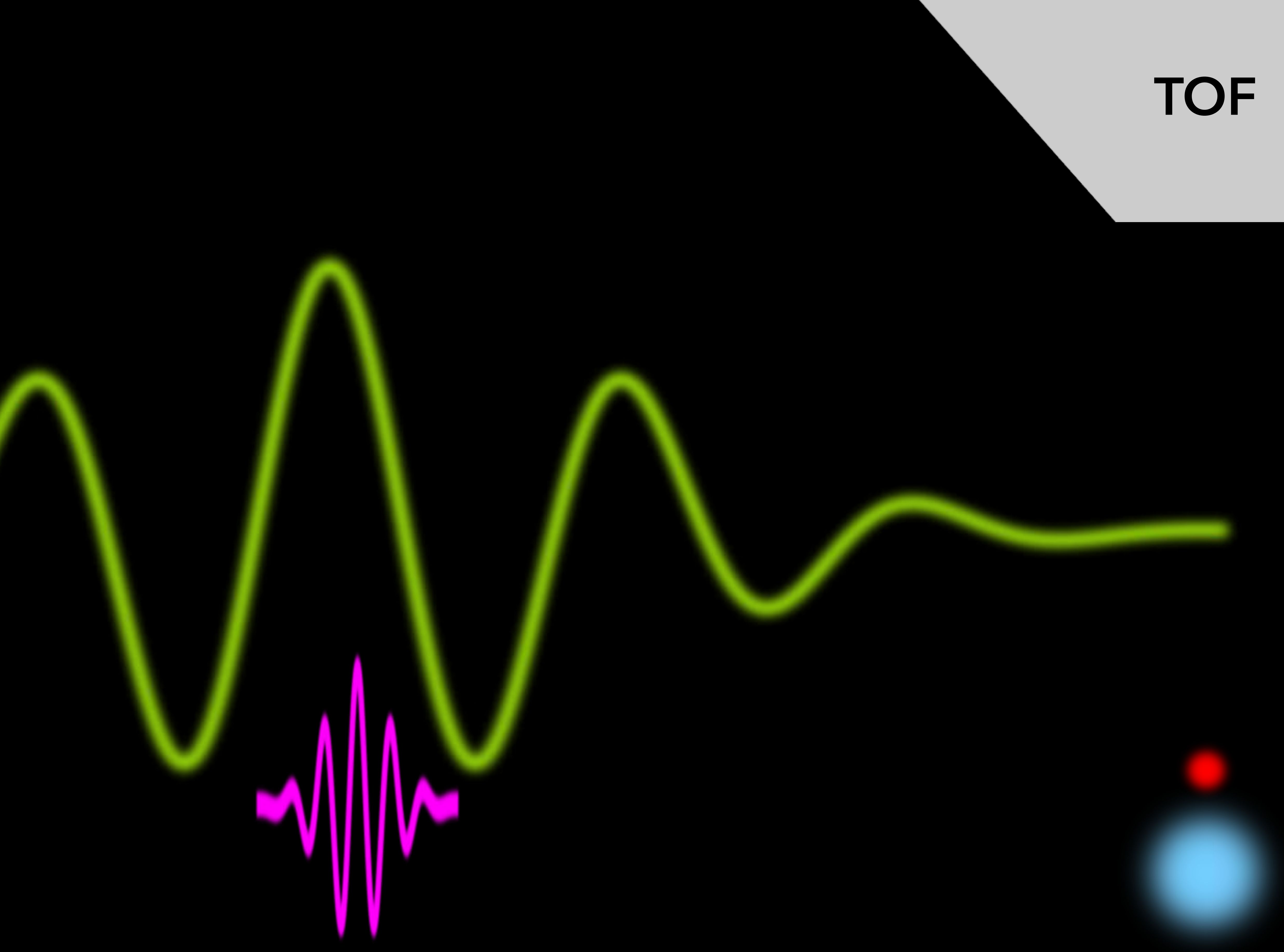
$$E_{\max} = I_p + 3.17U_p$$

$$U_p = \frac{e^2E^2}{2m\omega^2}$$



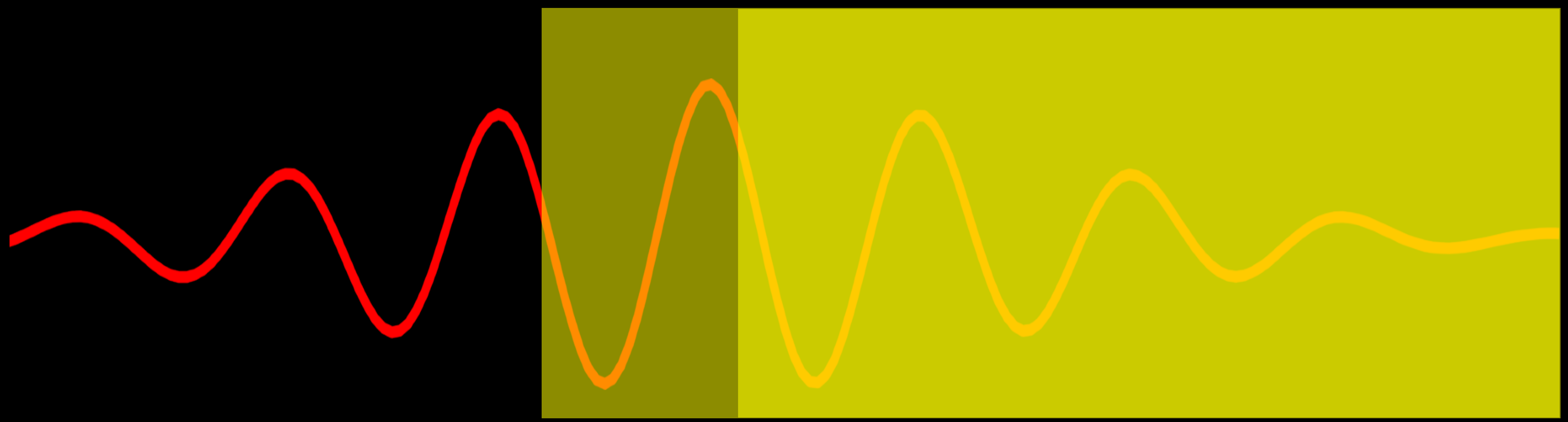
- Each of the main half-cycles of the pulse generates an XUV burst, producing a train of attosecond pulses
  - There are different schemes for isolating just one of them
- Amplitude gating: if a short enough pulse is used to drive HHG, the highest frequencies will only be generated during one half-cycle of the pulse

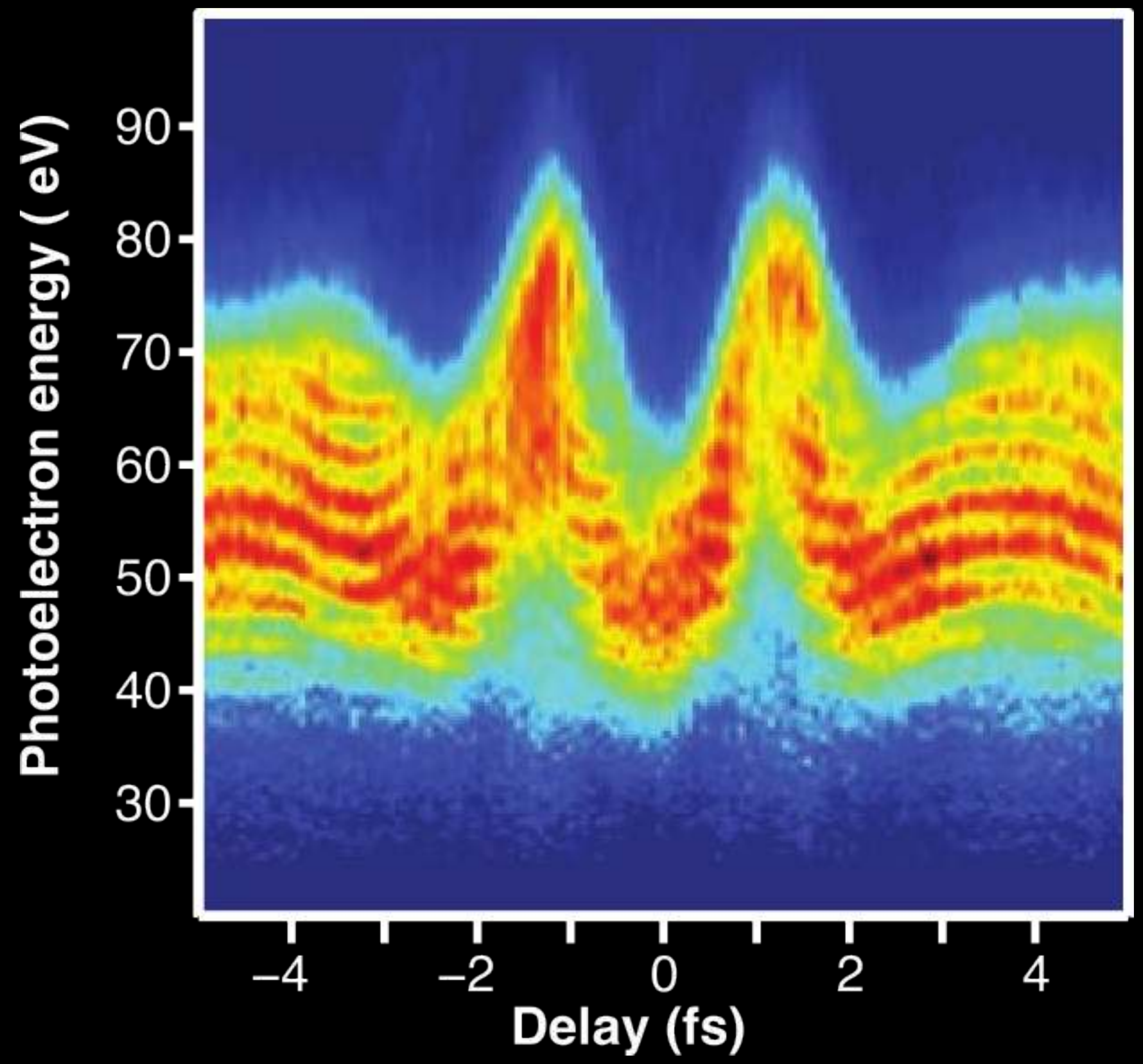
attosecond pulse islolation



✦ The instant of ionization and the optical waveform determine the final momentum of the electron

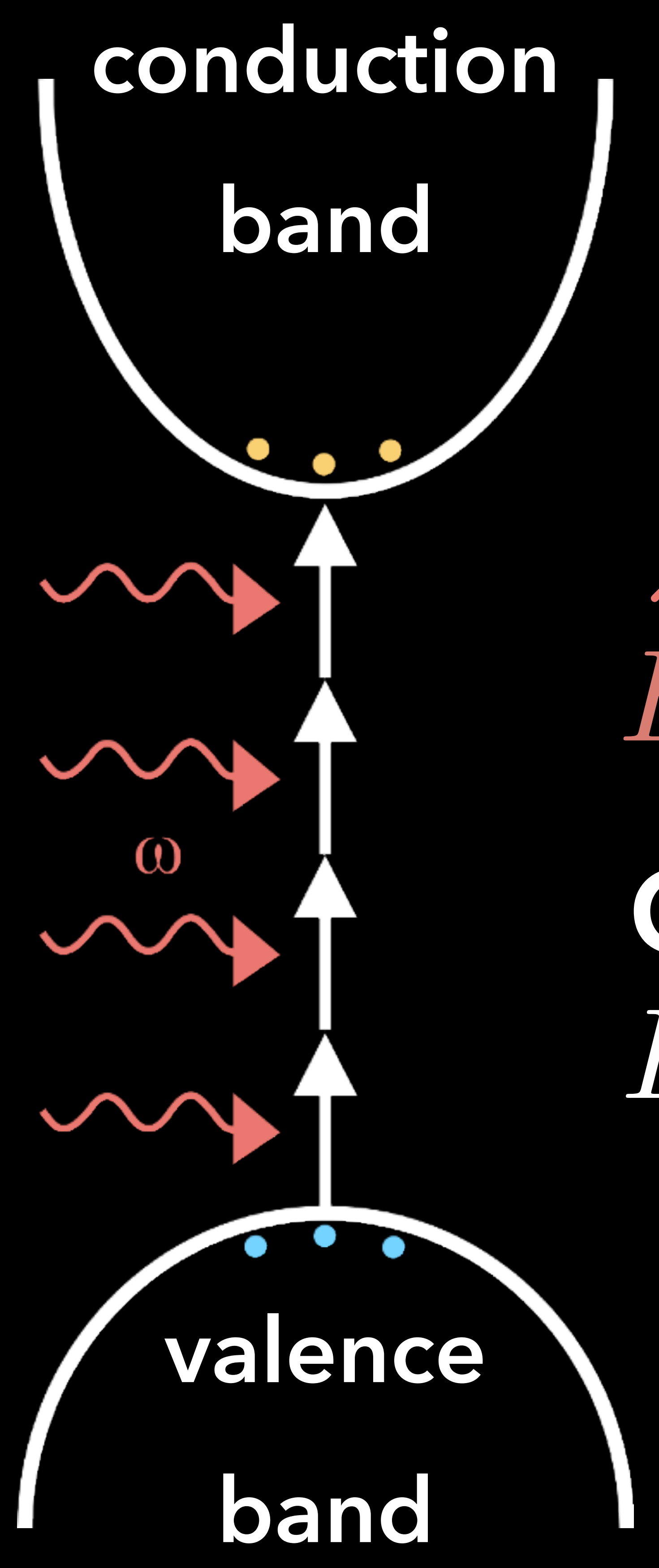
$$v(t) \propto \int_t^{\infty} \mathrm{d}t' E(t')$$





- The streaking spectrogram captures the vector potential of the laser pulse
    - A retrieval is performed to obtain the electric field
- The conceptual simplicity of this technique has made it the "gold standard" of optical waveform sampling

Goulielmakis et al., *Science* **320**:1614 (2008)

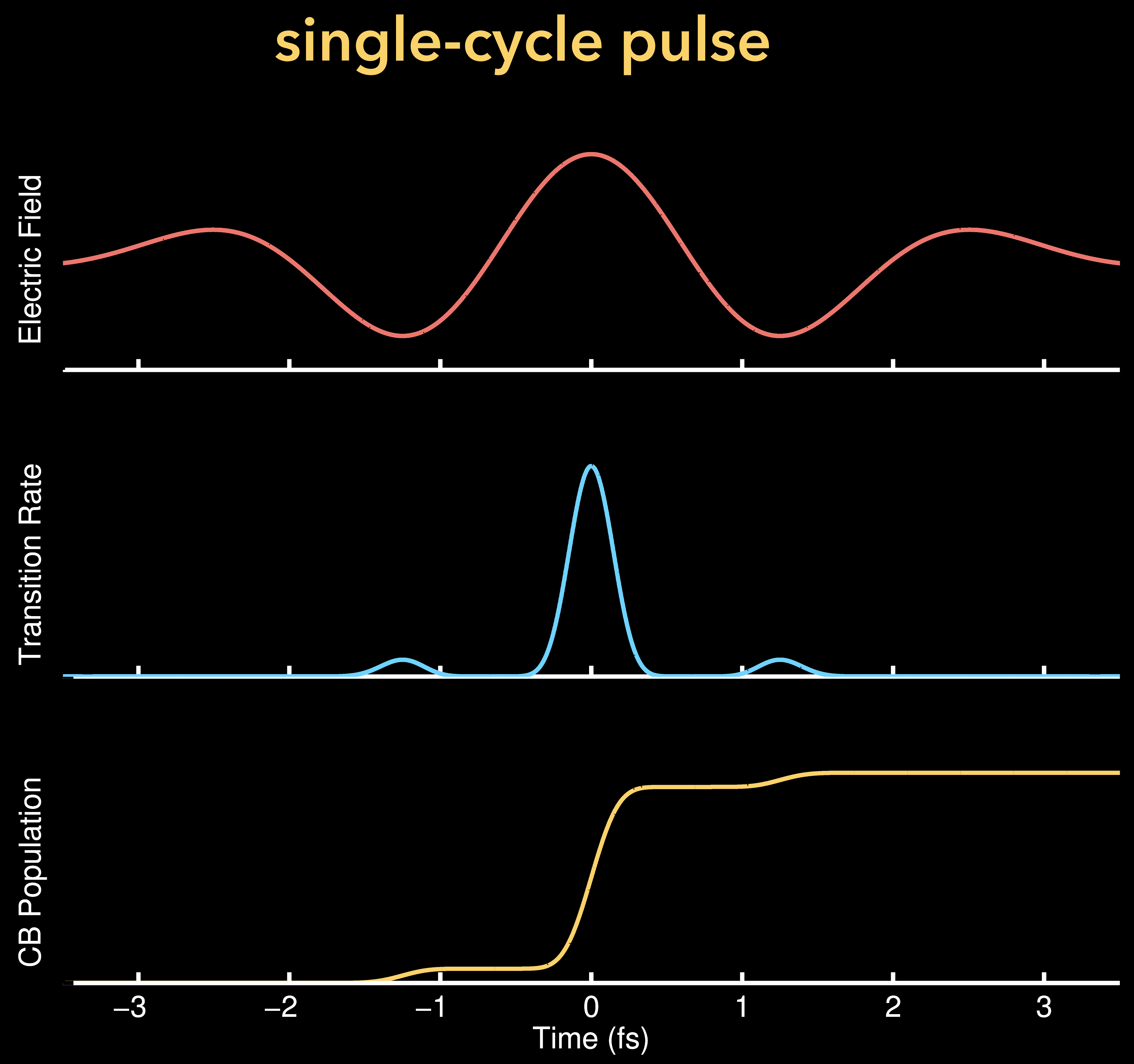


$\lambda = 750 \text{ nm}$

$E_{\text{phot}} = 1.65 \text{ eV}$

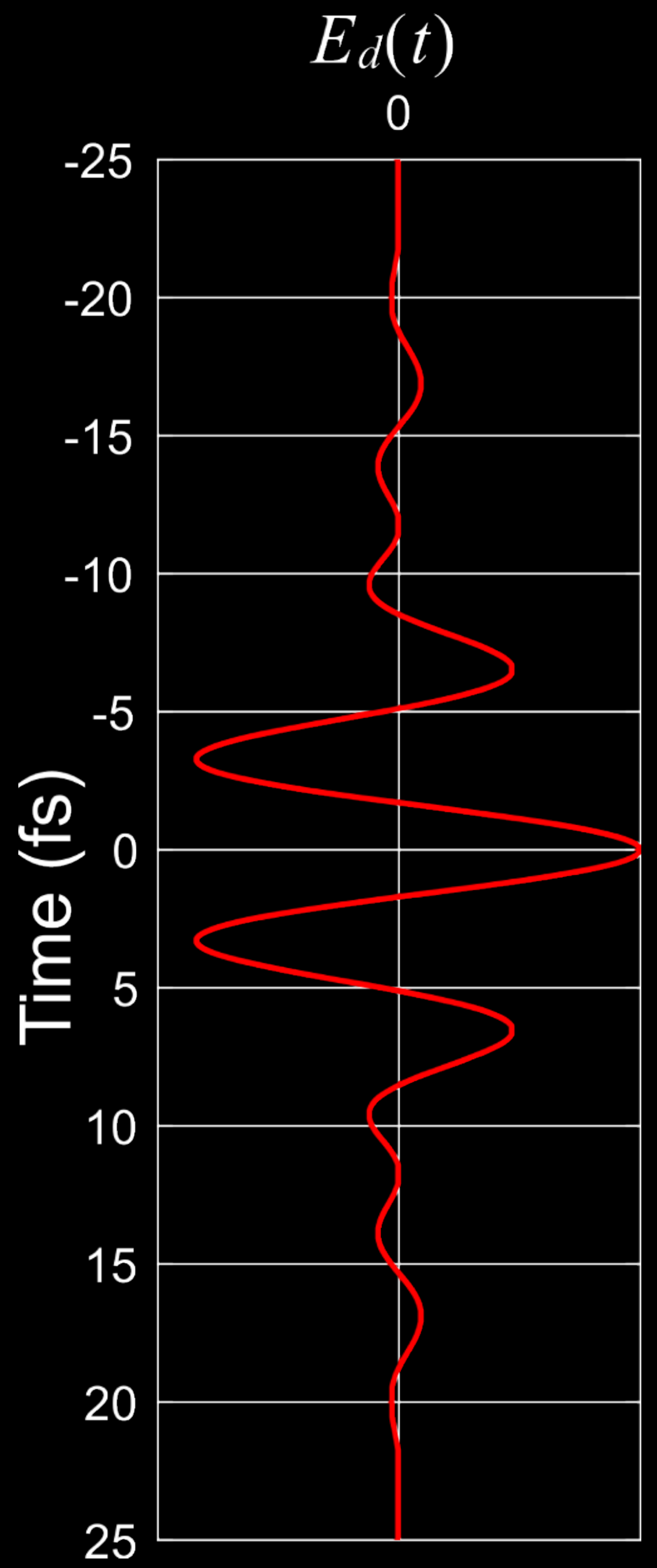
Quartz

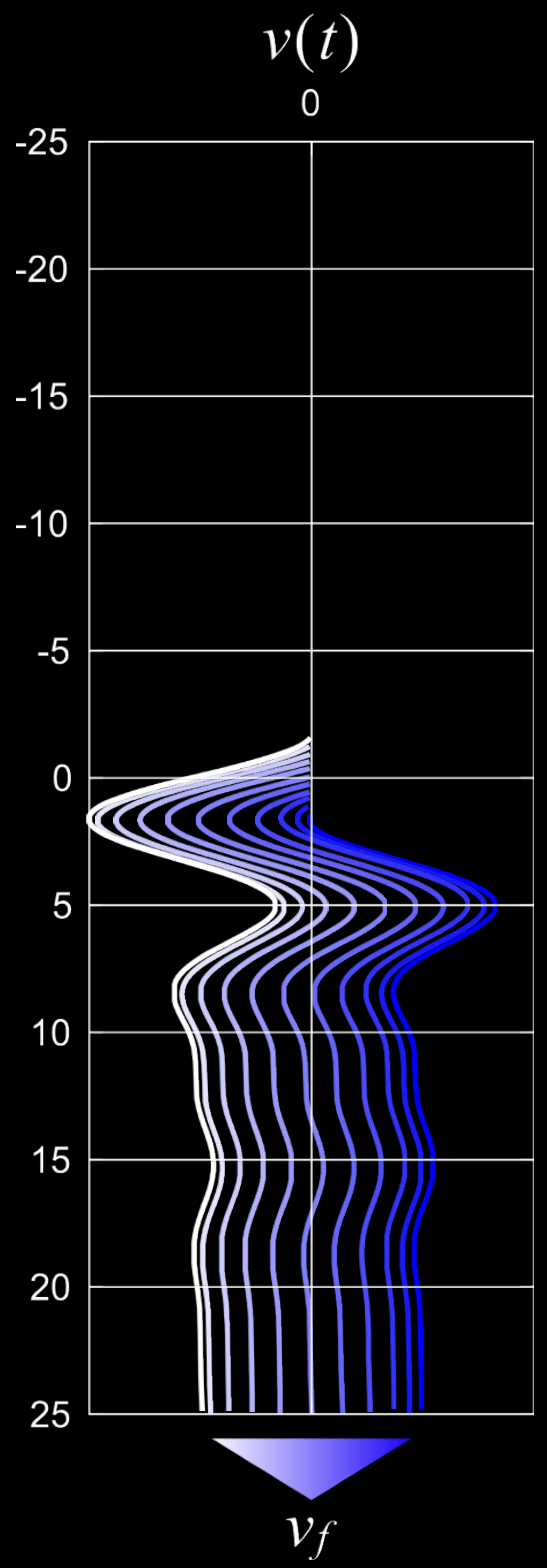
$E_g = 9.3 \text{ eV}$



- The separation between energy bands in solids can be comparable to that between energy levels in atoms
- For a sufficiently short pulse, the transition rate from the valence band into the conduction band can be predominantly confined in time to a sub-femtosecond burst

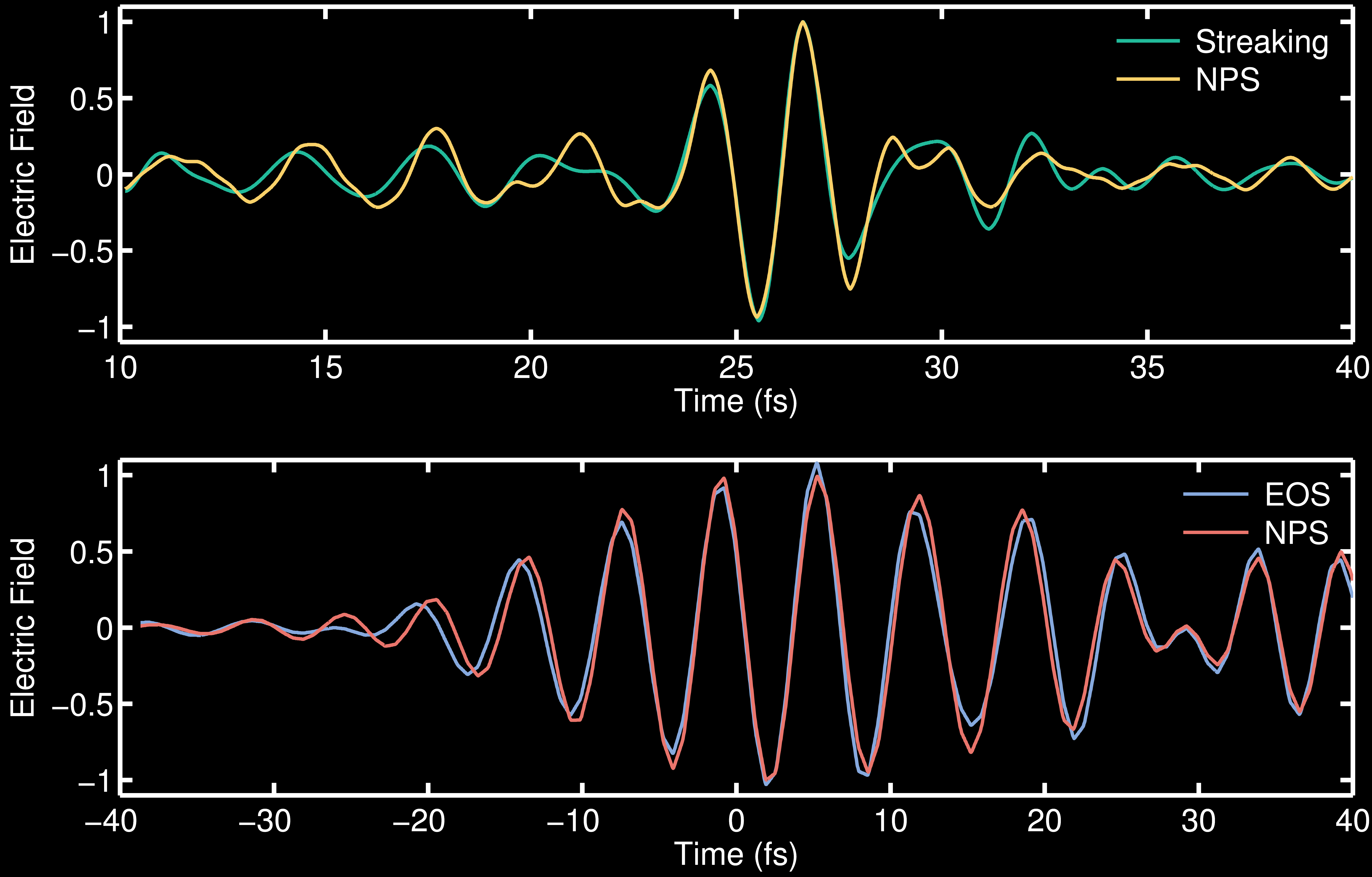
- In a similar manner as attosecond streaking, the instant of the electronic transition and the optical waveform determine the final crystal momentum of the electron
- This is valid for measuring optical waveforms that are weak relative to the injection pulse





*calculation by Nick Karpowicz

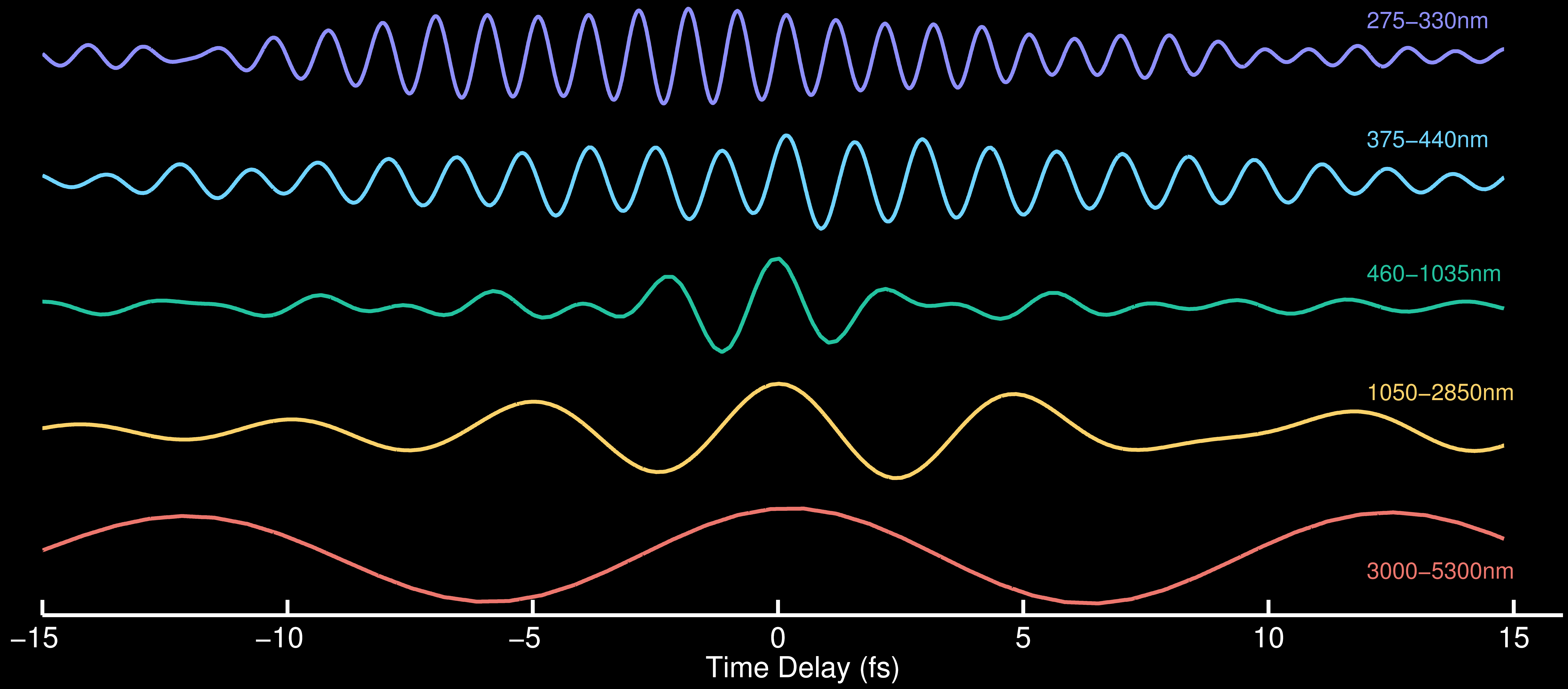
Sederberg et al., *Nature Comm.* **11**:430 (2020)



- The NPS signal was benchmarked against attosecond streaking and electro-optic sampling

benchmarking

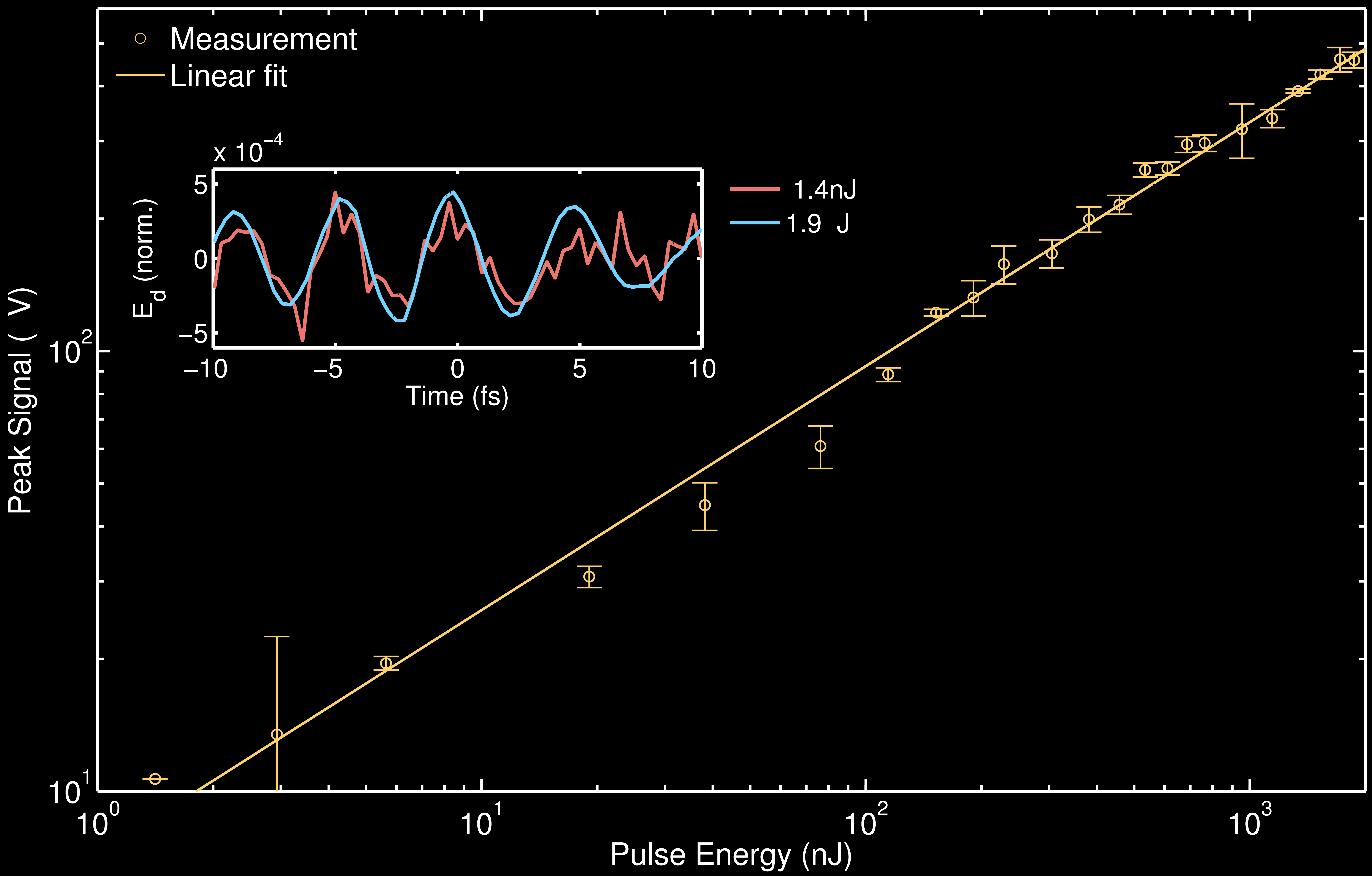
Sederberg et al., *Nature Comm.* **11**:430 (2020)

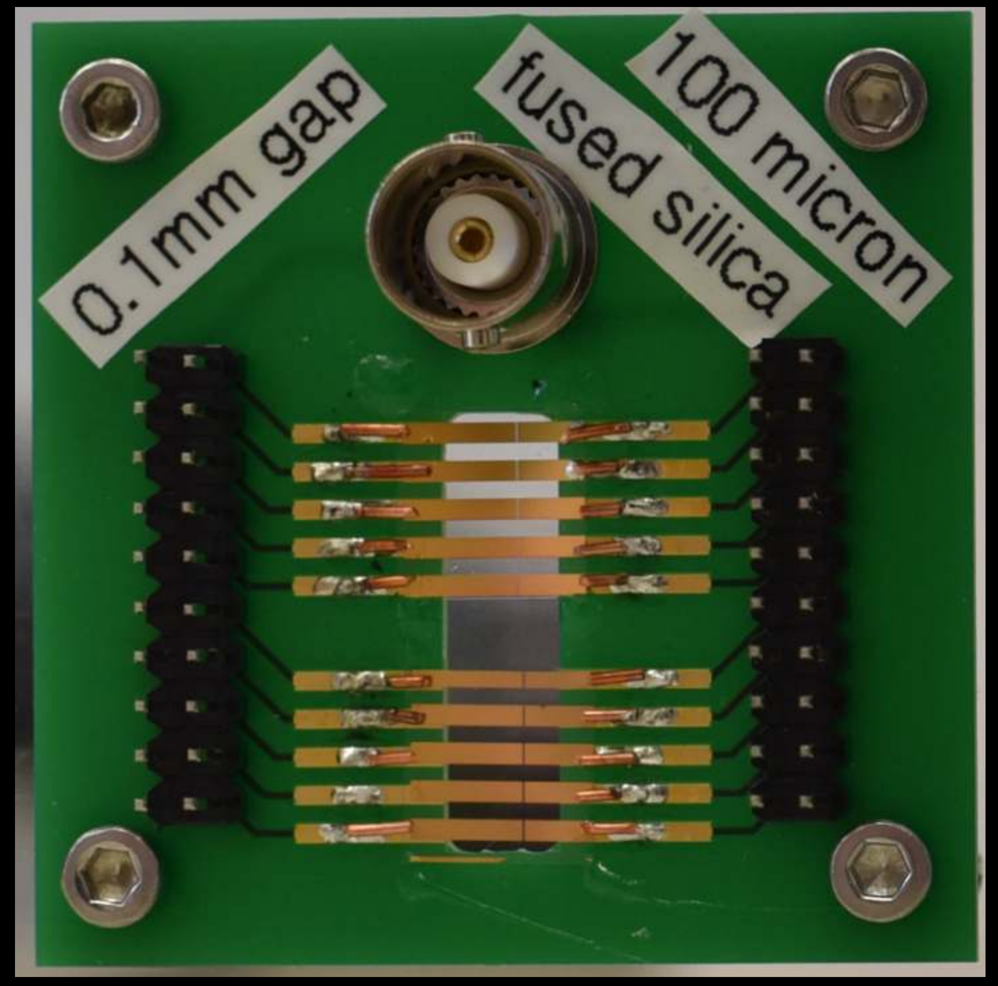


✦ **Detection over more than 1 PHz of continuous bandwidth**

exploring parameters

Sederberg et al., *Nature Comm.* **11**:430 (2020)





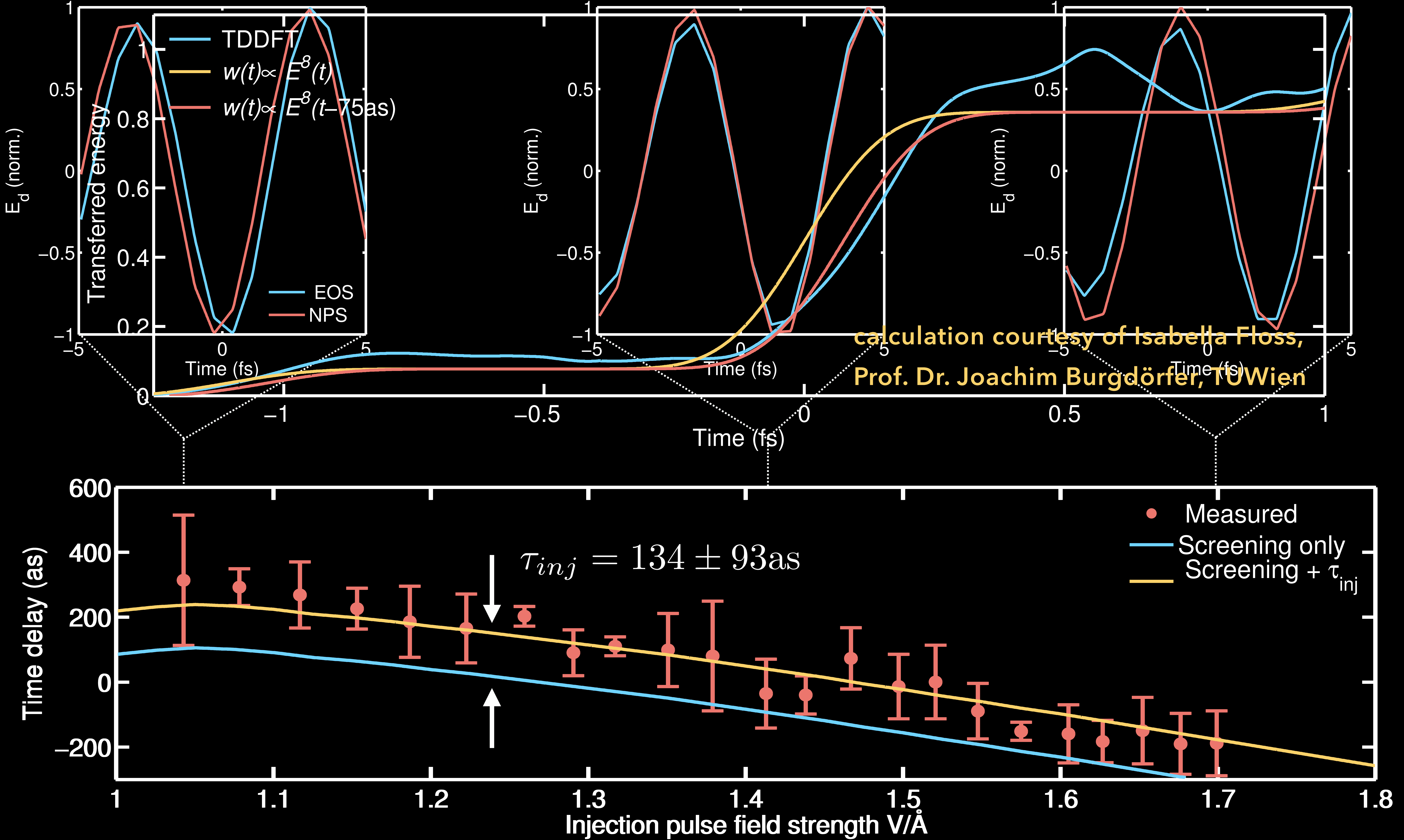
- ✦ **nJ sensitivity**
- ✦ **dynamic range >1000**
- ✦ **SNR > 8000**
- ✦ **could be scaled further**

dynamic range

Sederberg et al., *Nature Comm.* **11**:430 (2020)

- Measuring time delays in photoemission has been one application of attosecond spectroscopy
- Doing so requires a precise timing reference with which to compare the delayed signal
- In our case, we chose to use a complementary waveform sampling technique called electro-optic sampling
    - The detection process is based on perturbative nonlinear optics and is therefore near-instantaneous
- However, the detection process for nonlinear photoconductive sampling involves an electronic transition
- In our first application of this new technique, we chose to measure the time delay in the arrival of an electron in the conduction band after being excited by an optical pulse

Sederberg et al., *Nature Comm.* **11**:430 (2020)



✦ **Attosecond spectroscopy in ambient conditions**

time delays (in solids)

- Finer control over light enables finer control over the state of electrons
  - It provides direct insight into electron coherence in solids and enables the development of new experimental tools

- Applying few-cycle pulses to large-bandgap solids enables waveform sampling with performance metrics that surpass those of conventional XUV techniques
  - Developing attosecond techniques that operate in ambient conditions will broaden its scope of applications

- Spatial structure can be transferred from light to currents
  - We used this to develop a new magnetic field source for "pumping" magnetization dynamics (still in progress)
  - Can we also use it to control quantum systems and quasiparticles?

SFU

thank you!

MPQ

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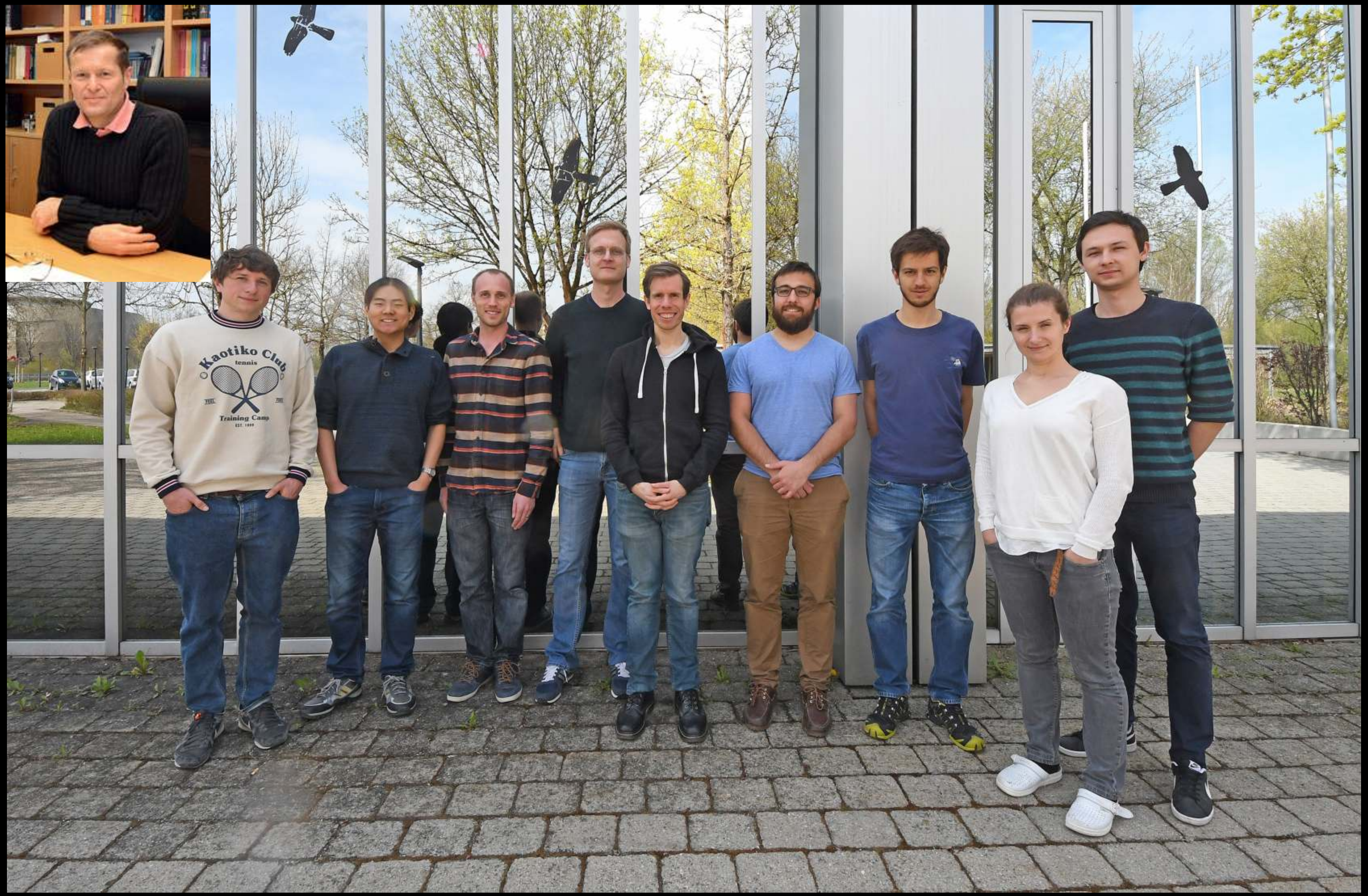

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