

What we can learn about super-intense interactions from intermediate intensities

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**National Research
Council**

University of Ottawa



50 Years ago: *L. V. Keldysh, Sov. Phys. JETP 20, 1307 (1965).*

'Ionization in the field of a strong electromagnetic wave''

$$\gamma^2 = IP / 2U_p$$

where

$$U_p = q^2 E_0^2 / (4m\omega^2)$$

if $\gamma < 1$, multiphoton ionization can be approximated as tunnelling.

U_p energy of the classical motion of an electron in IR field

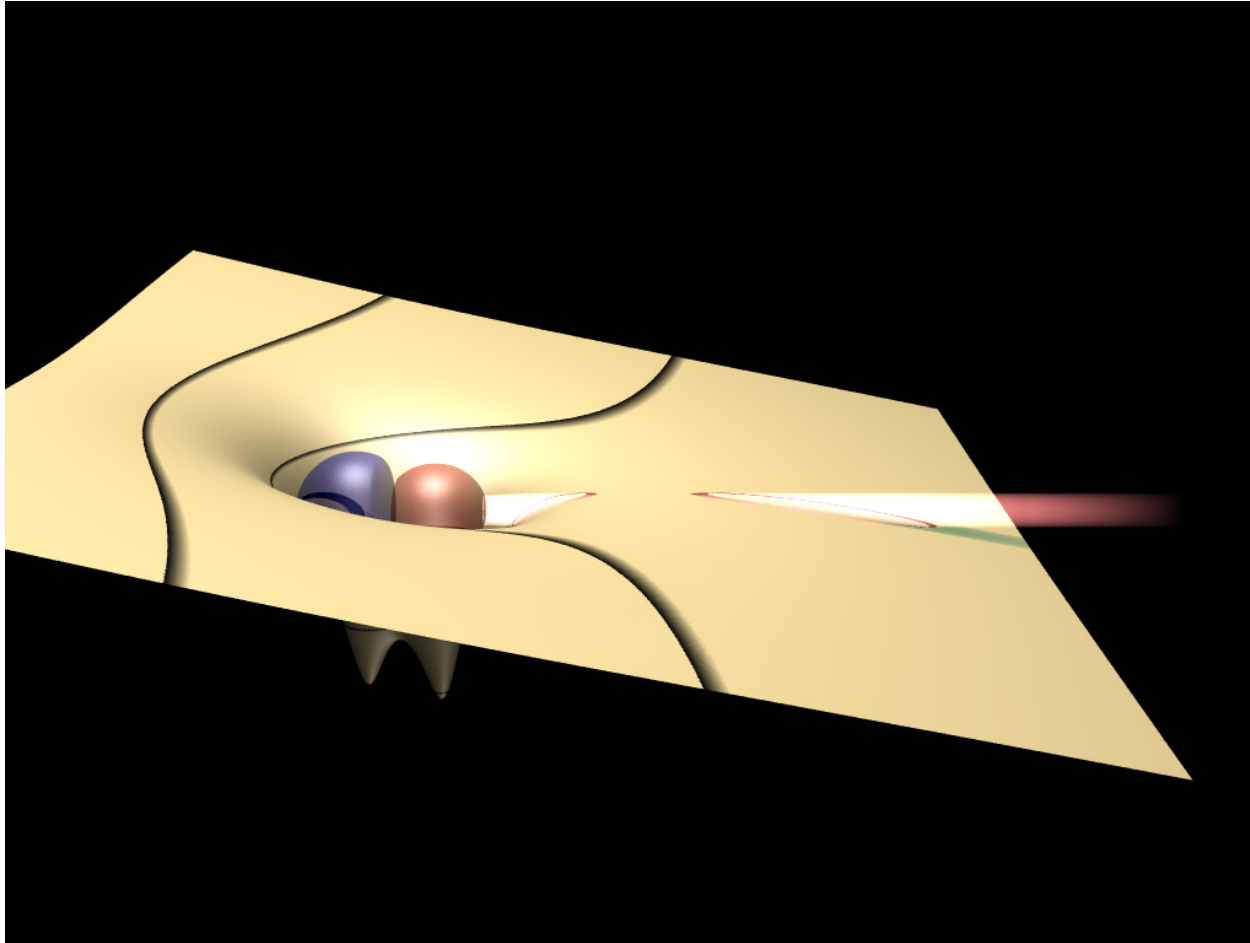
$U_p = 6$ eV for $\lambda = 800$ nm; $I = 10^{14}$ W/cm²

$U_p = 2.3$ eV for $\lambda = 3.5$ μ m; $I = 2 \times 10^{12}$ W/cm²

**Citations:
English 2,800
Russian 1,250**



Keldysh's was an astonishing and important statement



A beam splitter!

A static theory describes a sub-cycle phenomenon!

Keldysh treated atoms and solids together.

Electron ionization

$$F = ma$$

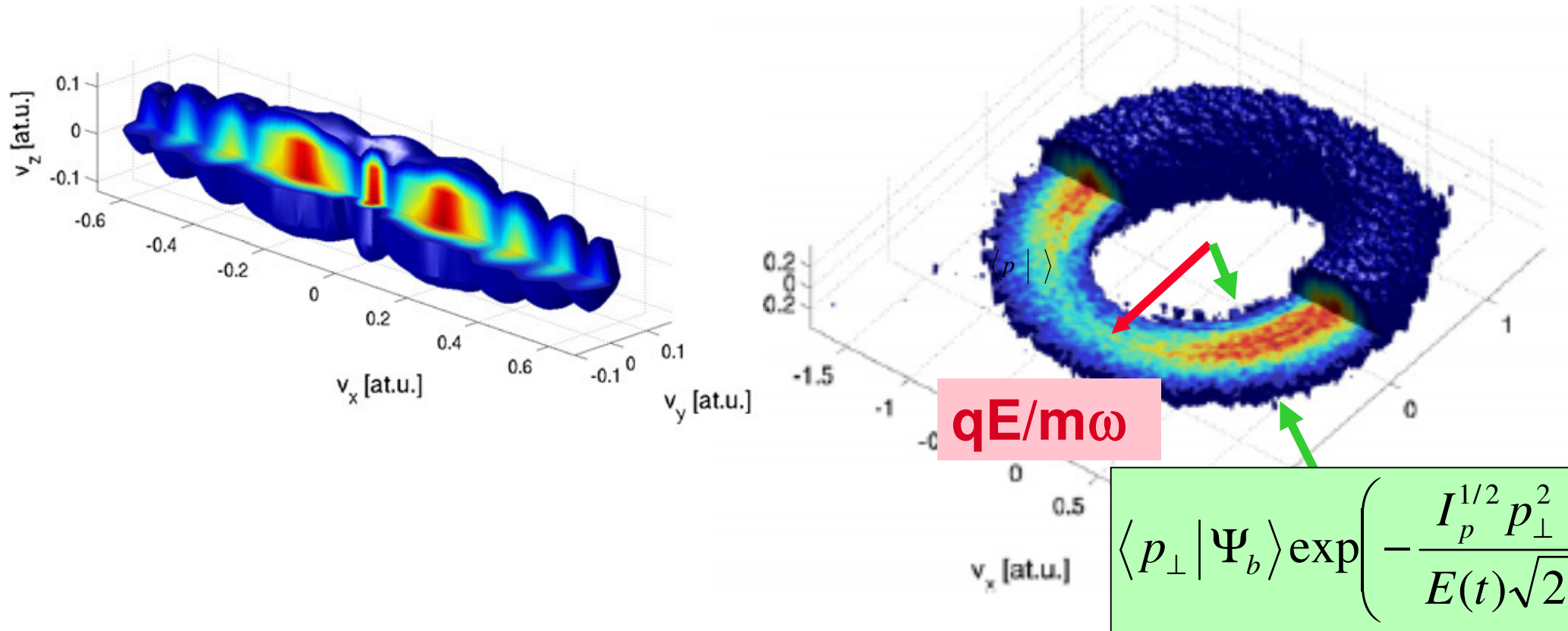
$$E(t) = E_0 \cos(\omega t) + \epsilon E_0 \sin(\omega t)$$

$v(t') = 0$ where t' is the time of tunnelling

**For 800nm
light and
 $I \sim 10^{15}$ W/cm²,
 $U_p \sim 60$ eV**



Electron tomography measurement of the electron momentum distribution -- ATI



$$\langle KE_{\text{lin}} \rangle \sim 0.1 U_{p, \text{lin}}$$

$$\langle KE_{\text{cir}} \rangle \sim U_{p, \text{circ}} \sim 2U_{p, \text{lin}}$$

Outline:

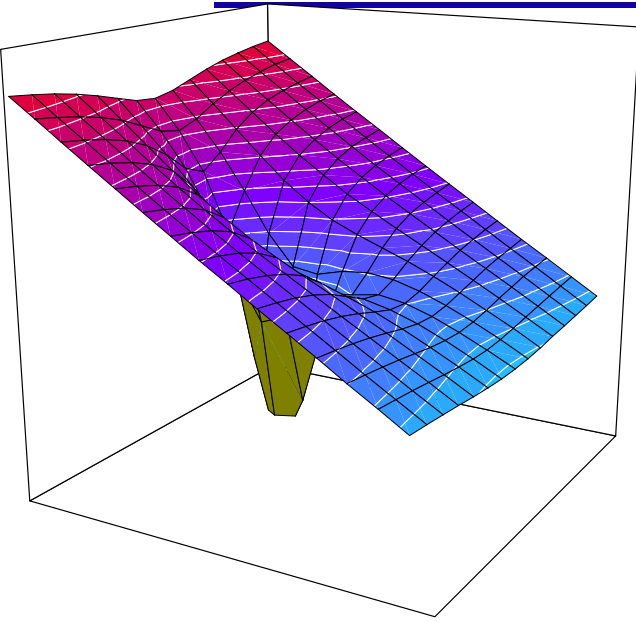
-- Measuring the lateral momentum.

“What happens to the photon’s momentum during ATI?”

-- Unconventional time-resolved measurement that can be extended to super-high intensities.



The non-classical behaviour: What determines the perpendicular momentum?



$$\omega(t) = 4\omega_0 \left[\frac{E_i}{E_h} \right]^{5/2} \frac{E_a}{E(t)} \exp \left[-\frac{2}{3} \left[\frac{E_i}{E_h} \right]^{3/2} \frac{E_a}{E(t)} \right]$$

$$E_{i,\text{eff}} = E_i + p_{\perp}^2/2m$$

Substituting $E_{i,\text{eff}}$ for E_i in the tunnelling formula

Any component of a wave function where p is not parallel to E is “wasted”. An electron with $p_{\perp} \neq 0$ experiences a larger effective barrier.

Initial conditions following tunneling

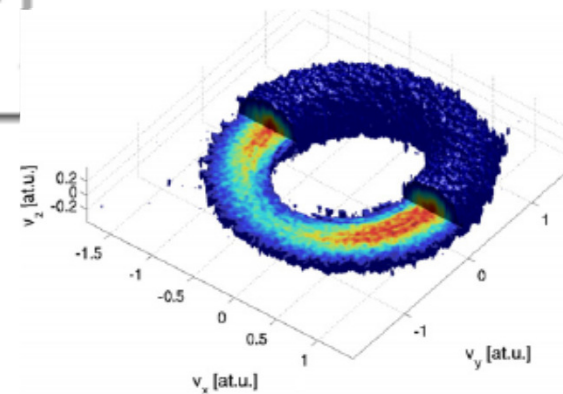
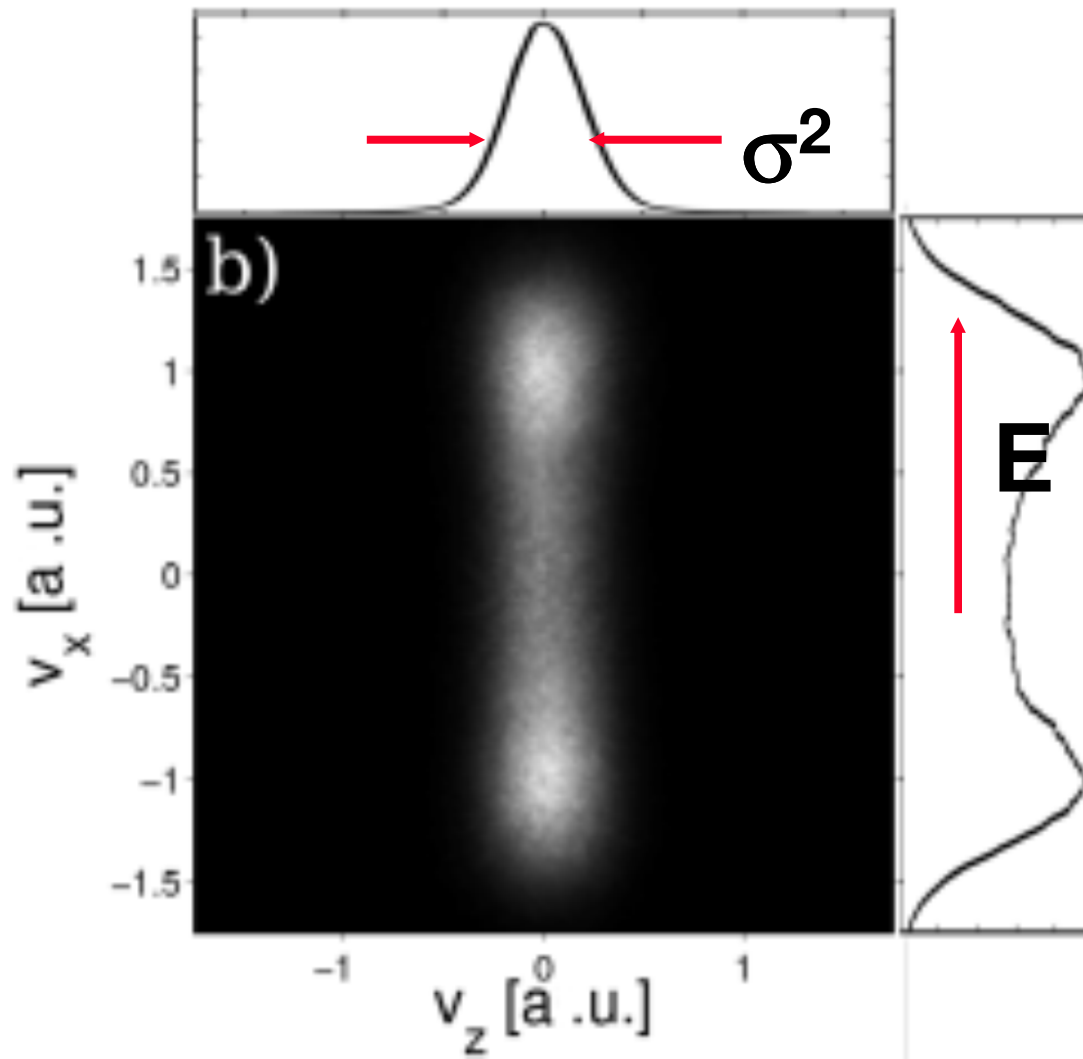
$$\omega(t) = [A] \exp \left\{ -\frac{2}{3E(t)} 2\sqrt{2} E_i^{3/2} \right\}$$

$$\omega(t, p_{\perp}) = [A] \exp \left\{ -\frac{2}{3E(t)} 2\sqrt{2} \left[E_i^{3/2} + \frac{3}{2} E_i^{1/2} dE_i \right] \right\}$$

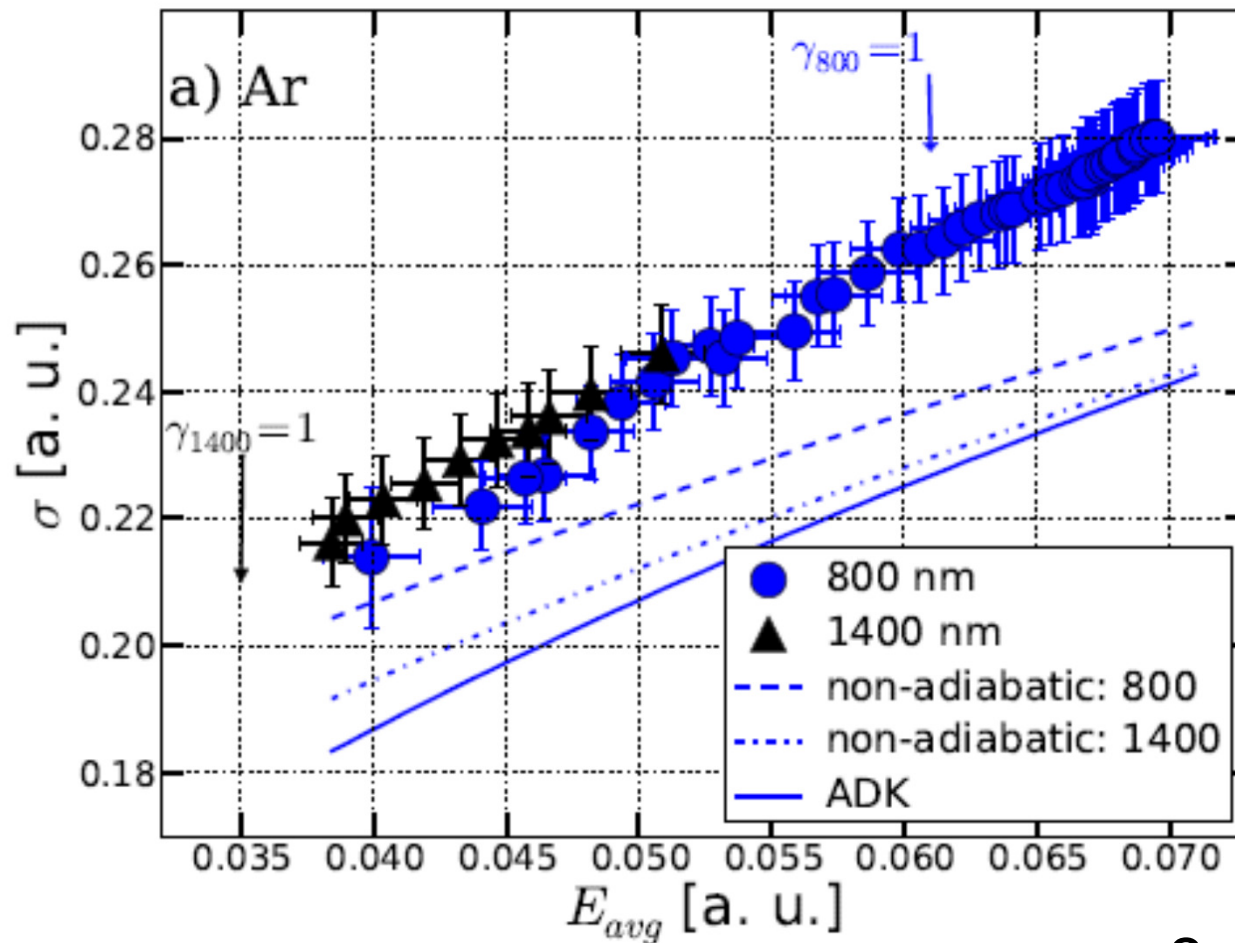
$$\omega(t, p_{\perp}) = \omega(t, 0) \exp \left\{ -\frac{\sqrt{2} p_{\perp}^2 E_i^{1/2}}{E(t)} \right\}$$

As E_i increases V_{\perp} also increases because $E(t)$ increases more quickly than $E_i^{1/2}$.

Confirming the lateral momentum.



The Gaussian width as a function of field -- Ar

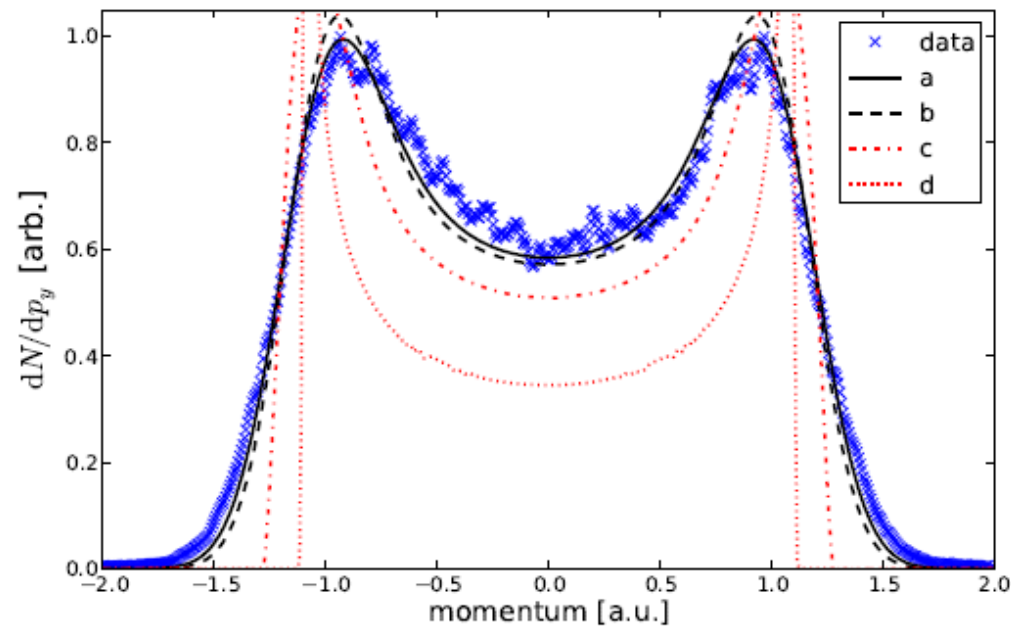


$$\sigma^2 = E/\sqrt{2Ip}.$$

Arissian et al, *PRL* **105**, 133002 (2010)

Precise *in-situ* measurement of laser pulse intensity using strong field ionization

C. Smeenk,^{1,*} J. Z. Salvail,¹ L. Arissian,^{1,2,3} P. B. Corkum,¹ C. T. Hebeisen,¹ and A. Staudte¹



$$V = \frac{qE_0}{m\omega} + \frac{\sigma}{m}$$

What about the photon's momentum? Can it be observed?

Energy balance

$$n\hbar\omega = I_p + K + U_p$$

I_p --- ionization potential

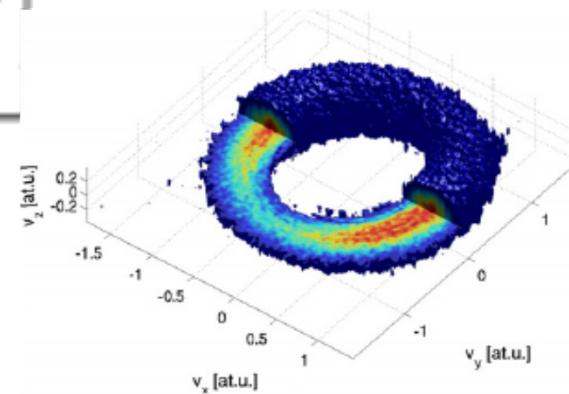
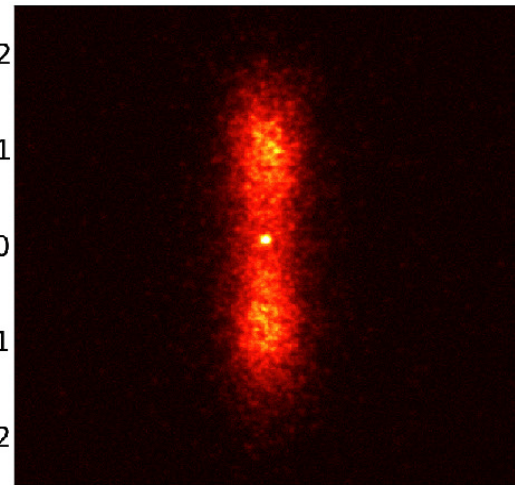
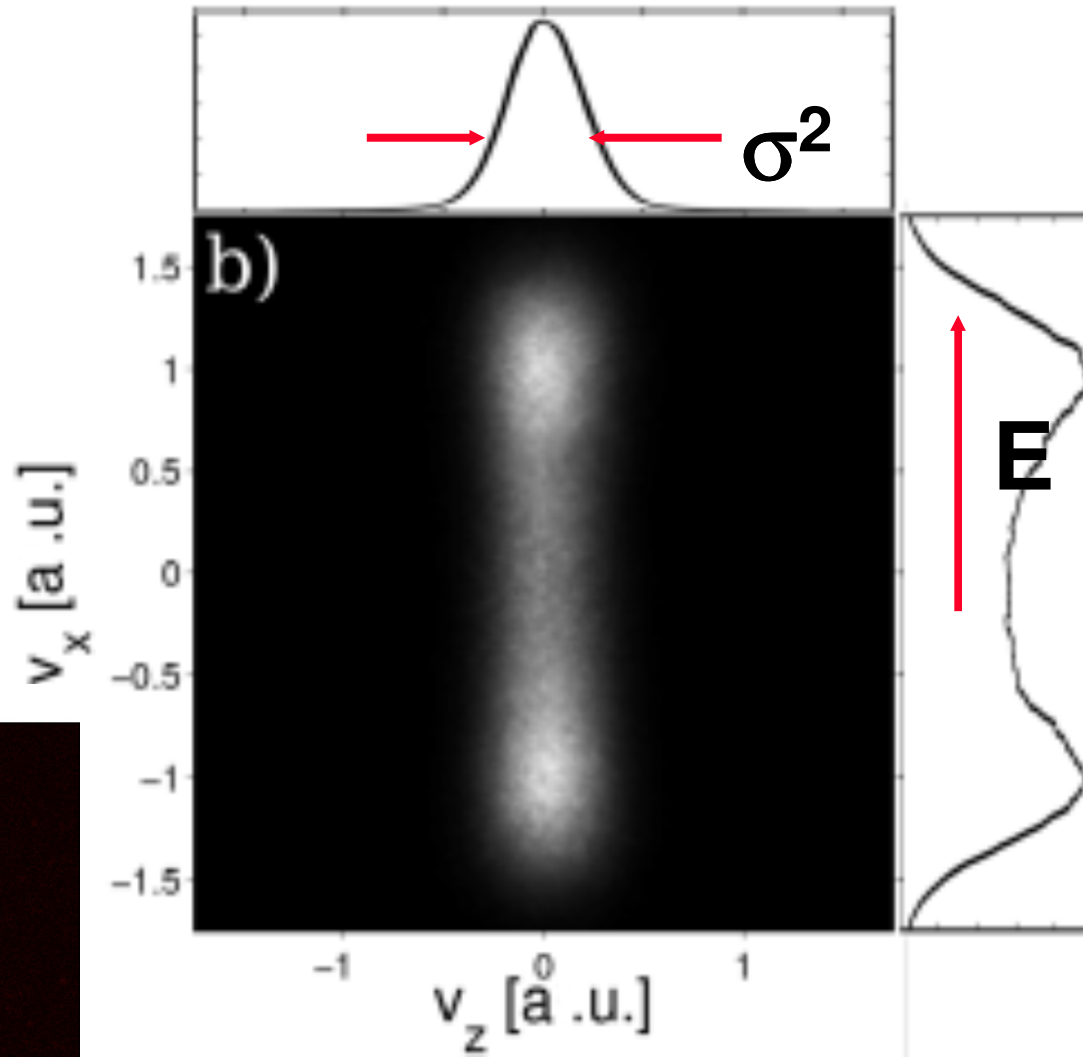
K --- drift (or ATI) energy

U_p --- oscillation or ponderomotive energy

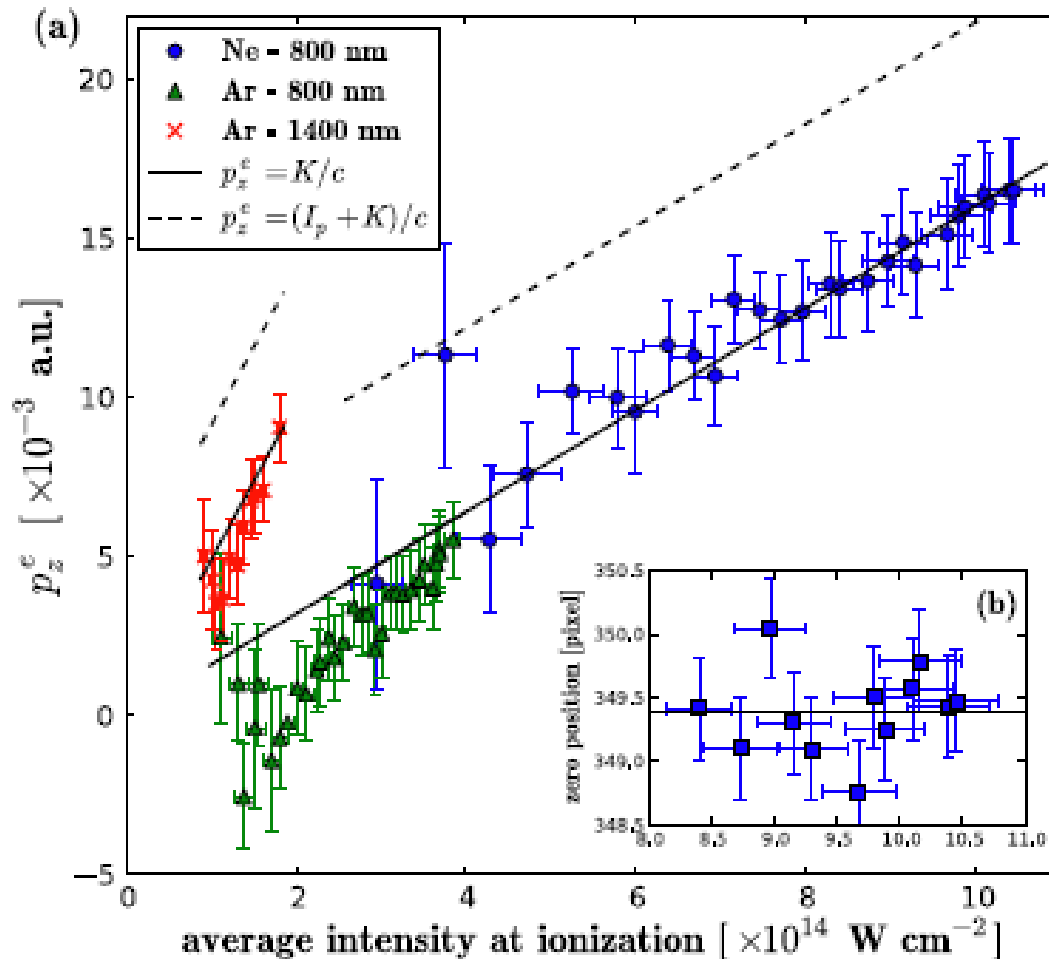
How is the photon's momentum shared?



Confirming the lateral momentum.



We measure the momentum given to the electron by the absorbed photons



$$n\hbar\omega = I_p + K + U_p$$

- U_p/c is given back to photons through their shifted frequency.
- I_p/c is given to the ion
- K/c is given to the electrons (ie, the plasma).

During filamentation, the absorbed photon momentum produces a small current – larger for circular than linear.

But plasma waves created during ionization are more important.

Comment 1:

Andre Bandrauk and Stephane Chelkowski have shown that:

- The ion only gets 0.7 IP/c during MPI.
- Momentum sharing is very different for single photon ionization. The electron gains more than $8\hbar\omega/5c$.
- *Clearly there is an unexplored transition between the two.*



Comment 2:

- Electron, ion re-interaction is key.
- We chose circular polarized light so the electron would not interact with the ion after ionization.
- The ion will gain much more for linear polarization.
- There is an unexplored transition between linear and circular.
- For linear polarization, sharing will depend on intensity

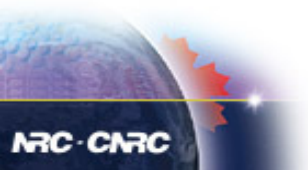


Comment 3:

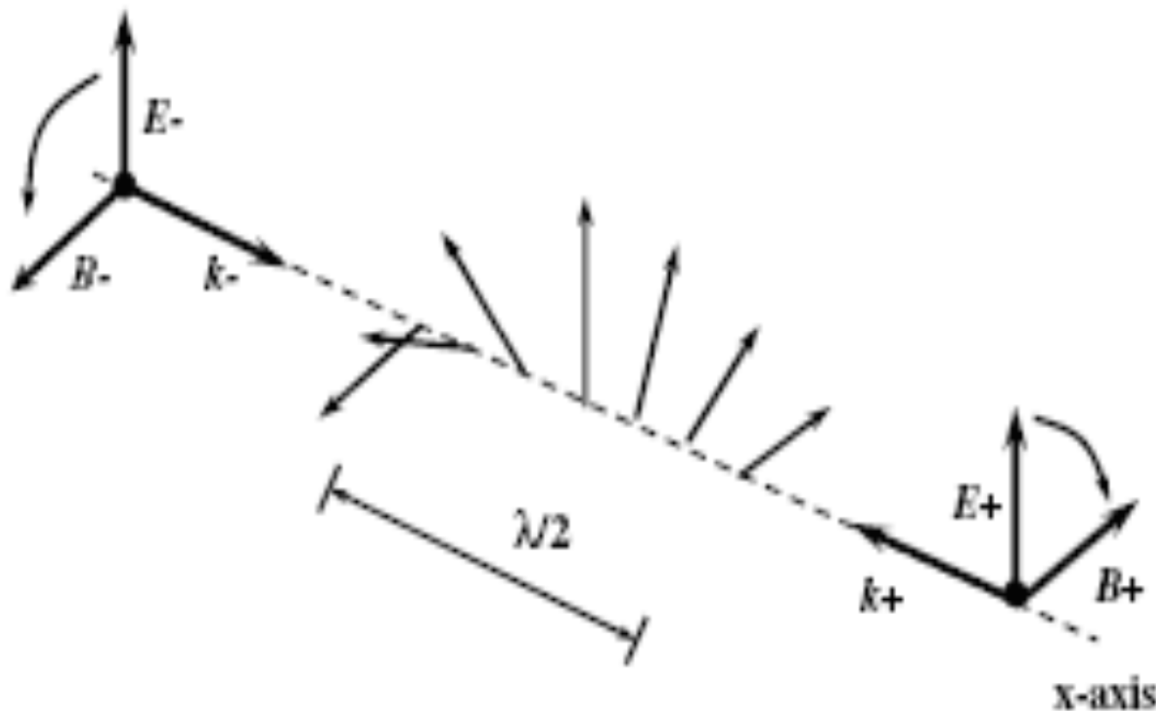
The electron's lateral momentum will become large for high IP ions.

Re-collision will disappear – the electron will miss the ion.

However the influence of the photon momentum can be canceled allowing super-intense re-collision.



The momentum shift can be overcome with counter propagating pulses of the same handedness



The strong magnetic field restricts quantum diffusion

100 MeV recollisions seems possible! But is it interesting? High energy electron collisions have been studied for decades.

Outline:

-- Measuring the lateral momentum.

“What happens to the photon’s momentum during ATI?”

-- Unconventional time-resolved measurement that can be extended to super-high intensities.

For conventional optics, we require nonlinearities for all time resolved measurements.

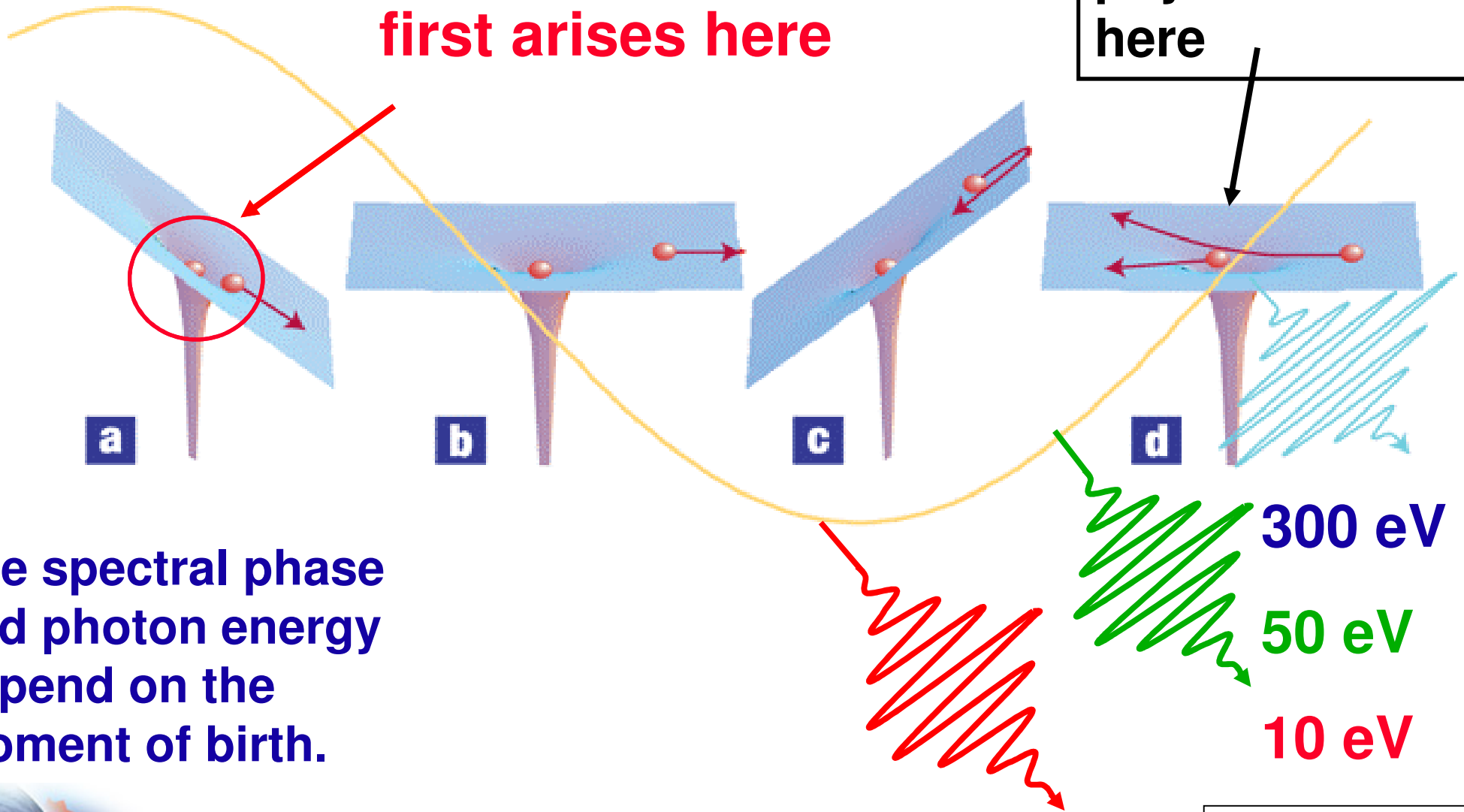
Semi-classical physics and re-collision is a “new” nonlinear appropriate for high fields.



Reminder of atto-physics

**Attoseconds
first arises here**

**Mapped by
classical
physics to
here**



**The spectral phase
and photon energy
depend on the
moment of birth.**

PRL 71, 1994
(1993)



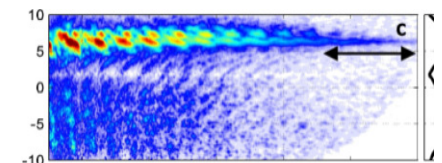
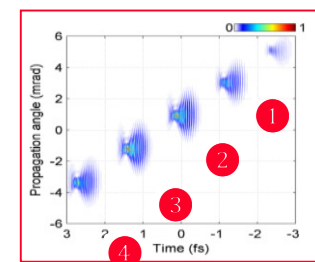
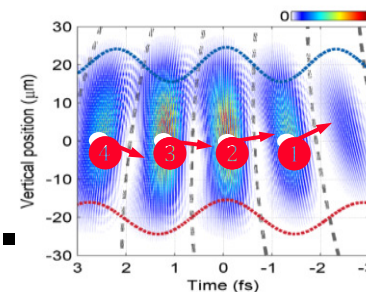
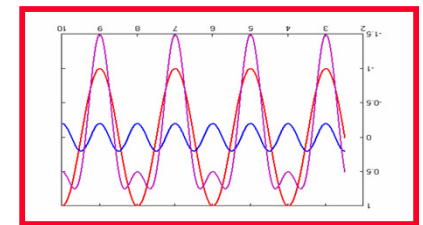
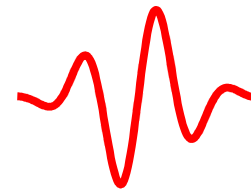
A brief aside:

• *All highly nonlinear interactions allow **sub-cycle response**. Tunneling and re-collision are just the first to be applied.*

*If sub-cycle response is possible, **sub-wavelength** should be also-- a new approach to nano-technology and imaging.*

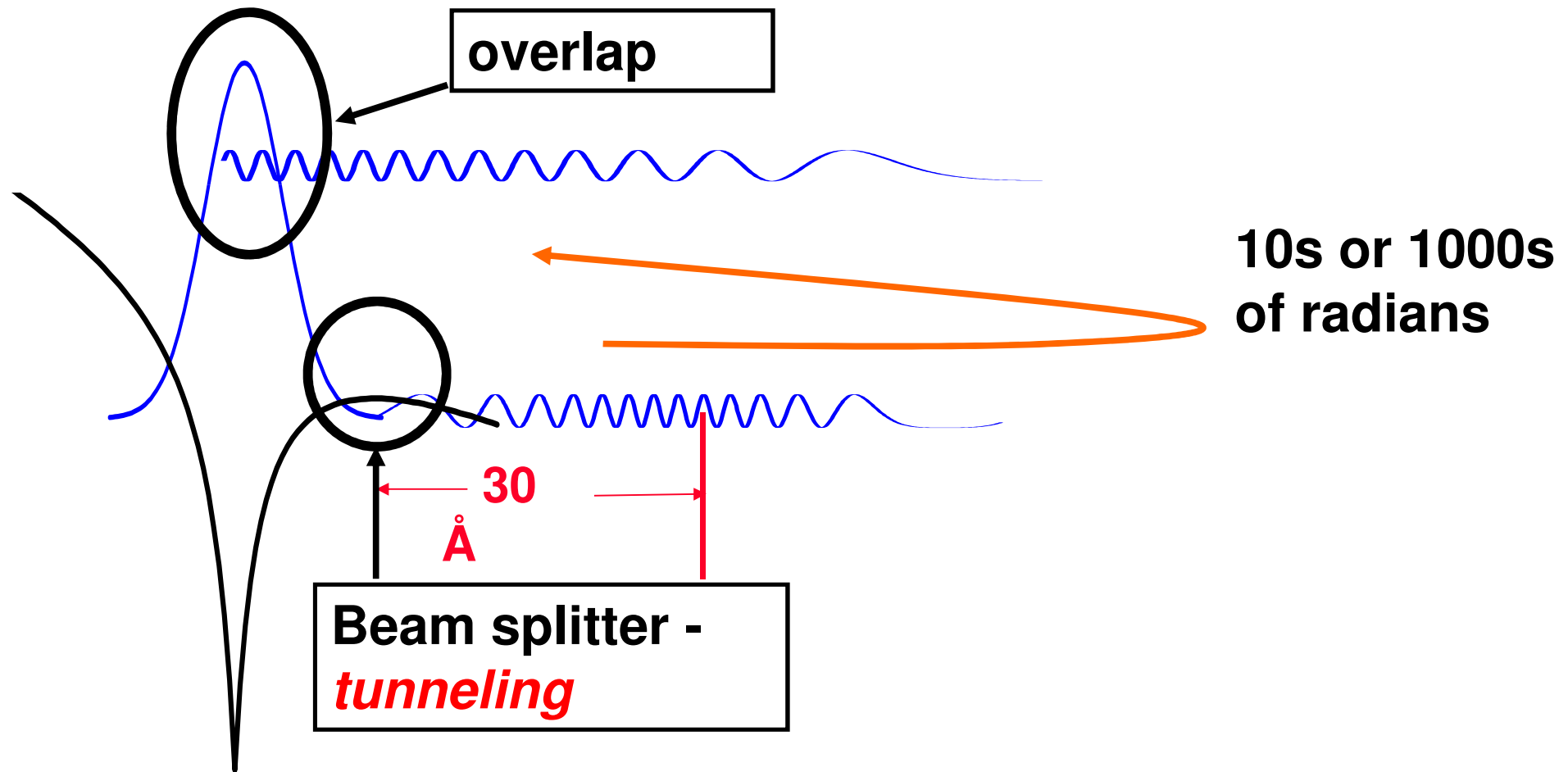
“Gating” methods to produce single pulses — all require CEP control

- Use a near single cycle pulse.
- Use a pulse with time-dependent polarization.
- Augment these using second harmonic.
- Use a space-time coupled pulse.
- Rapid ionization destroys phase matching.

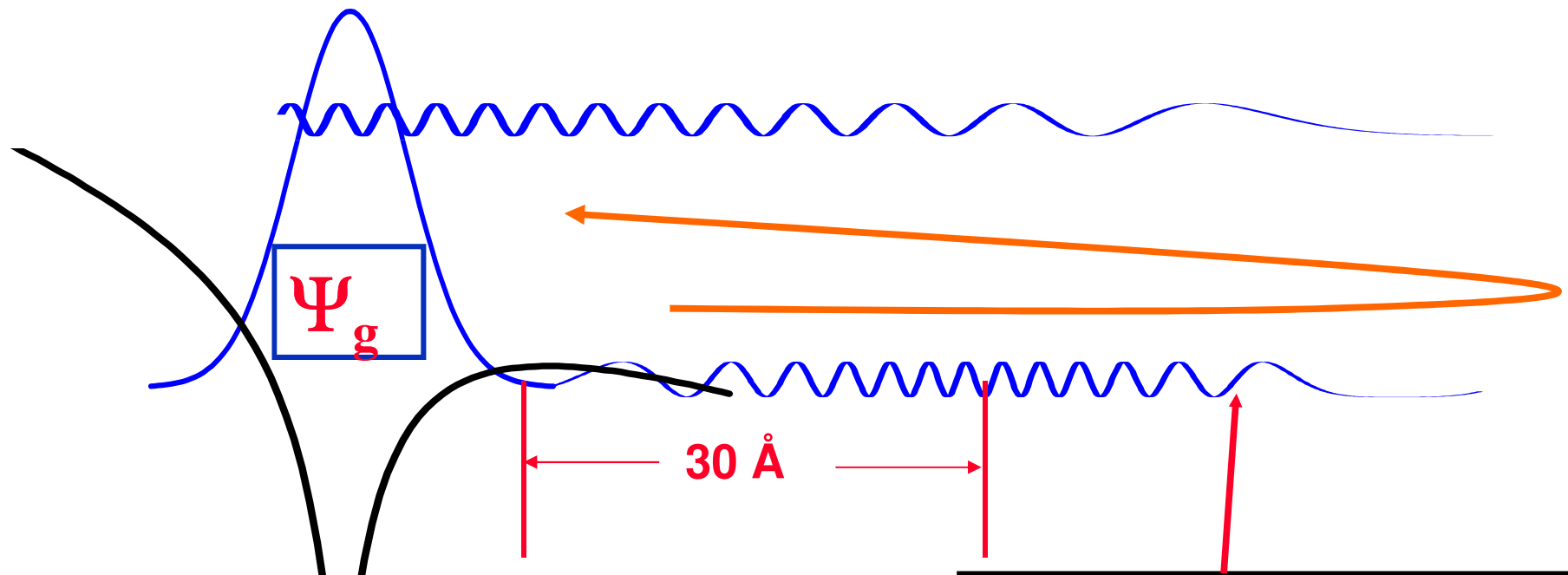


All of these methods work, but 1, 2 and 3 are most developed – they only rely on the single atom response.

A quantum perspective (SFA) -- A sheared nonlinear interferometer



Ways to Control the interferometer



The added degree of freedom – **a second dimension** – allows measurement of **orbitals** and of the **attosecond pulse**

Move the arm

- Length → add a 2nd color
- Angle → a 2nd color beam at an angle or with a different polarization ---.

High harmonics and attosecond pulses.

Attosecond pulses, repeated each $\frac{1}{2}$ period, create a series of femtosecond duration harmonics. They can extend to more than 1 keV if needed.

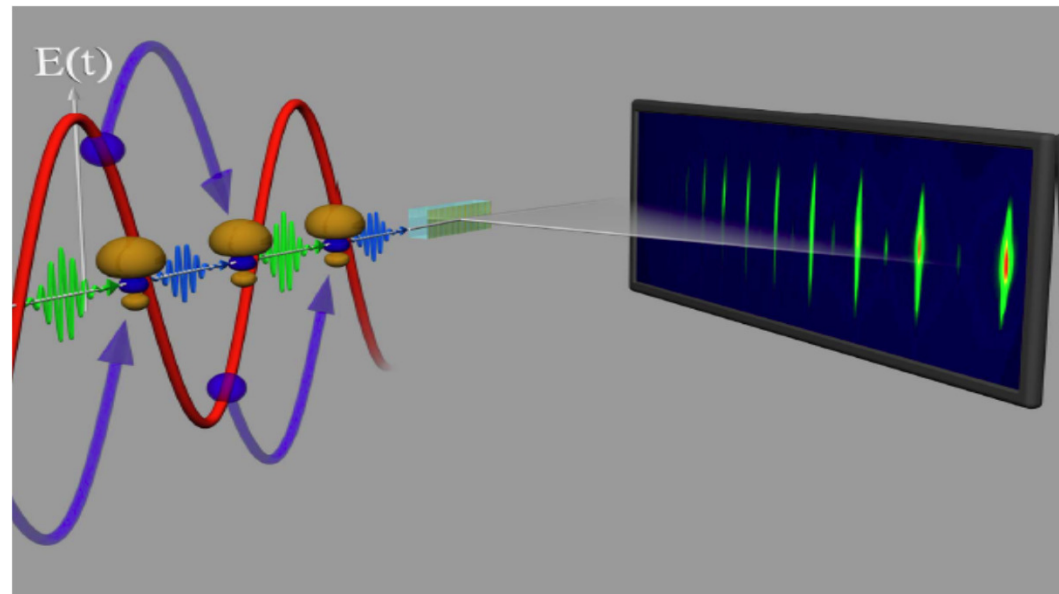
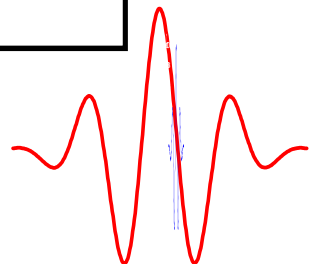
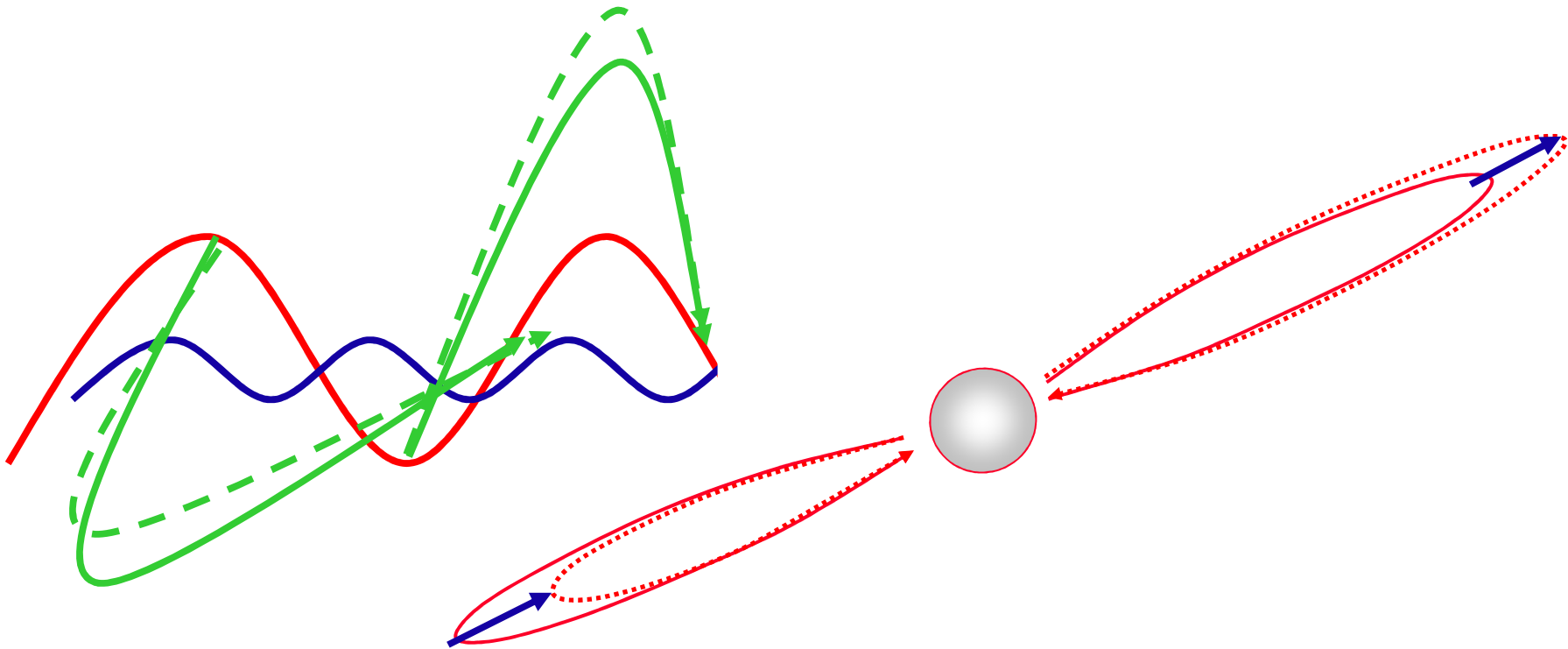


Illustration of HHS on a polar molecule.



For a long pulse -- odd harmonics

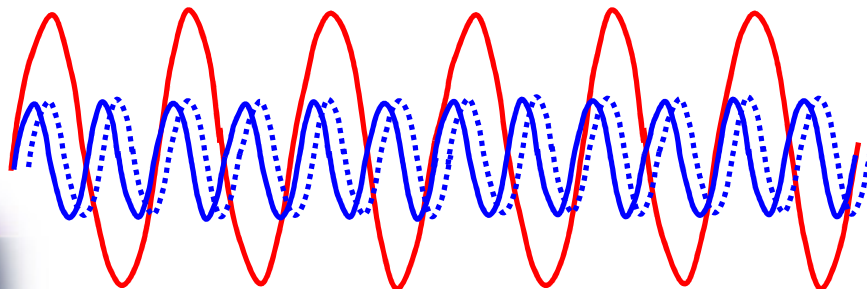
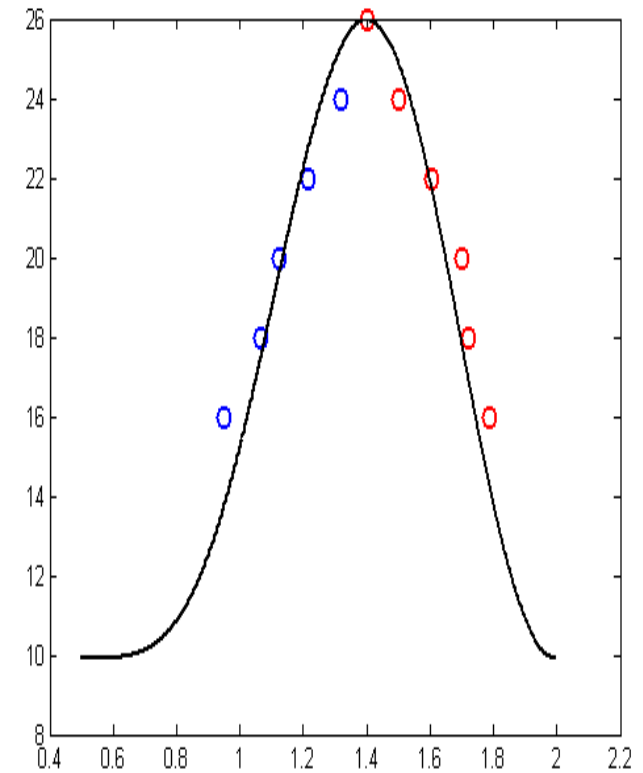
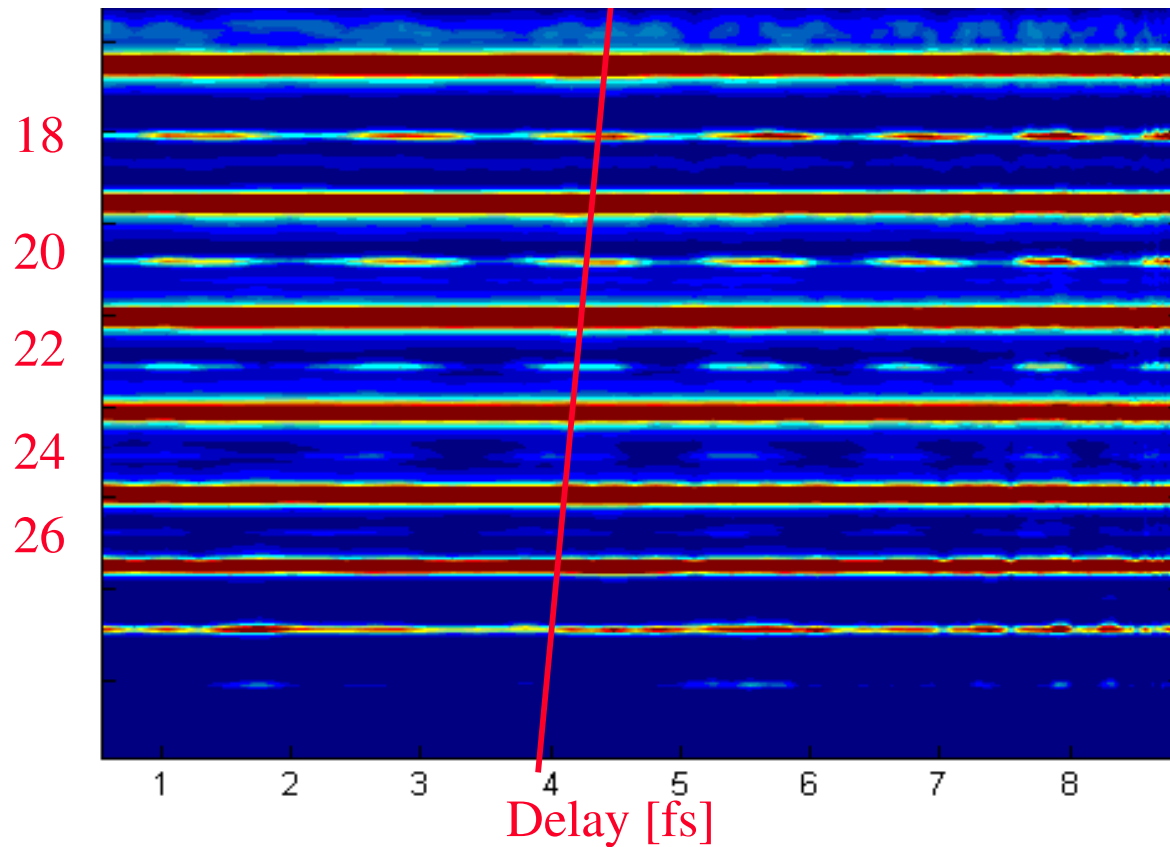


A (weak) 2ω field breaks symmetry, generating even harmonics

Each moment of birth (re-collision) has an optimum phase difference (θ) between ω and 2ω

What Phase difference moves the interferometer arms optimally?

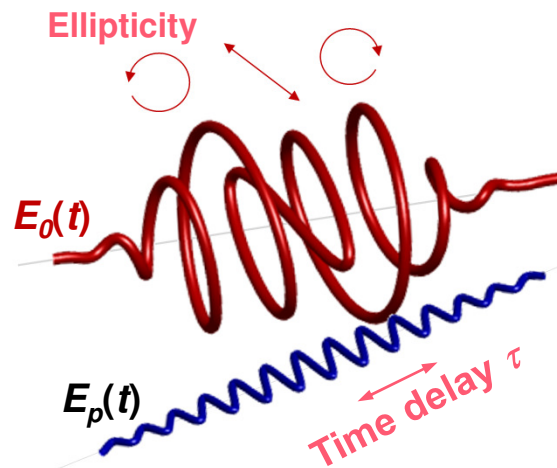
Harmonic order



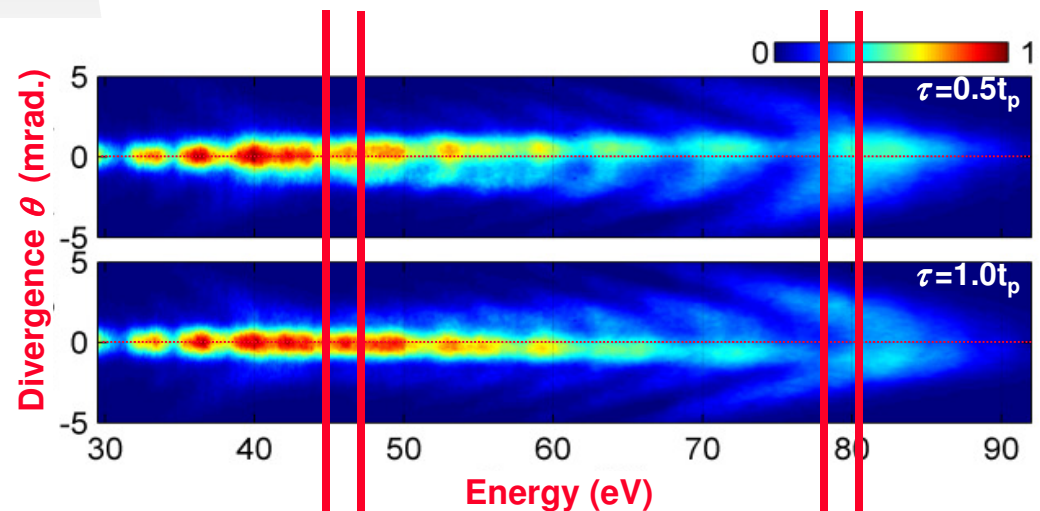
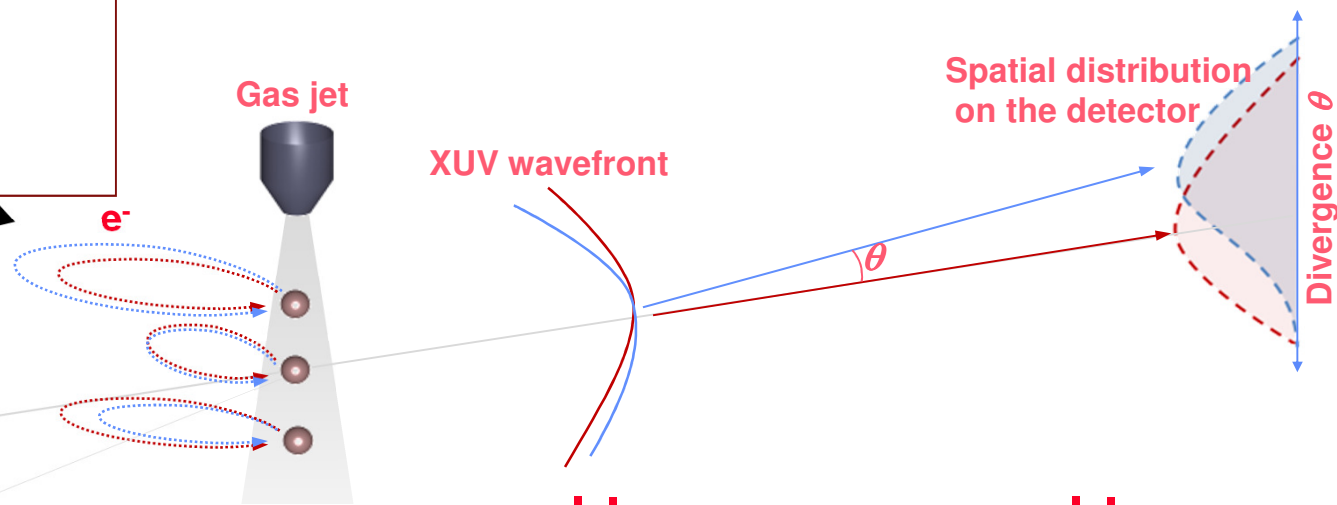
Dudovich et al
Nature Physics
2, 781 (2006).

All-optical, space-time measurement of attosecond pulses

The 2ω beam creates a delay dependent phase shear



Note: $I_{2\omega} \sim 10^{-3} I_{\omega}$

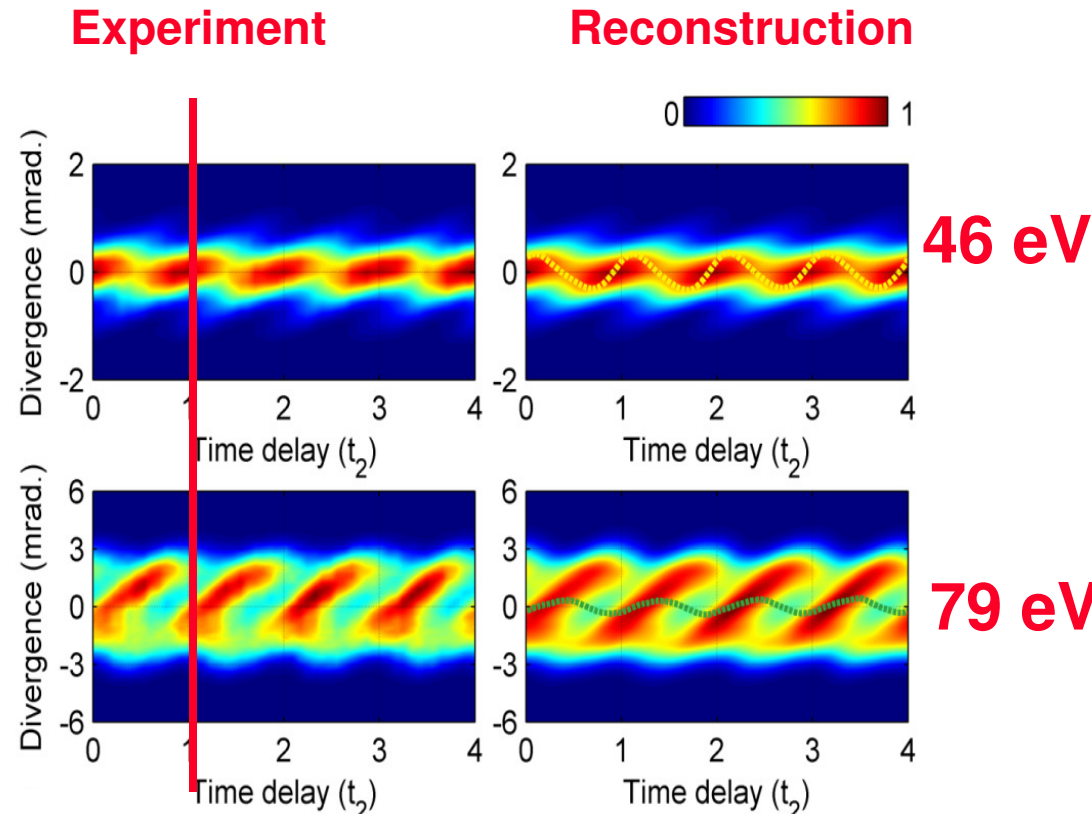


The 2ω beam creates a spatially dependent phase modulation – a phase gate

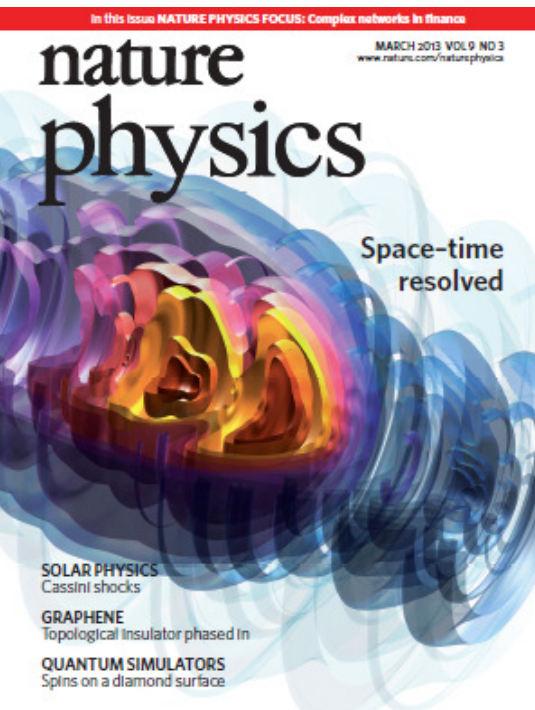
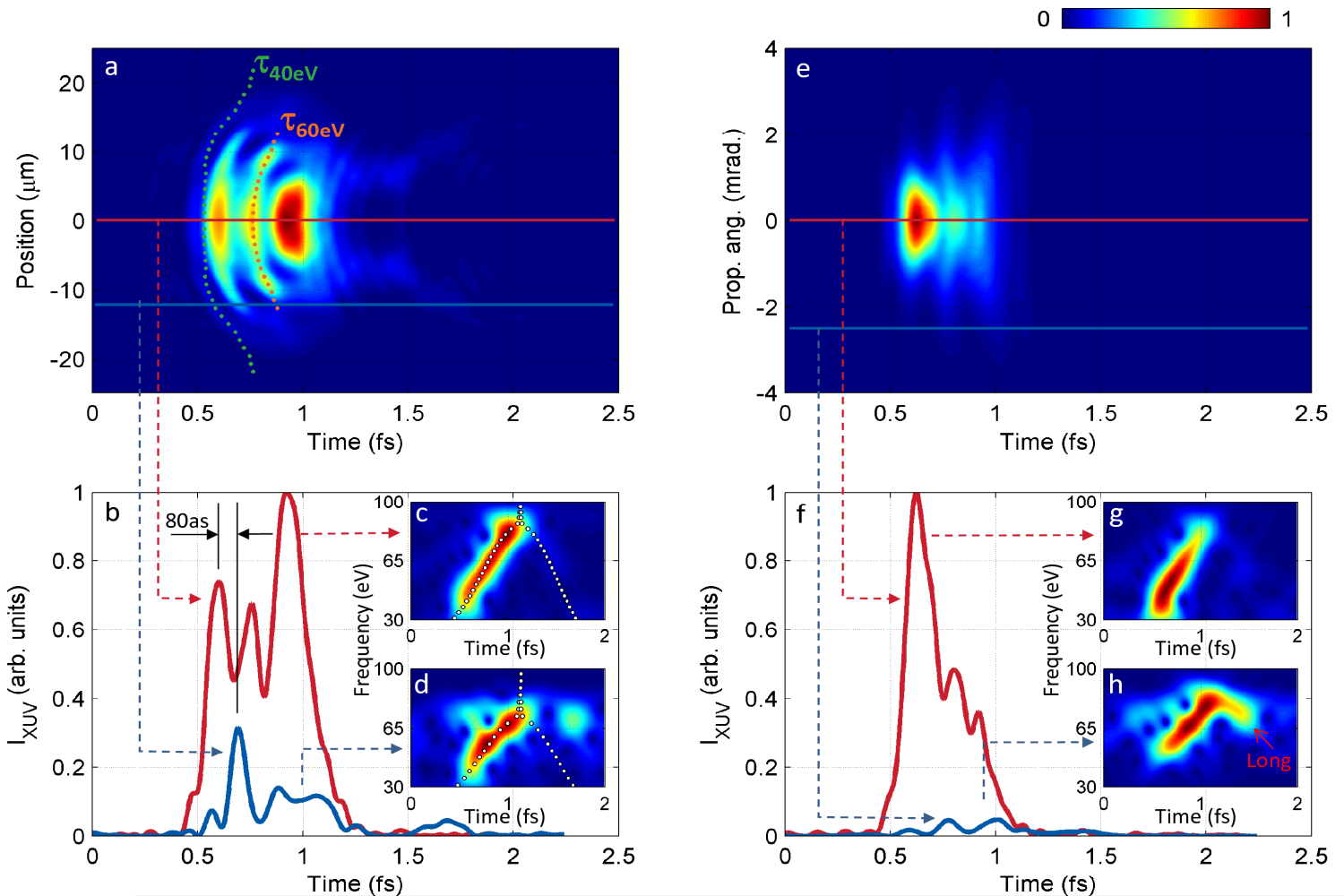
A FROG retrieval algorithm reconstructs the spatial structure of a pulse.

The phase dependence of the maximum deflection determines the spectral phase.

Frequency dependent far field pattern



An attosecond pulse emerging from the nonlinear medium (left) and in the far field (right).

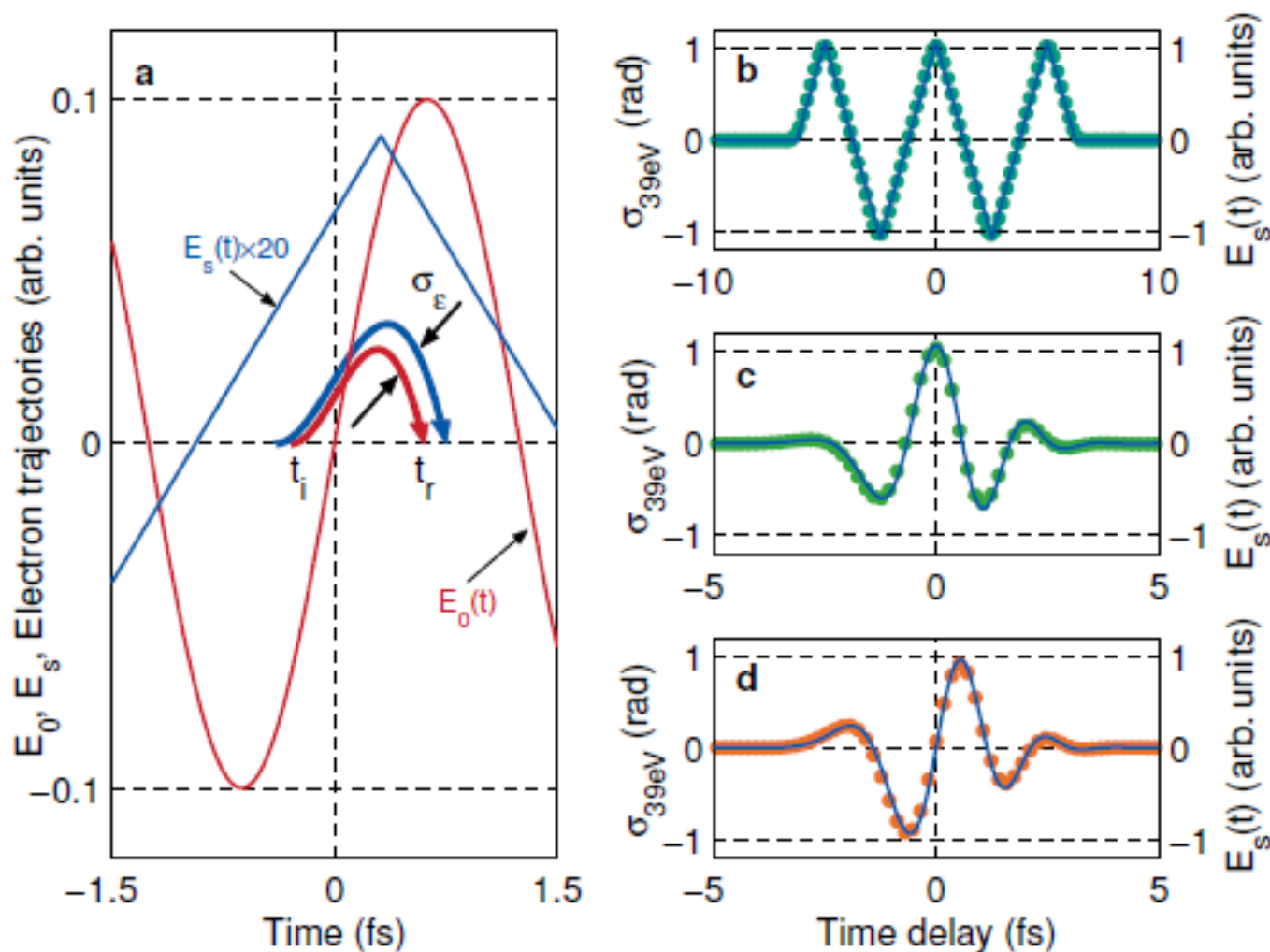


The first spatial-temporal reconstruction of an attosecond pulse

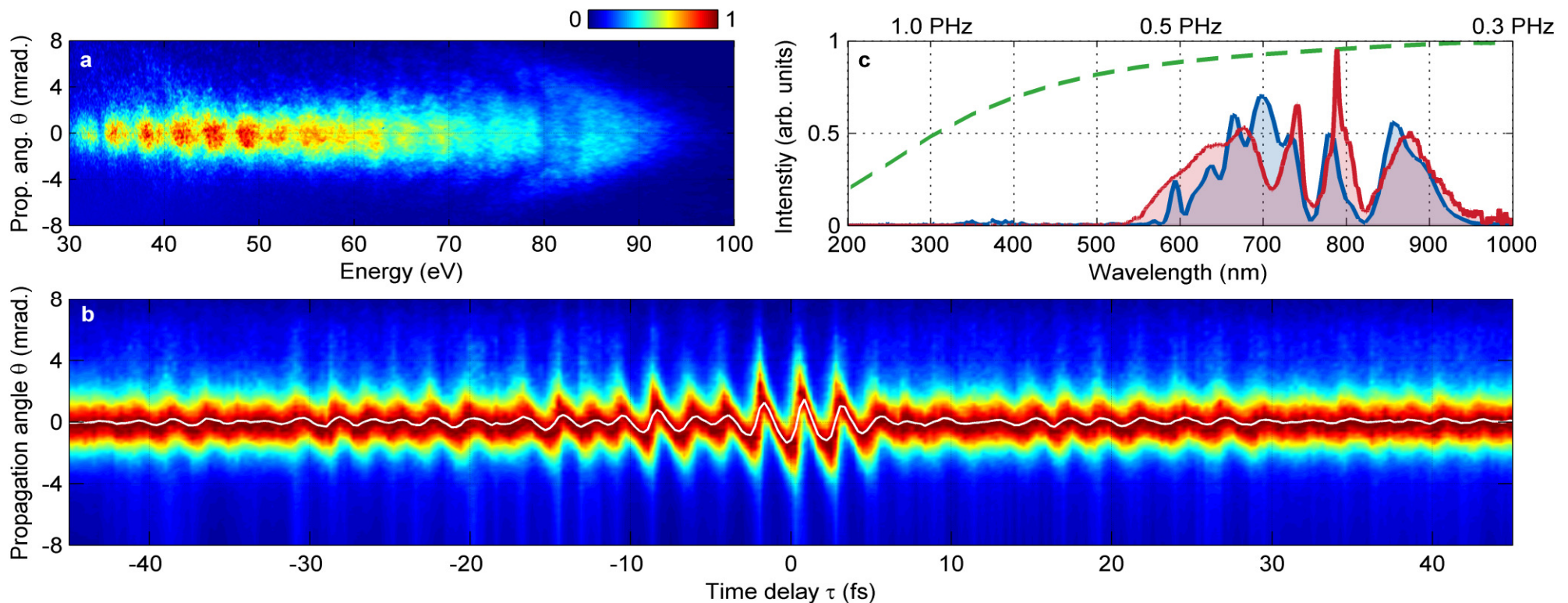
But it is not only the attosecond pulse that is measured. The time dependent field is measured at the same time.



The electron is only affected between ionization and re-collision – offering **sub-optical-cycle gate**.



The time-dependent electric field of an 800 nm deflecting pulse



A petahertz all-optical oscilloscope

Generality of insitu measurement

Weak fields *hardly modified strong field processes*, yet **they leave an indelible imprint**, allowing measurement.

We should apply insitu measurement to solids.

Was Keldysh correct? Can we reuniting solids and gases?



Note: Quéré has generalized this idea by modulating a plasma surface with a pre-pulse. In this way he images harmonic created at plasma surfaces.



Final comments:

Even at 10^{23} W/cm², laser electric fields are small on the particle or nuclear physics scale.

Our aim could be to introduce a systematic means to measure nuclear dynamics.



Returning to Keldysh:

During the last 50 years, strong field atomic and solid state physics diverged

Atomic physics developed HHG, attophysics, double ionization, etc.

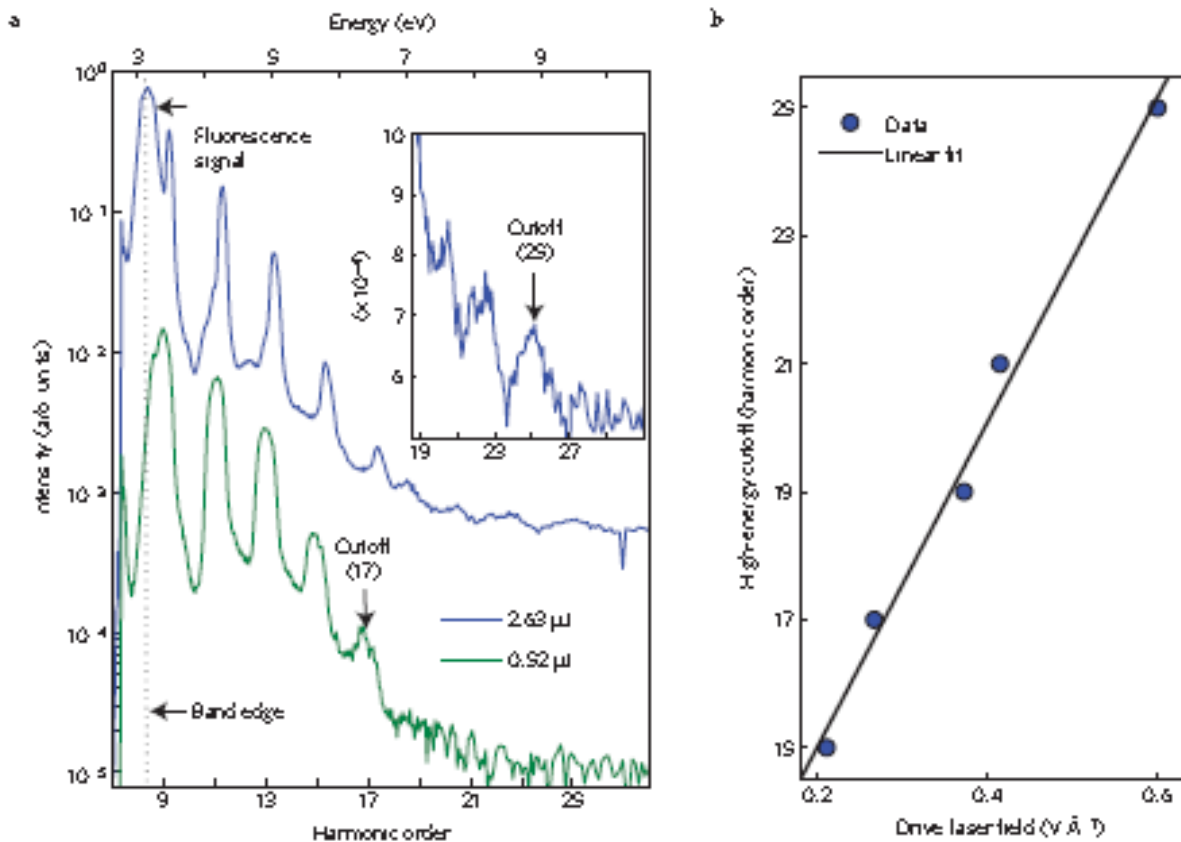
Solid state physics developed the technology of laser materials processing.

Will physics allow atoms and solids to reunite?

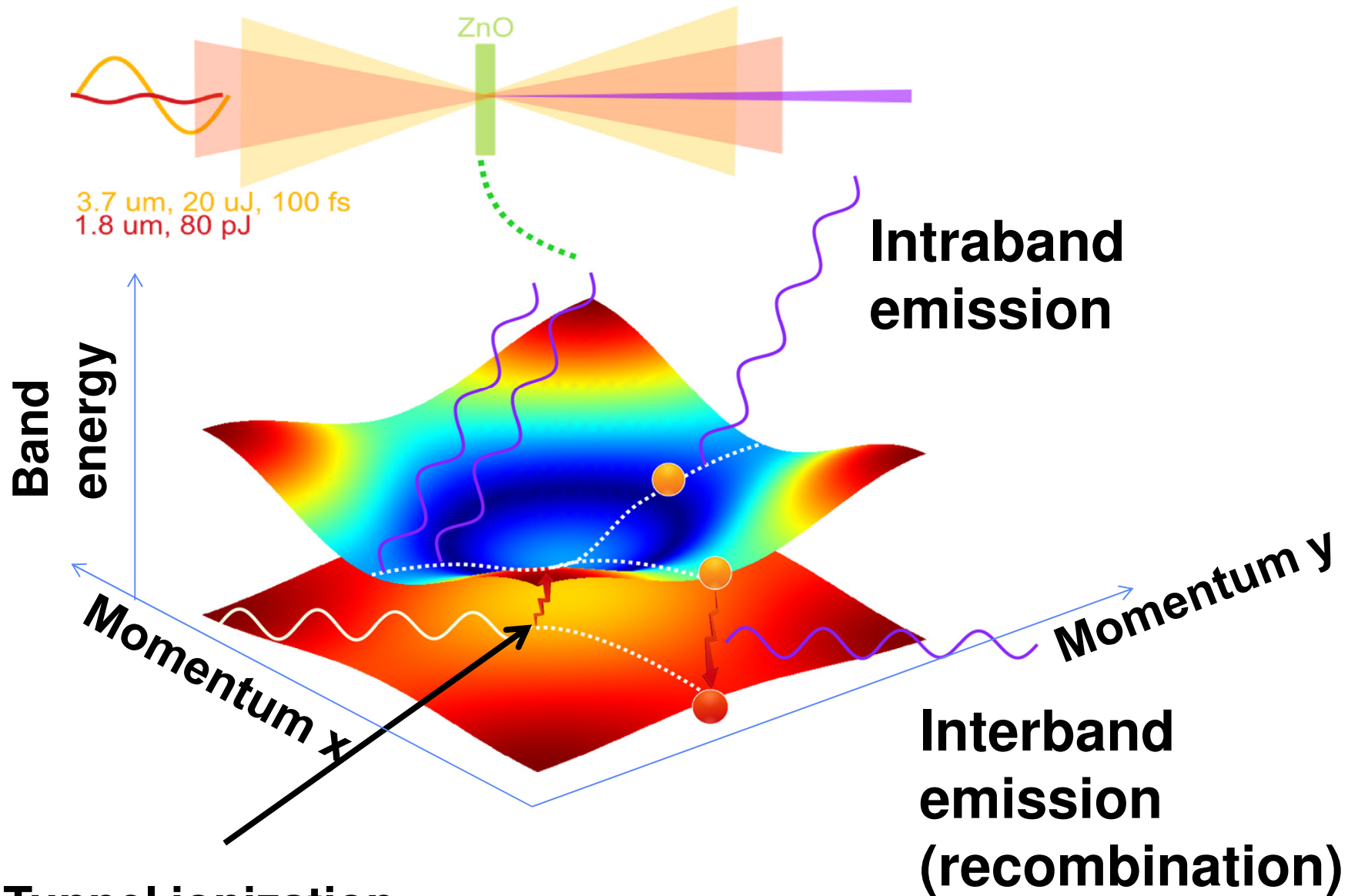


Observation of high-order harmonic generation in a bulk crystal

Shambhu Ghimire¹, Anthony D. DiChiara², Emily Sistrunk², Pierre Agostini², Louis F. DiMauro² and David A. Reis^{1,3*}



Concept of Theory and Experiment



Tunnel ionization
(Keldysh 1965)

PRL 113, 073901 (2014)

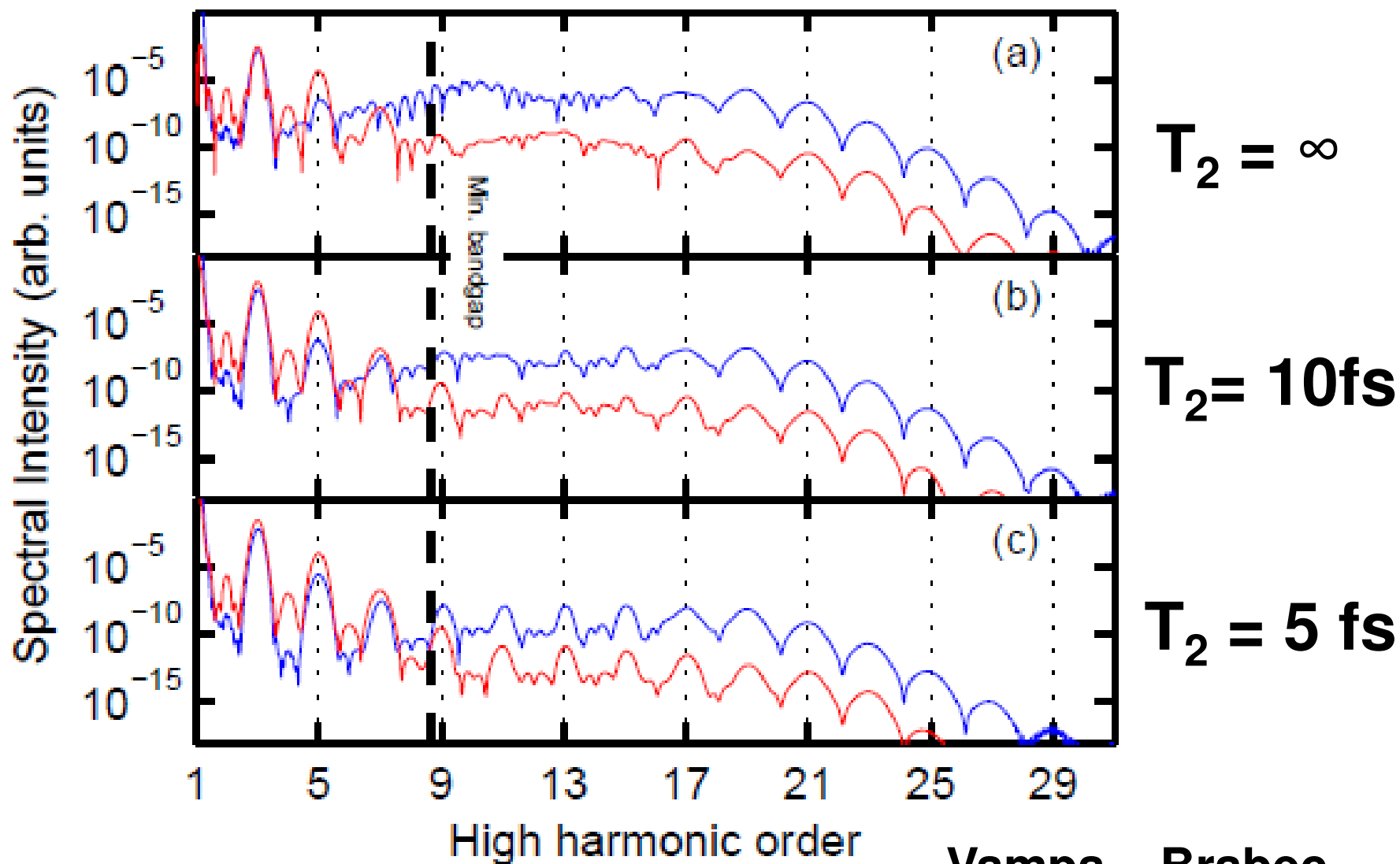


The model -- Giulio Vampa and Thomas Brabec

- Two bands
- 3-D
- Band parameters from ZnO
- Require dephasing of <5 fs for experimental-like harmonics
- Broad ellipticity dependence, similar to experiment.

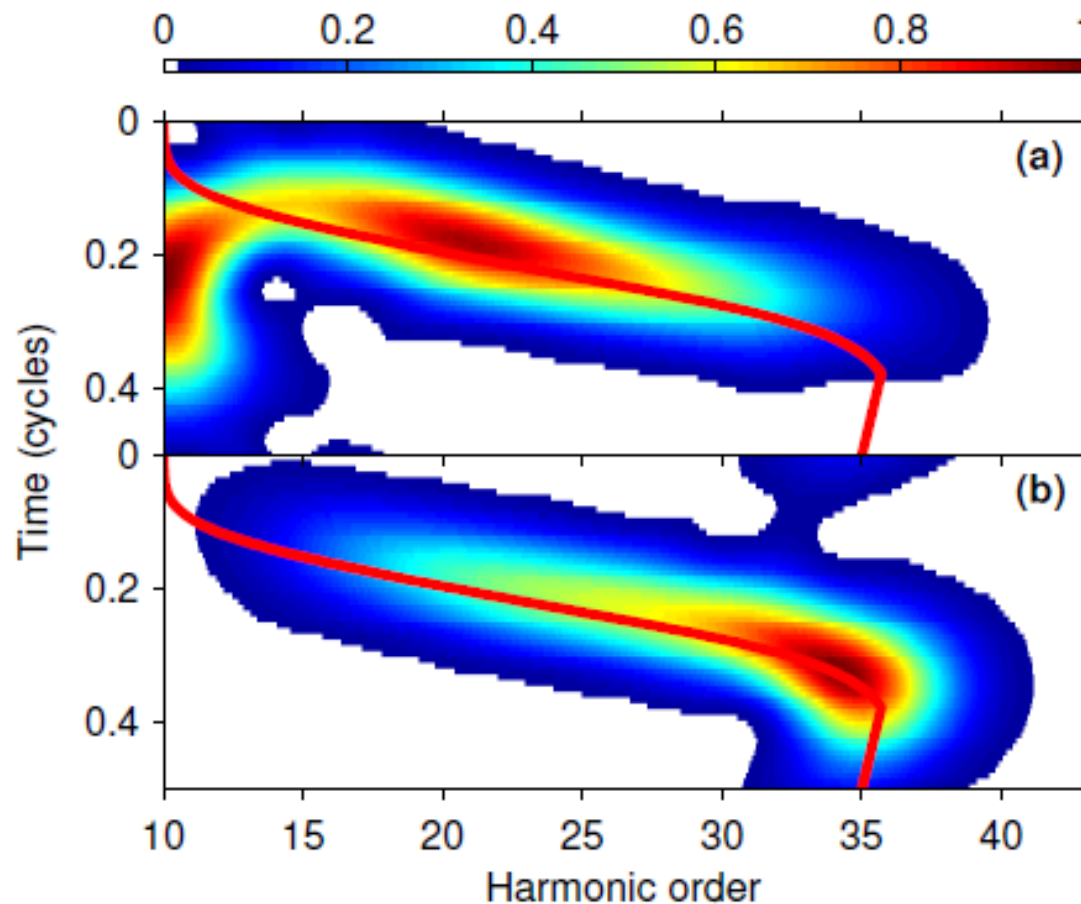


Interband and intraband harmonics from solids – ZnO₂ at 3.5 micron.



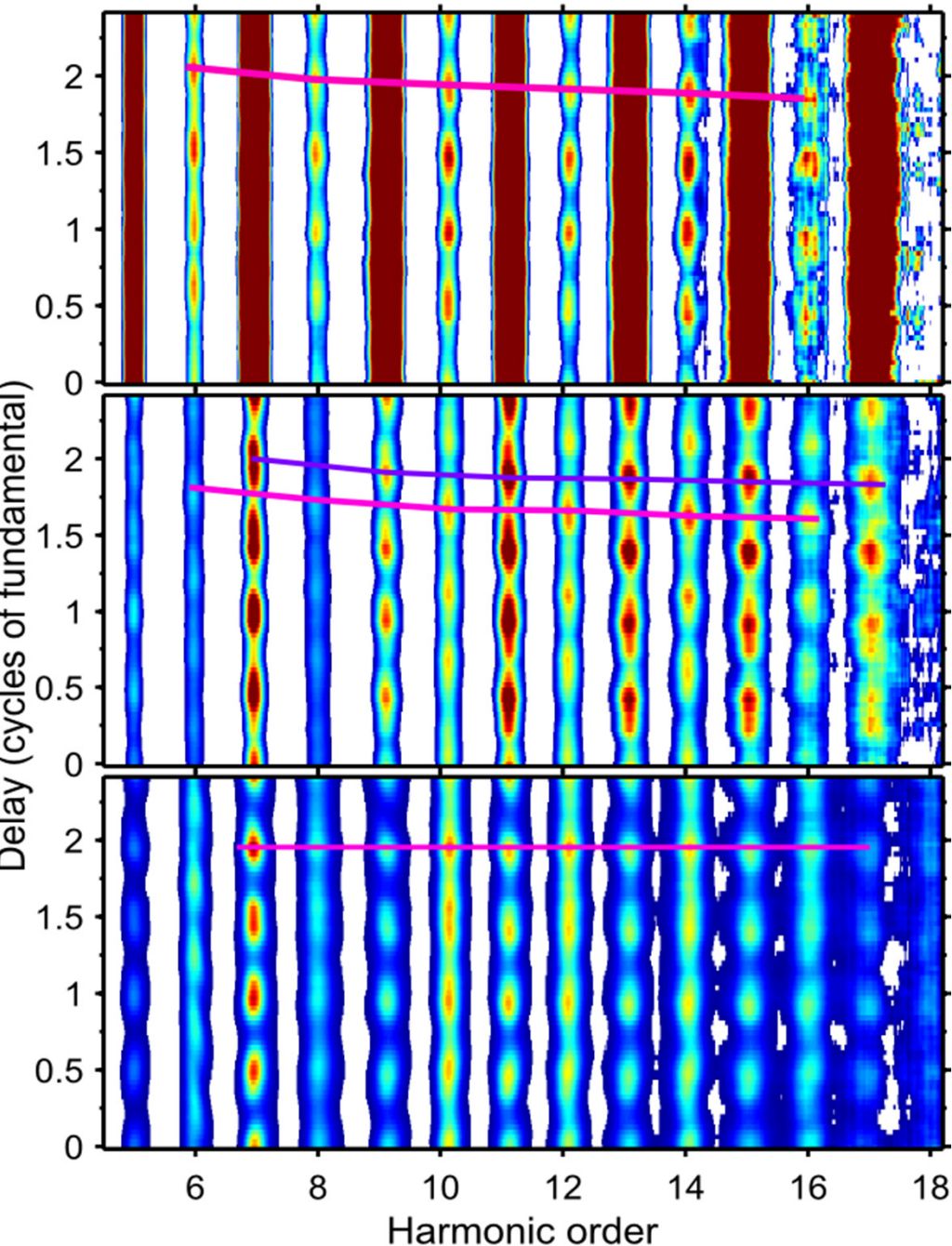
Vampa – Brabec,
Submitted to PRL

Emission time from intraband term (a) and the interband term (b)



**Recollision
seems
important
for both!**

Perturbing and Controlling HHG in solids

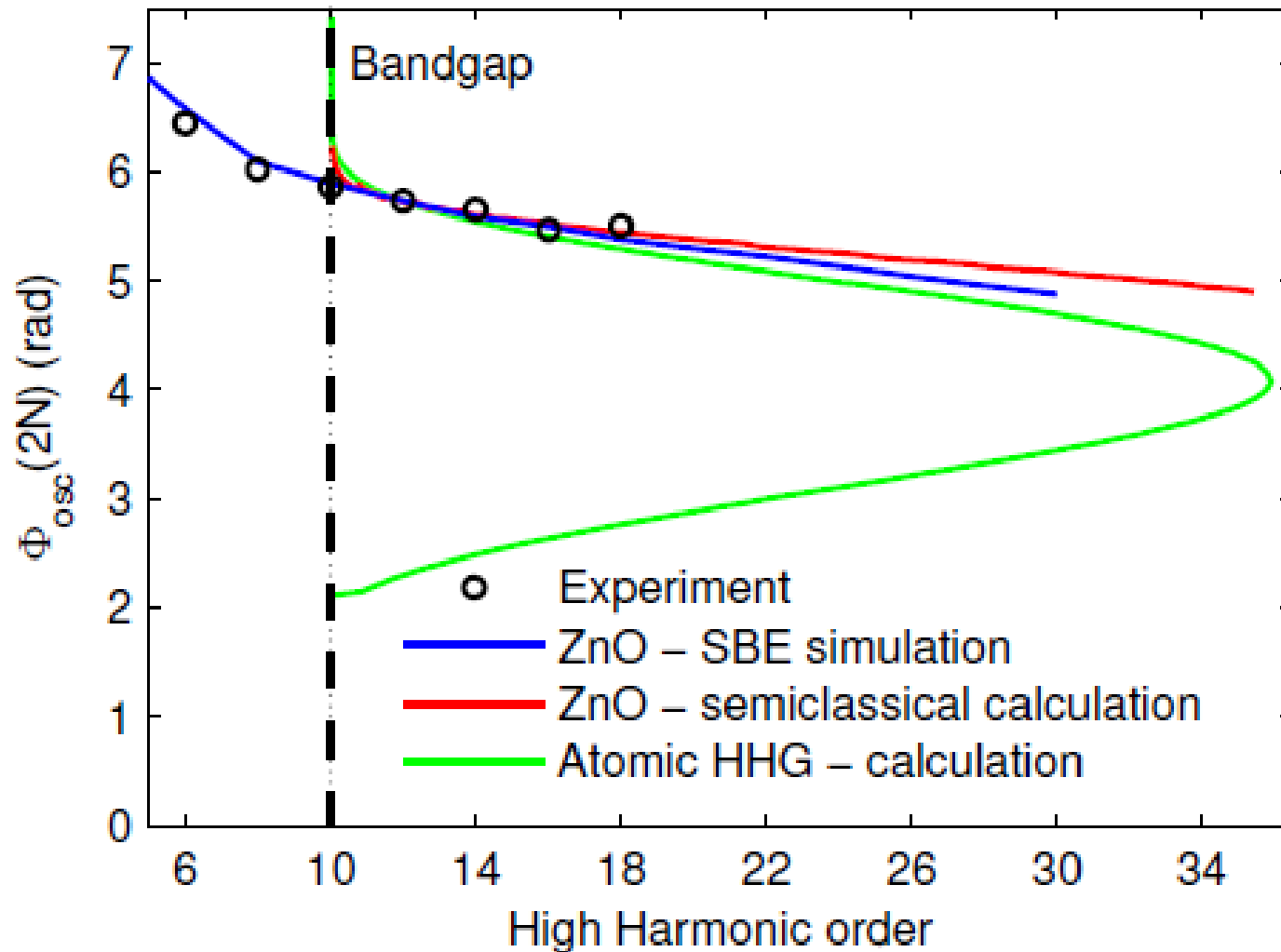


**Weak 2ω (2×10^{-6});
trajectories are
perturbed**

**Stronger 2ω ($\sim 10^{-4}$);
Phase modified by π**

**Strong 2ω ($\sim 10^{-2}$);
ionization occurs
each $\frac{1}{2}$ cycle.**

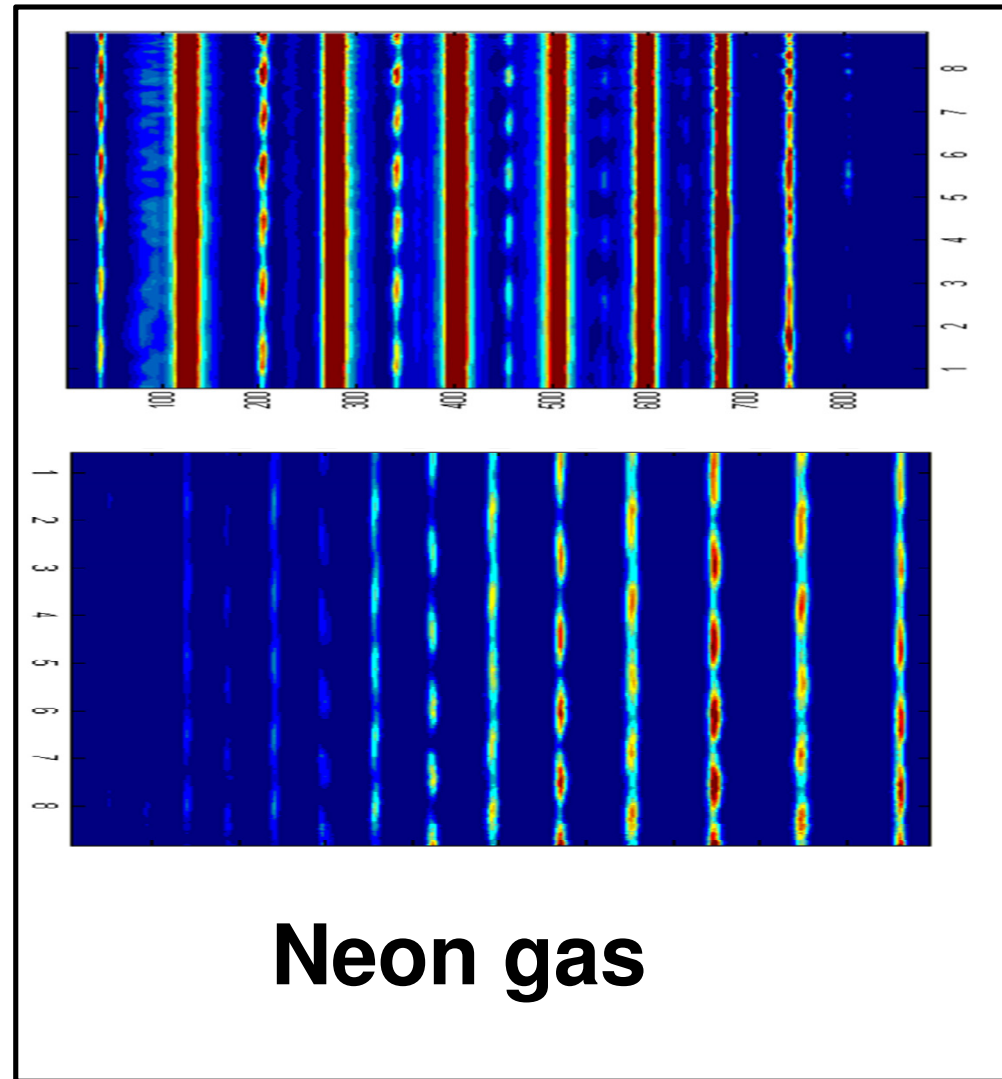
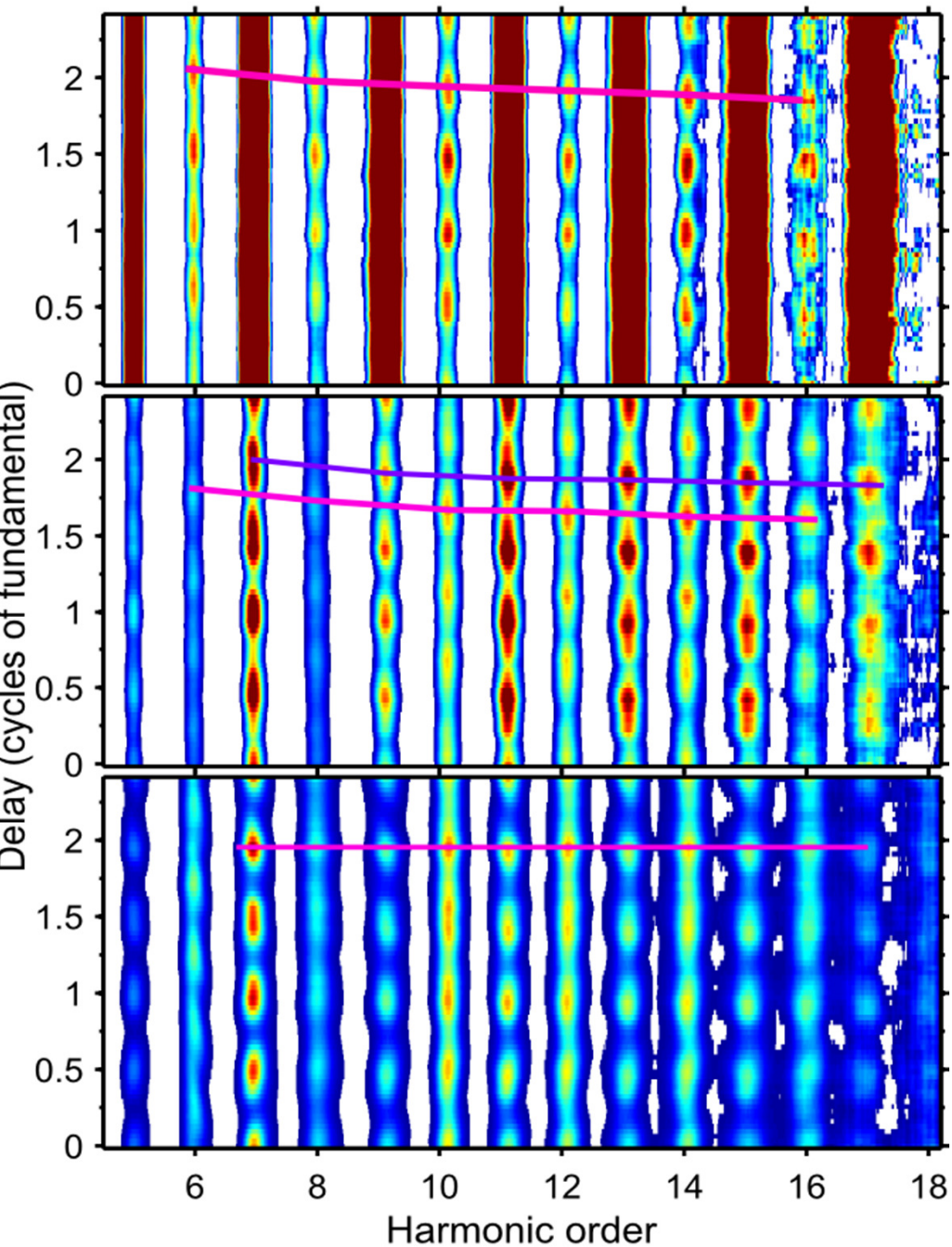
The spectral phase of solid harmonics



For the atomic calculation, the intensity is scaled to make the cut offs agree

No long trajectory contribution is found in the experiment

Perturbing and Controlling HHG in solids



From Nature Physics, 2,
281, (2006)

Final comment:

Even at 10^{22} or 10^{23} W/cm², laser electric fields are small on the particle or nuclear physics scale.

What we can offer super-intense science could be a systematic method for measuring nuclear dynamics.

As we have demonstrated, insitu methods allow ultrafast dynamics to be resolved and they help us differentiate between theories.



JASLab, 2013

Canada, China, France, Germany, Israel, Italy, Korea, USA



Funds:

- NSERC
- CRC
- CFI
- NRC
- Ontario, Premier's Fund
- US AFOSR
- US Army
- US DARPA