Shaping the collective interaction of relativistic electrons with matter

Physics and Applications of High Brightness Beams

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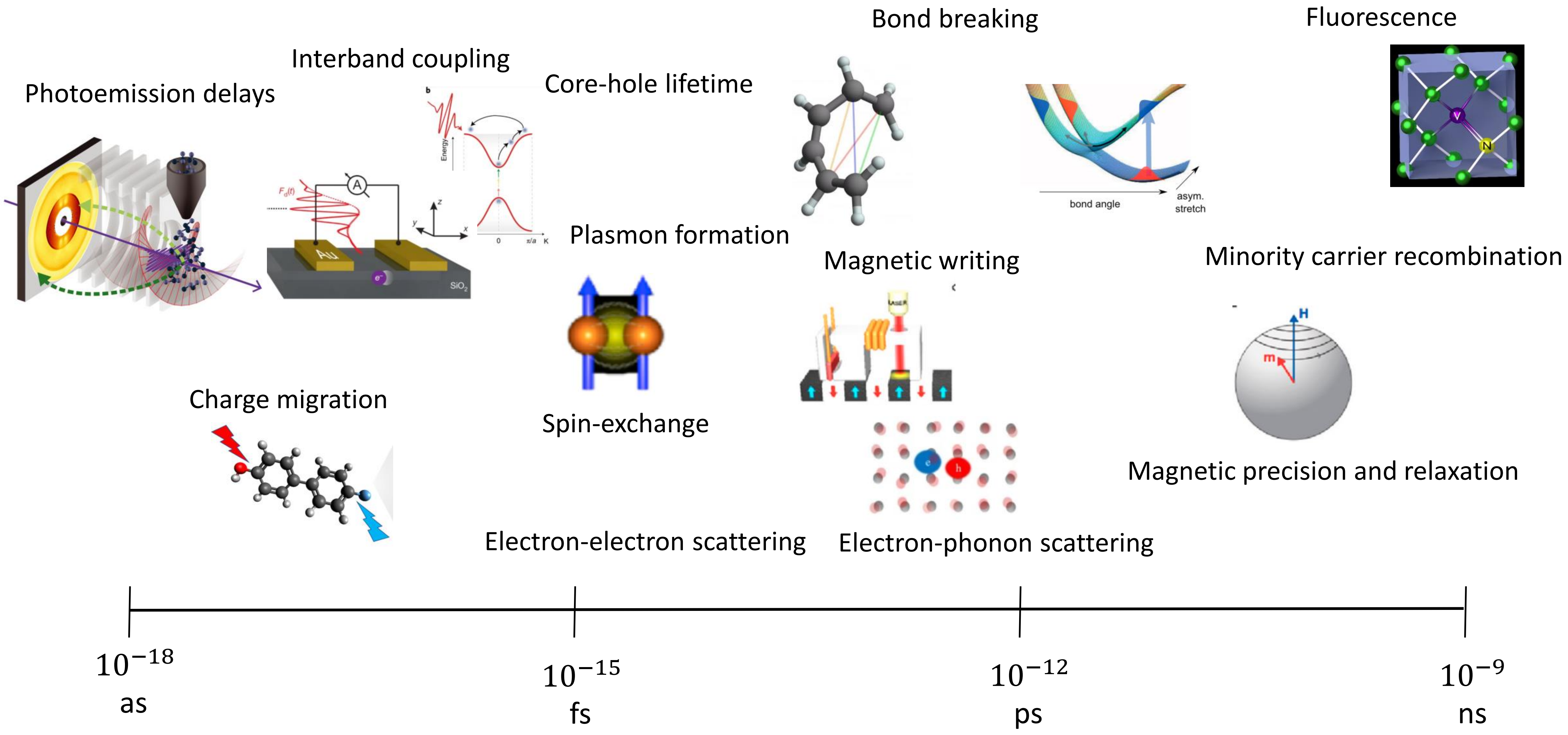
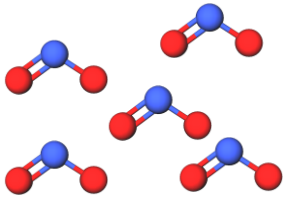
Stanford University | U.S. DEPARTMENT OF ENERGY

The world in motion: ultrafast science



The pump-probe paradigm

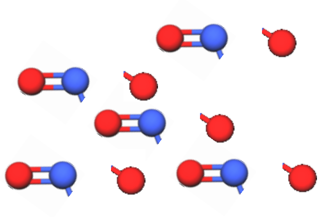
(1) Pump



(2)

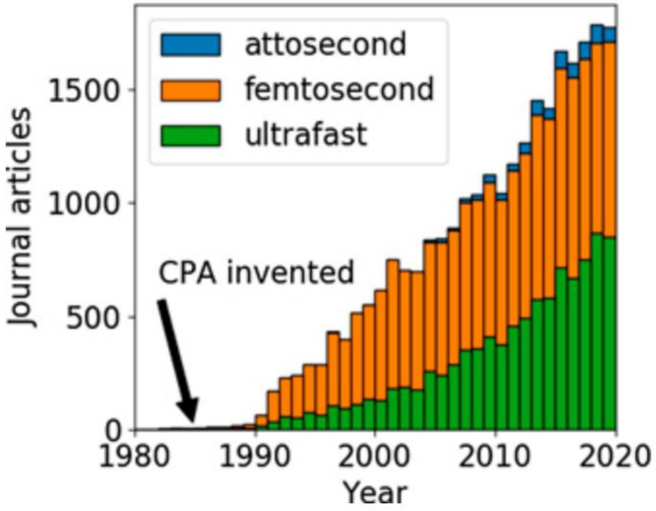
(controlled delay while system evolves)

(3) Probe



(4)

(Repeat with new delay)

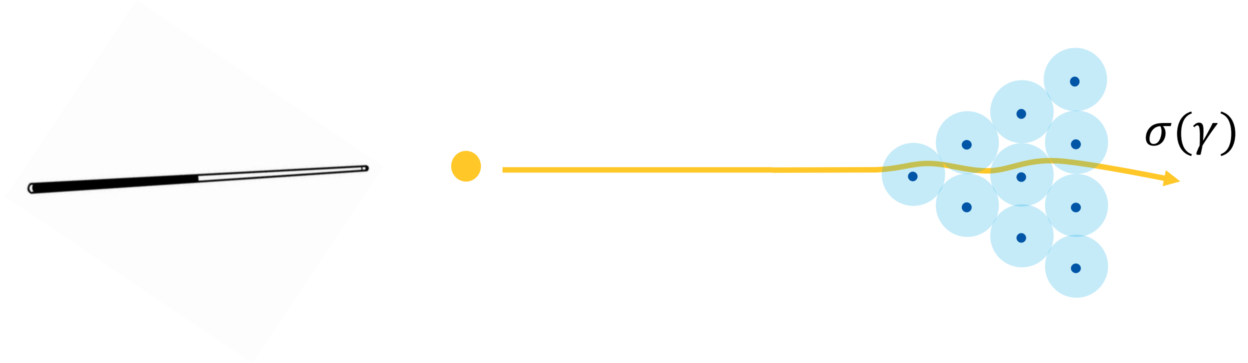


The probe may be, for example: XFEL, laser, or electron beam; but the pump is almost always a laser

Lloyd-Hughes, Hegmann et. al. “The 2021 ultrafast spectroscopic probes of condensed matter roadmap” J.o.P. Condensed Matter (2021)

Interaction of electrons with matter

In traditional usage interactions are governed by a *cross section* σ, which can be used to give the probability of an incident electron making a certain *collision*.



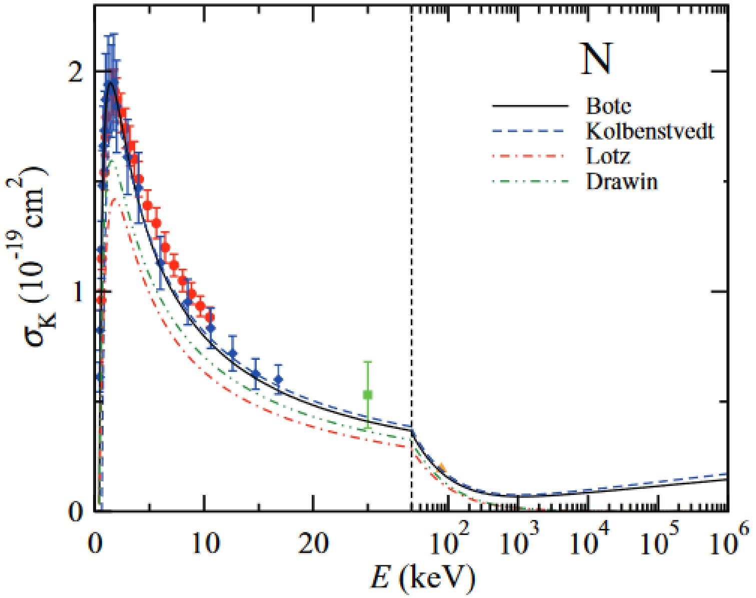
E.g. The Bethe cross section for high energies is:

$$\sigma_i^B = 2\frac{\pi q^4}{m_e v^2}(2j+1)\frac{b_i}{E_j}\left(\left[\ln\{(\gamma^2-1)\}-\beta^2\right]+\ln\left\{\frac{c_i m_e c^2}{2E_j}\right\}\right)$$

The fraction of impact ionized molecules in a sample is then:

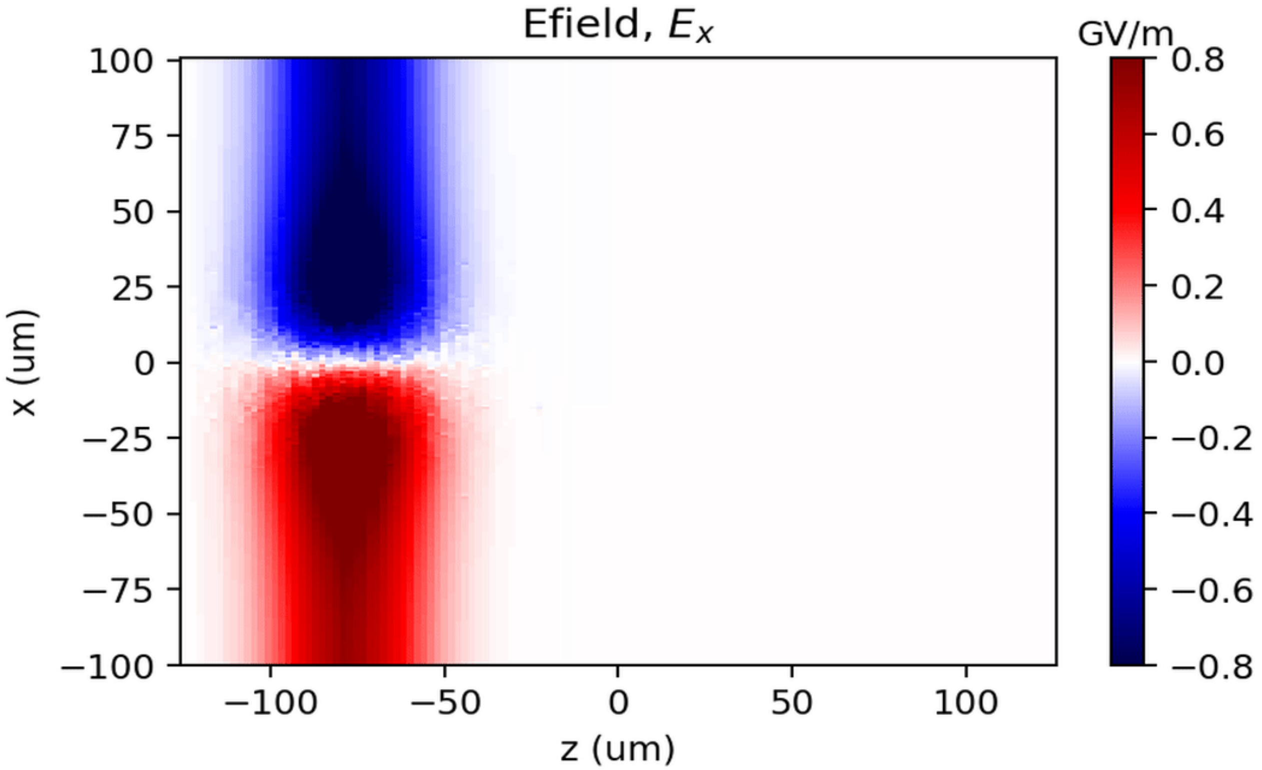
$$\sigma_i^B \frac{N_e}{\pi\sigma_r^2} \approx 10^{-6}$$

K shell impact ionization in Nitrogen



Llovet, X. “Cross sections for inner-shell ionization by electron impact” J. Phys. Chem. Ref. Data (2014)

Gaussian beam passing by a dielectric



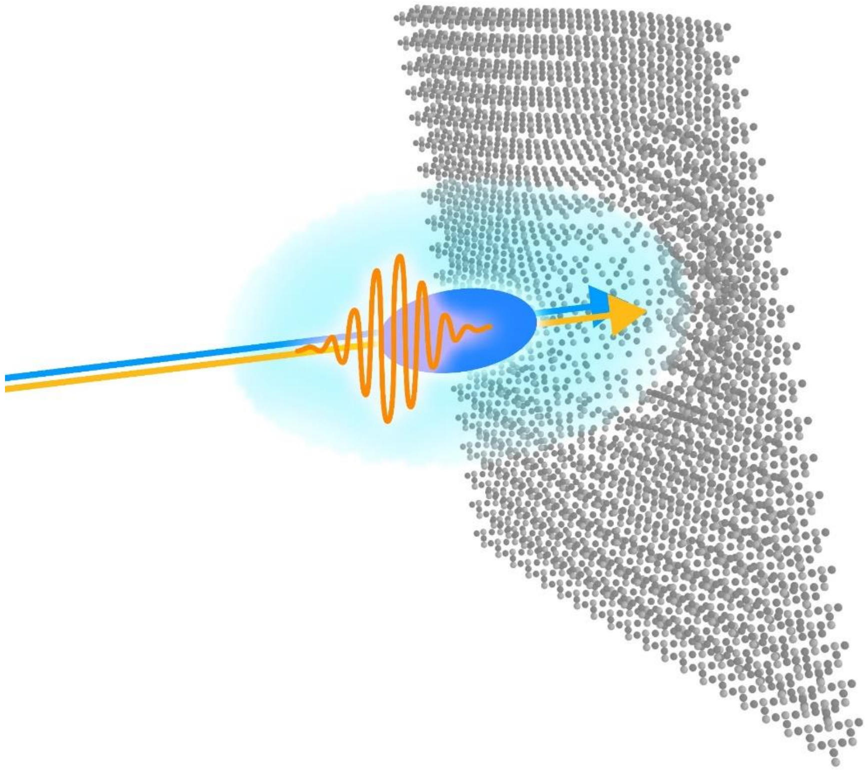
Note:
$\epsilon(\omega) = \text{const}$

We can observe the classical interaction with a dielectric block:

- Shielding/polarization
- Reflections
- Diffraction

Ref: Landau Lifschitz electrodynamics of continuous media, Chapter XIV, Section 116

PEPPEx: Photon-electron pump-probe experiment

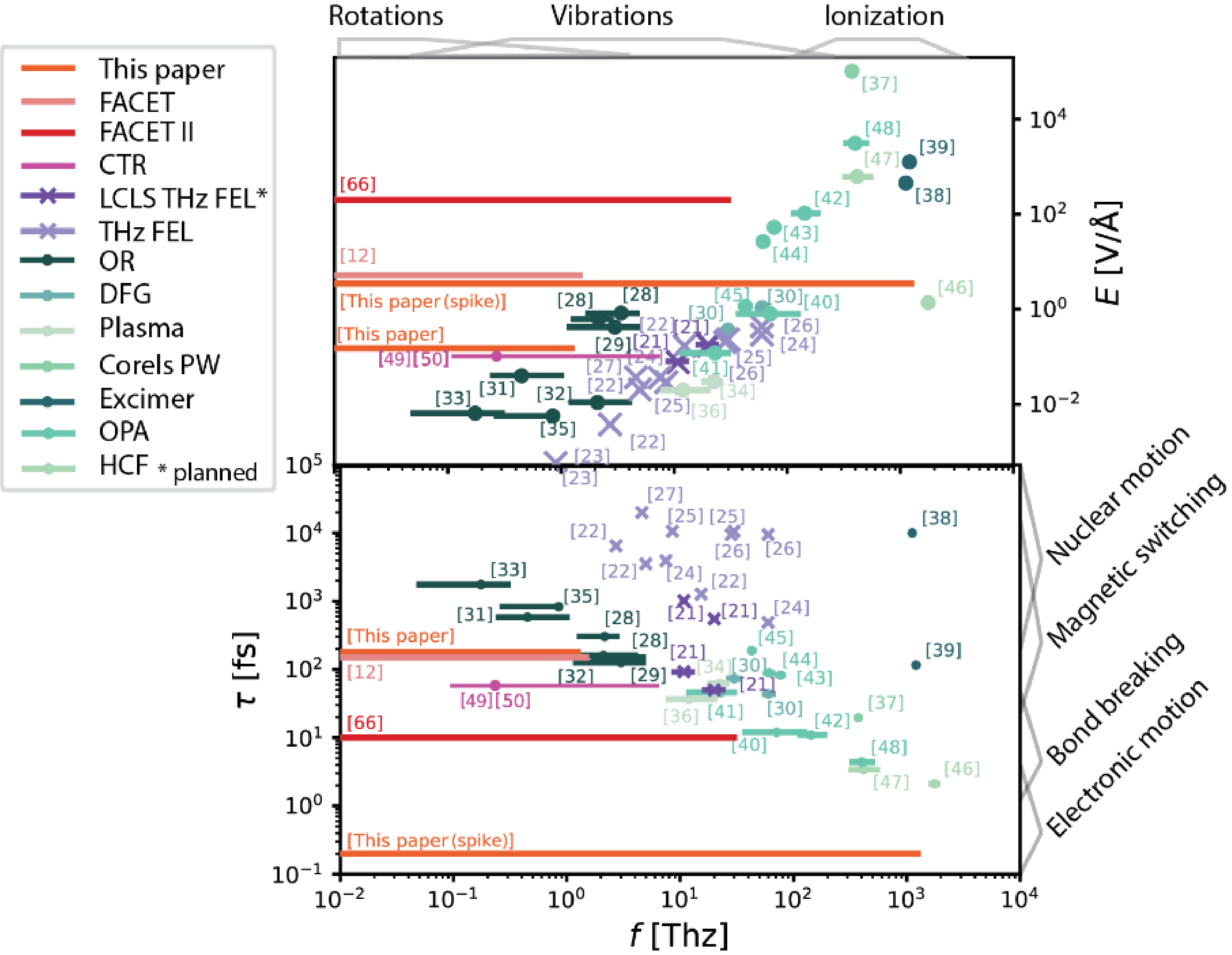


Intense electron beam excites novel ultrafast phenomena

Synchronous laser probes the resulting dynamics

D.Cesar et. al. “Ultrafast quantum dynamics driven by the strong space charge field of a relativistic electron beam” Optica (2023). Cartoon by G.Stewart (SLAC).

Space charge vs lasers

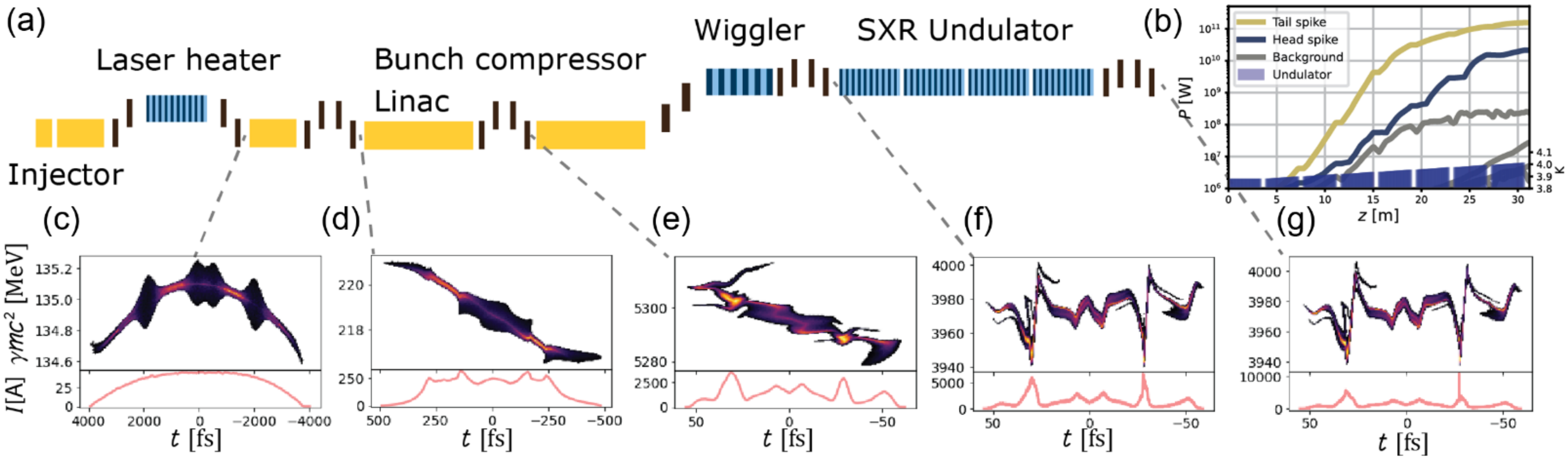


The space charge field is, compared to a laser:

- **Shorter** (for the same wavelength)
- **Stronger** (in THz and EUV regions)
- **Unipolar** (strong momentum transfer)
- **Intrinsically synchronized** and/or mutually coherent with laser pulse

D.Cesar et. al. “Ultrafast quantum dynamics driven by the strong space charge field of a relativistic electron beam” Optica (2023). Cartoon by G.Stewart (SLAC).

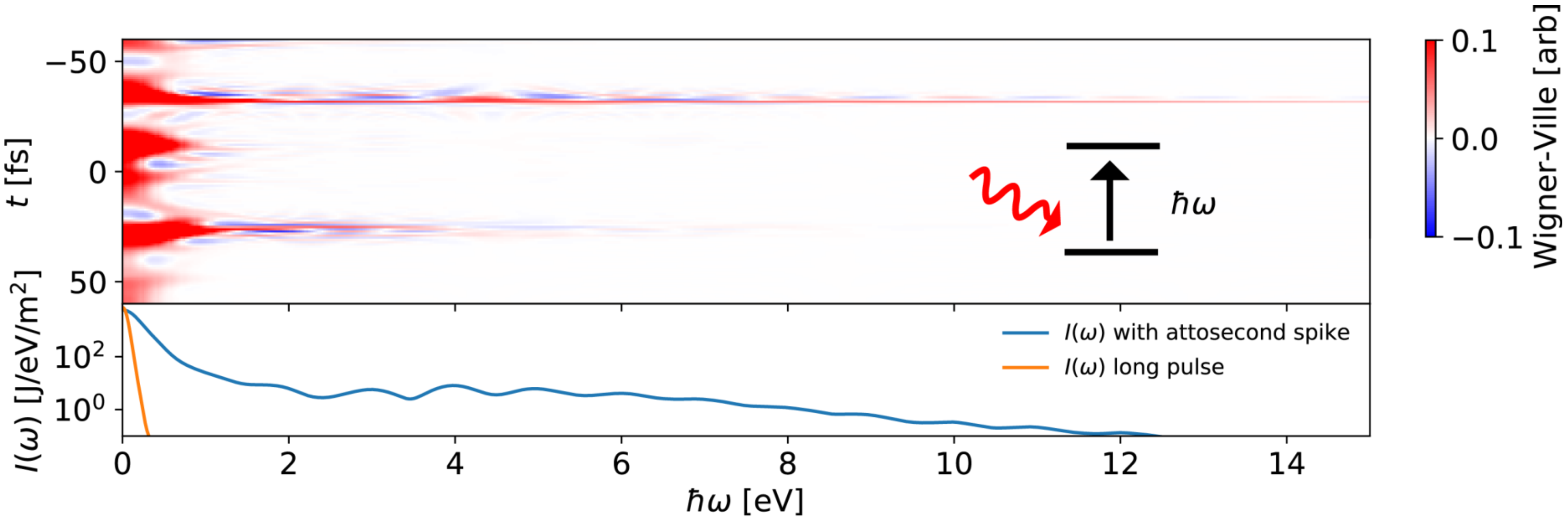
Pump-probe with attosecond current spikes



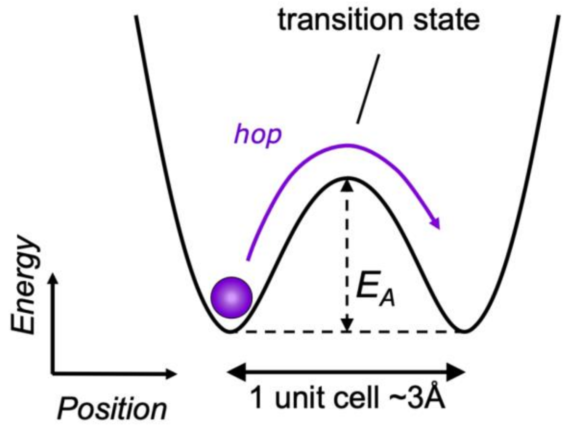
Laser heater shaping: seed the microbunching instability to shape the electron pulse

Cesar, D. *et al.* Electron beam shaping via laser heater temporal shaping. *Phys. Rev. Accel. Beams* **24**, 110703 (2021)

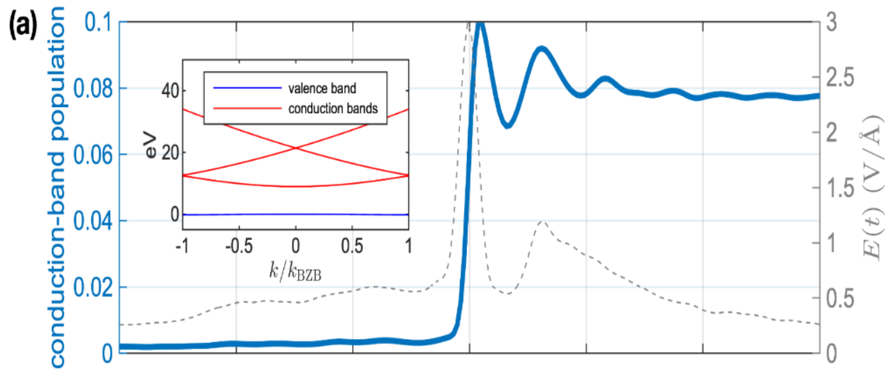
Quantum dynamics at different time scales



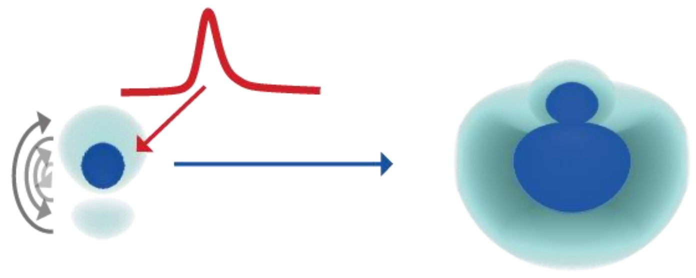
Thz driven ionic motion



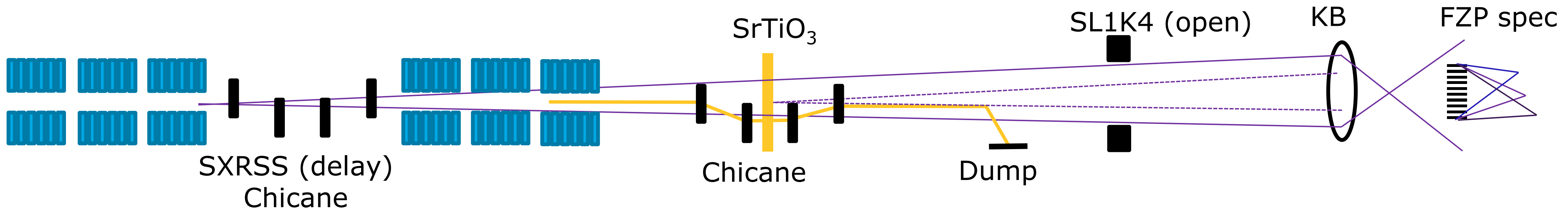
Interband and intraband electronics



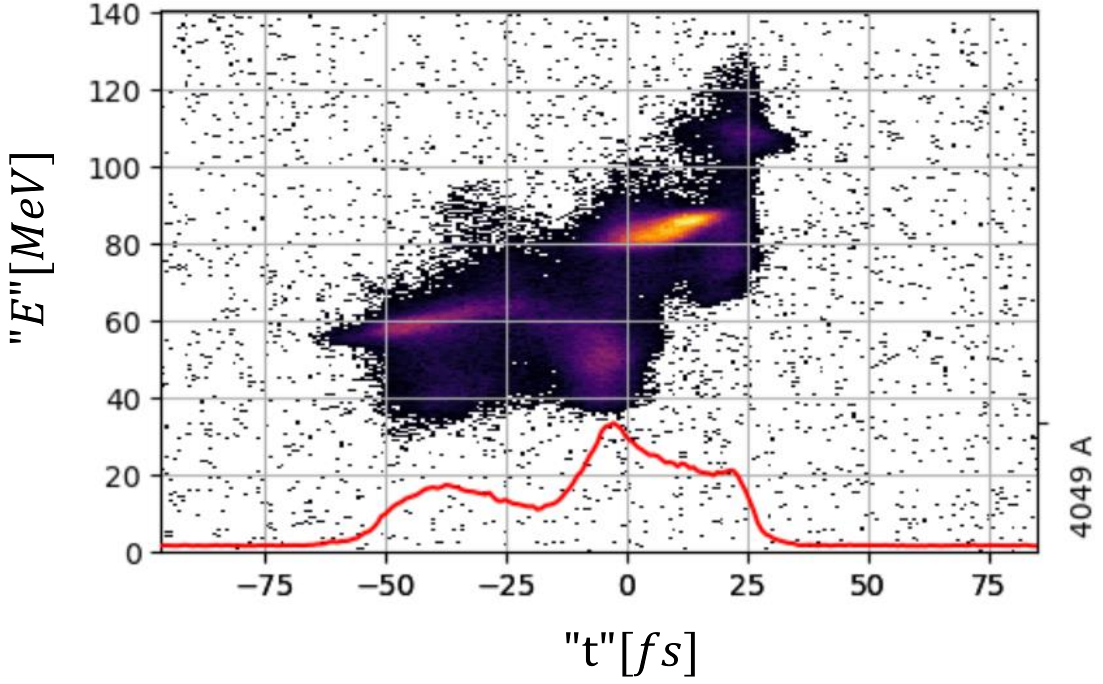
Impulsive ionization



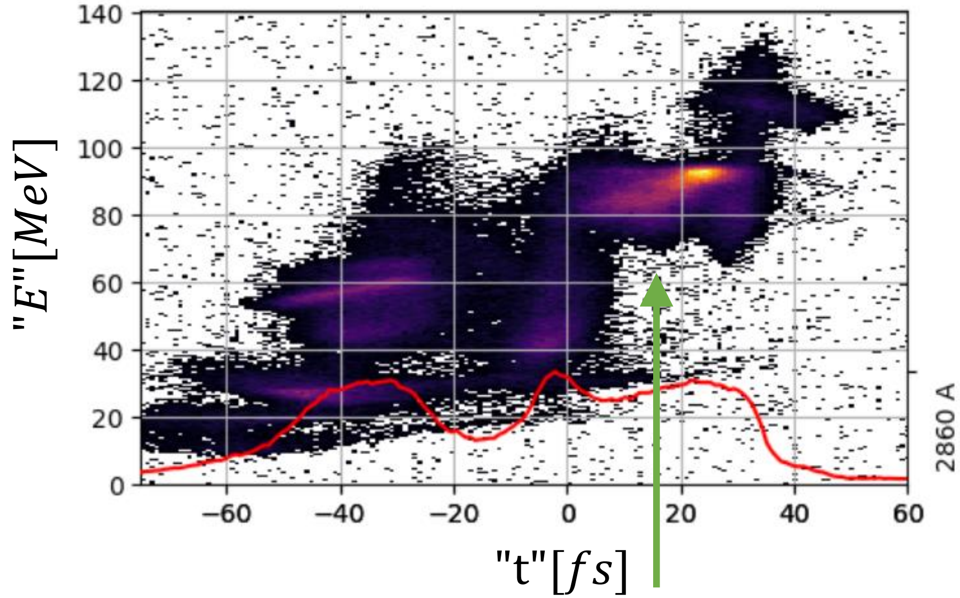
Preliminary data | temporal overlap



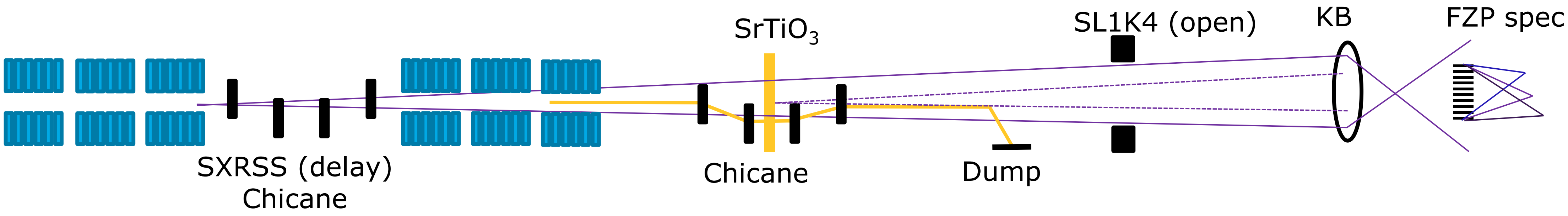
Electron beam pump

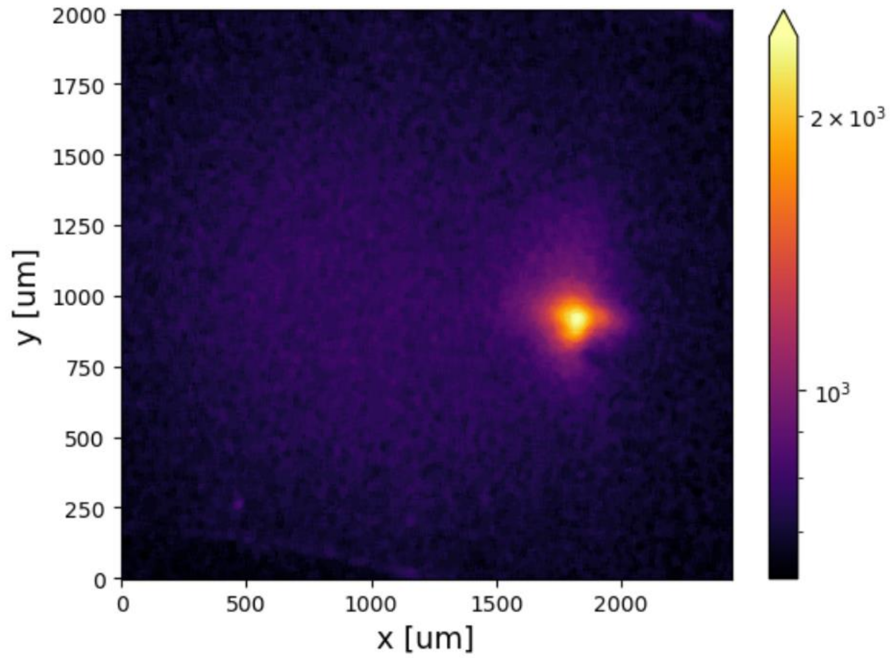


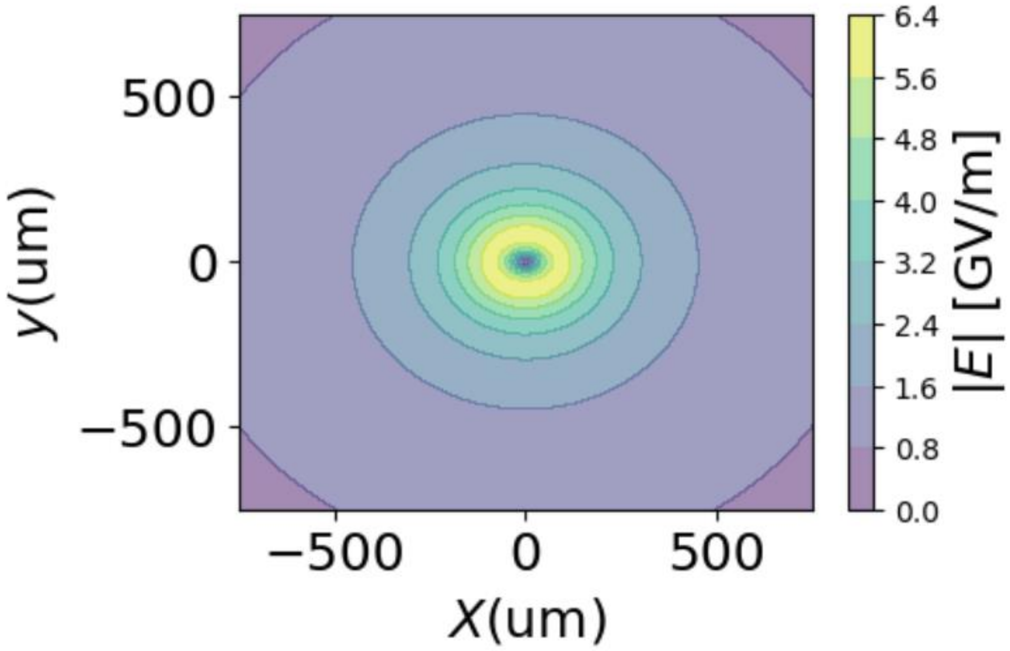
Re-amplified laser (14 fs delay)



Preliminary data | focal spot





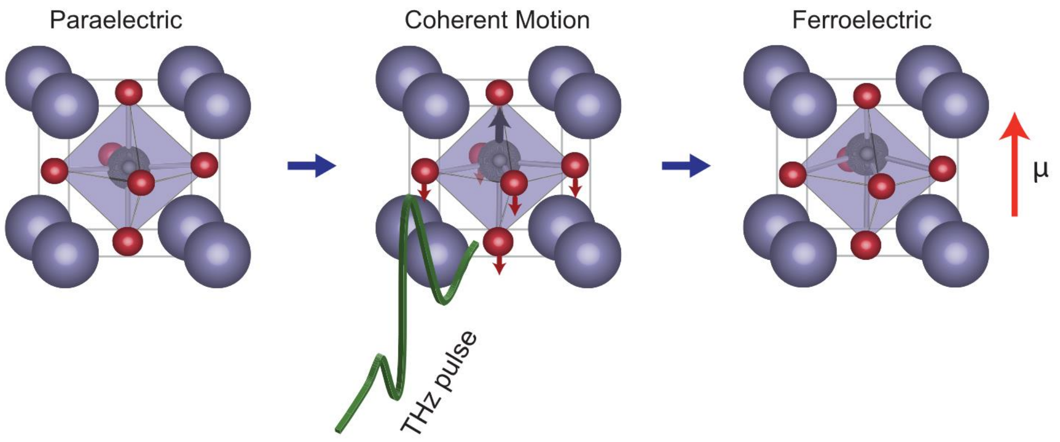


*Using projected beam size and 6kA peak current

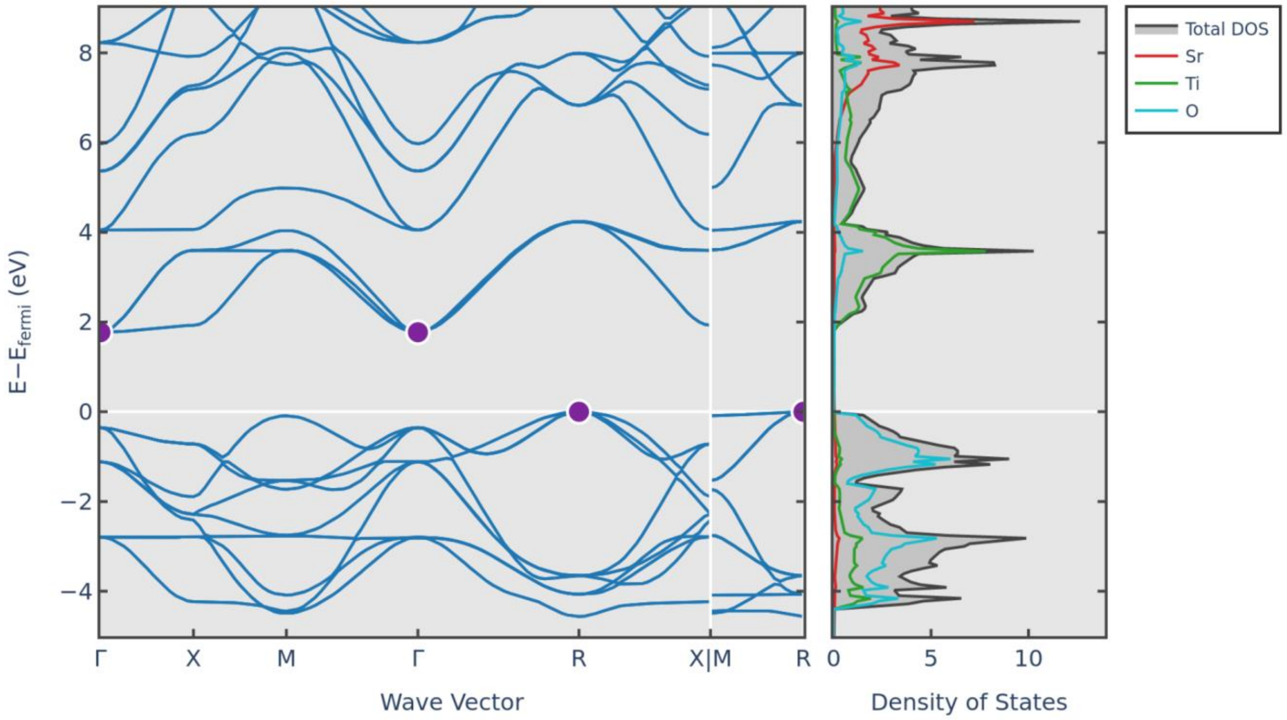
Preliminary data | Strontium Titanate

SrTiO3 is paraelectric ($\epsilon(\omega = 0) \approx 300$) which can become ferroelectric under a variety of perturbations

Thz excitation of the soft mode has been shown to induce transient ferroelectric state

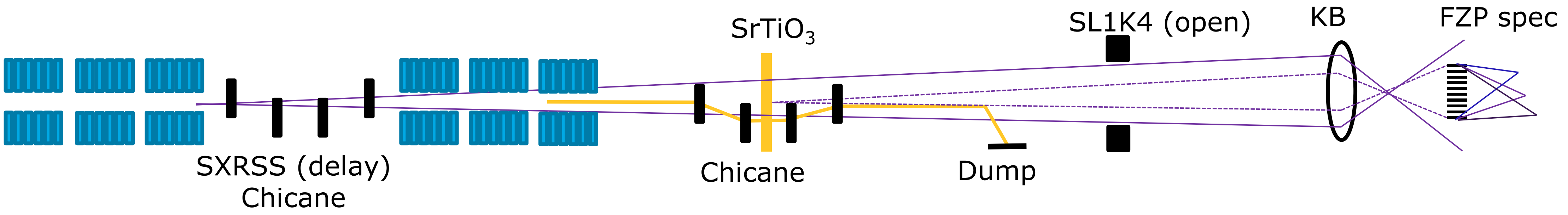


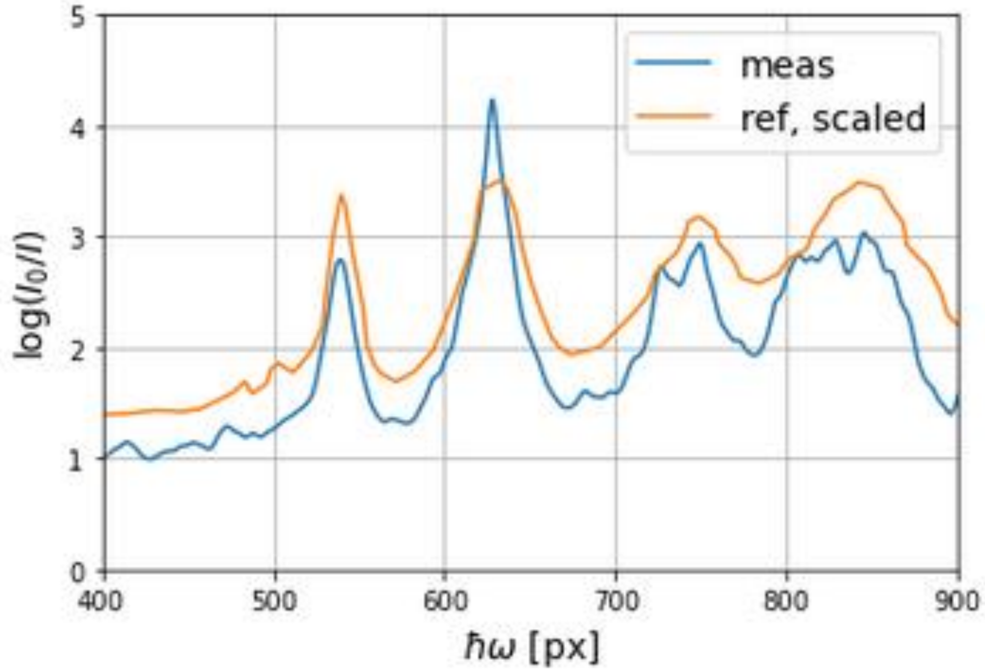
3.75 eV direct bandgap (3.25 indirect) allows a current spike to inject charge into the conduction band

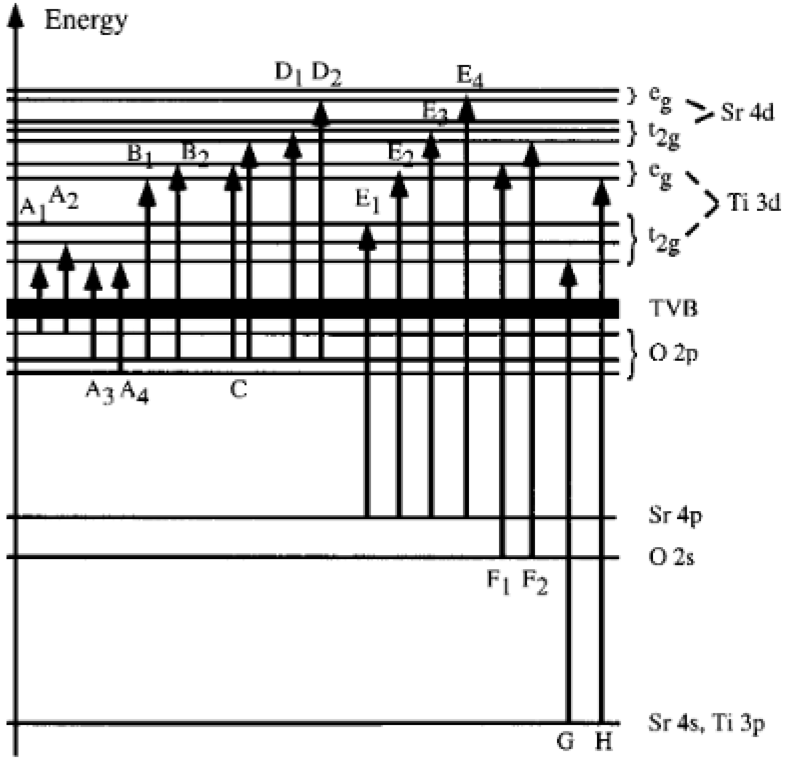


Materials project https://materialsproject.org/materials/mp-5229
Li, X. et al. Terahertz field–induced ferroelectricity in quantum paraelectric SrTiO3. *Science* 364, 1079–1082 (2019)
Nova, T. F., Disa, A. S., Fechner, M. & Cavalleri, A. Metastable ferroelectricity in optically strained SrTiO3. *Science* **364**, 1075–1079 (2019)
Kozina, M. *et al.* Terahertz-driven phonon upconversion in SrTiO3. *Nat. Phys.* **15**, 387–392 (2019).
Park, Sang Han et. al. Direct and real-time observation of hole transport dynamics in anatase TiO2 using X-ray free-electron laser Nat. Comm. (2022)

Preliminary data | transient absorption at Ti L edge

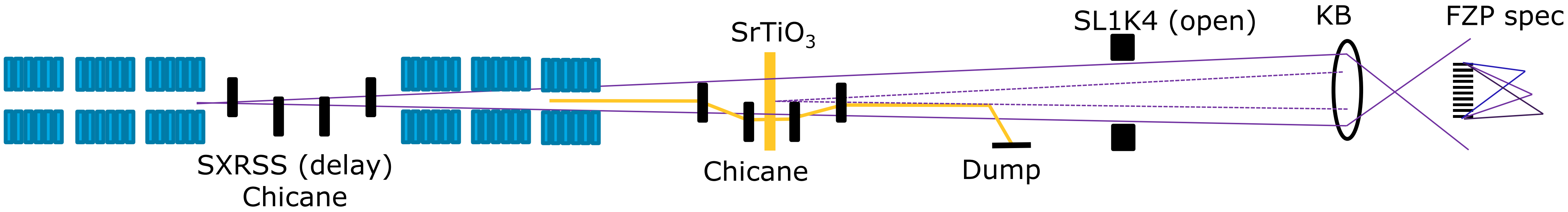


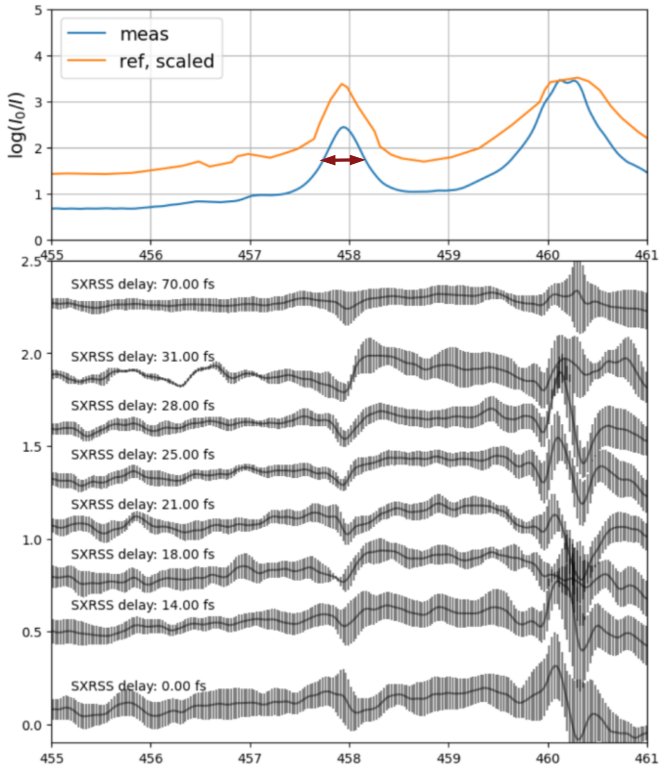


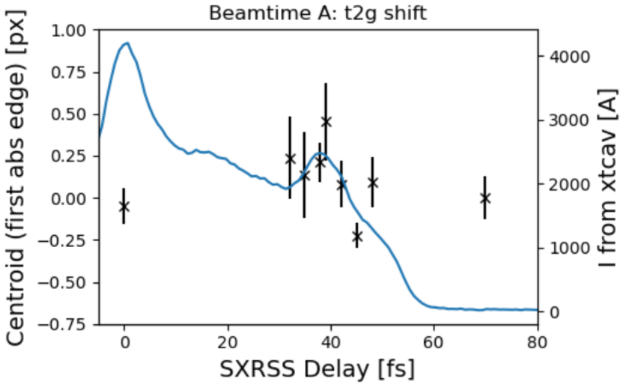


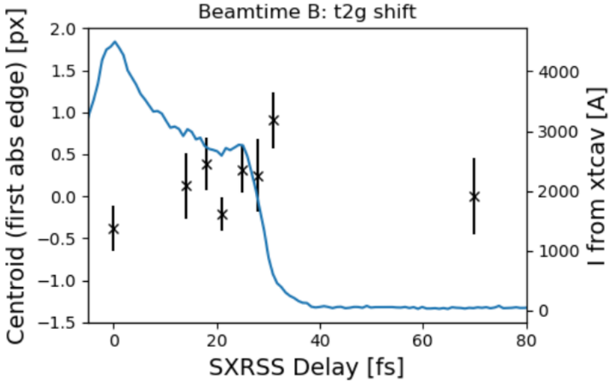
Benthem, K. "Bulk electron structure of SrTiO3 Experiment and theory" J. appl. phys. (2001)

Preliminary data | transient absorption





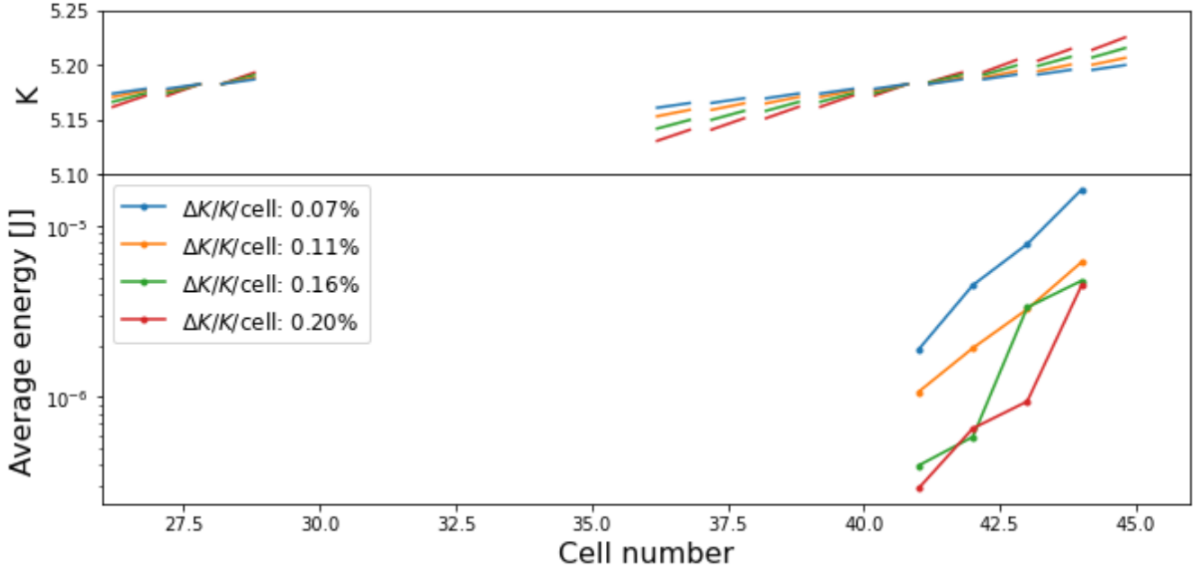


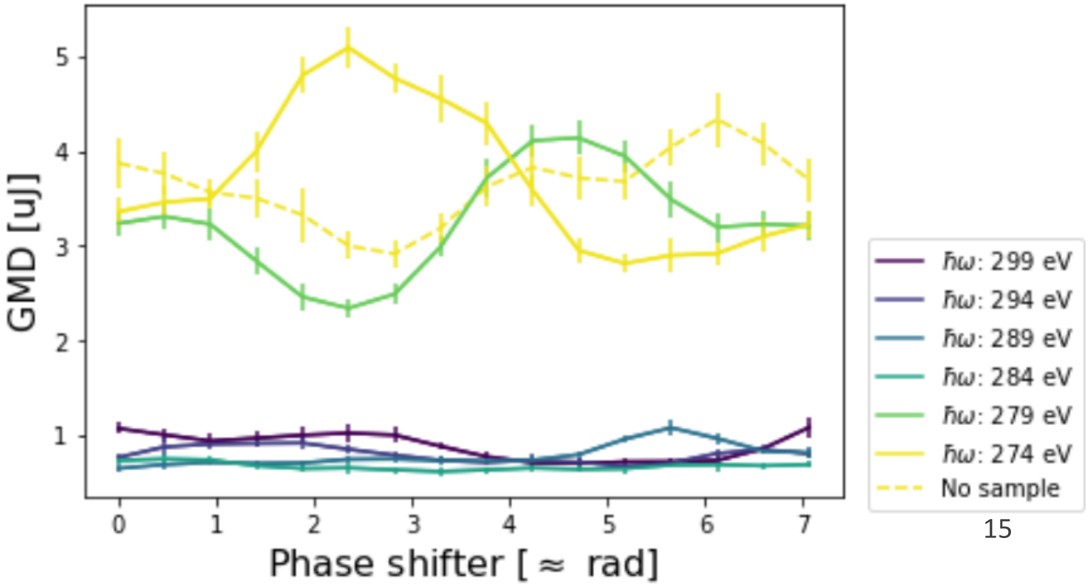


Caveats: Spectra are noisy and we have some spatial-overlap scans that we don’t yet understand

Preliminary data: Interference between xCTR and xFEL

1. The reverse taper micobunches the beam with low saturation power (xFEL)
2. The microbunched beam emits transition radiation (xCTR)
3. The total energy oscillates as a function of the phase shifter delay between the beam and the xFEL





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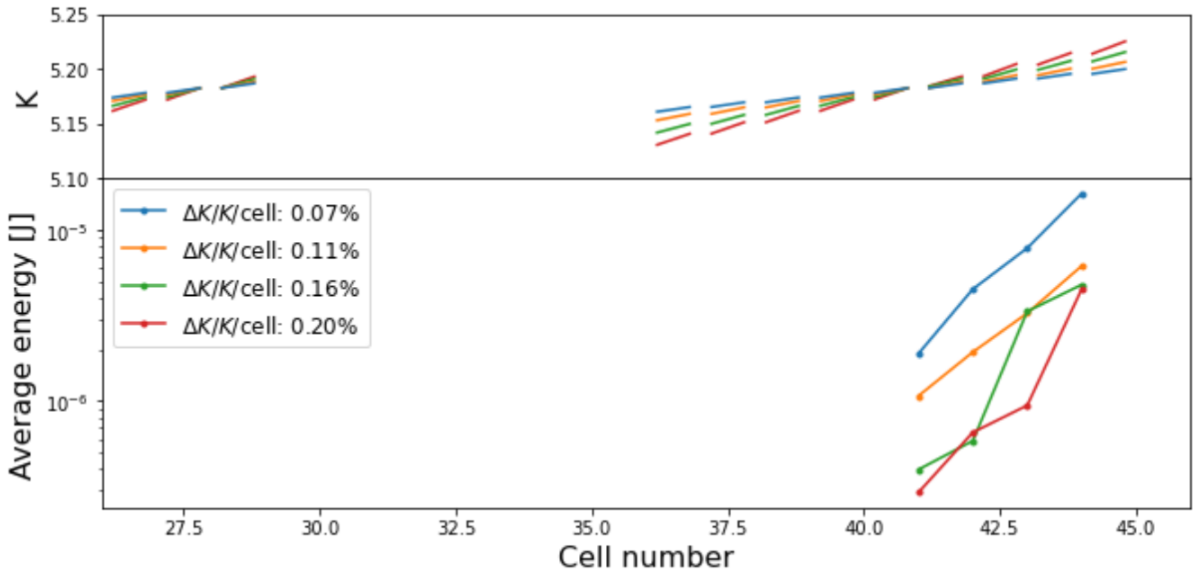

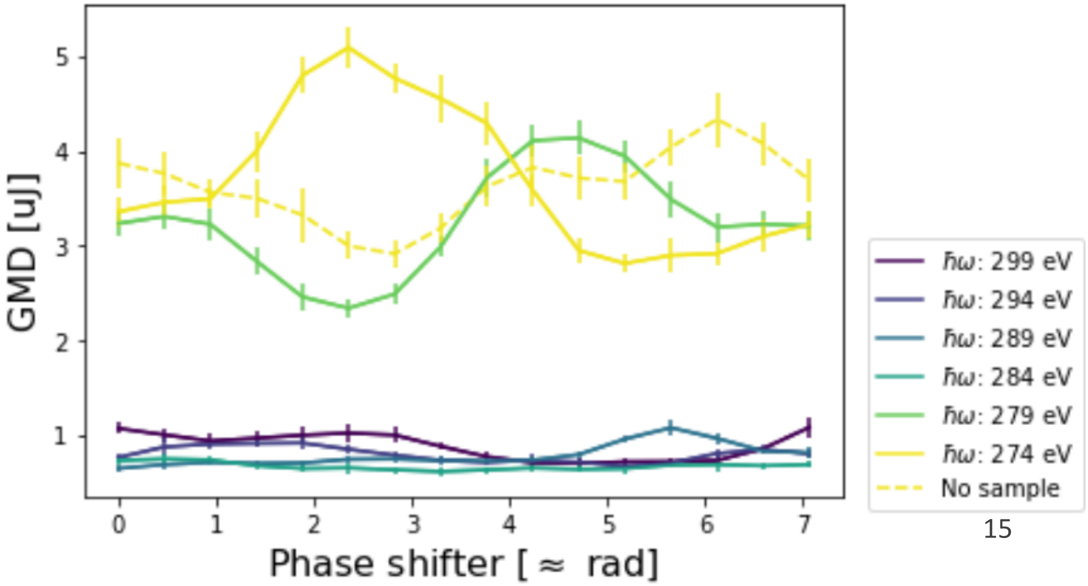

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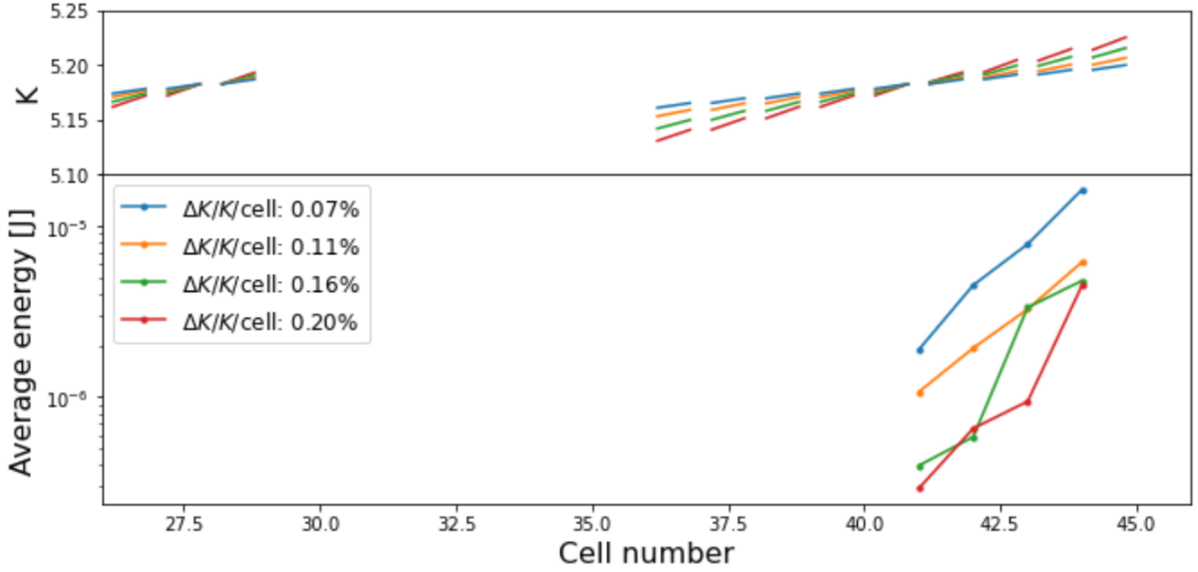

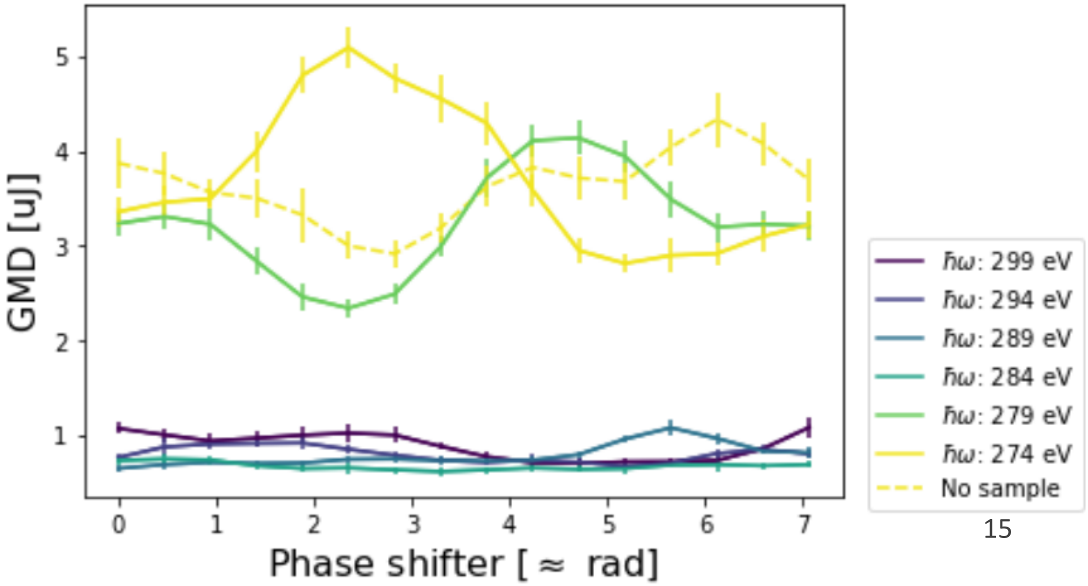

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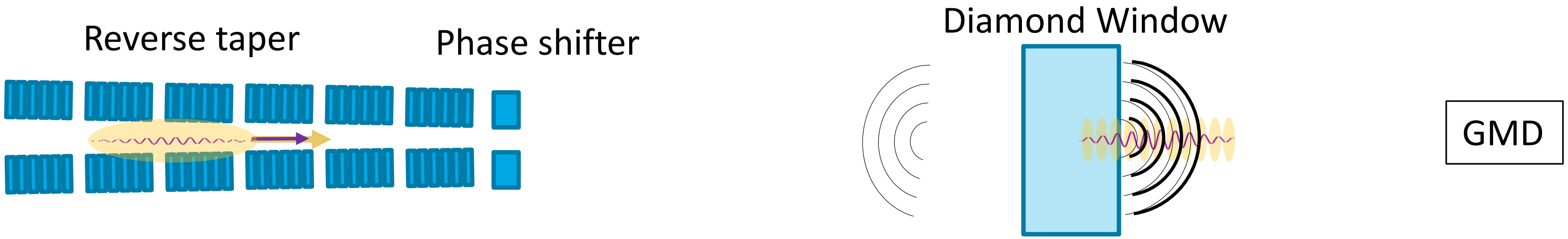

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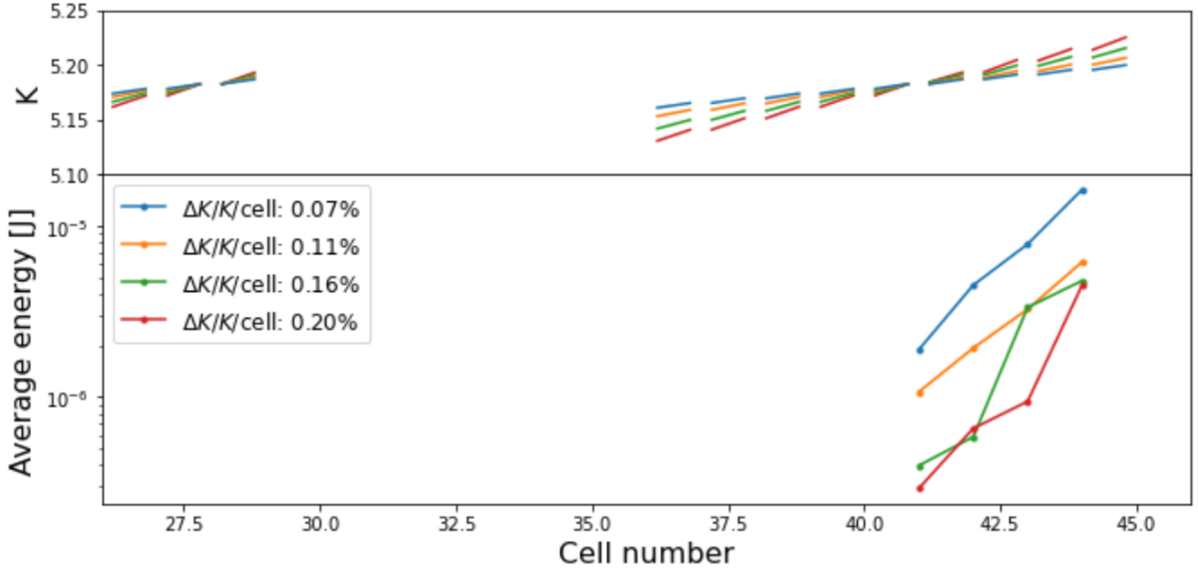

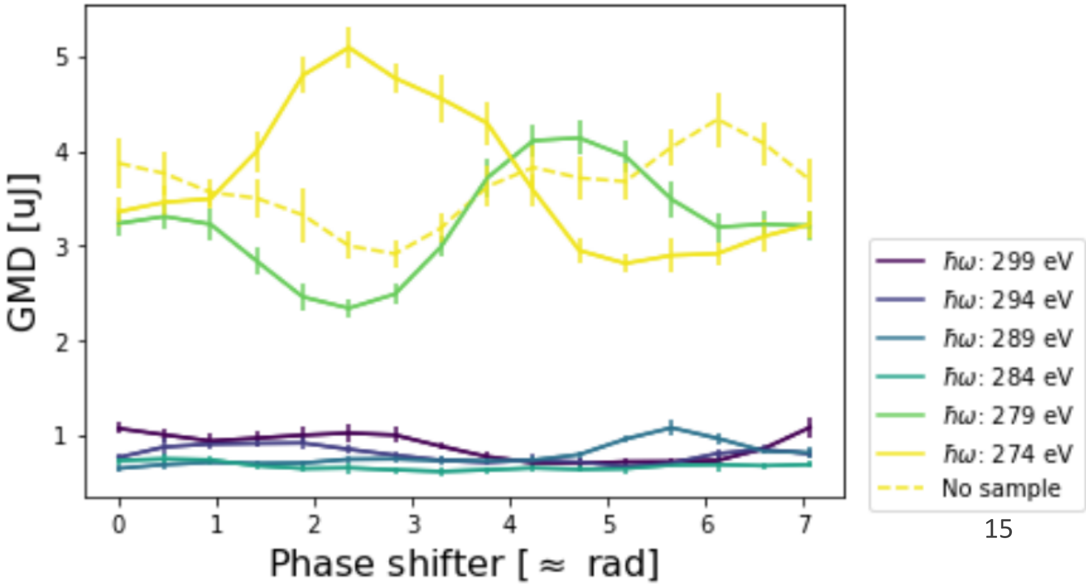

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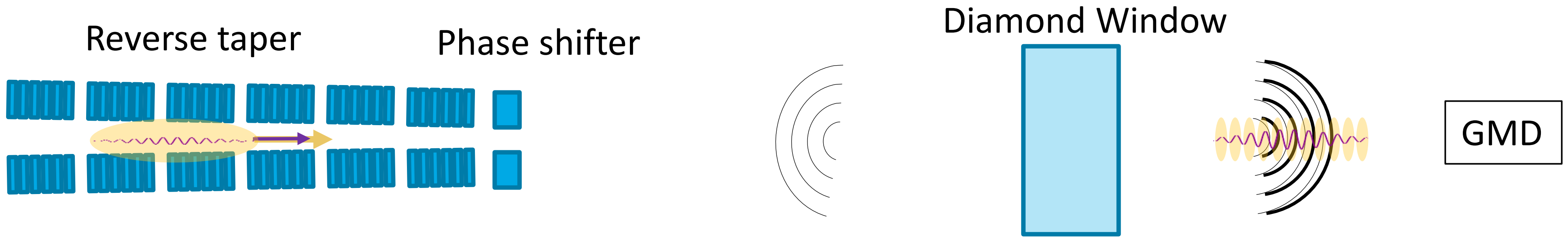

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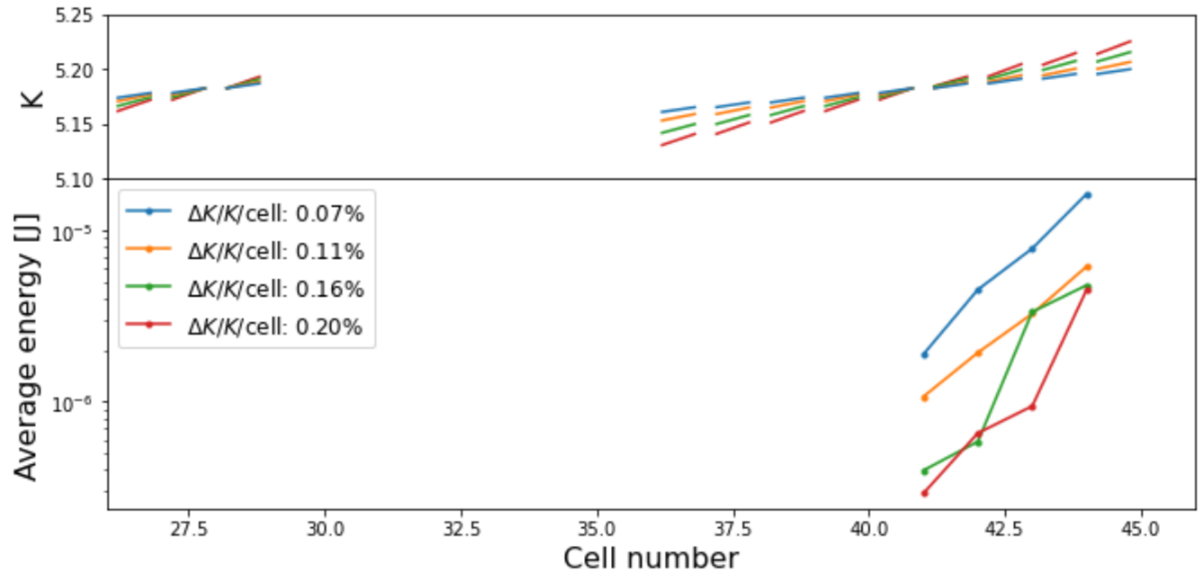

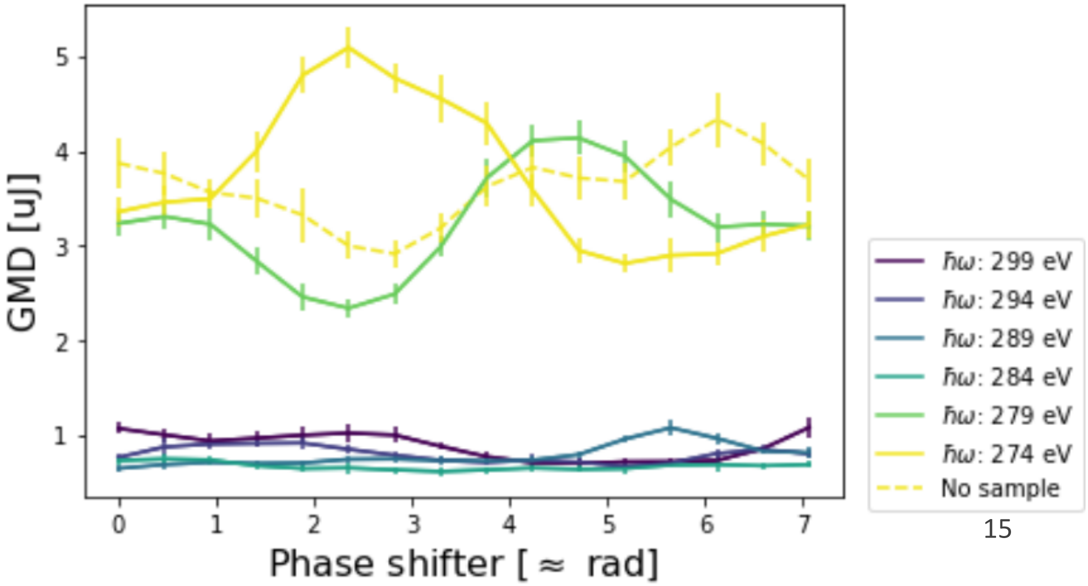

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Conclusions

The collective field of an electron beam can excite novel phenomena to help us explore the nature of ultrafast measurement.

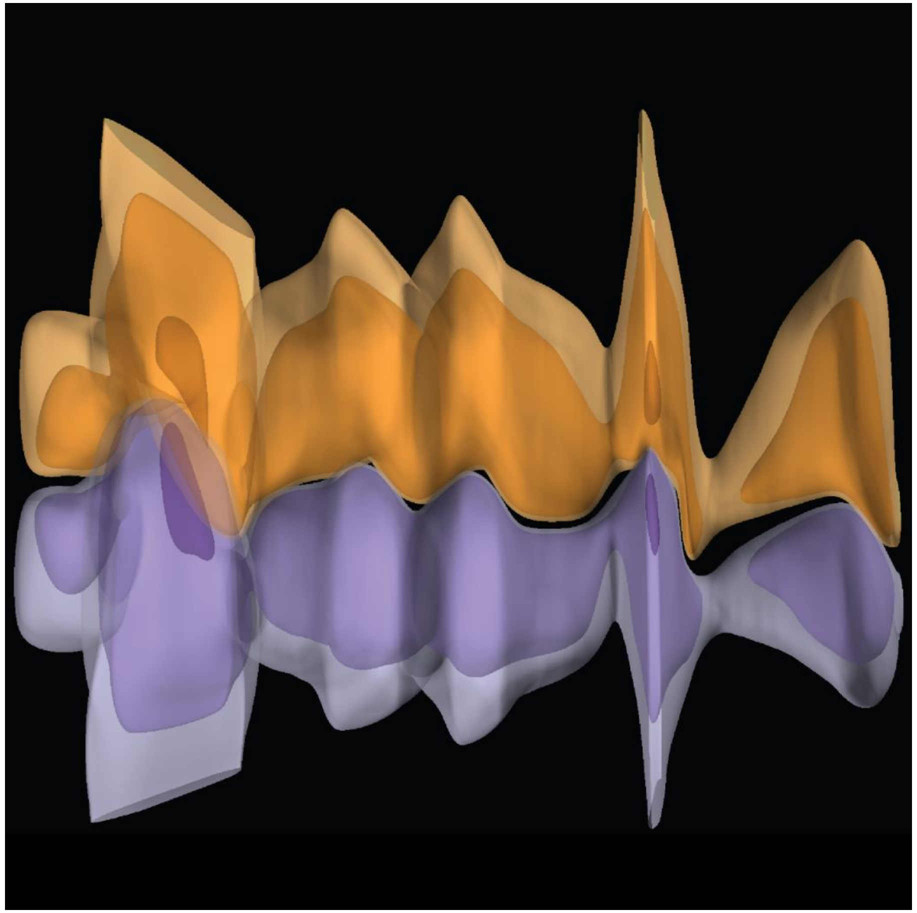
The beam field is:

- Strong, fast, and tightly focused
- Unipolar
- Intrinsically synchronized with a laser probe

It can be shaped to:

- Drive large scale ionic motion
- Excite valence electrons
- Enhance or suppress transition rates

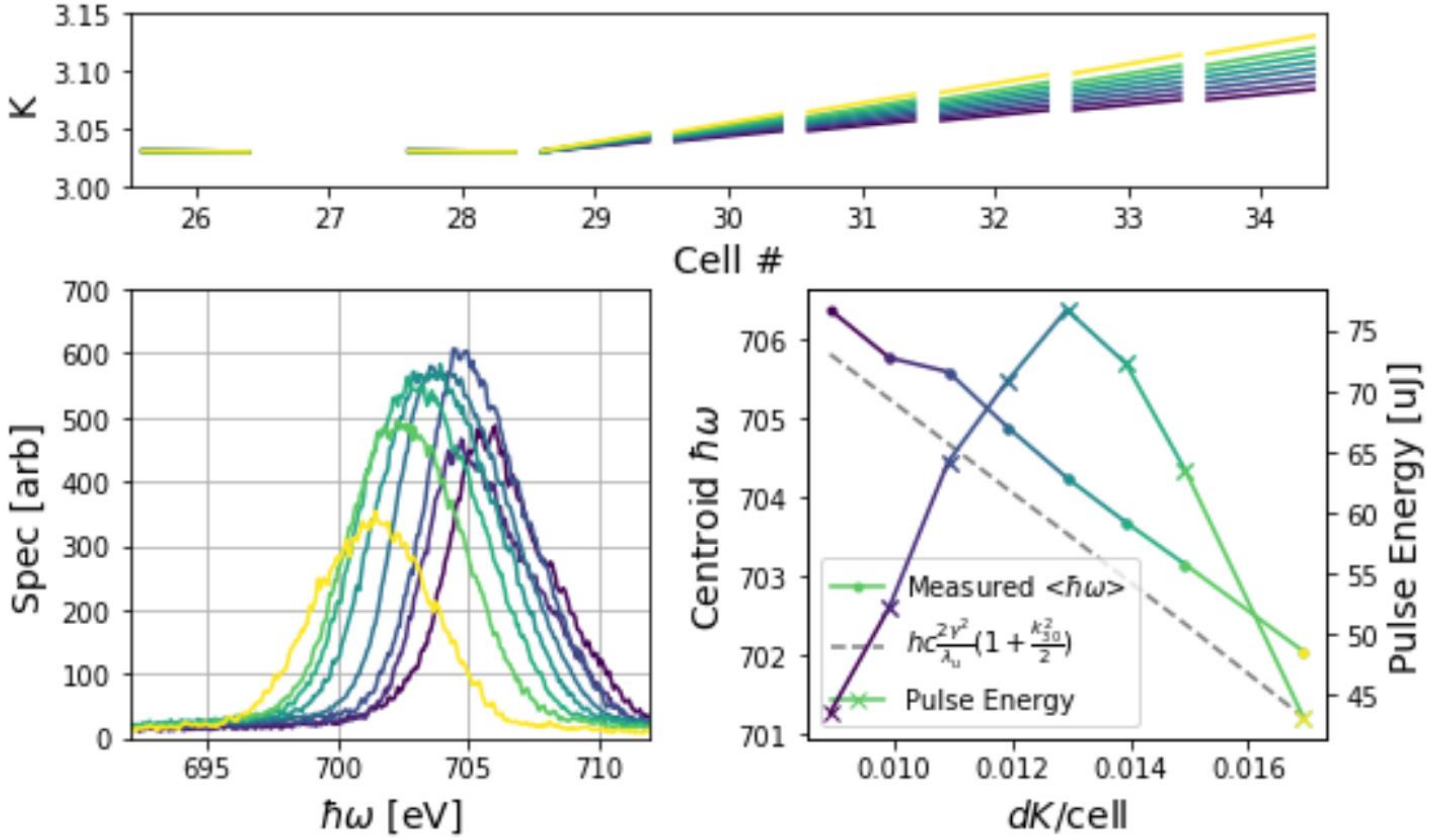
These dynamics are inherently a coherent superposition of a broadband array of states. This makes the following dynamics well suited for unambiguous ultrafast measurement.



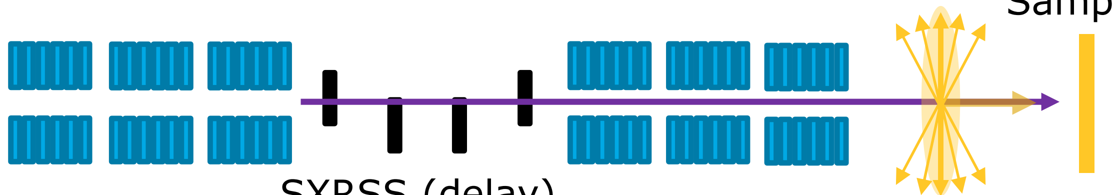
3D rendering of the space-charge field of a shaped electron beam

Backup slides

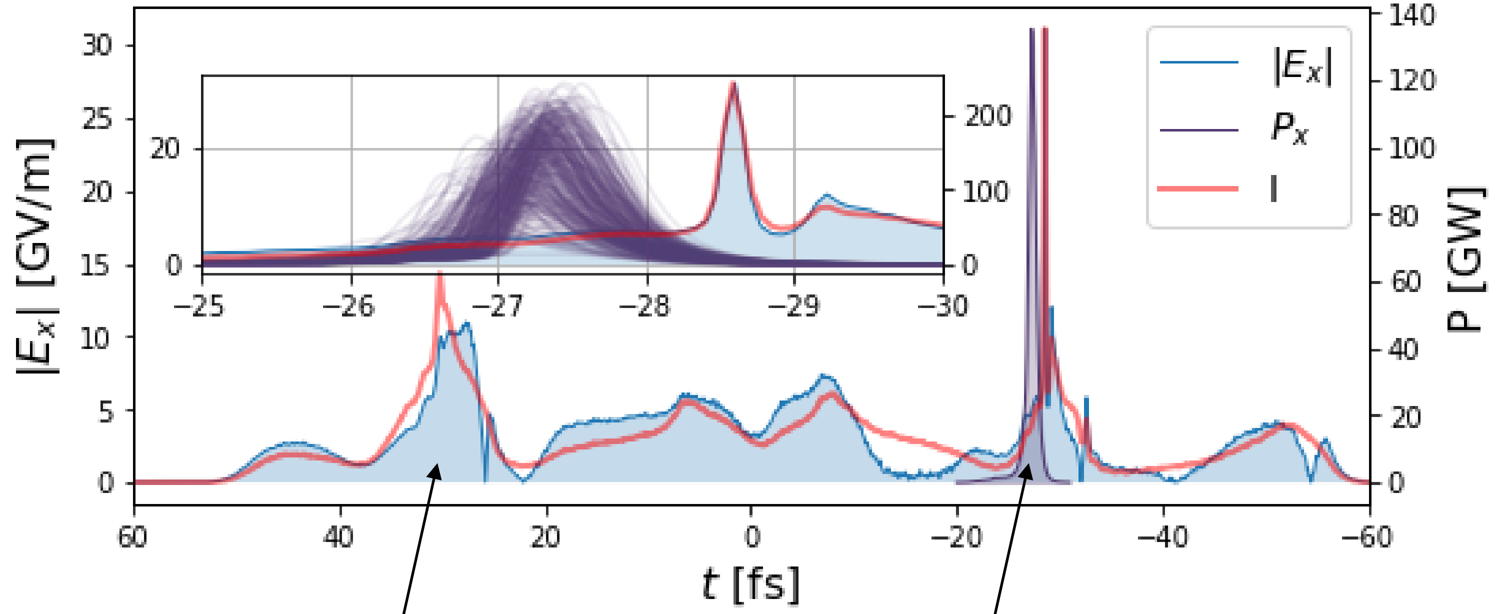
Chirp-taper matching



Attosecond pump-probe



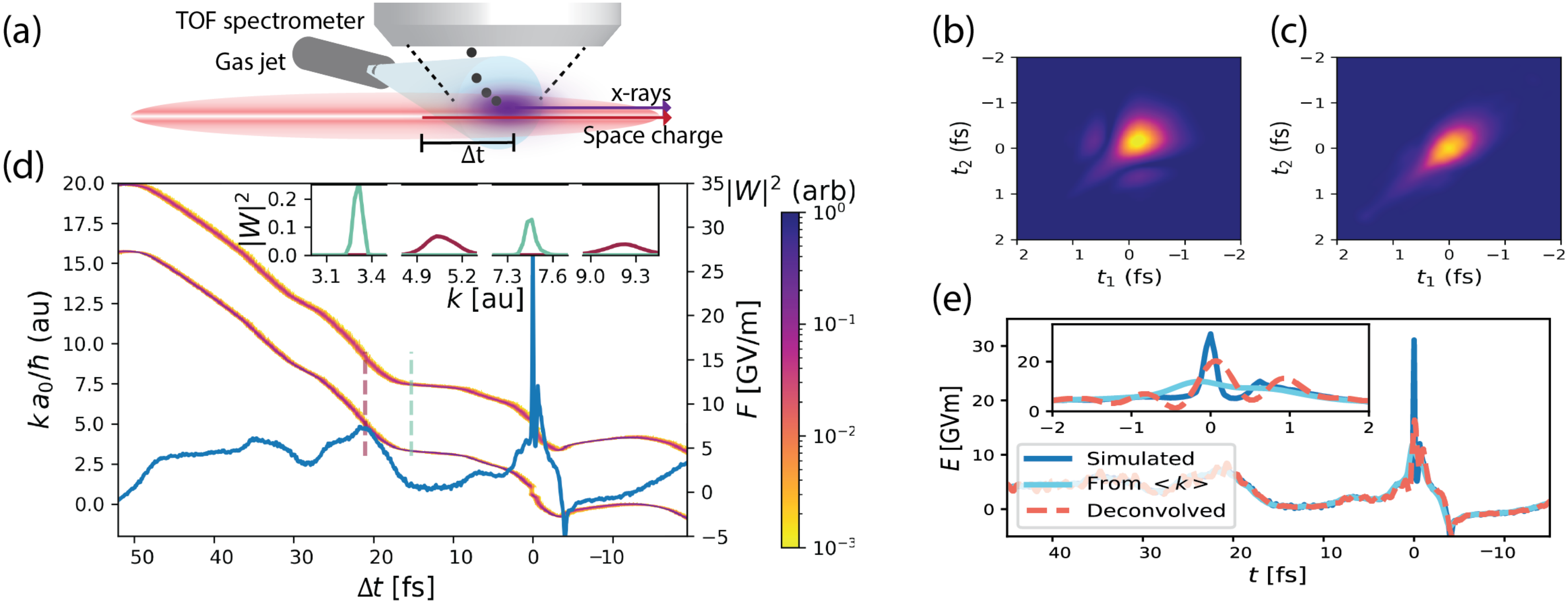
Electrons can be delayed to vary pump-probe delay



X-rays start here

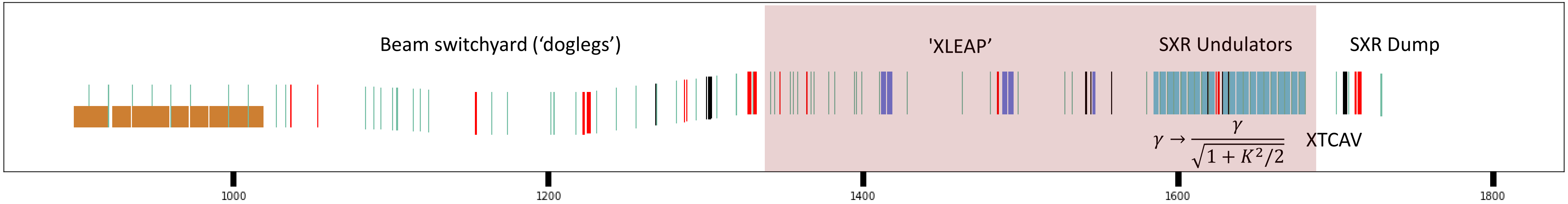
SXRSS delays to desired pump-probe delay

Ultrafast pulse metrology



D.Cesar et. al. “Ultrafast quantum dynamics driven by the strong space charge field of a relativistic electron beam”

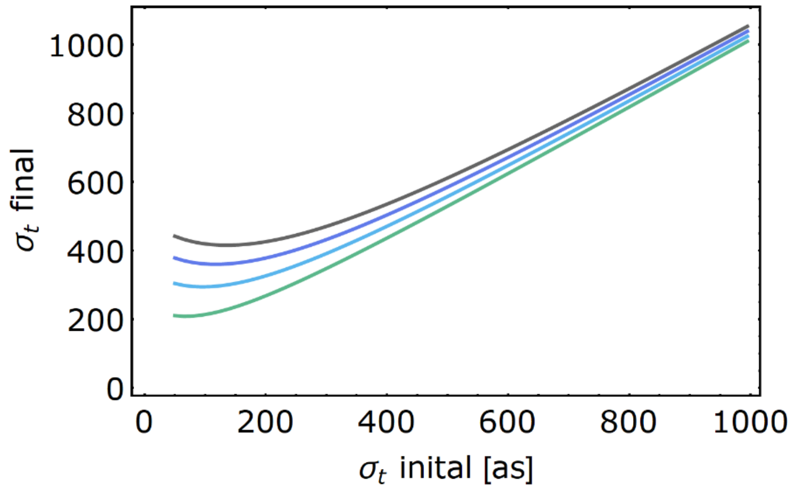
What is the limit of beam compression?

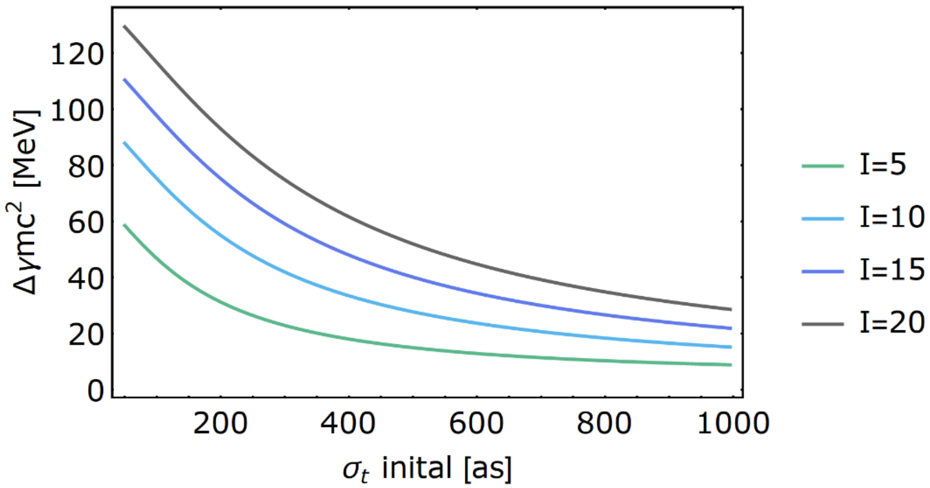


If $\lambda(z) = \frac{3N_e}{4\,L^3}\left(L^2 - z^2\right) for\ |z| < L$:

Then dynamics are entirely linear (see Chao et al slac pub 9189)

$$\Gamma = 3\ N_e \frac{r_e\left(\ln\left(\frac{b}{a}\right) + \frac{1}{2}\right)}{5^{3/2}\beta^3\gamma^3}, \text{then defocusing happens in S} \approx \frac{2\beta^3\gamma^3\sigma_z}{\sqrt{\sigma_{\Delta\gamma}^2 + 2\frac{\beta^4\gamma^4\Gamma}{\sigma_z}}}$$

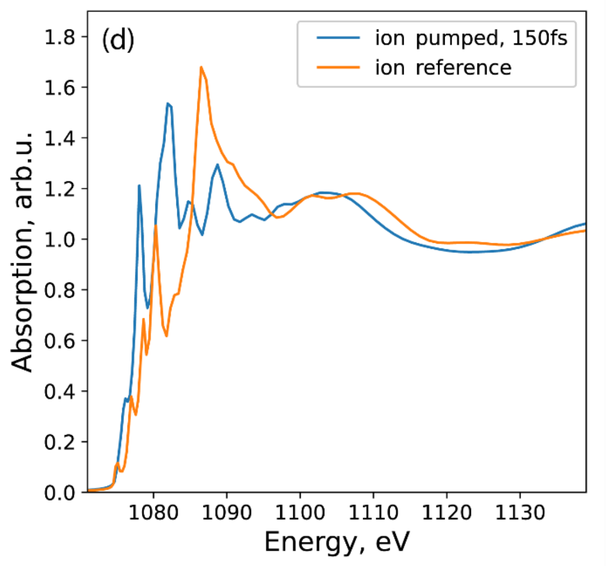




* rms conversion to Gaussian requires interpretation

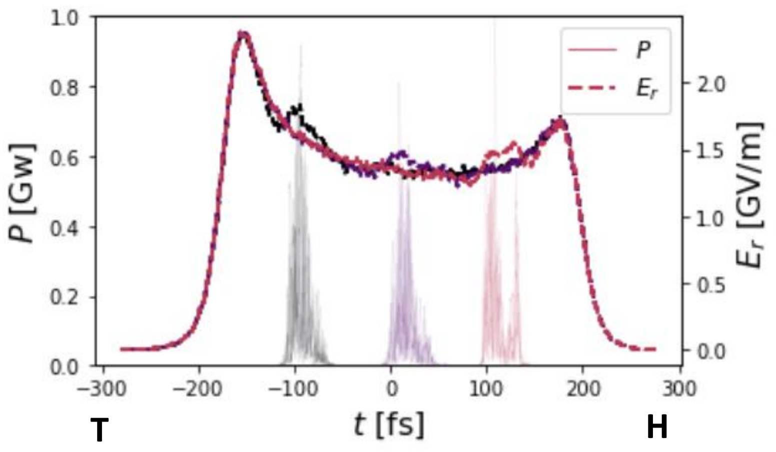
Probing ionic motion in β − Alumina

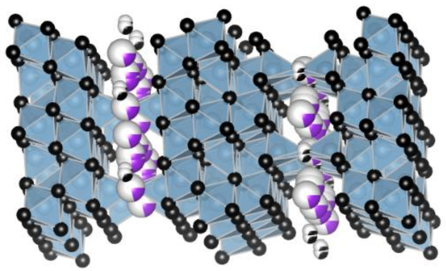
Soft x-ray absorption (FEFF)

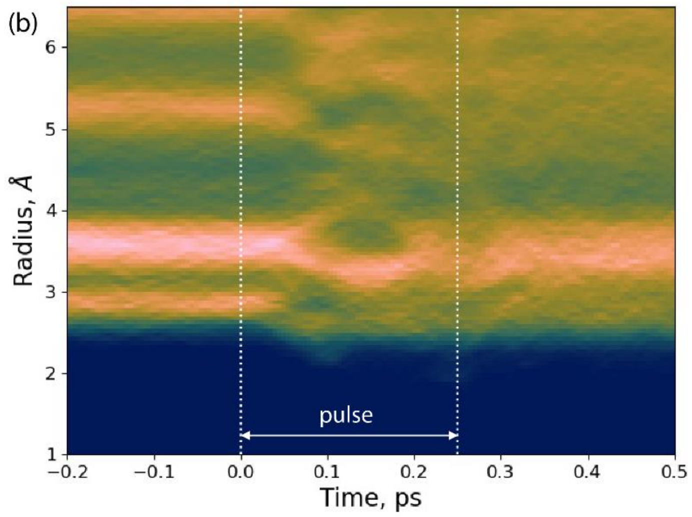


To probe the dynamics we can generate short x-ray pulses using an emittance spoiler

- x-rays always move faster than beam
- ~1.5 eV full bandwidth (without sxrss)





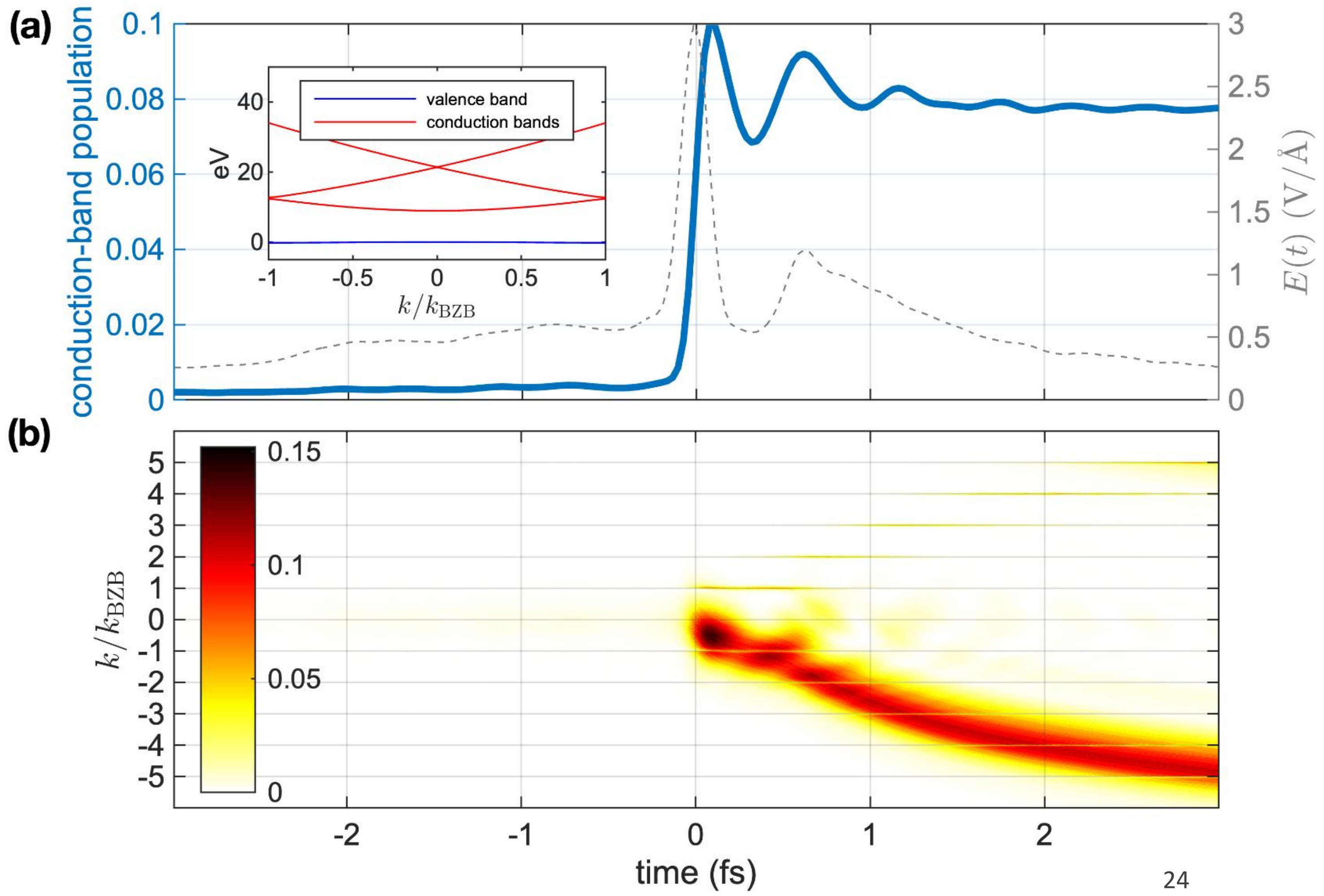


Electronic dynamics in a model dielectric

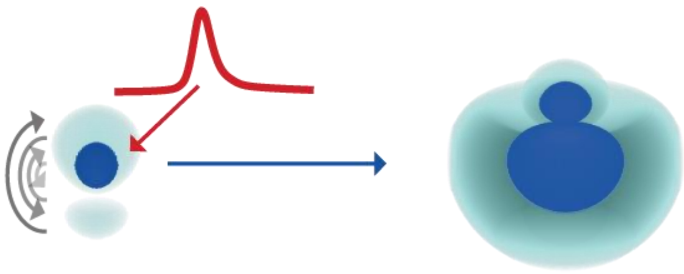
1D material with 9eV band gap

- Current spike causes sudden population transfer to conduction band

- Subsequent field rapidly accelerates conduction band electrons across multiple BZB

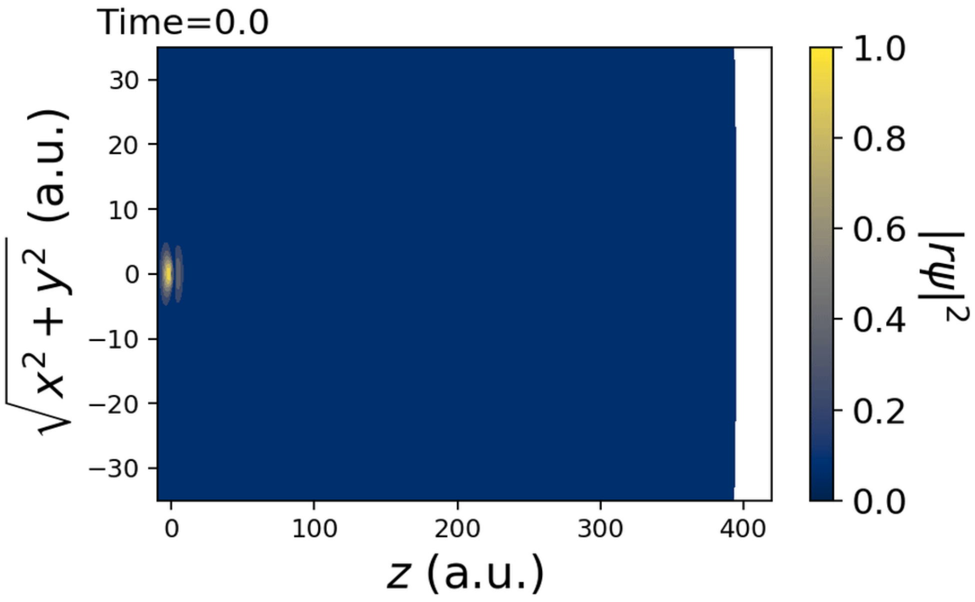


Impulsive ionization



Short, high-contrast, current spikes can kick electron in the sudden approximation:

$$|\psi(k)\rangle \rightarrow |\psi(k + A_0)\rangle \text{ (length gauge)}$$
$$\text{Provided: } \tau \ll 2\pi/\mathrm{IP}$$

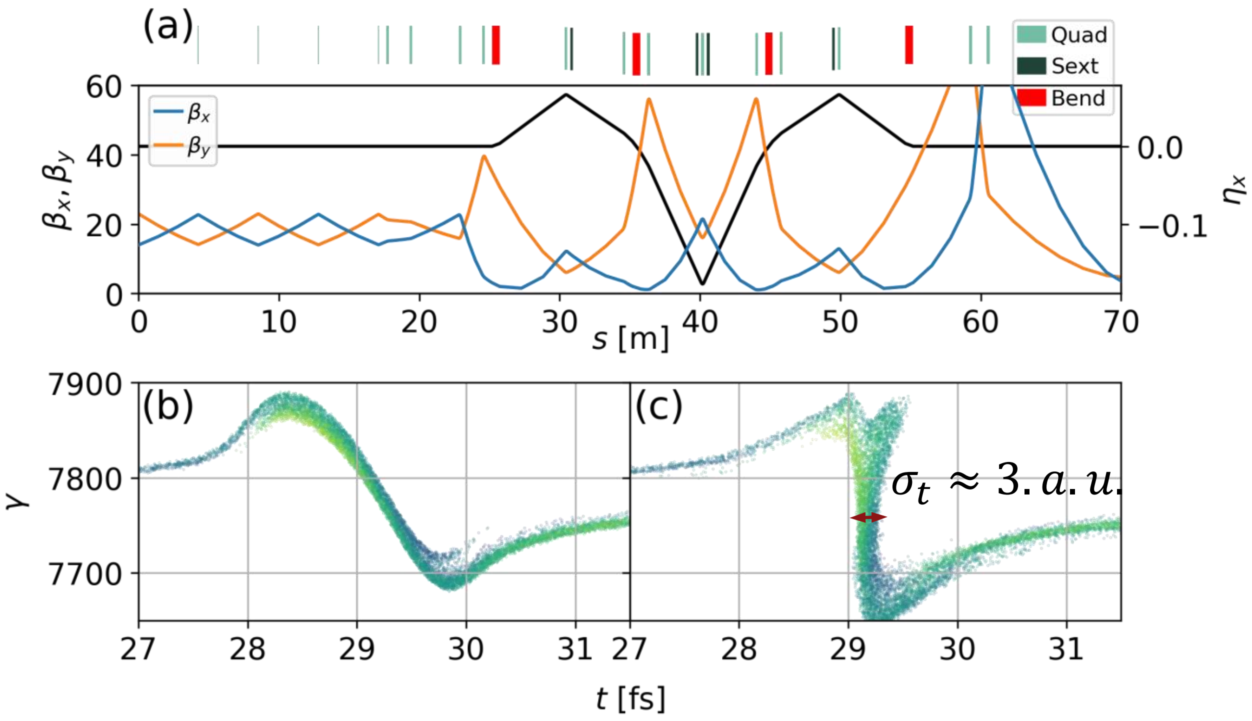


Simulation:

TDSE with classical field in code qprop
Velocity gauge

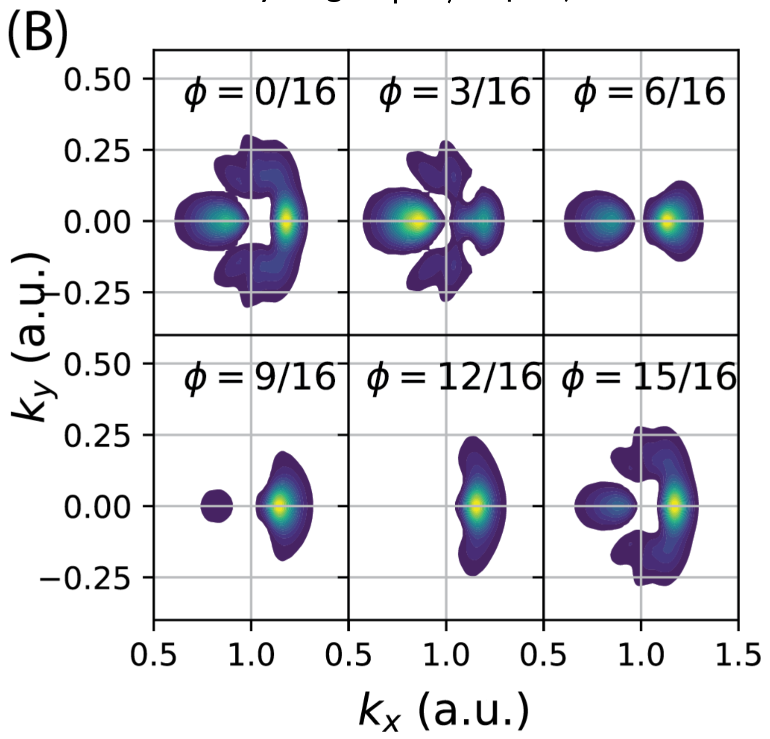
Nearly impulsive ionization

A chicane with anomalous dispersion can compress the space-charge chirp of the electron beam into a high contrast, attosecond, current spike.



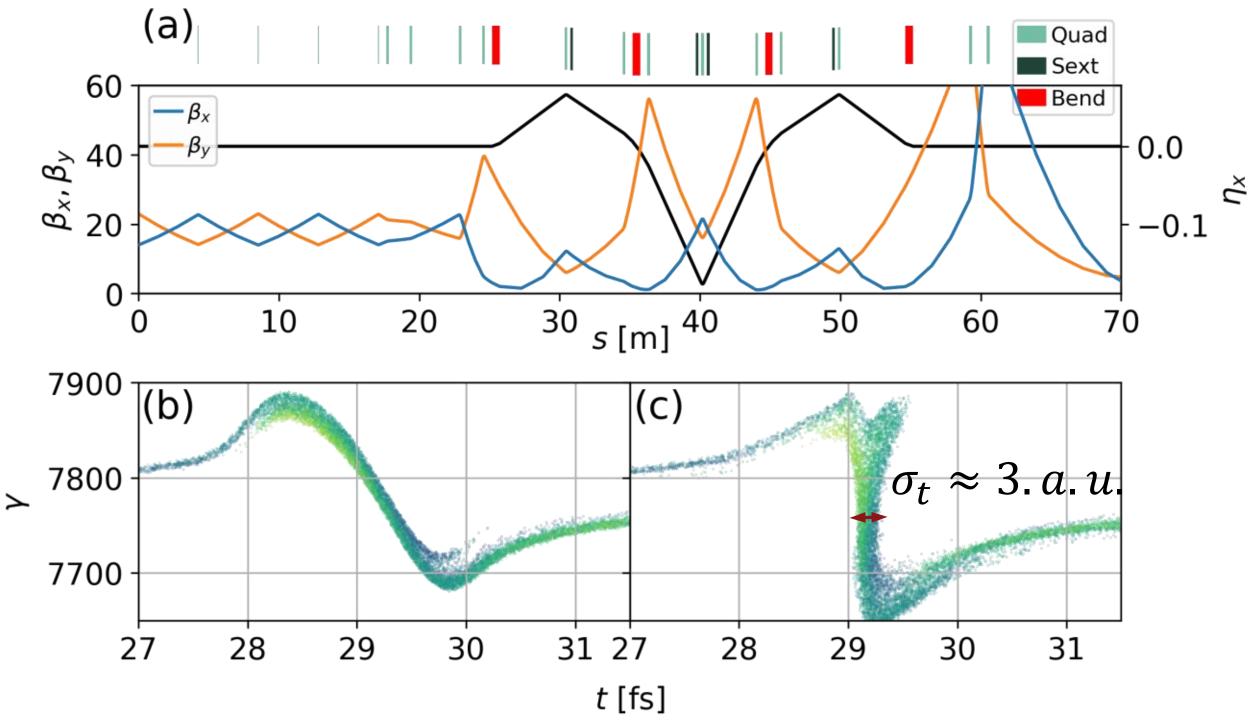
As a consequence, the free electron is nearly a copy of the bound state momentum wave-packet.

Initial condition:
Hydrogen $|2P\rangle + |3D\rangle e^{i\phi}$



Towards ultrafast control

A chicane with anomalous dispersion can compress the space-charge chirp of the electron beam into a high contrast, attosecond, current spike.



The pedestal of the pulse introduces dynamics (i.e. stark splitting) before ionization

Initial condition:
Hydrogen $|2P\rangle$