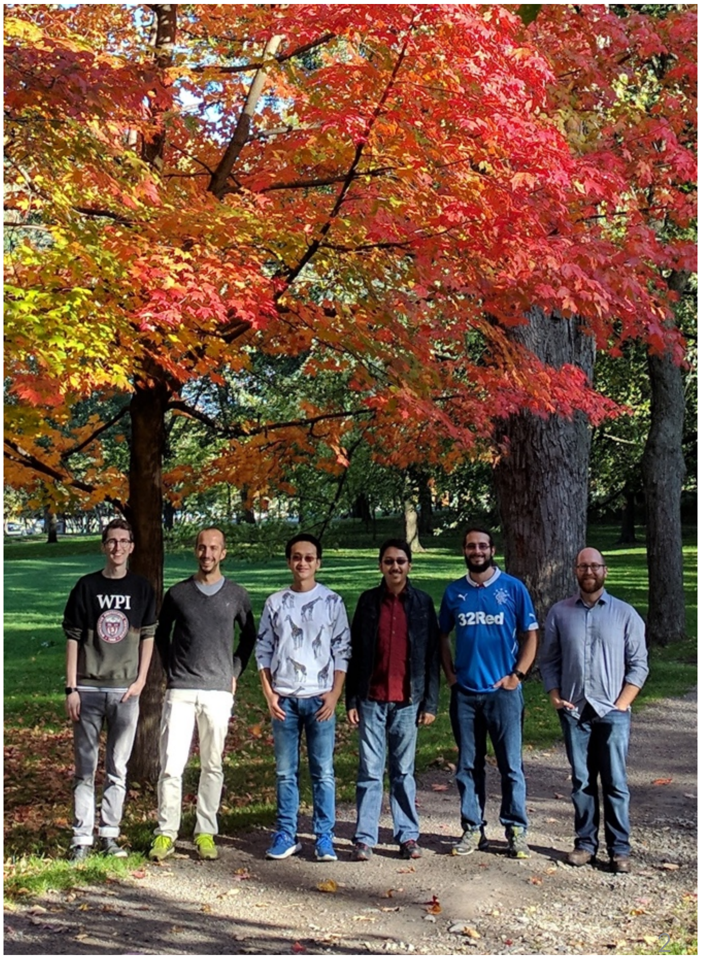
Extreme THz Light-Matter Interactions

David G. Cooke

Dept. of Physics, McGill University, Montreal, QC Canada

ULTRAFAST THZ SCIENCE AT MCGILL



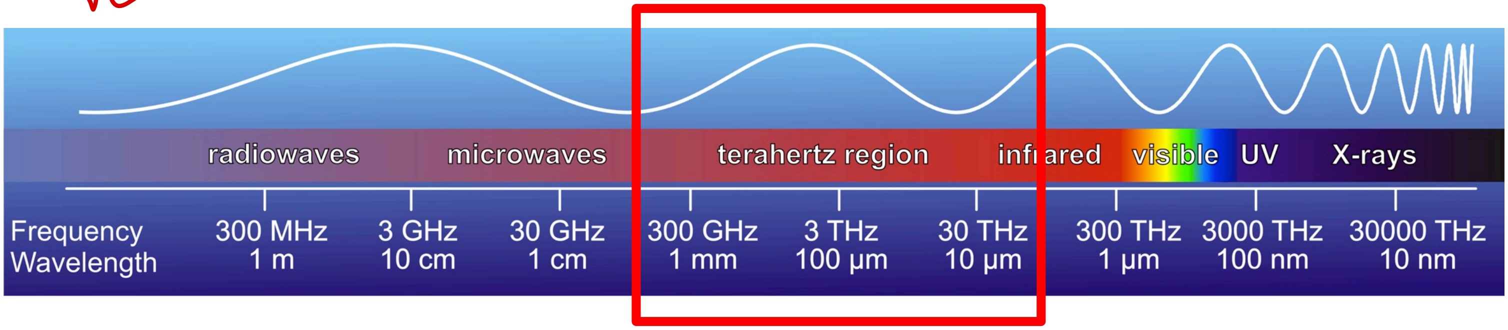
CURRENT RESEARCH THEMES:

1. Ultrafast carrier dynamics in condensed matter
2. THz light-driven coherent electron pulses
3. Dynamic photonics in the THz band

Outline

- **Strong field THz light-matter interactions**
  - Laser-based sources of strong field THz pulses
  - Sub-cycle control of charge and spin degrees of freedom
  - Strong, shaped THz fields for control of matter
- **Cold THz field emission of electrons from nanotips**
  - High bunch charge (> 10^5 electrons/pulse @ 1 kHz)
  - Few fs temporal jitter
  - High degree of transverse coherence / brightness
- **Particle acceleration using THz fields**
  - Why THz fields?
  - State of the art and outlook

ELECTROMAGNETIC SPECTRUM



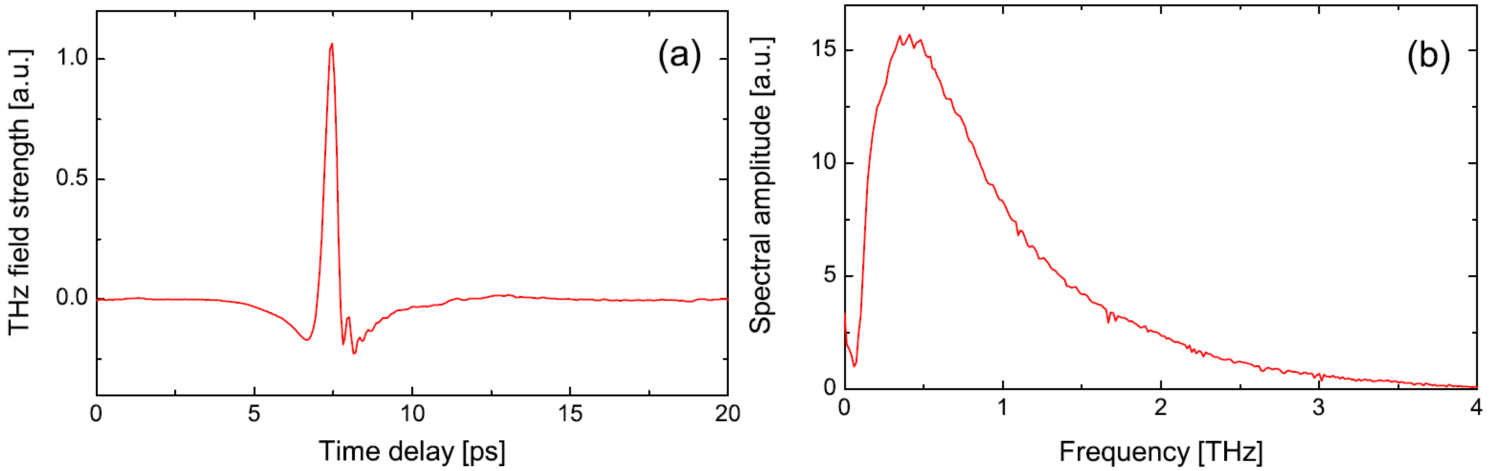
1 THz frequency light:

- 300 µm vacuum wavelength (approx. thickness of the thickest human hair)
- period of 1 *picosecond* (1 trillionth of a second)
- Energy equivalent (1 THz):
  - 48 Kelvin ($E = k_B T$)
  - 33.3 cm^{-1}
  - 4.13 meV

THZ FIELD CONTROL OF MATTER

THZ PULSES

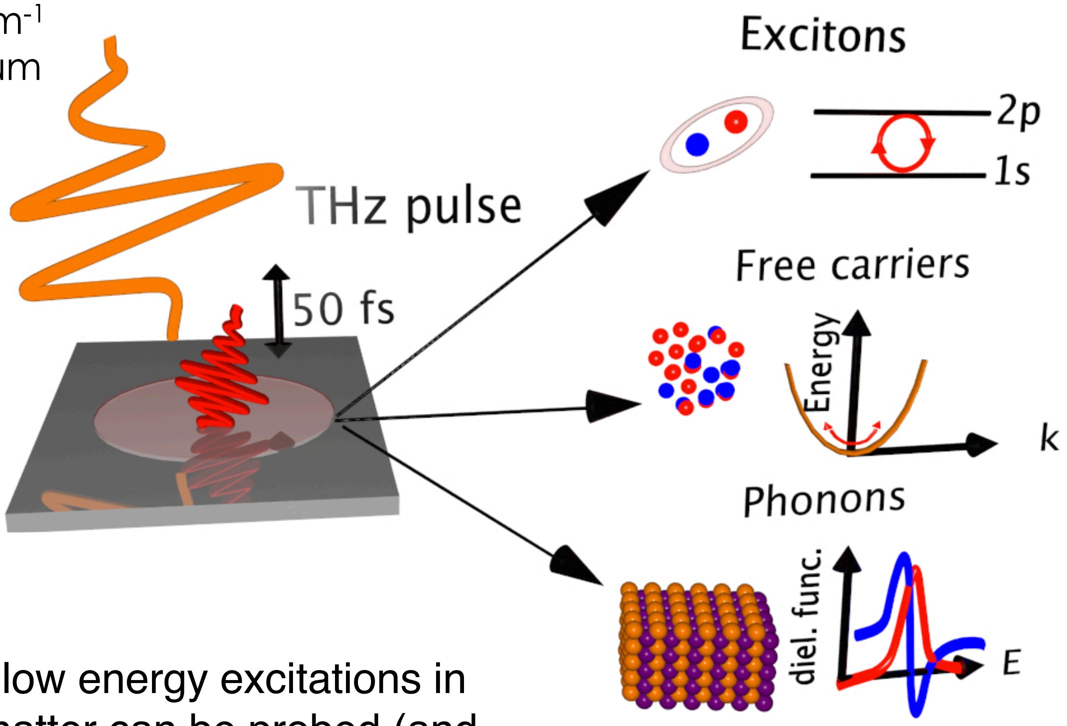
Single cycle of the electric field = shortest light pulse possible (at given f_c)



- Electric field of the pulse is measured coherently in the time domain.
- Spectra is obtained by Fourier transform.

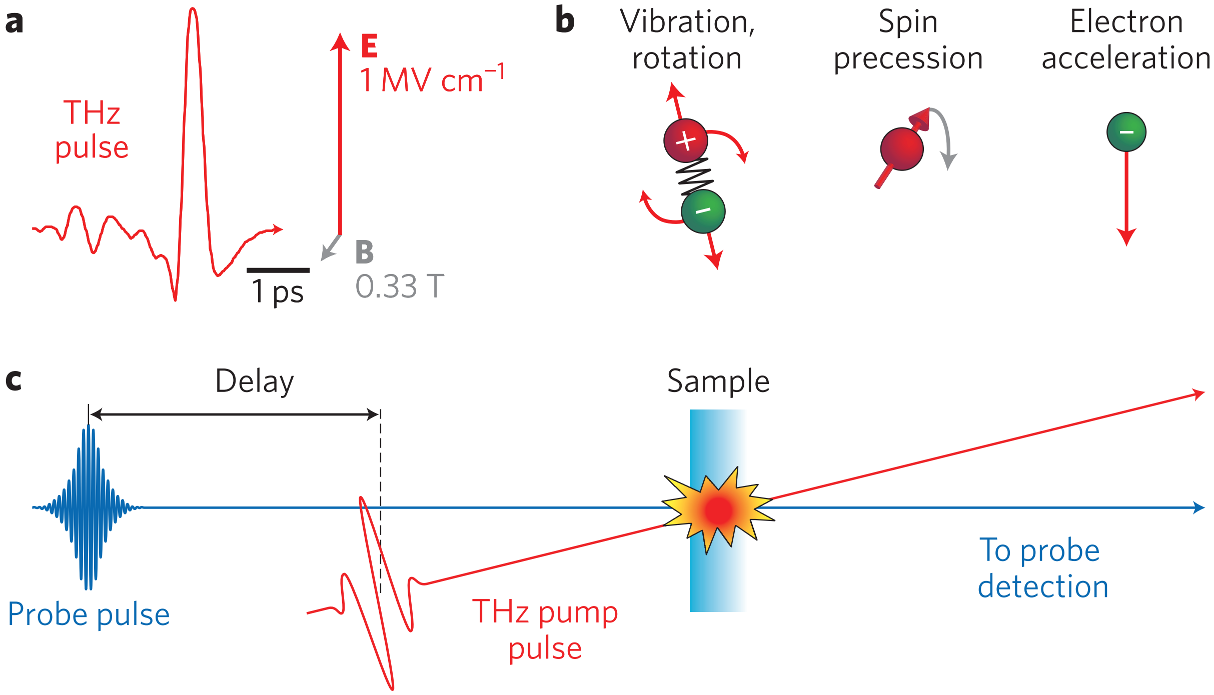
THz EXCITATIONS IN MATERIALS

1 THz = 4.13 meV
33 cm^{-1}
300 µm
48 K

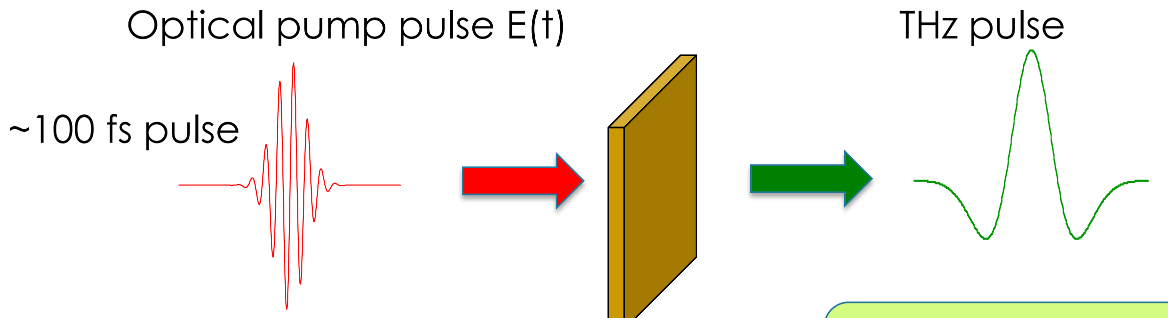


Dynamics of low energy excitations in condensed matter can be probed (and controlled) on sub-picosecond time scales.

THz FIELD CONTROL OF MATTER



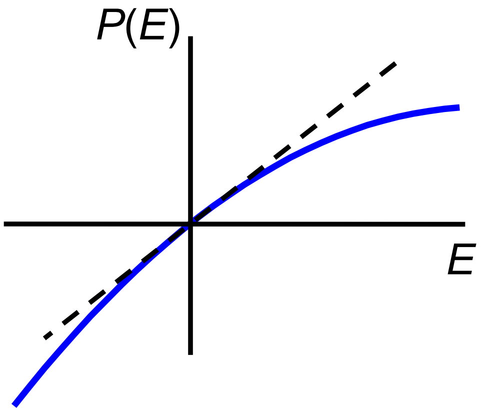
OPTICAL RECTIFICATION



$$E(t)=\varepsilon(t)\cos\omega_0 t$$

$$\mathbf{P}_{\mathrm{NL}}^{(2)} \sim E^2(t)$$

$$E_{rad}(t) \propto \frac{\partial^2 \mathbf{P}_{\mathrm{NL}}^{(2)}(t)}{\partial t^2}$$

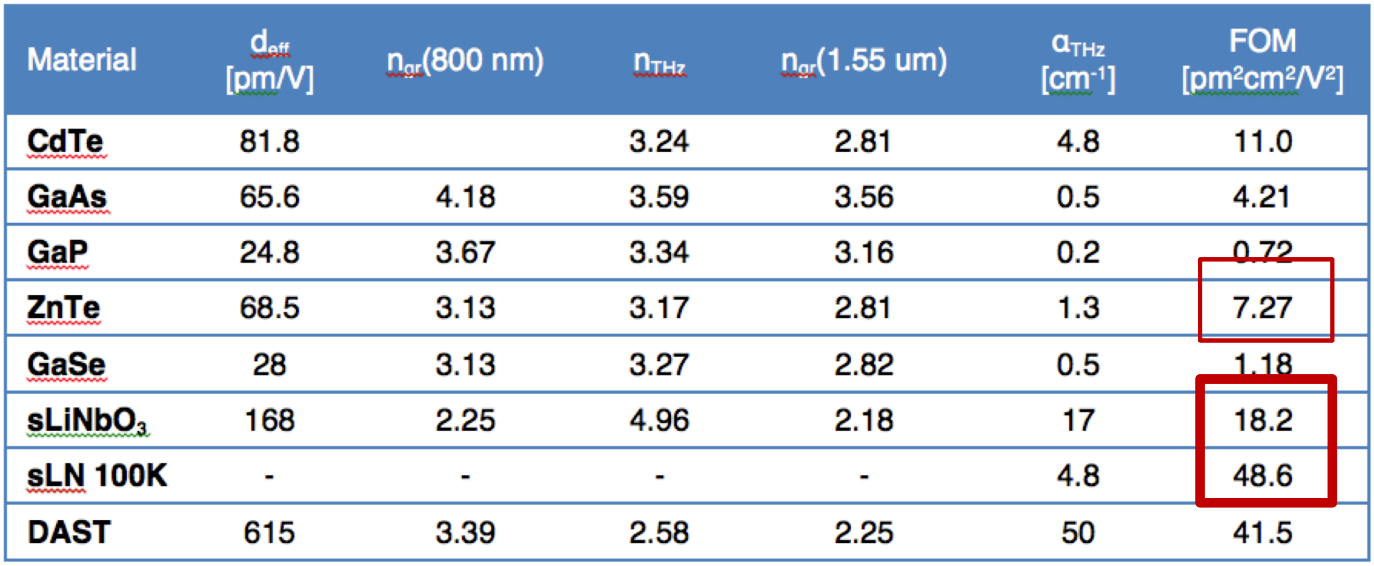


$$\sim \underbrace{\varepsilon^2(t)\cos 2\omega_0 t}_{\text{SHG}} + \underbrace{\varepsilon^2(t)}_{\textbf{THz}}$$

Rectified the carrier wave of light.
"Optical rectification"

NONLINEAR CRYSTALS

- High nonlinearity
- Good phase matching (THz wave front same v as pump)
- Low absorption of pump and THz

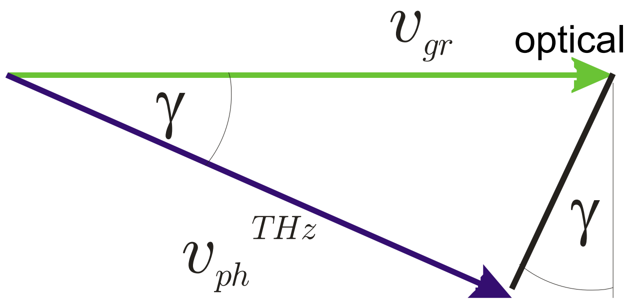


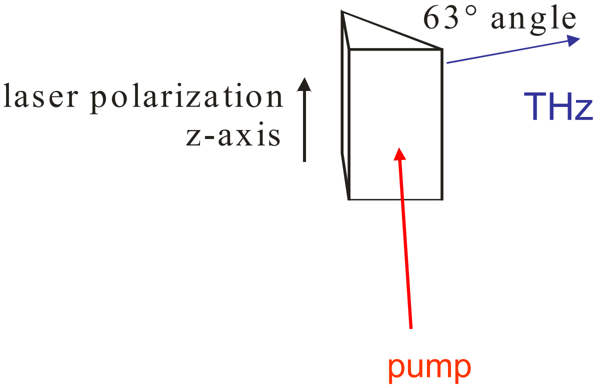
Material	d_{eff} [pm/V]	n_{gr}(800 nm)	n_{THz}	n_{gr}(1.55 um)	α_{THz} [cm^{-1}]	FOM [pm^{2}cm^2/V^2]
CdTe	81.8		3.24	2.81	4.8	11.0
GaAs	65.6	4.18	3.59	3.56	0.5	4.21
GaP	24.8	3.67	3.34	3.16	0.2	0.72
ZnTe	68.5	3.13	3.17	2.81	1.3	7.27
GaSe	28	3.13	3.27	2.82	0.5	1.18
sLiNbO$_3$	168	2.25	4.96	2.18	17	18.2
sLN 100K	-	-	-	-	4.8	48.6
DAST	615	3.39	2.58	2.25	50	41.5

Lithium niobate looks great, but terrible phase matching…

TILTED PULSE FRONT OPTICAL RECTIFICATION

phase matching by pulse front tilting



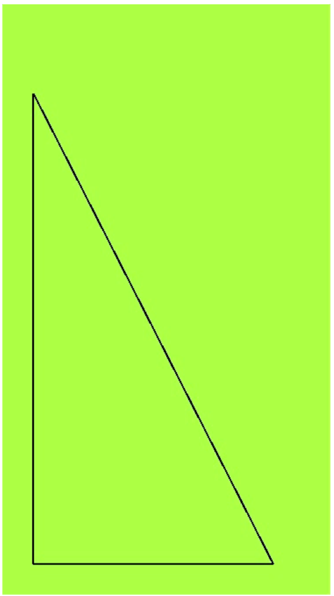


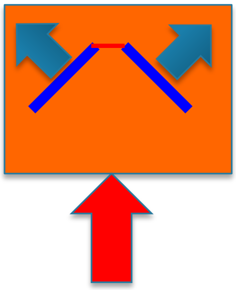
$$\cos\gamma = \frac{n_{vis}}{n_{THz}} = \frac{2.2}{4.9} \Rightarrow \gamma = 63\,\text{deg}$$

Can then achieve phase matching in $LiNbO_3$

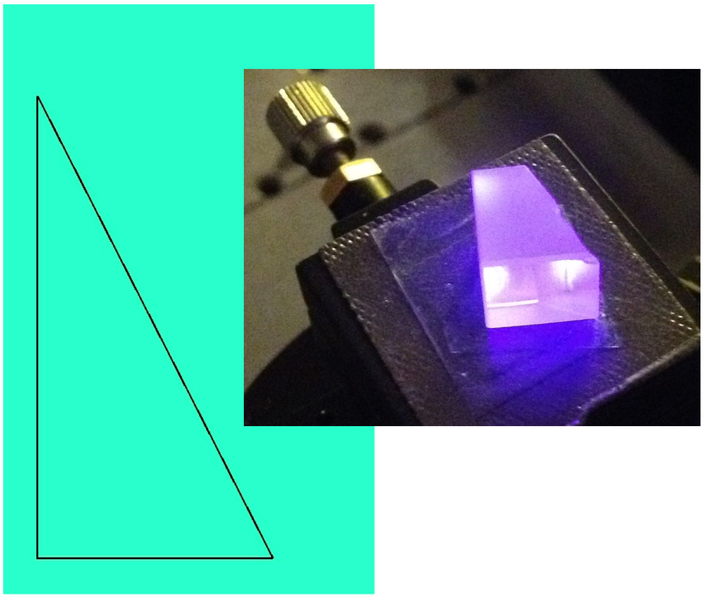
Appl. Phys. Lett. 83, 3000 (2003), Appl. Phys. B 78, 593 (2004)

Simulations courtesy of Matthias Hoffman, SLAC





plane wave excite



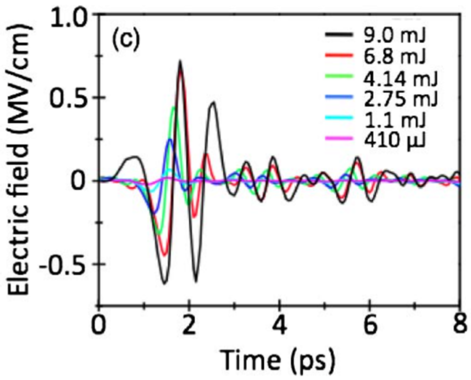
Intense THz emission

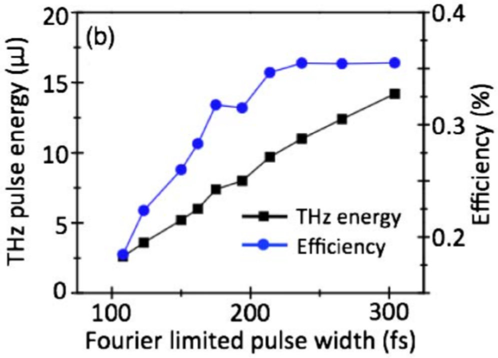
“tilted pulse front”

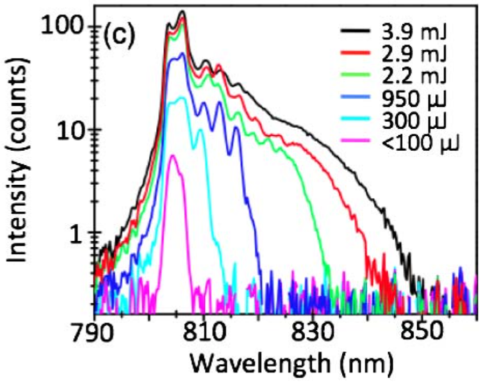
Limitations of tilted-pulse-front

Blanchard, Cooke et al., Opt. Lett. 39, 4333 (2015)

700 kV/cm peak field strength.
MV/cm possible with better focusing.



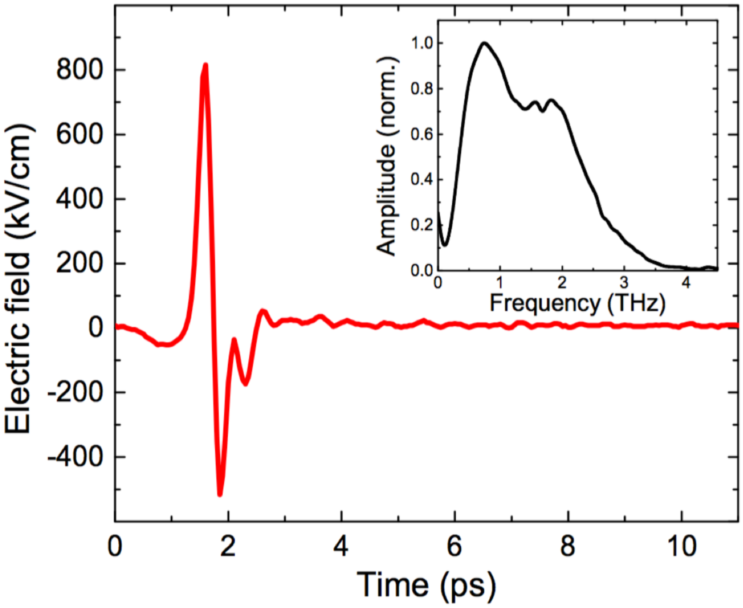




More efficient with pump pulse durations > 200 fs.

Huge red-shifting of pump pulse due to cascaded nonlinear interaction.

INTENSE THZ PULSES



Tilted pulse front optical rectification

- Pulse energy ~ 1 μJ
- Peak E-field ~800 kV/cm
- Peak B-field ~ 0.26 Tesla
- Intensity = 800 MW/cm^2

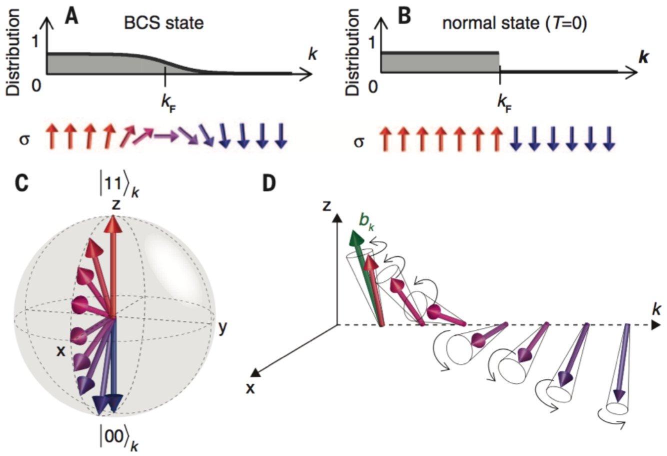
THz FIELD CONTROL OF THE SOLID STATE

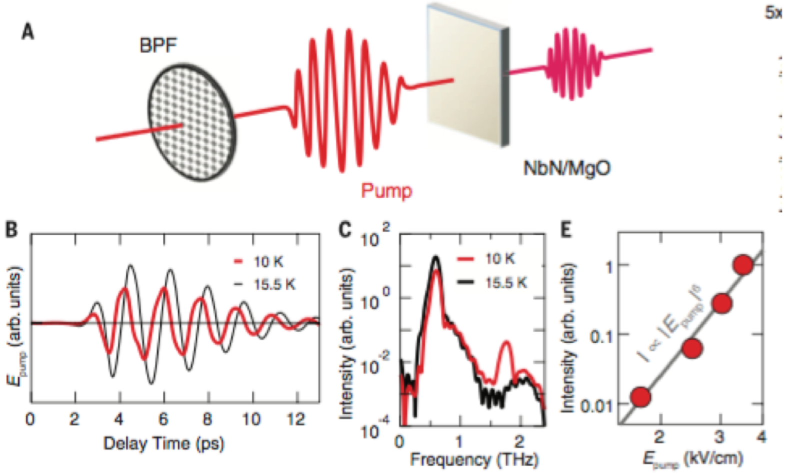
- Energy scales of “interesting” materials are in the milli-eV range
  - superconductivity
  - charge/spin density waves
  - magnons
  - excitons
  - cavity exciton polaritons
  - polarons
  - Etc…

- Coherent control of these excitations requires shaped, near resonant and strong THz field transients.

Coherent control of superconductivity

Matsunaga, Shimano et al., Science 2014





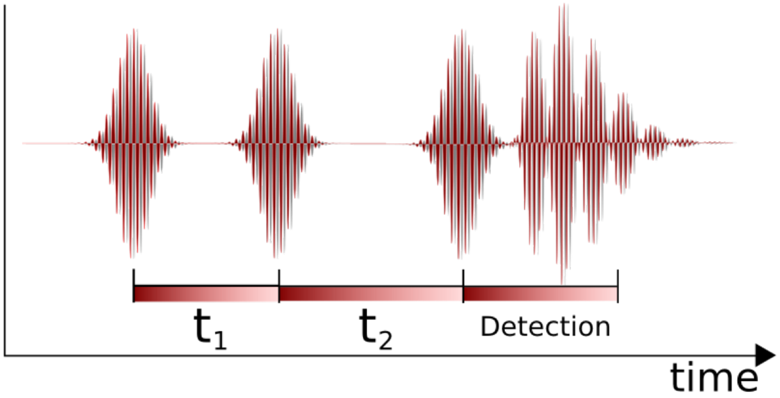
$$\boldsymbol{j}_{\mathrm{NL}}(t) \propto \frac{e^2\Delta}{U}\boldsymbol{A}(t)\boxed{\delta\Delta(t)}$$

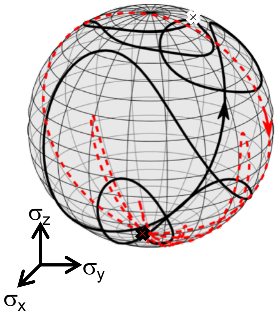
Dynamic modulation of order parameter

- Multiple-pulses can manipulate the order parameter in more complex ways

Shaped THz fields to coherently manipulate superconducting order parameter

THz multi-dimensional spectroscopy



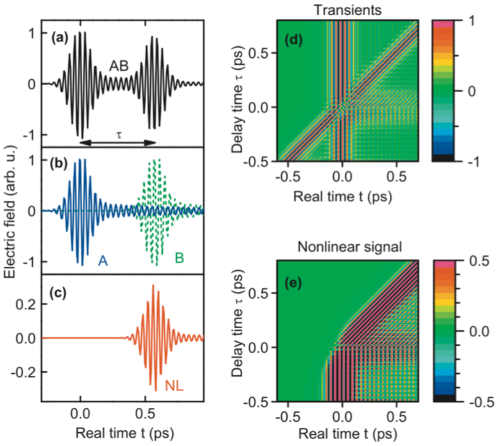


Coherent state manipulation.

Frequency range	Relevant excitations
Radio(MHz-GHz)	Nuclear spin, magnetic moments
Infrared (mid-IR)	Vibrational
Optical	Electronic, excitonic
1 – 10 THz	Collective excitations, spin/charge density waves, superconductivity, excitons, magnons, phonons, molecular rotations…

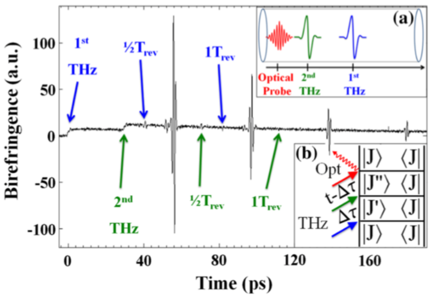
Previous work: 2D THz spectroscopy

Quantum wells



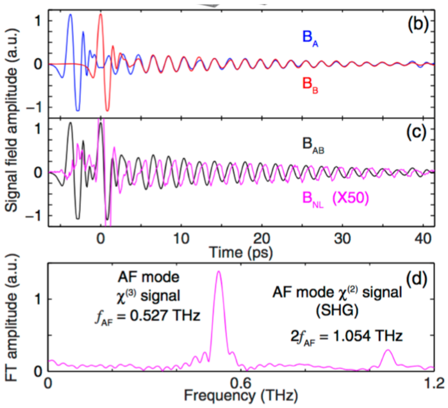
T. Kuehn, T. Elsaesser et al., J. Chem. Phys 2009

Molecular rotations



S. Fleischer, K. Nelson et al., PRL 2012

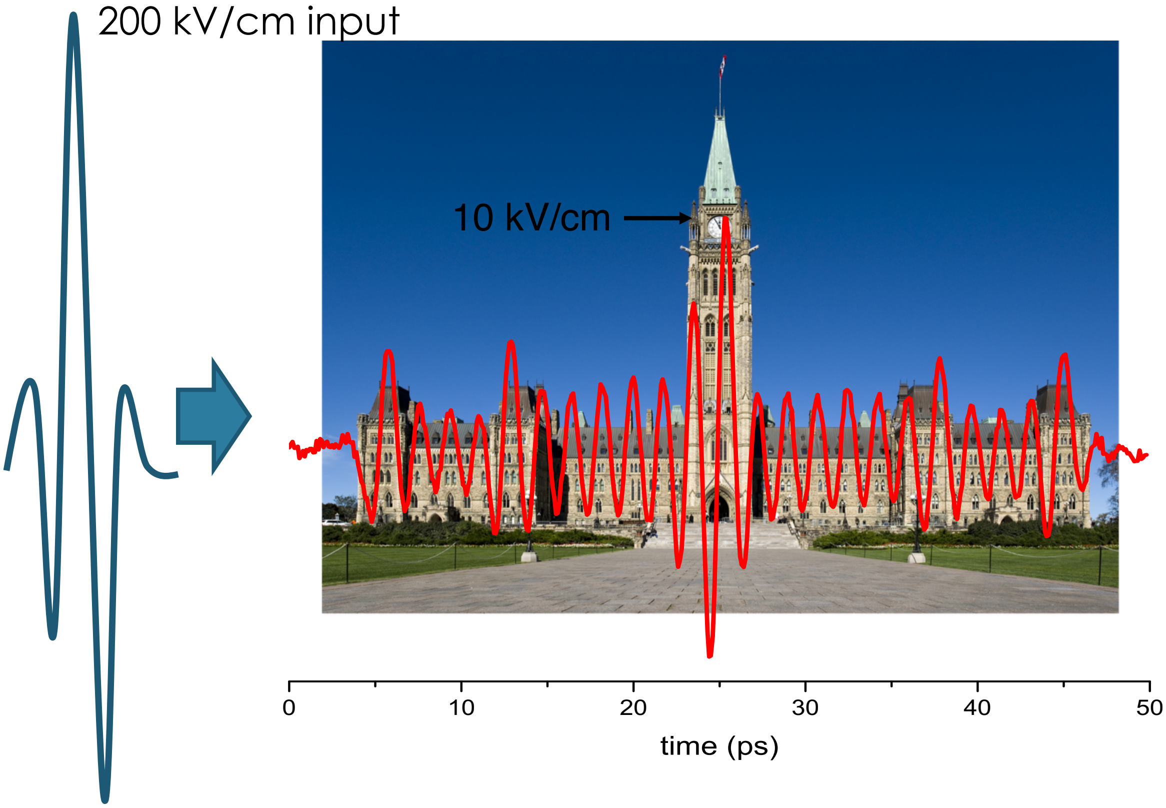
Collective spin waves



J. Lu, K. Nelson et al., PRL 2017

TEMPORAL SHAPING THZ LIGHT

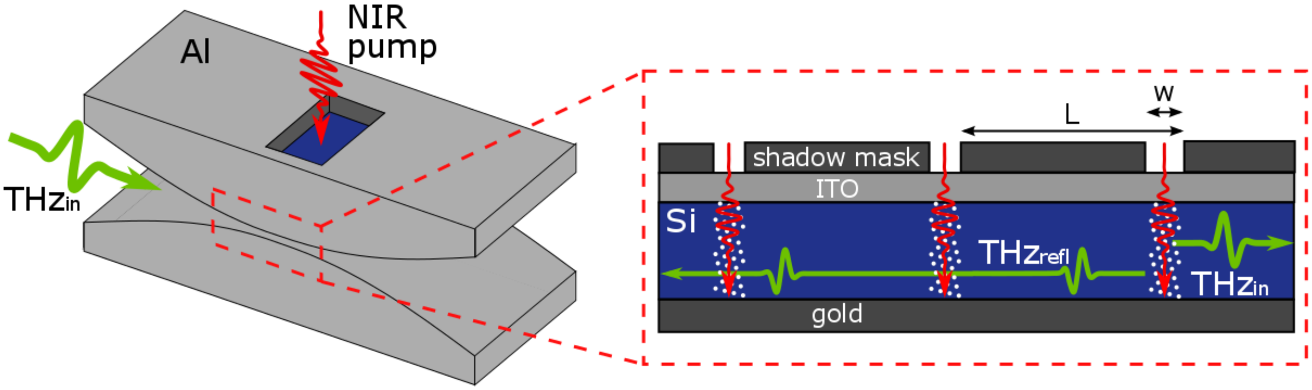
New capability: Direct and arbitrary shaping of kV/cm THz light pulses



JACQUES—CARTIER BRIDGE, MTL

Dynamic THz photonics

A system for advanced pulse shaping and dynamic photonics



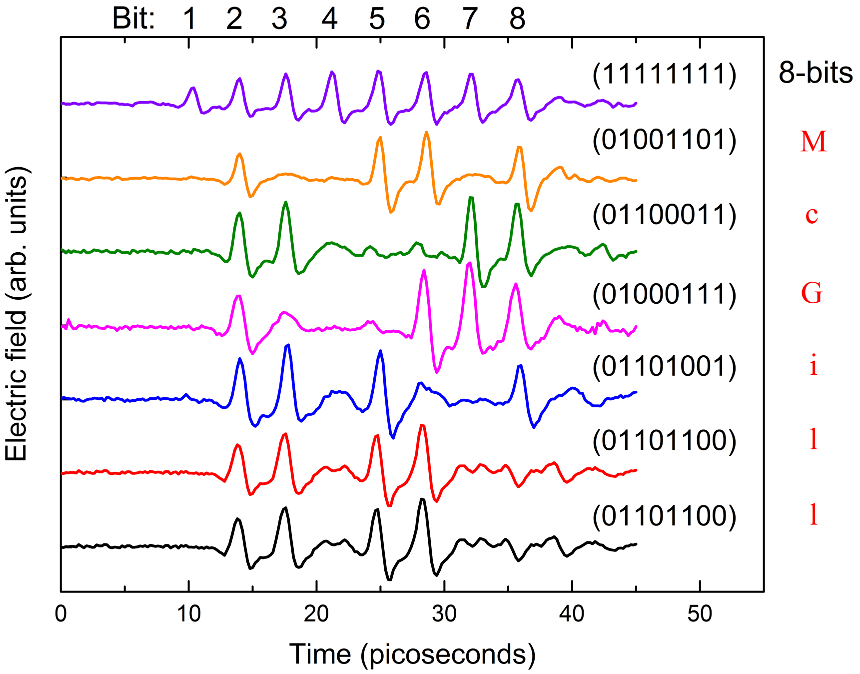
fs creation of metal-dielectric structures patterned in HRFZ silicon slab

Playground for THz pulse shaping and manipulation in space and time.

Not the only way! Can also pattern the pump + antenna or optical rectification.

1. Liu, Park, Weiner, IEEE J. Sel. Top. Quant. Electron. **2**, 709 (1996)
2. Ahn, Efimov, **Averitt** and Taylor, Opt. Express **11**, 2486 (2003).

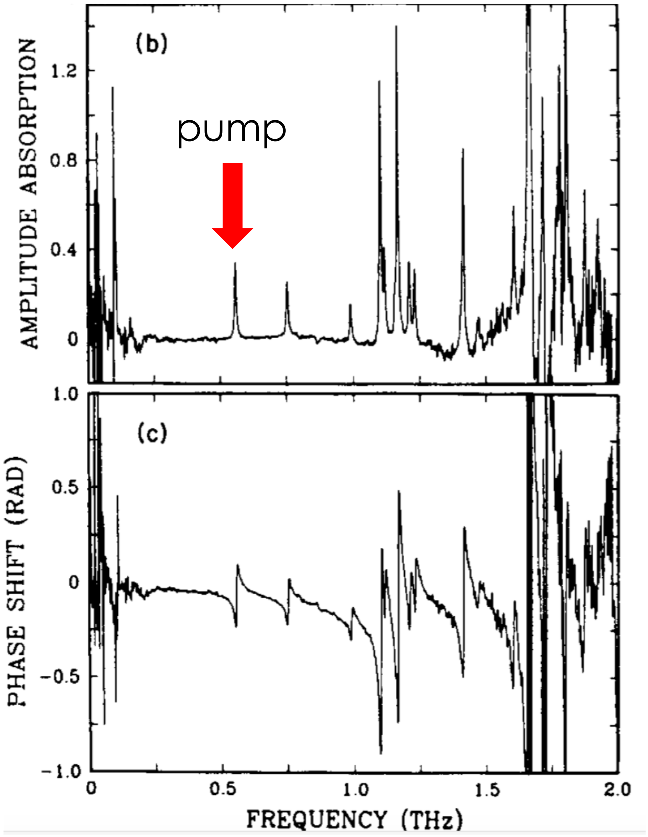
THz waveform synthesis



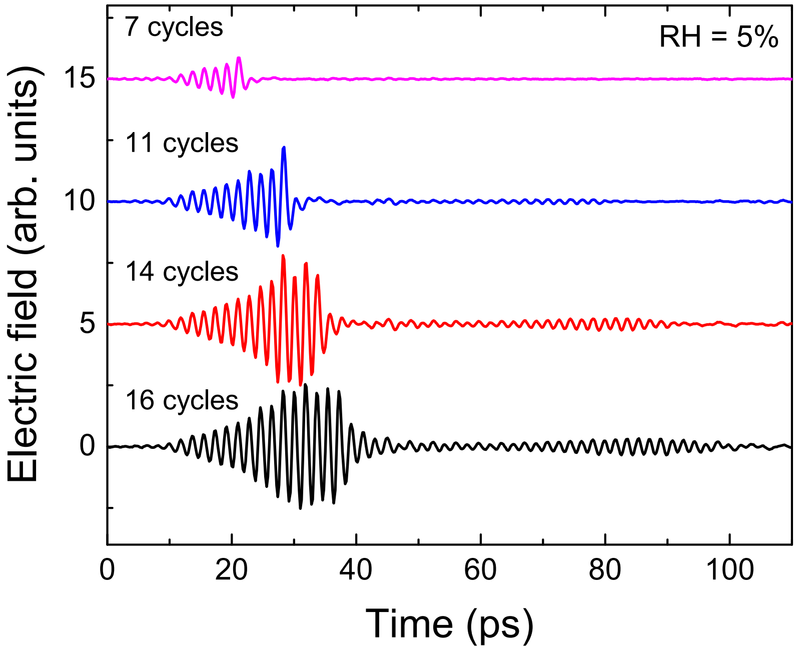
Pretty much any pulse shape is possible now and can be switched pulse-to-pulse.

“SWITCHED” ROTATIONAL WAVE PACKET

H_2O



1. Slowly align molecules in resonant field.
2. Rapid switch off, free induction decay.

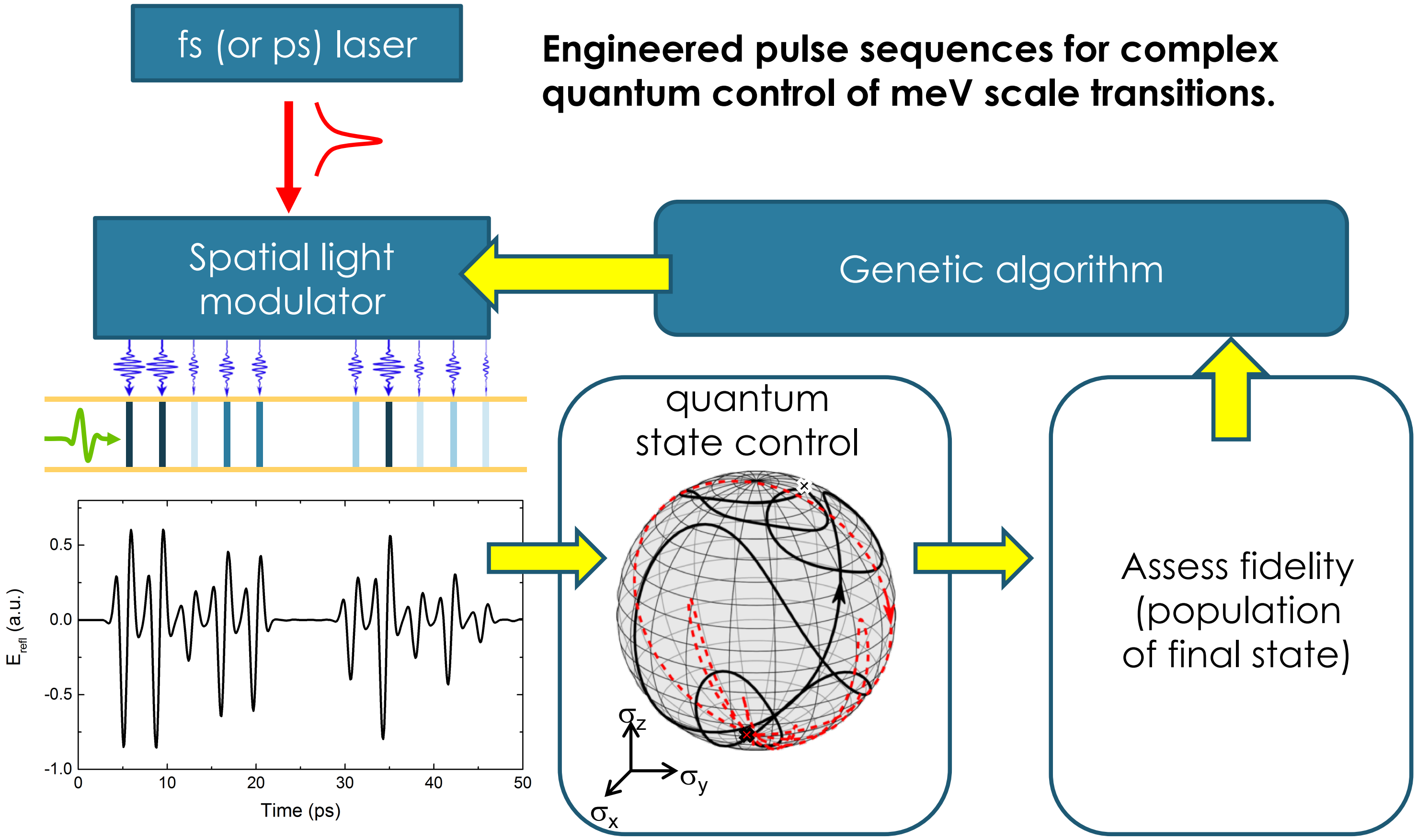


Revival observed, field free alignment.

L. Gingras, D. Cooke et. al., unpublished

OPTIMIZED THZ COHERENT CONTROL

Engineered pulse sequences for complex quantum control of meV scale transitions.



fs (or ps) laser

Spatial light modulator

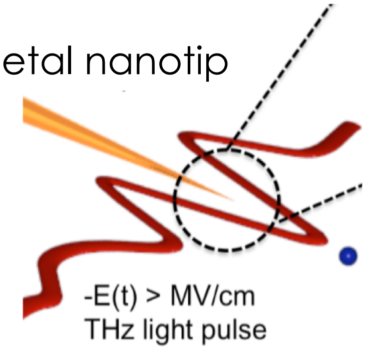
Genetic algorithm

quantum state control

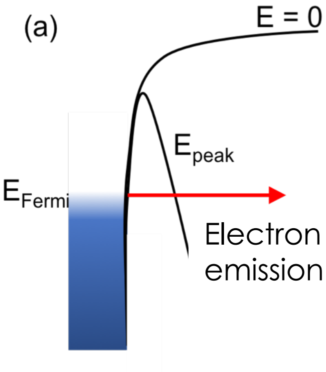
Assess fidelity (population of final state)

THZ FIELD EMISSION FROM NANOTIPS

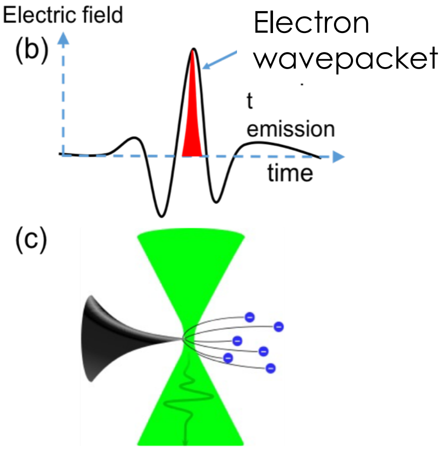
Femtosecond electron emission



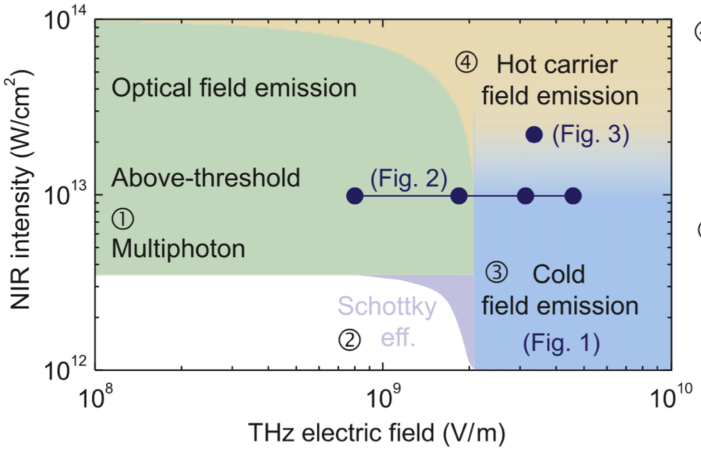
Metal nanotip

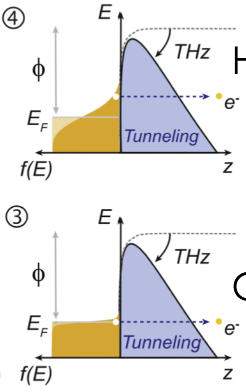


Electron emission



Electron wavepacket



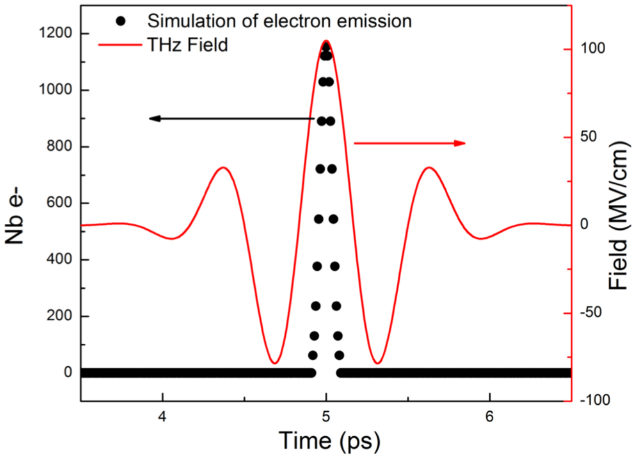


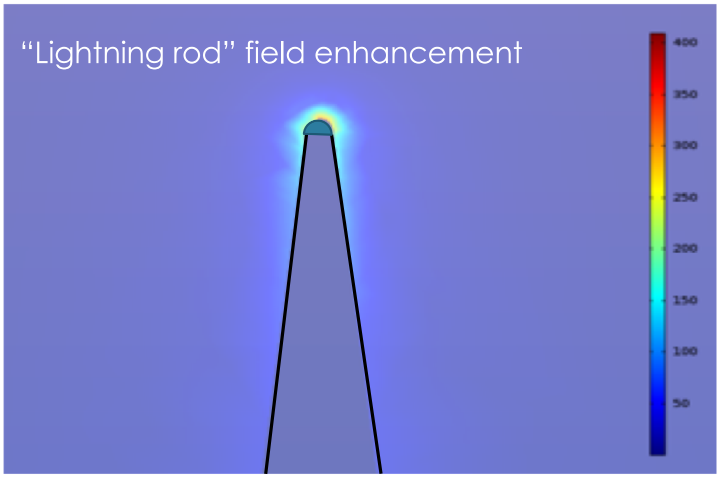
Hot field emission

Cold field emission

Herink et al., N. J. Phys. 16, 123005 (2014)

Particle acceleration in the half-cycle




“Lightning rod” field enhancement

Decays exponentially over ~ tip radius
Finite element simulations: γ ~ 350

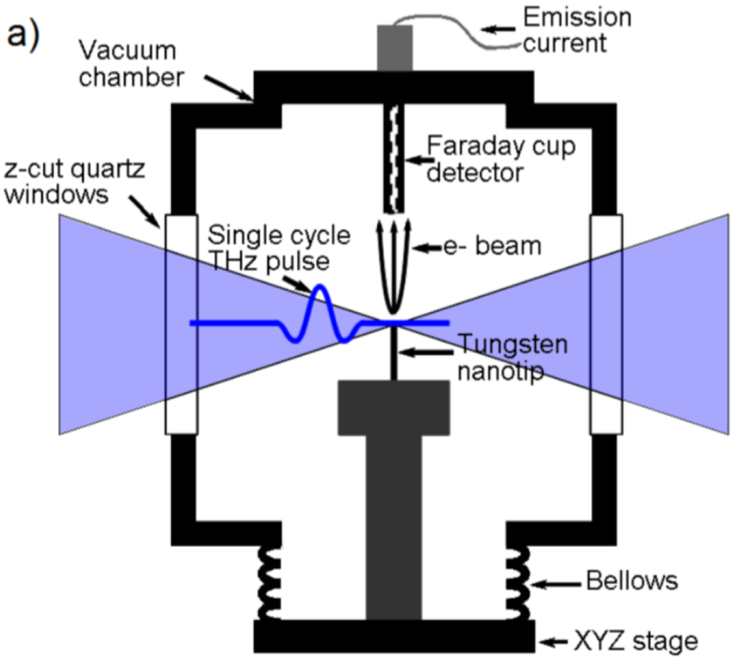
Local field drives electron motion:

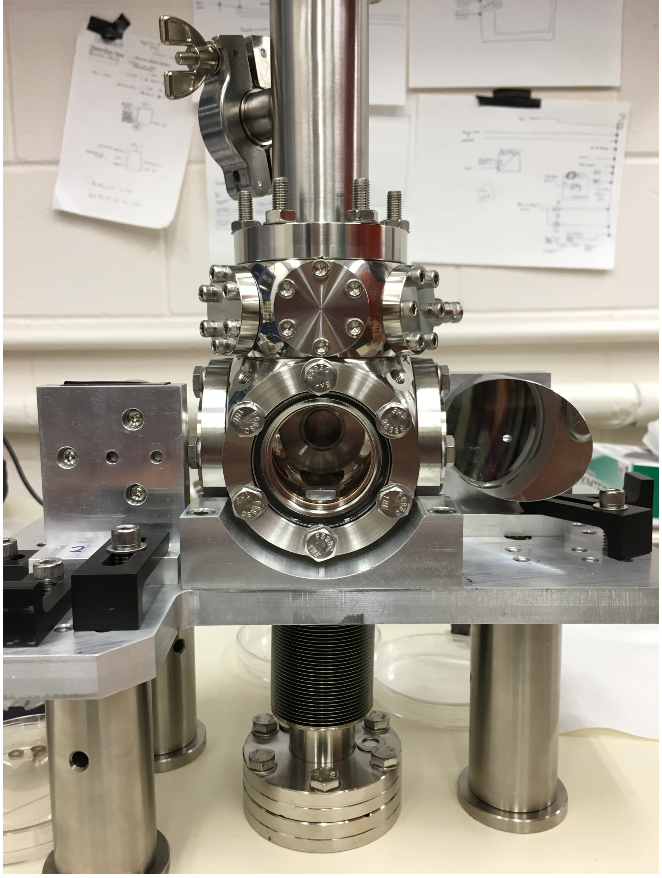
$E_{incident}$ = 400 kV/cm

E_{local} = 15 GV/m!

Electrons experience accelerations of 10^{21} m/s^2, reaching ~ 1 keV in < 10 fs (50 nm)!

Electron emission setup

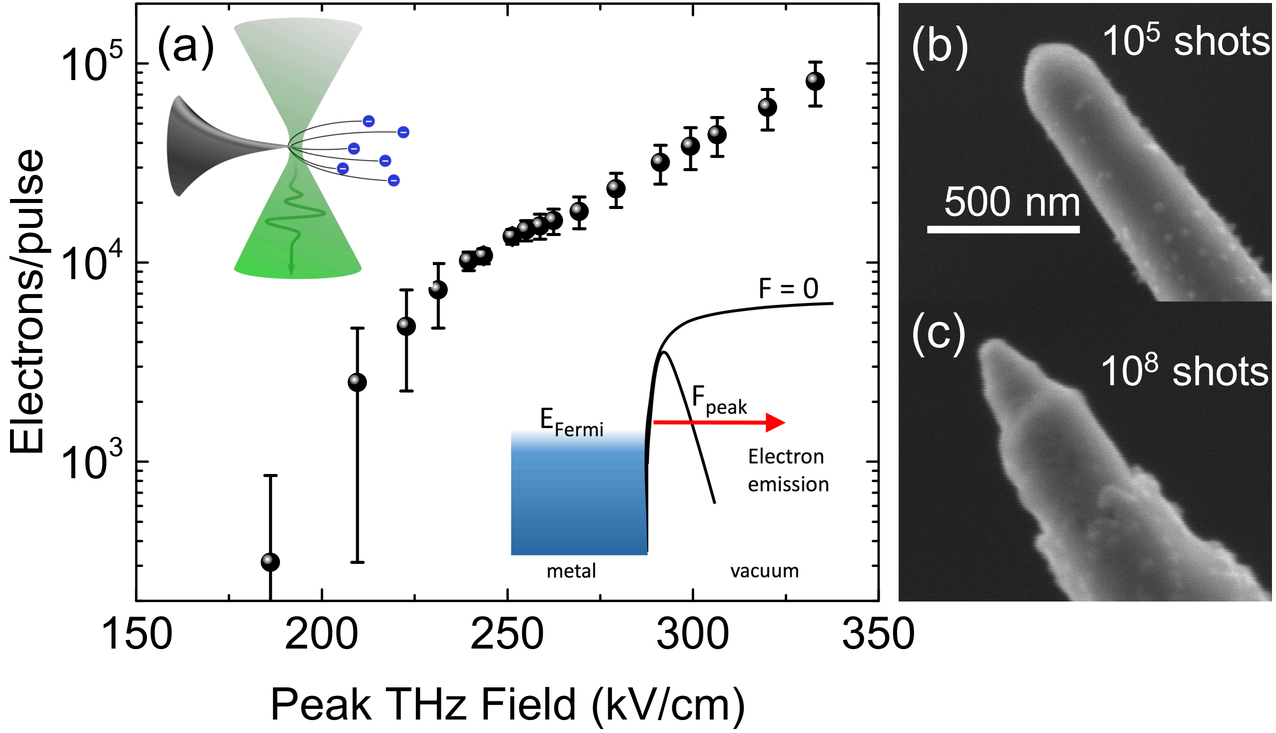




femtoCoulomb bunch charges

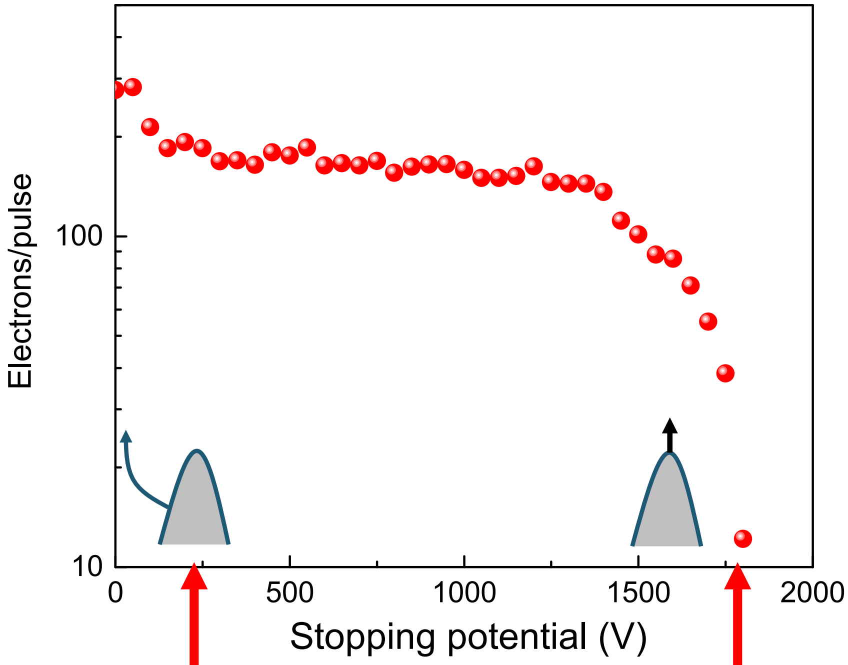
10^5 electrons per bunch achieved

THz field conditioning



Latest results: ~500,000 electrons/pulse

Energy spectrum



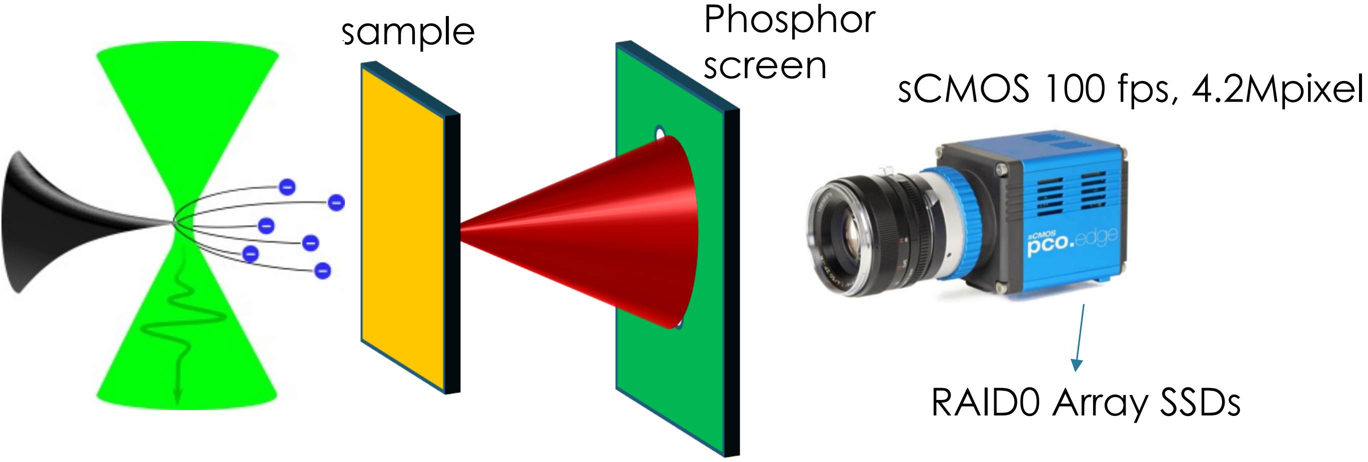
Low energy electrons
- high surface area, low field enhancement, low directionality

Electrons emitted from tip apex

Energies up to 2 keV in ~ 100 nm

Tip contamination is an issue. In-situ heating installed.

Single-shot electron diffraction/microscopy



- Investigate non-repetitive structural effects (grain boundaries, meta-stability, etc.. OR soft/organic compounds that degrade.

- > 10^5 electrons in a bunch required to resolve a diffraction image: *Daoud, Floettmann and R. J. D. Miller, Struct. Dyn.* ***4****, 044016 (2017).*

- THz measured nanotip bunch charge is ~10^5 ! Possible to resolve single shot diffraction image.

Two-pulse electron correlation spectroscopy (TP-ECS)

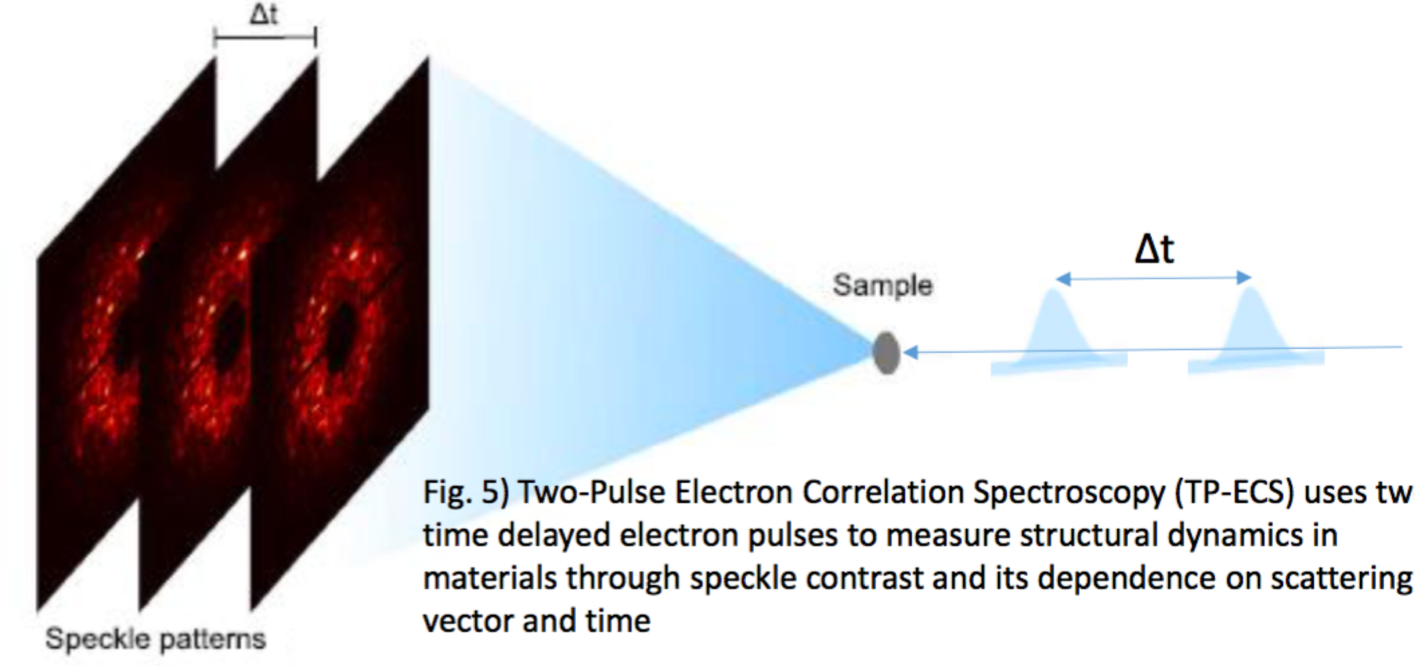


Fig. 5) Two-Pulse Electron Correlation Spectroscopy (TP-ECS) uses two time delayed electron pulses to measure structural dynamics in materials through speckle contrast and its dependence on scattering vector and time

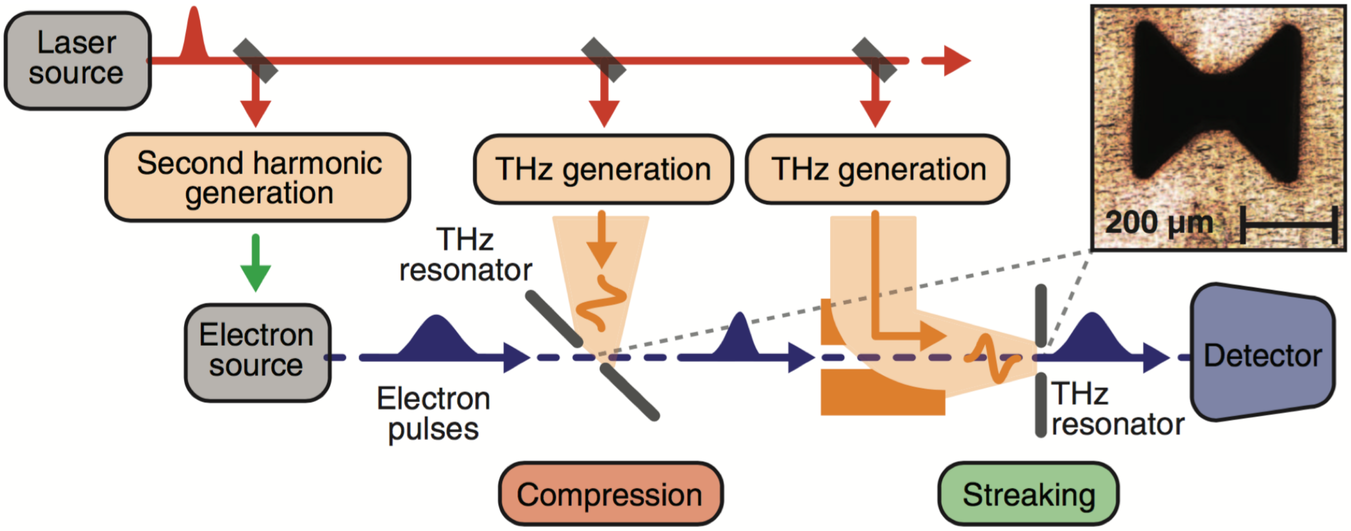
- Electron equivalent to X-ray Pulse Correlation Spectroscopy (XPCS) – Mark Sutton

- Ground (or excited) state atomic and electronic structural dynamics with sub-Angstrom, few fs resolution.

Optical field electron wave packet control

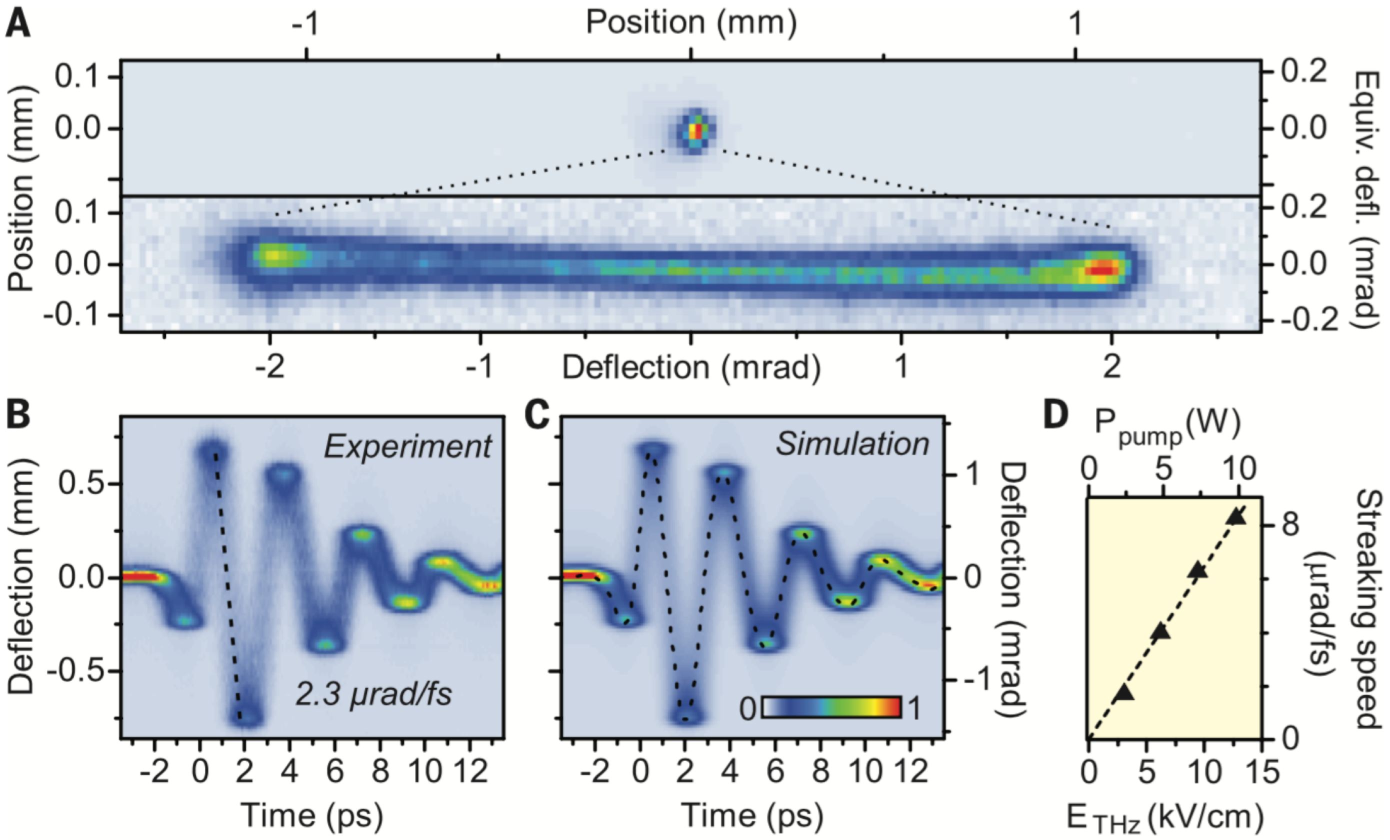
All-optical control and metrology of electron pulses

C. Kealhofer,[1,2] W. Schneider,[1,2] D. Ehberger,[1,2] A. Ryabov,[1,2] F. Krausz,[1,2]* P. Baum[1,2]*

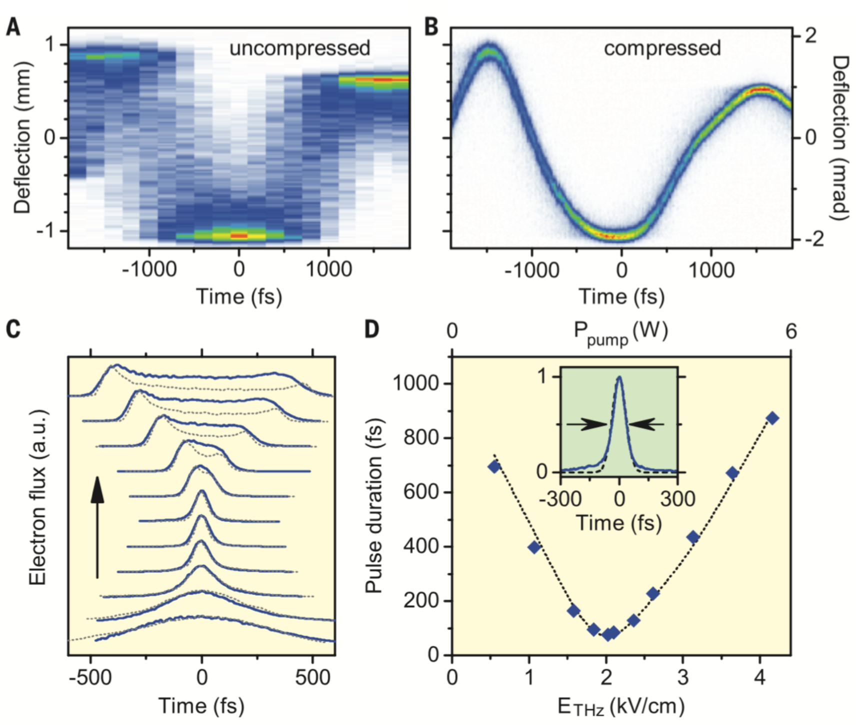


Science, **352**, 429 (2016).

THz electron streaking



THz compression



Energy imparted by THz pulse:
g_E = 50 eV/ps @ 10 kV/cm
incident peak THz field.

Minimum electron pulse duration achieved at sample is 4.5 fs!

This was single to few electrons/bunch. Can we do 10^5 electrons?

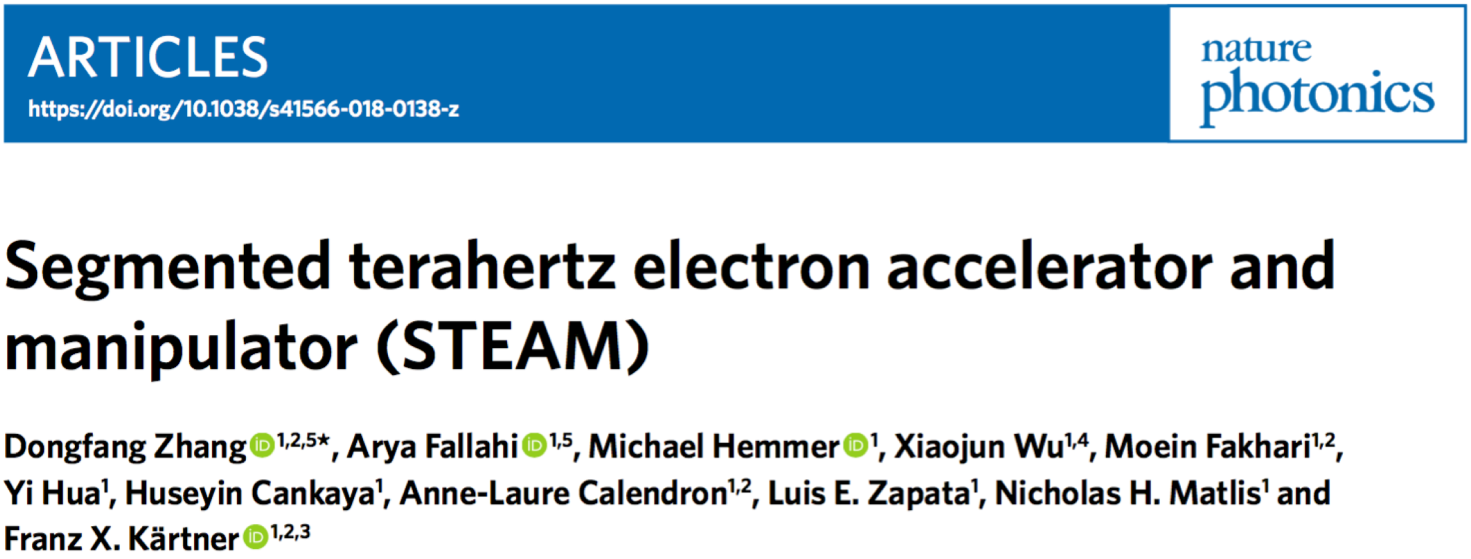
Goal: All-THz driven UED instrument

THZ PARTICLE ACCELERATION

Post-doctoral position available!

Why use THz fields?

- Maximum acceleration gradients limited by field induced breakdown.
- Breakdown field scales as 1/(pulse duration)^6
  - GV/m fields are accessible yielding compact devices
- Fields are intrinsically phase stable and synchronized to temporal jitter of < 2 fs rms (typical).
- Compatible with high brightness sources (e.g. nanotips)
- Size of structures are mm scale (easy to make and support high (picoCoulomb) bunch charge.
- Potentially save some $$$.



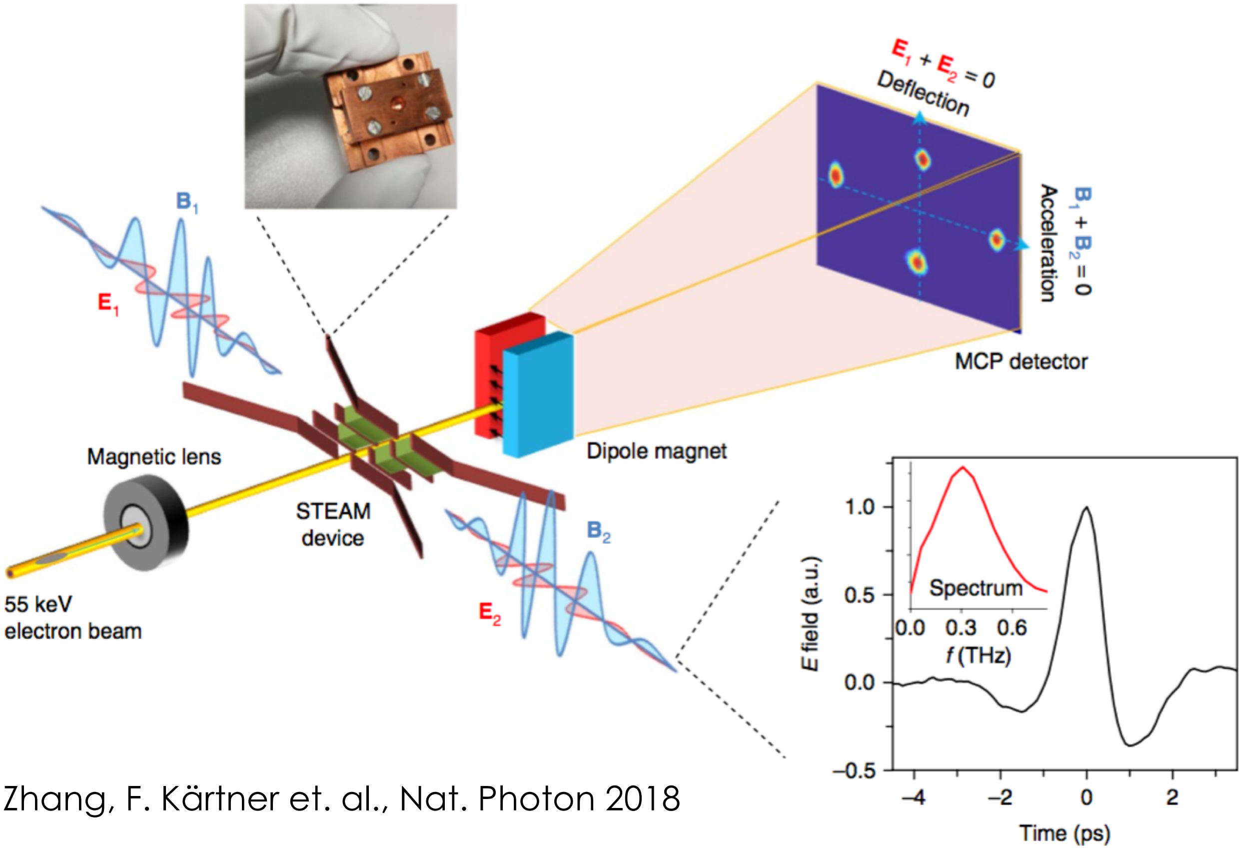
ARTICLES
https://doi.org/10.1038/s41566-018-0138-z
nature photonics

Segmented terahertz electron accelerator and manipulator (STEAM)

Dongfang Zhang[1,2,5]*, Arya Fallahi[1,5], Michael Hemmer[1], Xiaojun Wu[1,4], Moein Fakhari[1,2], Yi Hua[1], Huseyin Cankaya[1], Anne-Laure Calendron[1,2], Luis E. Zapata[1], Nicholas H. Matlis[1] and Franz X. Kärtner[1,2,3]

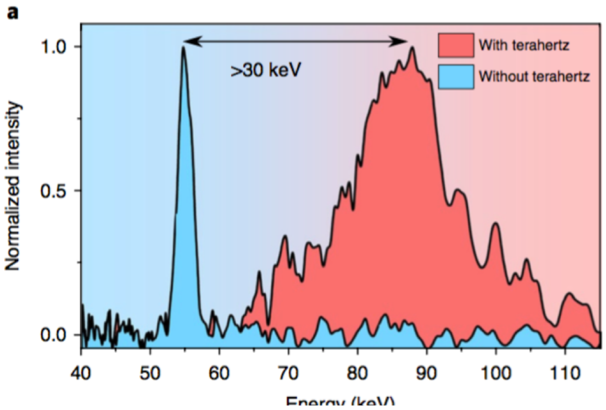
Published in April 2018

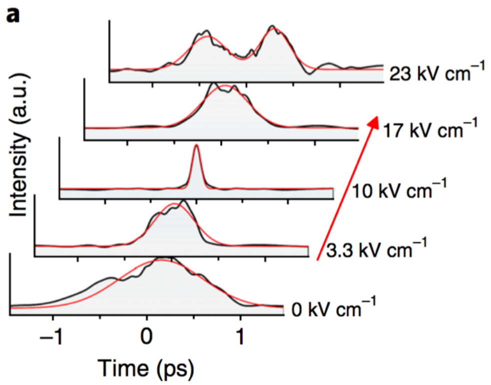
STEAM power

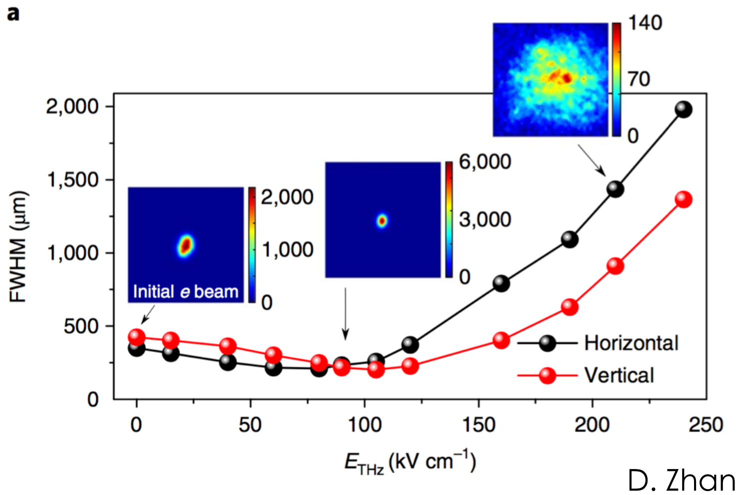


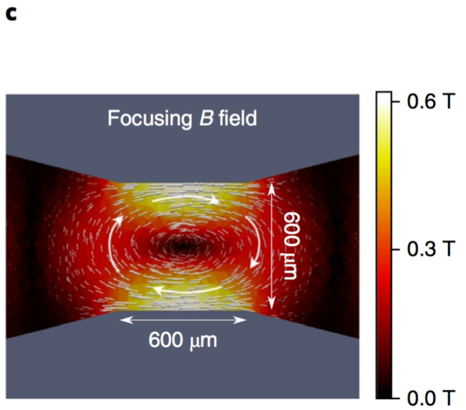
D. Zhang, F. Kärtner et. al., Nat. Photon 2018

Boosting, compression, lensing









D. Zhang, F. Kärtner et. al., Nat. Photon 2018

Conclusions

- Currently a renaissance in THz tool development.
- 10's of MV/cm peak THz source + pulse shaping for **multi-pulse NMR spectroscopy in the THz band**.
- Cold field emission of electrons from nanotips promising for next generation ultrafast instrumentation.
- **THz control of particle beams is here** and developing rapidly. Huge payoff if it can be made to work.

Thanks

McGill
Lauren Gingras, PhD
Dominique Matte, PhD
David Valverde-Chavez, PhD
Yang Lan, PhD
Benjamin Dringoli, MSc
Bradley Siwick
Peter Grutter
Mark Sutton

Northwestern University
Costas Stoumpos
Mercouri Kanatzidis

Lund University
Villy Sundstrom
Arkady Yartsev
Carlito Ponseca Jr.

University of Ottawa
Jean-Michel Menard
Wei Cui
Aidan Schiff-Kearn

NIST
Paul Haney

NSERC CRSNG
INNOVATION.CA
CANADA FOUNDATION FOR INNOVATION | FONDATION CANADIENNE POUR L'INNOVATION
Économie, Innovation et Exportations Québec
Fonds de recherche sur la nature et les technologies Québec