



**Training &
Experience Hub
Berlin**

Quantum Materials: from ARPES to trARPES



16/09/2026
online



PHOTONICS PUBLIC PRIVATE PARTNERSHIP
This project has received funding from the European Union Horizon Europe research and innovation program under grant agreement No 101135838.

Ultrafast lasers and HHG sources

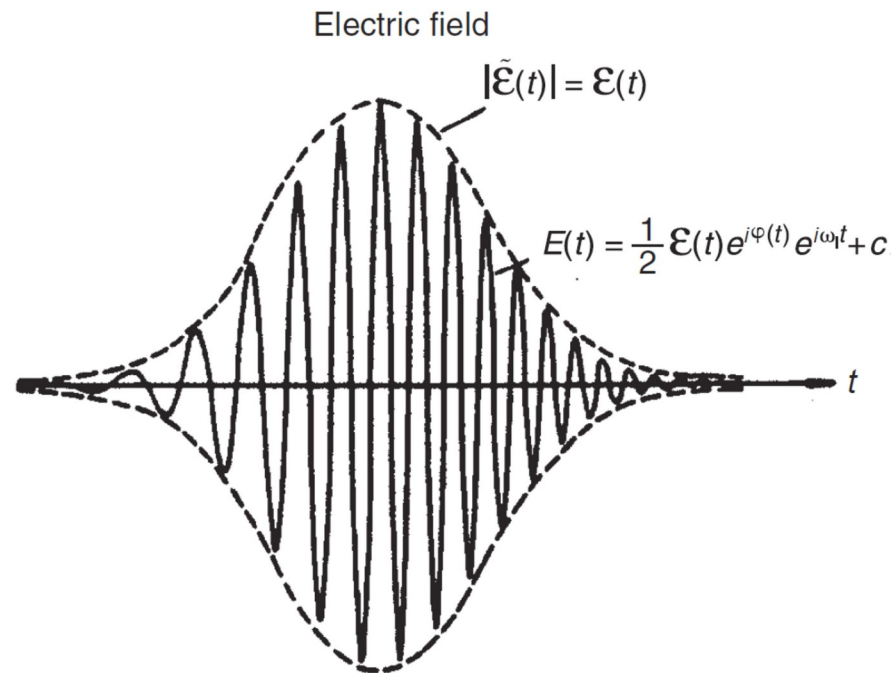
Oleg Kornilov



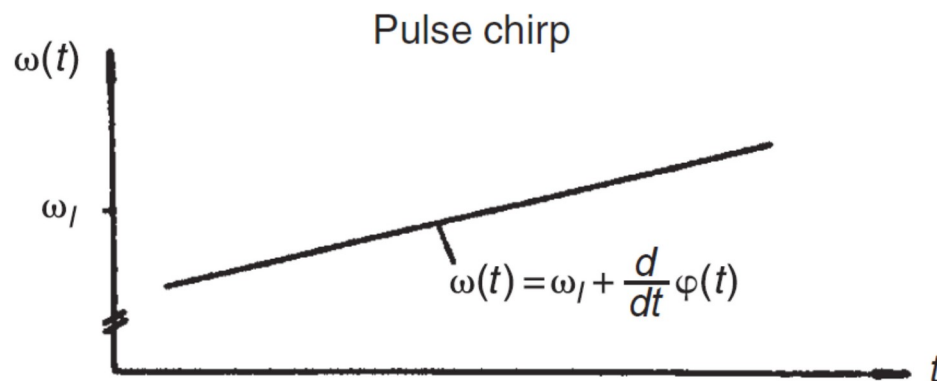
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www.lasers4.eu

Basic ultrafast laser concepts



The laser electric field $E(t)$ is described as the product of a real envelope function $\varepsilon(t)$ and an oscillatory term depending on the time-dependent phase $\varphi(t)$ and the carrier frequency ω_l .



The laser chirp is determined by the time-dependent phase $\varphi(t)$.

Basic ultrafast laser concepts

Frequency domain representation of laser field:

$$\tilde{E}(\omega) = \int_{-\infty}^{\infty} E(t) e^{-i\omega t} dt = |\tilde{E}(\omega)| e^{i\varphi(\omega)}$$

$$E(t) = \frac{1}{2\pi} \int_{-\infty}^{\infty} \tilde{E}(\omega) e^{i\omega t} d\omega$$

Alternative description using only positive frequencies

$$\tilde{E}^+(\omega) = \begin{cases} \tilde{E}(\omega), & \text{for } \omega \geq 0 \\ 0, & \text{for } \omega < 0 \end{cases}$$

J.-C. Diels and W. Rudolph, *Ultrashort Laser Pulse Phenomena*, (Academic Press, 2006)

Fourier-limited pulse durations

$$\Delta\omega_p\tau_p = 2\pi\Delta\nu_p\tau_p \geq 2\pi c_B$$

Examples of standard pulse profiles. The spectral values given are for unmodulated pulses. Note that the Gaussian is the shape with the minimum product of mean square deviation of the intensity and spectral intensity.

Shape	Intensity profile $I(t)$	τ_p FWHM	Spectral profile $S(\Omega)$	$\Delta\omega_p$ FWHM	c_B	$\langle\tau_p\rangle\langle\Delta\Omega_p\rangle$ MSQ
Gauss	$e^{-2(t/\tau_G)^2}$	$1.177\tau_G$	$e^{-\left(\frac{\Omega\tau_G}{2}\right)^2}$	$2.355/\tau_G$	0.441	0.5
Sech	$\text{sech}^2(t/\tau_s)$	$1.763\tau_s$	$\text{sech}^2\frac{\pi\Omega\tau_s}{2}$	$1.122/\tau_s$	0.315	0.525
Lorentz	$[1 + (t/\tau_L)^2]^{-2}$	$1.287\tau_L$	$e^{-2 \Omega \tau_L}$	$0.693/\tau_L$	0.142	0.7
Asym. sech	$\left[e^{t/\tau_a} + e^{-3t/\tau_a}\right]^{-2}$	$1.043\tau_a$	$\text{sech}\frac{\pi\Omega\tau_a}{2}$	$1.677/\tau_a$	0.278	
Square	1 for $ t/\tau_r \leq 1$, 0 elsewhere	τ_r	$\text{sinc}^2(\Omega\tau_r)$	$2.78/\tau_r$	0.443	3.27

J.-C. Diels and W. Rudolph, *Ultrashort Laser Pulse Phenomena*, (Academic Press, 2006)

Influence of chirp

Time-dependent variations of the time-dependent phase $\varphi(t)$ can lead to pulses longer than the Fourier-limit

carrier envelope

phase

instantaneous frequency

linear chirp

N.B. positive chirp

means $d\omega/dt > 0$

$$\varphi(t) = \varphi_0 + \left(\frac{\partial \varphi(t)}{\partial t} \right)_{t=t_0} (t - t_0) + \frac{1}{2} \left(\frac{\partial^2 \varphi(t)}{\partial t^2} \right)_{t=t_0} (t - t_0)^2 + \frac{1}{6} \left(\frac{\partial^3 \varphi(t)}{\partial t^3} \right)_{t=t_0} (t - t_0)^3 + \dots$$

group delay

group delay dispersion (GDD)

$$\varphi(\omega) = \varphi_0 + \left(\frac{\partial \varphi(\omega)}{\partial \omega} \right)_{\omega=\omega_0} (\omega - \omega_0) + \frac{1}{2} \left(\frac{\partial^2 \varphi(\omega)}{\partial \omega^2} \right)_{\omega=\omega_0} (\omega - \omega_0)^2 + \frac{1}{6} \left(\frac{\partial^3 \varphi(\omega)}{\partial \omega^3} \right)_{\omega=\omega_0} (\omega - \omega_0)^3 + \dots$$

N.B. positive GDD; red before blue

Phase velocity and group velocity

$$v_{phase} = \lambda v = \frac{\lambda \omega}{2\pi} = \frac{\omega}{k}$$

ω **laser frequency**
k **laser wavevector**

$$v_{group} = \frac{\partial \omega}{\partial k}$$

Propagation through a medium with refractive index $n(\omega)$

$$k = \frac{\omega}{v_{phase}} = \frac{\omega n(\omega)}{c}, \text{ so } \frac{dk}{d\omega} = \left(n + \omega \frac{dn}{d\omega} \right) / c$$

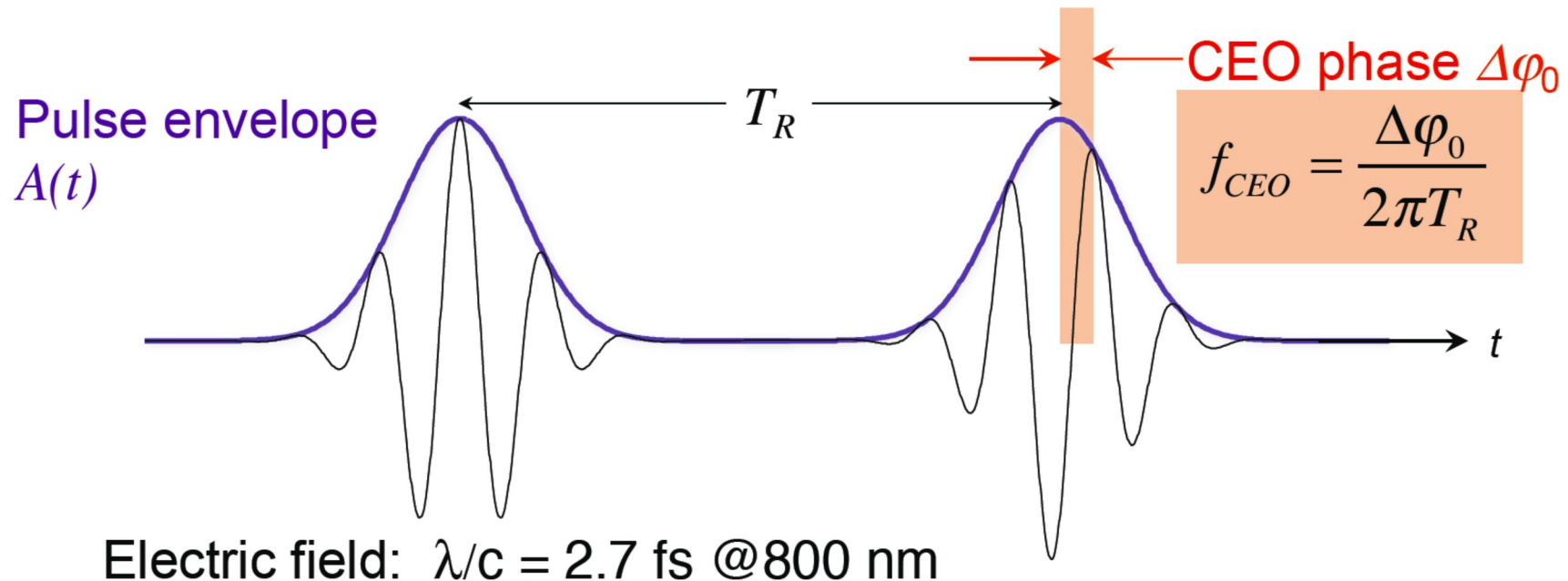
$$v_{group} = v_{phase} / \left(1 + \frac{\omega}{n} \frac{dn}{d\omega} \right)$$

The phase and group velocity differ in a dispersive medium

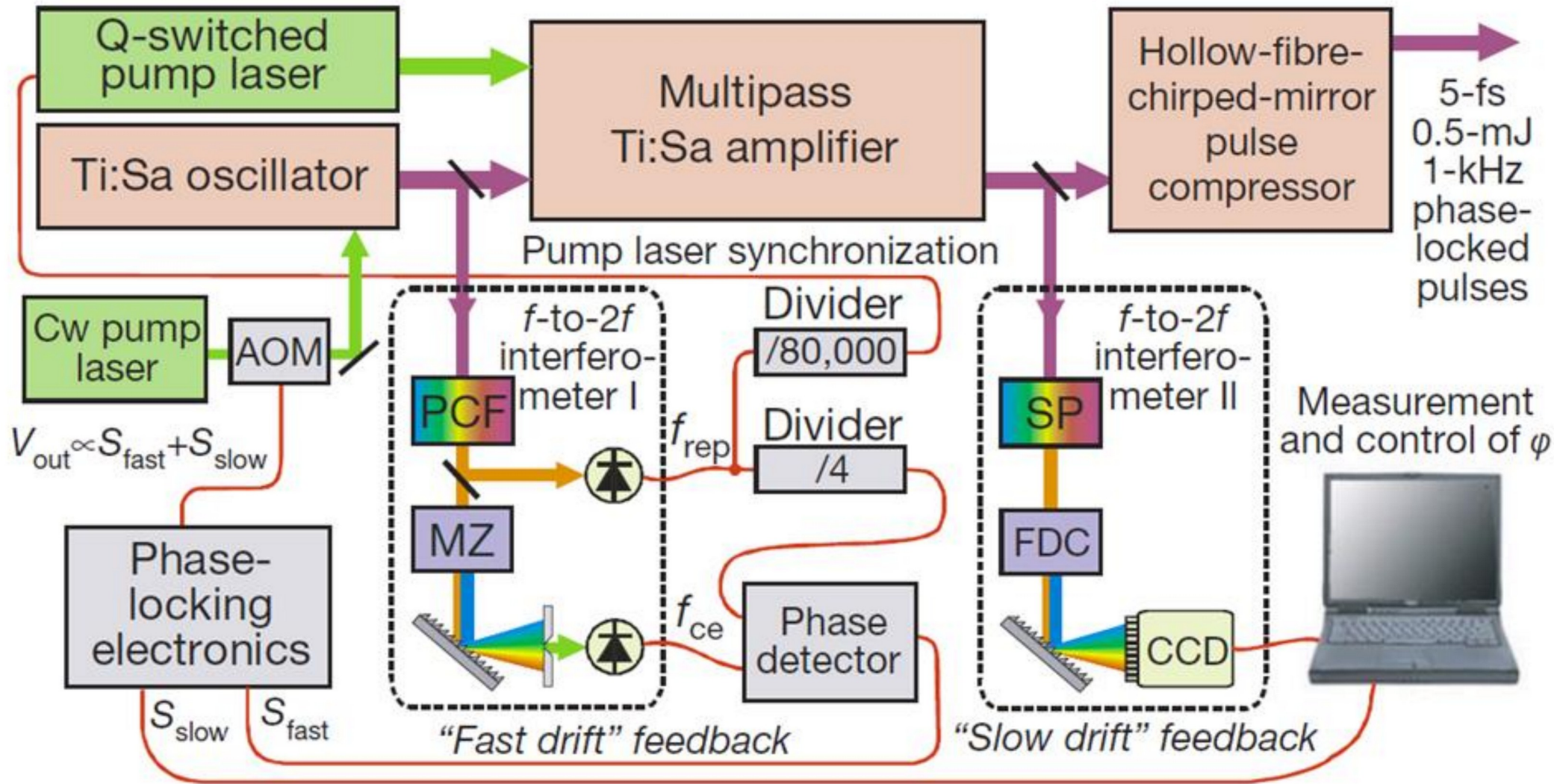
Carrier-Envelope Phase

carrier envelope
phase

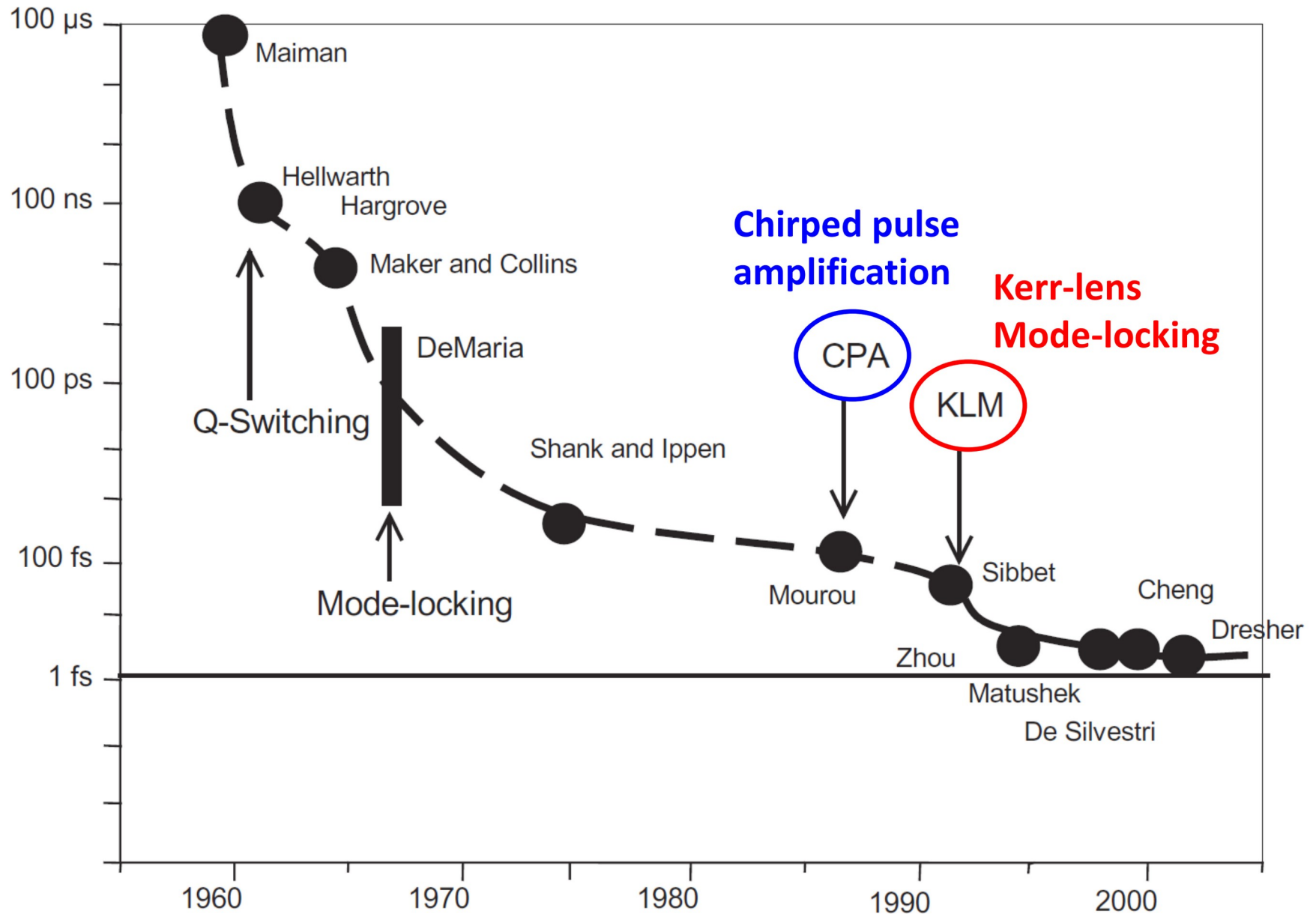
$$\varphi(t) = \varphi_0 + \left(\frac{\partial \varphi(t)}{\partial t} \right)_{t=t_0} (t - t_0) + \frac{1}{2} \left(\frac{\partial^2 \varphi(t)}{\partial t^2} \right)_{t=t_0} (t - t_0)^2 + \frac{1}{6} \left(\frac{\partial^3 \varphi(t)}{\partial t^3} \right)_{t=t_0} (t - t_0)^3 + \dots$$



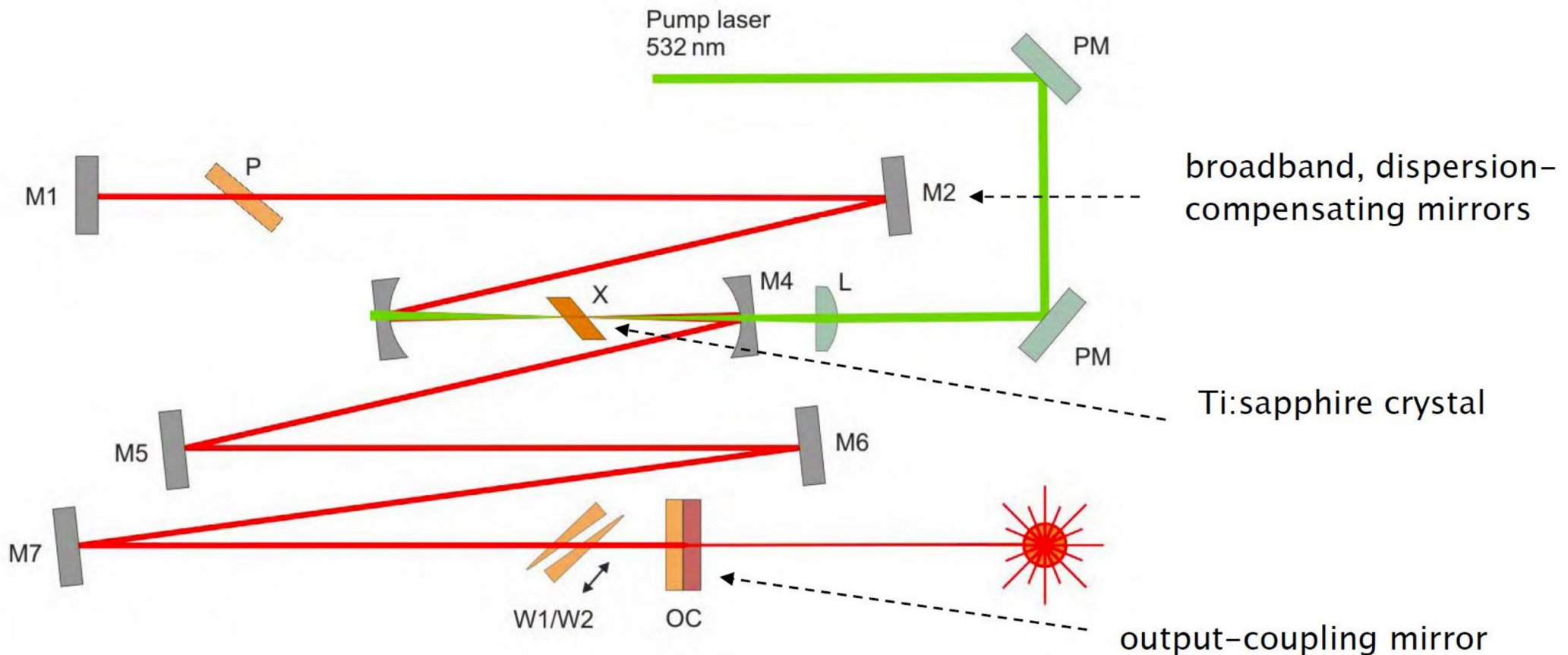
A state-of-the-art laser system for attosecond science - 1



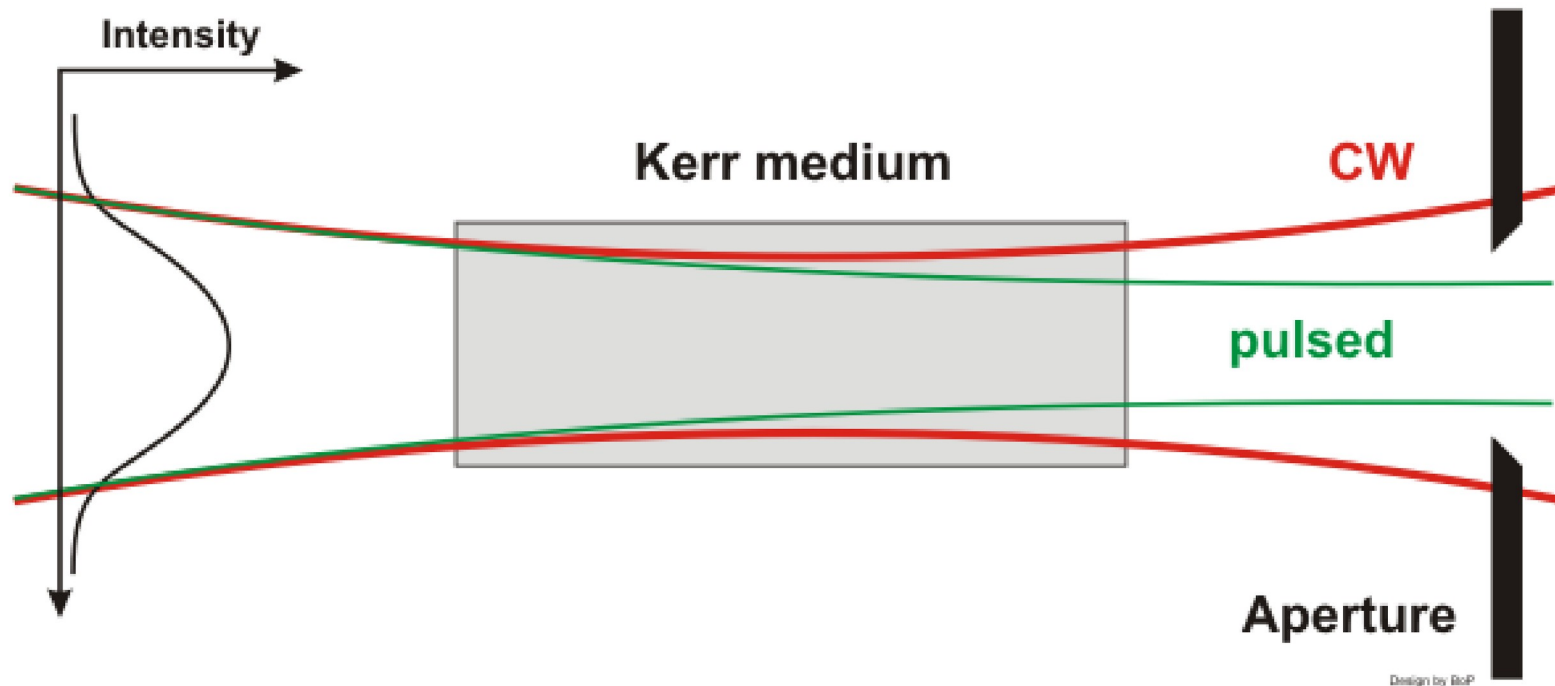
Basic operation of a mode-locked Ti:Sapphire oscillator



Basic operation of a mode-locked Ti:Sapphire oscillator



Kerr-lens mode-locking



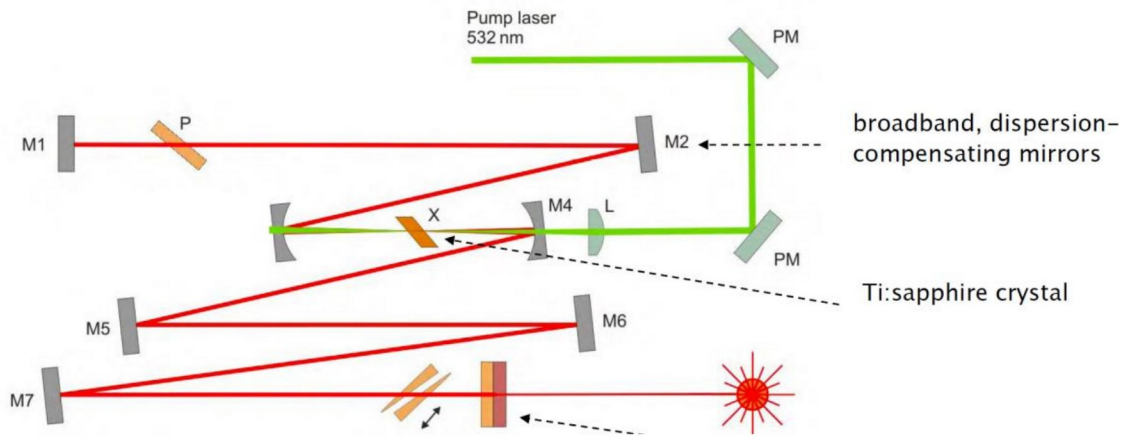
Kerr-lens mode-locking exploits the non-linearity of the refractive index

$$n(I) = n_0 + n_2 I$$

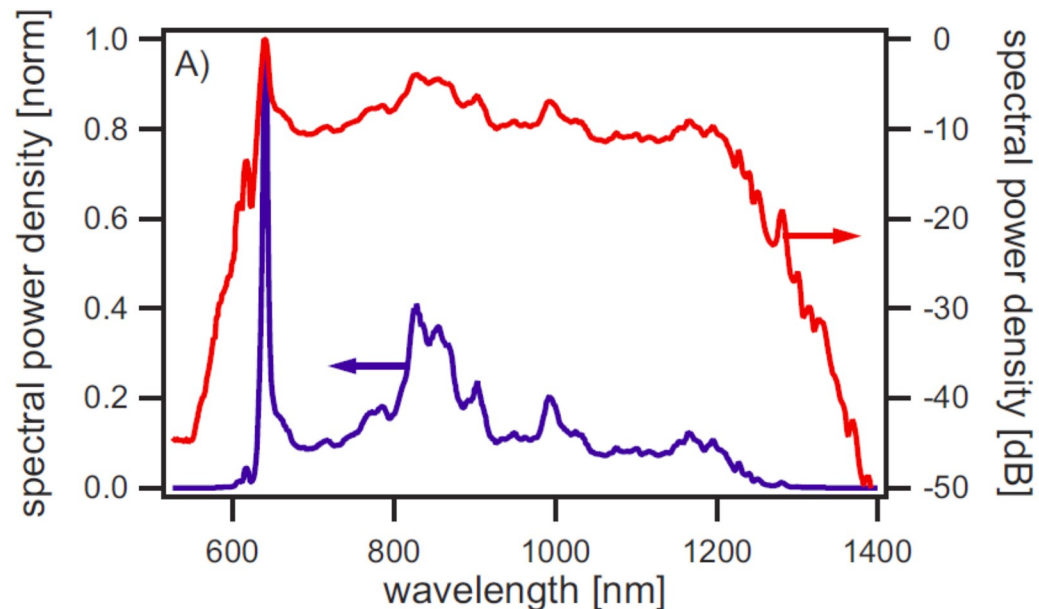
Focal length of the Kerr lens $f = \frac{w_0^2}{4n_2 d I_0}$ w_0 = beam radius
d = thickness

http://www.rp-photonics.com/kerr_lens.html

Example of an octave-spanning oscillator

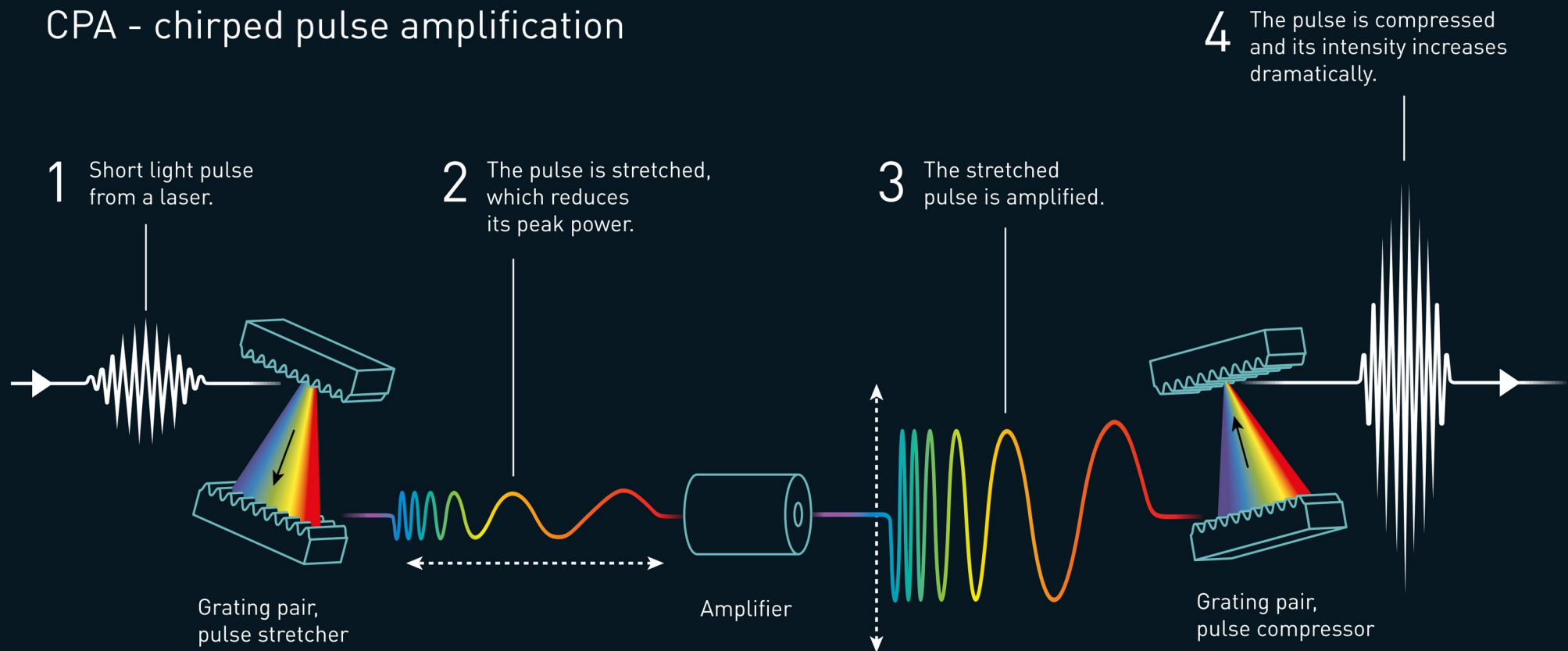


Using an octave-spanning oscillator 4-fs pulses can be generated and the stage is set for stabilization of the carrier envelope phase



Pulse energy: Chirped pulse amplification (CPA)

CPA - chirped pulse amplification



Nobel Prize 2018



Gérard Mourou



Donna Strickland

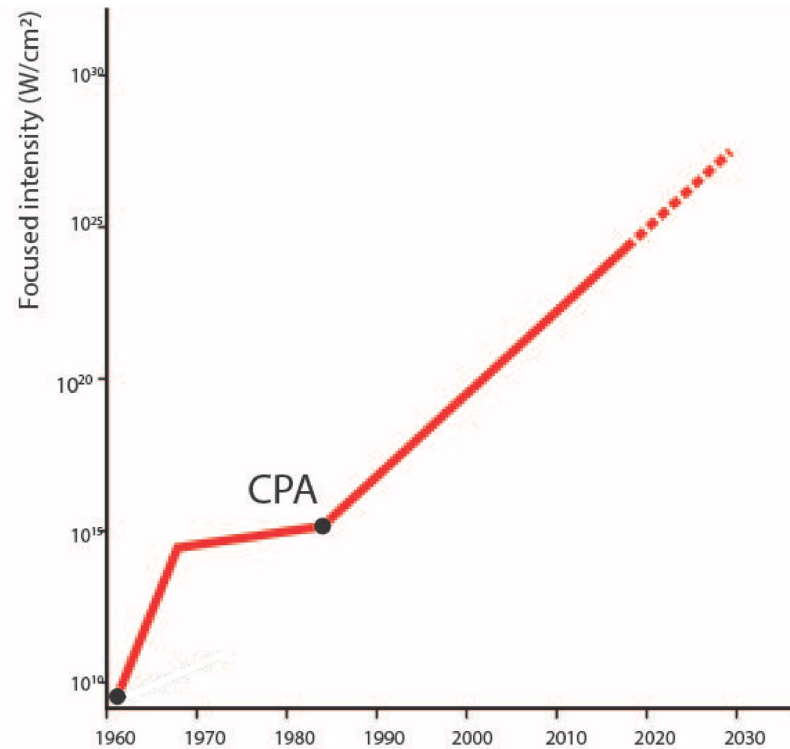


Figure 3. The history of pulsed laser technology in terms of peak intensity. After an initial increase in intensity during the 1960s, as described in the text, only an incremental increase occurred during more than a decade. The invention of the CPA technique for optical pulses by Strickland and Mourou in 1985 [45] dramatically changed the situation. The values on the y-axis are indicative because the intensity depends on how sharply the laser beam is focused.

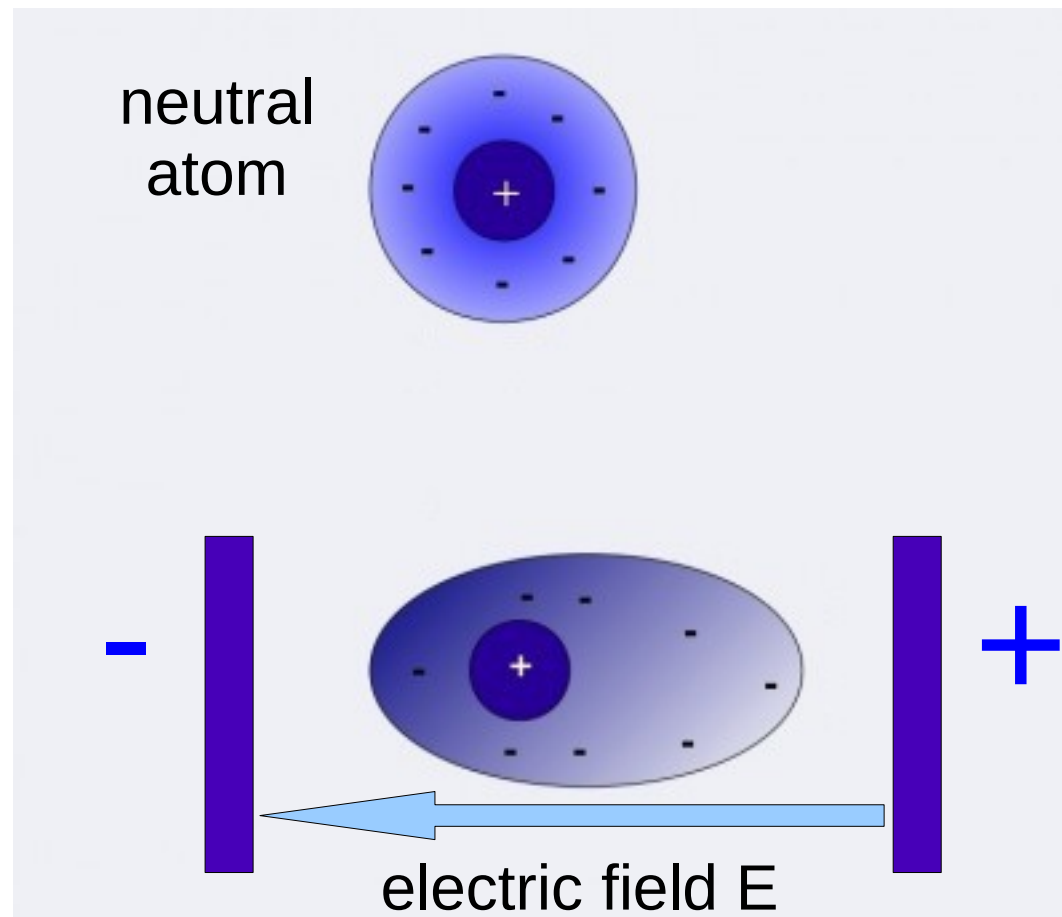


High-harmonic generation and production of attosecond pulses

1. Nonlinear susceptibility
2. Keldysh parameter
3. High-harmonic generation
4. Attosecond pulses



$$P = \chi^{(1)} E + \chi^{(2)} E^2 + \chi^{(3)} E^3 + \dots$$



P - polarization
E - electric field
 χ - susceptibility

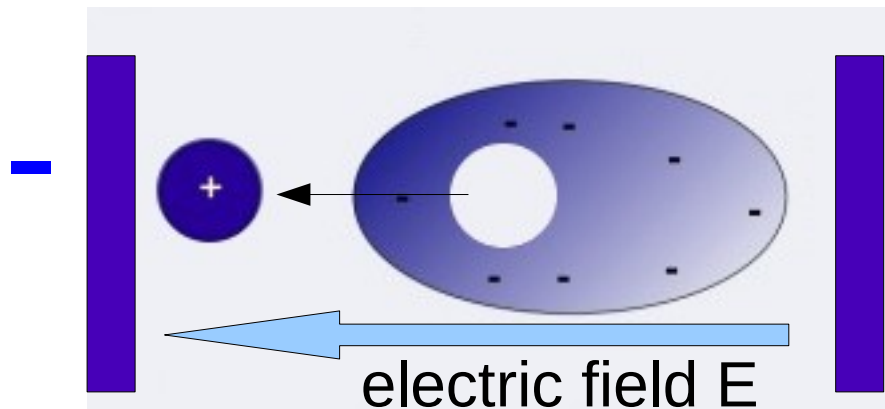
$$P = \chi^{(1)} E + \chi^{(2)} E^2 + \chi^{(3)} E^3 + \dots$$

Contributions from high orders are small,
typical nonlinear optics.



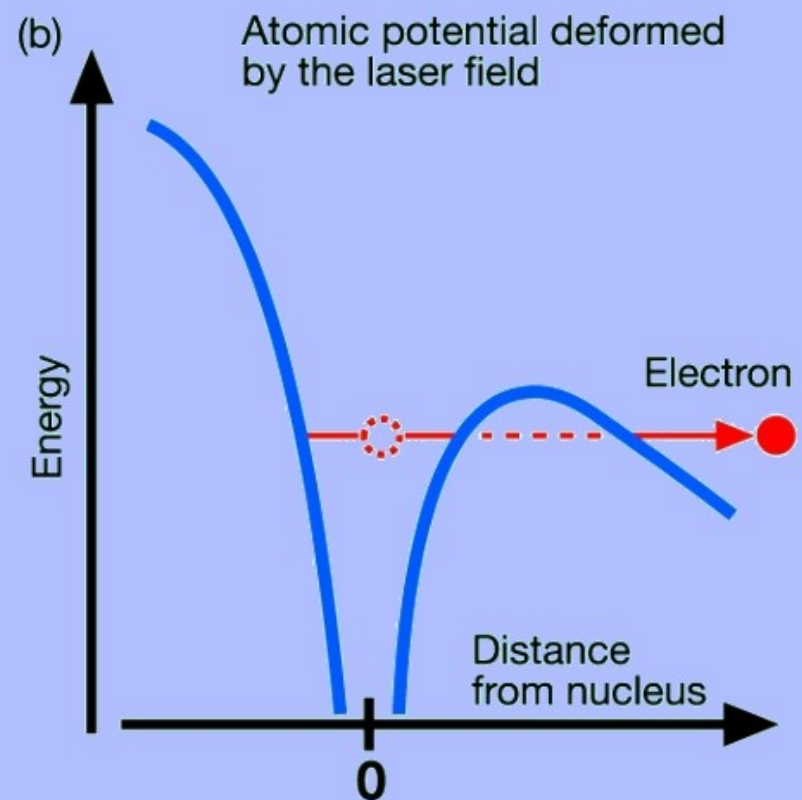
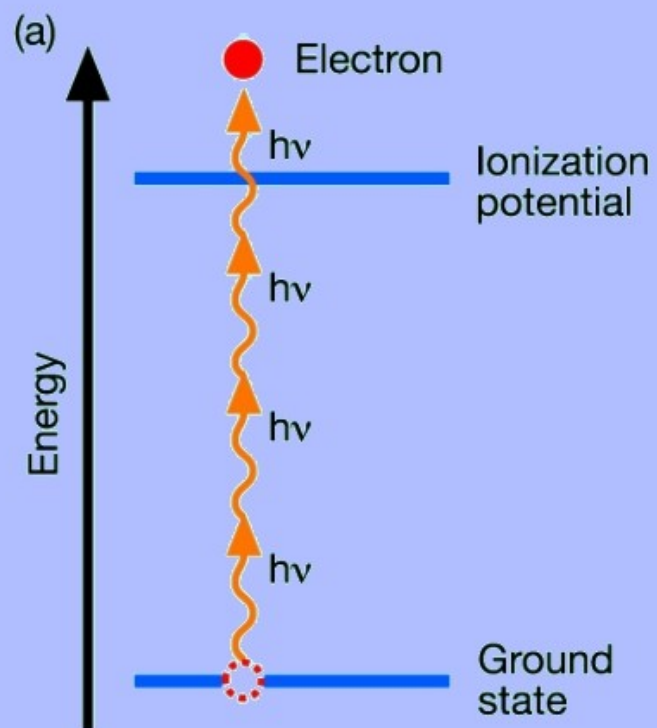
$$P = \chi^{(1)} E + \chi^{(2)} E^2 + \chi^{(3)} E^3 + \dots$$

The series is divergent!
Not a valid description.



VERY nonlinear optics – STRONG FIELD!





(Keldysh parameter)

momentum in the field of ion

$$\gamma = \omega \frac{\sqrt{2m_e V_{IP}}}{eE}$$

1/time

force of electric field

ω - electric field frequency

E – electric field amplitude

V_{IP} – ionization potential



(Keldysh parameter)

$$\gamma = \omega \frac{\sqrt{2m_e V_{IP}}}{eE}$$

multiphoton ionization
perturbative

$$\gamma \gg 1$$

tunneling ionization
non-perturbative

$$\gamma < 0.5$$



Tunneling ionization:

$$\gamma = \omega \frac{\sqrt{2m_e V_{IP}}}{eE} < 0.5$$

What laser do we need?

Typical:

$$\lambda = 633 \text{ nm} \rightarrow \omega = 3 \cdot 10^{15} \text{ Hz}$$

$$V_{IP} = 1 \text{ Ry} = 13.6 \text{ eV}$$

$$E \approx 10^{11} \text{ V/m}$$



Intensity
Poynting vector

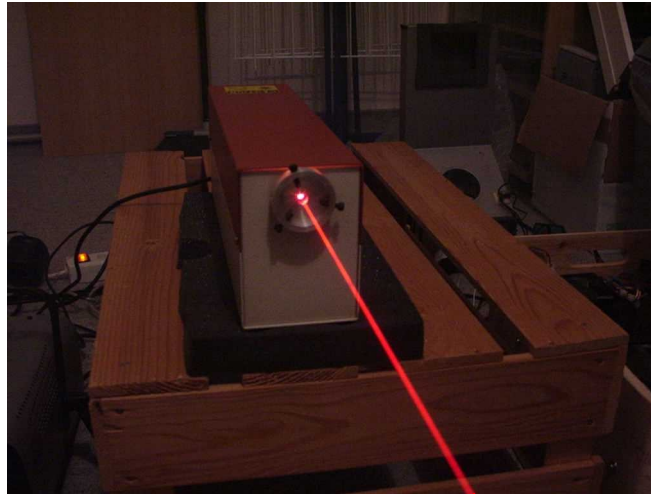
HeNe laser?

Typical:

Power: 5 mW

Focus: 20 μm

Intensity: $1.6 \times 10^7 \text{ W/m}^2$



$E \approx 10^5 \text{ V/m}$ – need 12 more **orders** to get 10^{11} V/m !



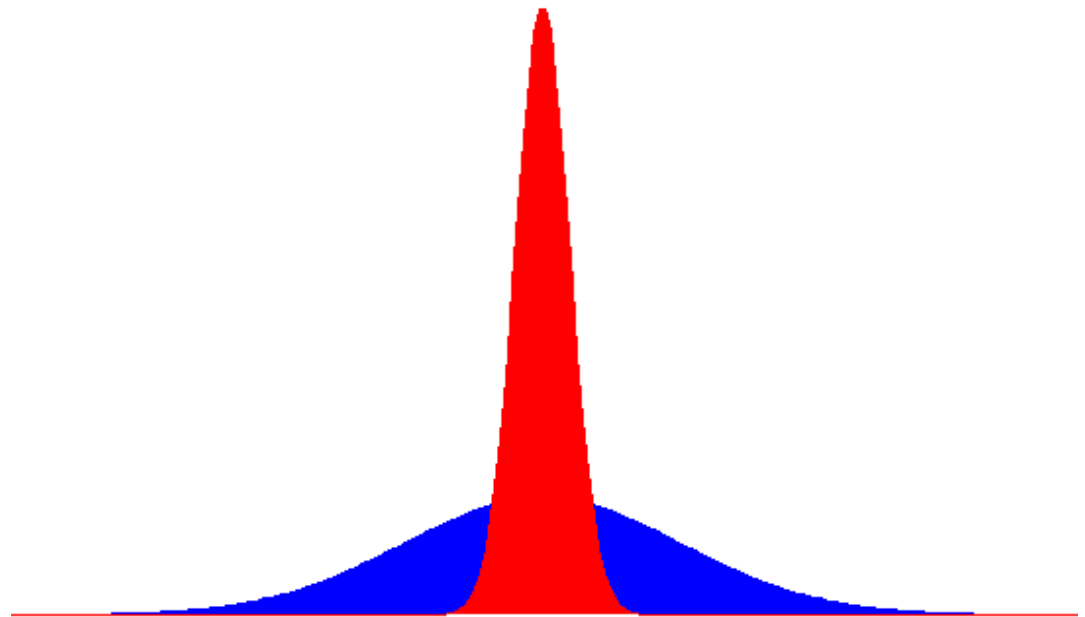
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The shorter the pulse, the stronger the field (same pulse energy)

Pulsed lasers!

Typical:
Power: 1W
Rep. rate: 1kHz
Pulse energy: 1mJ

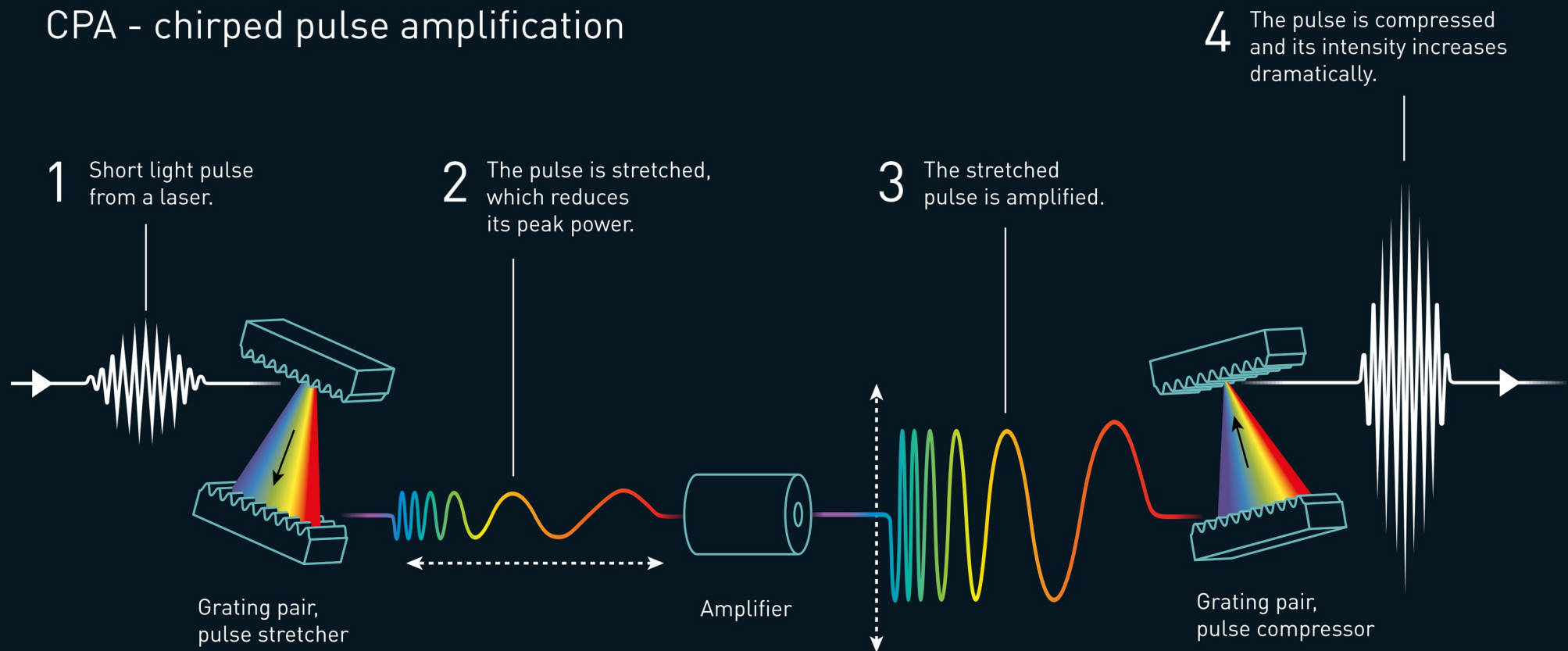


Need pulse duration of 200 fs to get 10^{11} V/m.

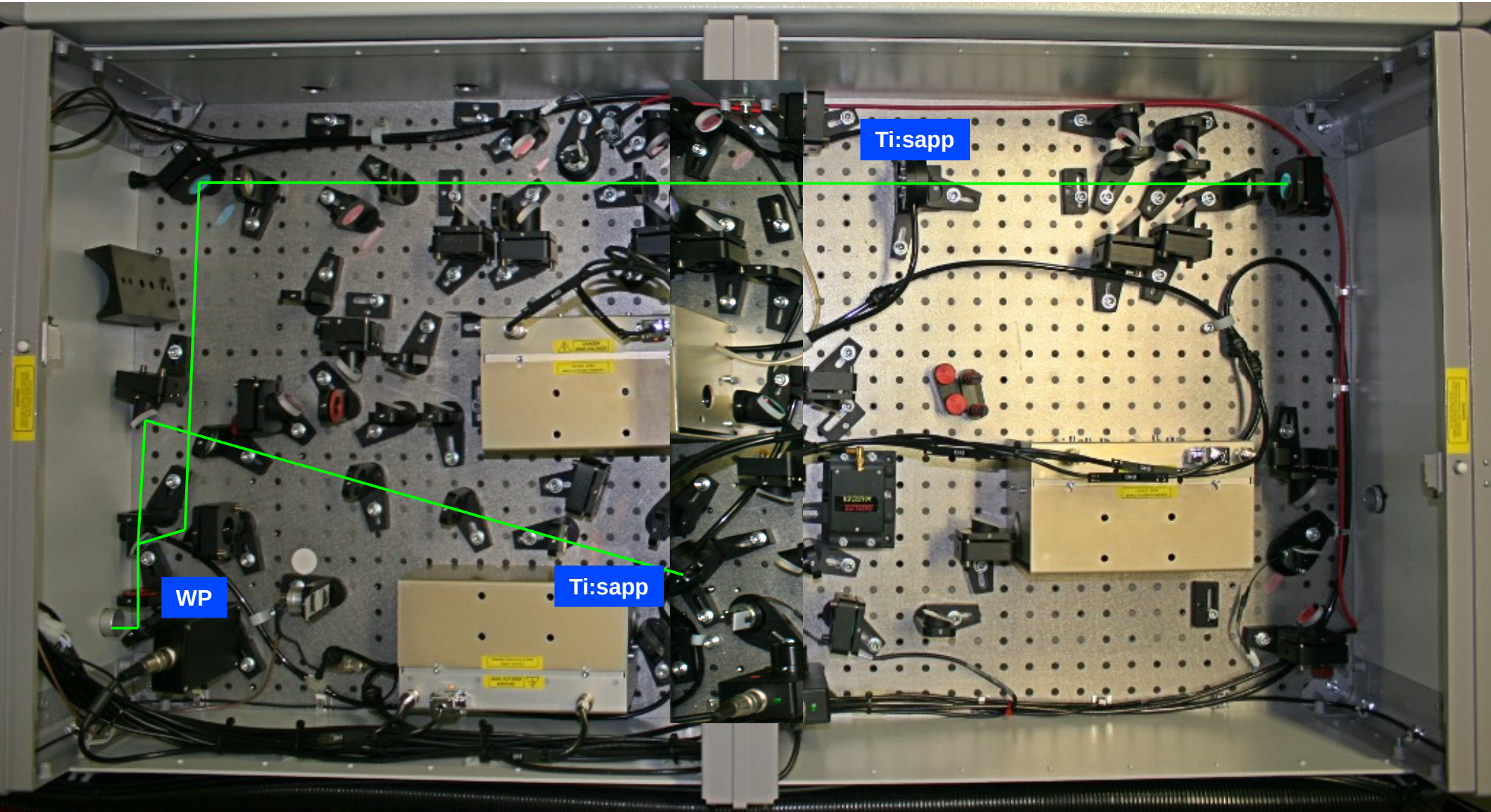
Usually cannot focus too tight, need about 20-50 fs pulses.

Pulse energy: Chirped pulse amplification (CPA)

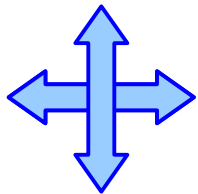
CPA - chirped pulse amplification



regen+booster amplifier



polarization



rep. rate

10 kHz

pulse duration

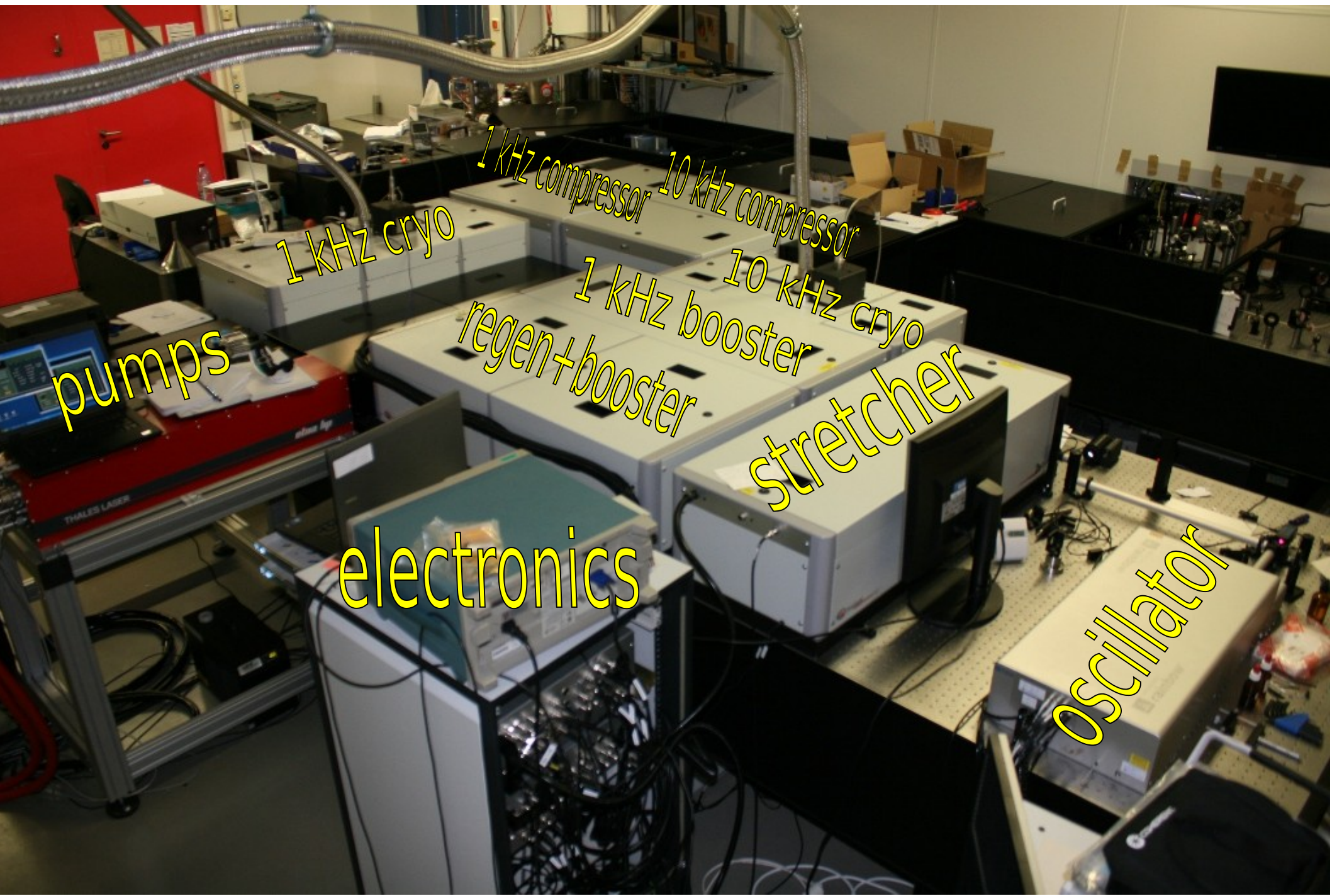


50 ns

trigger source

ETNA

Aurora laser



pumps

1 kHz cryo

1 kHz compressor

10 kHz compressor

1 kHz booster

10 kHz cryo

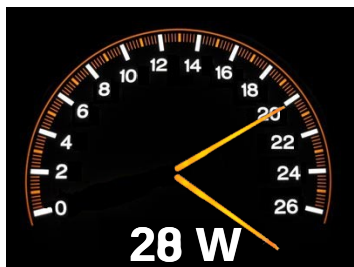
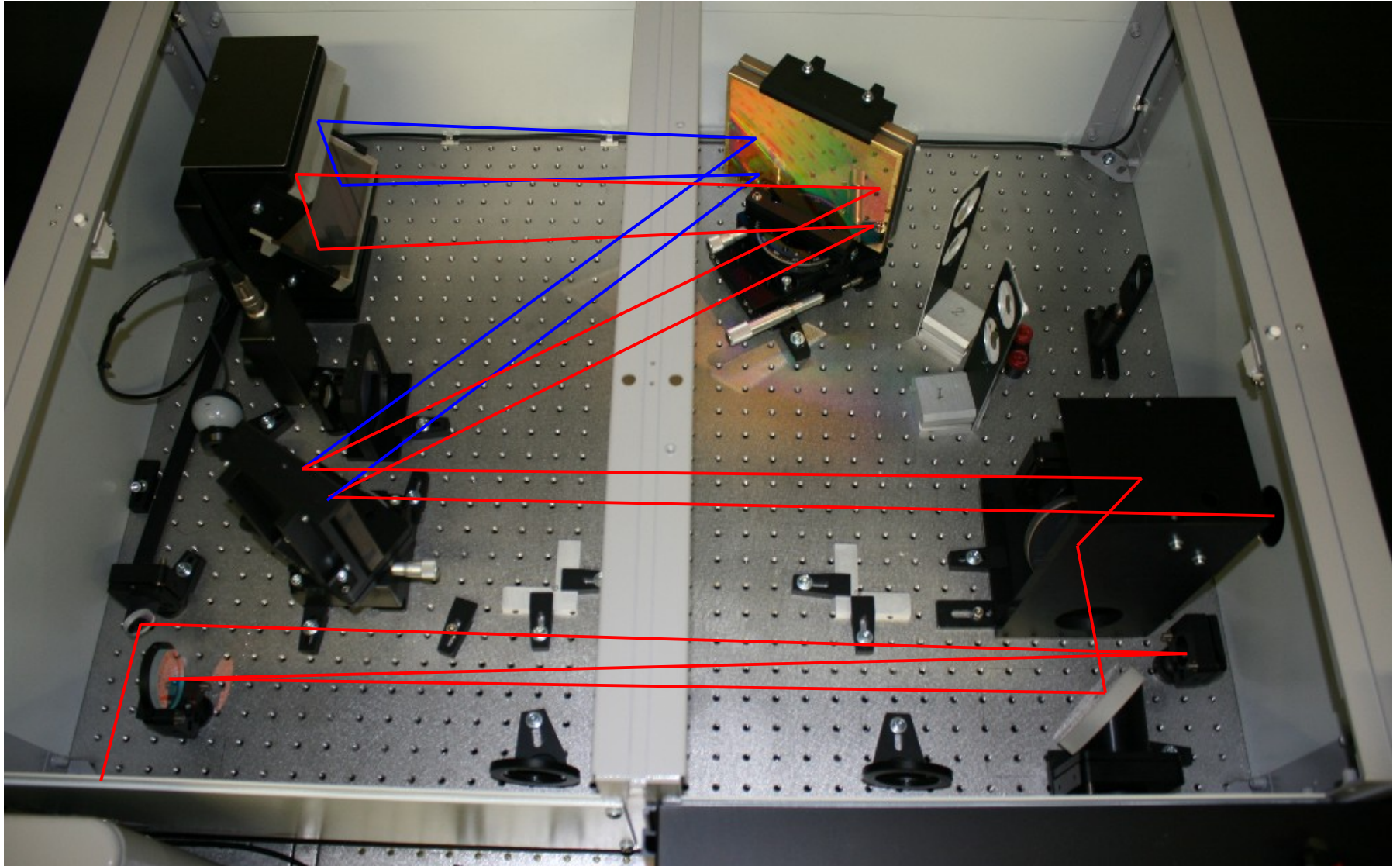
regen+booster

stretcher

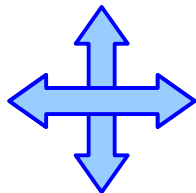
electronics

oscillator

10 kHz compressor



polarization



rep. rate

10 kHz

pulse duration

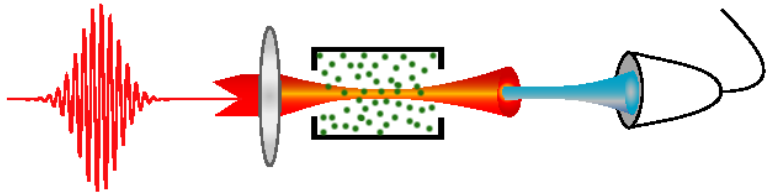


trigger source

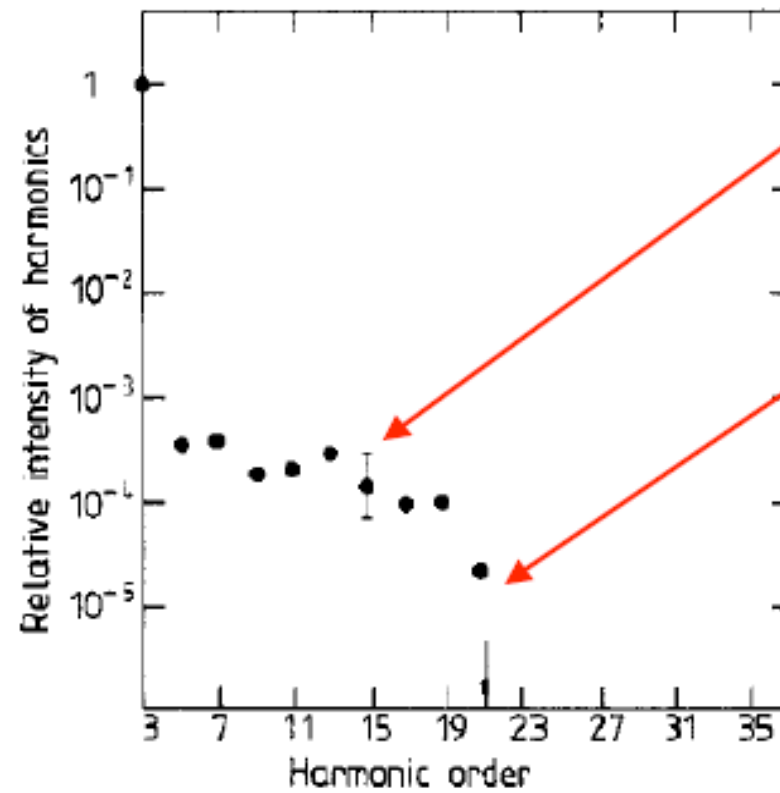
PC 2

High-order harmonic generation





only odd harmonics



plateau

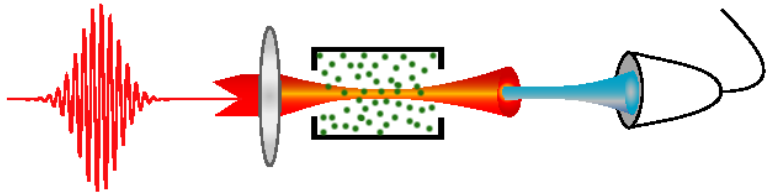
cut-off

Ferray *et al.* (1988)

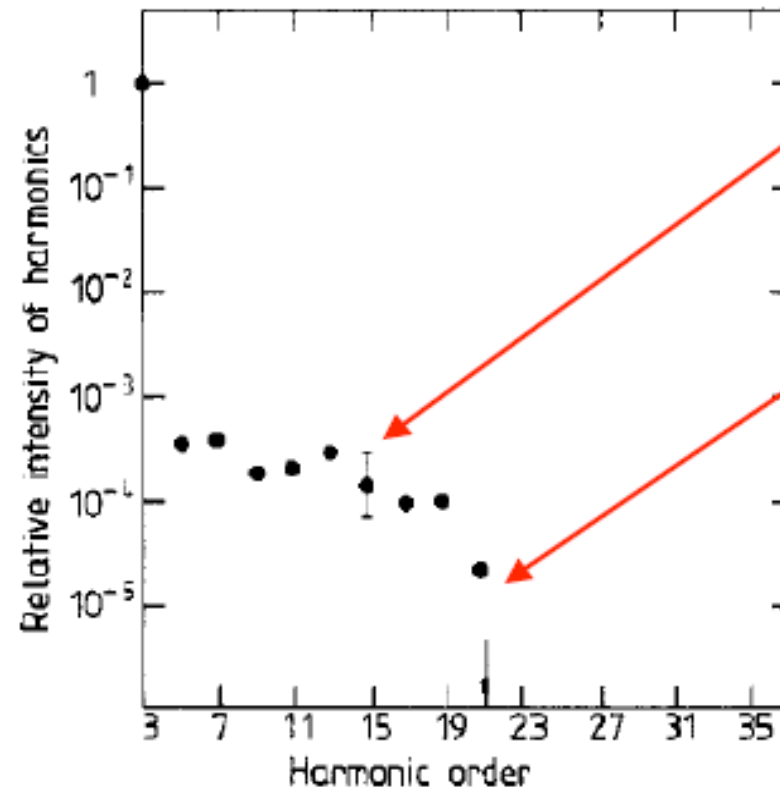


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only odd harmonics



Ferray *et al.* (1988)

plateau

cut-off

$$I_p + 3.17 U_p$$

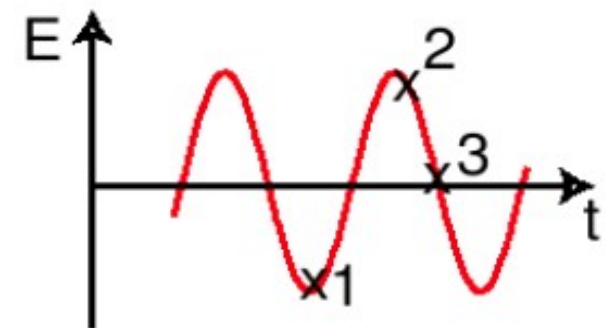
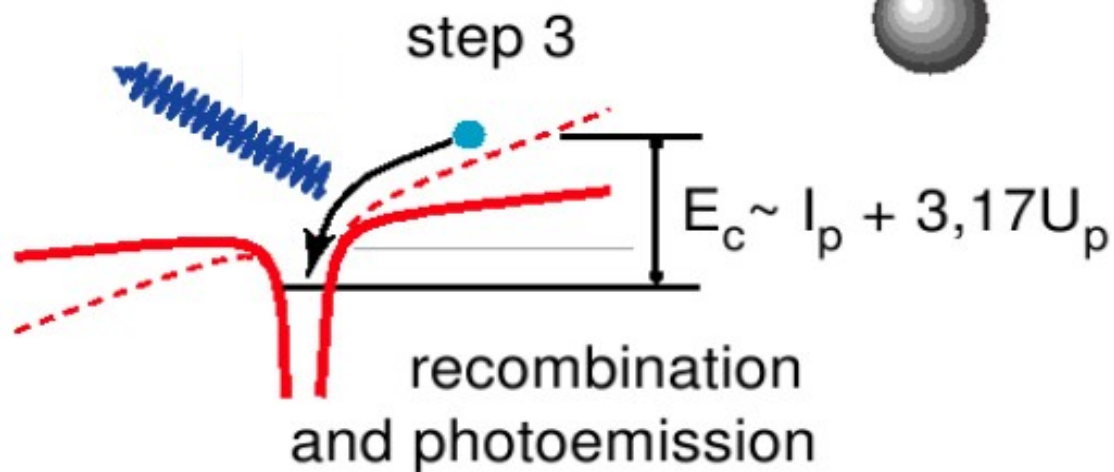
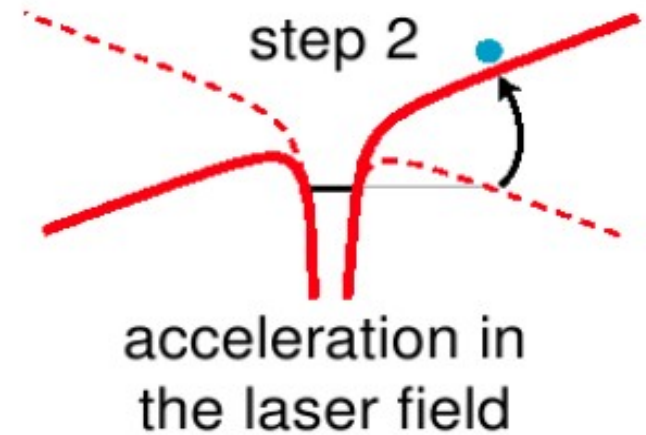
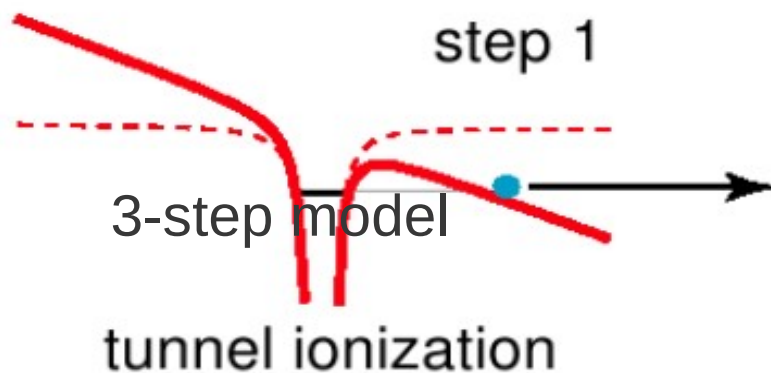
$$U_p = \frac{e^2 E^2}{4 m \omega}$$

Ponderomotive potential



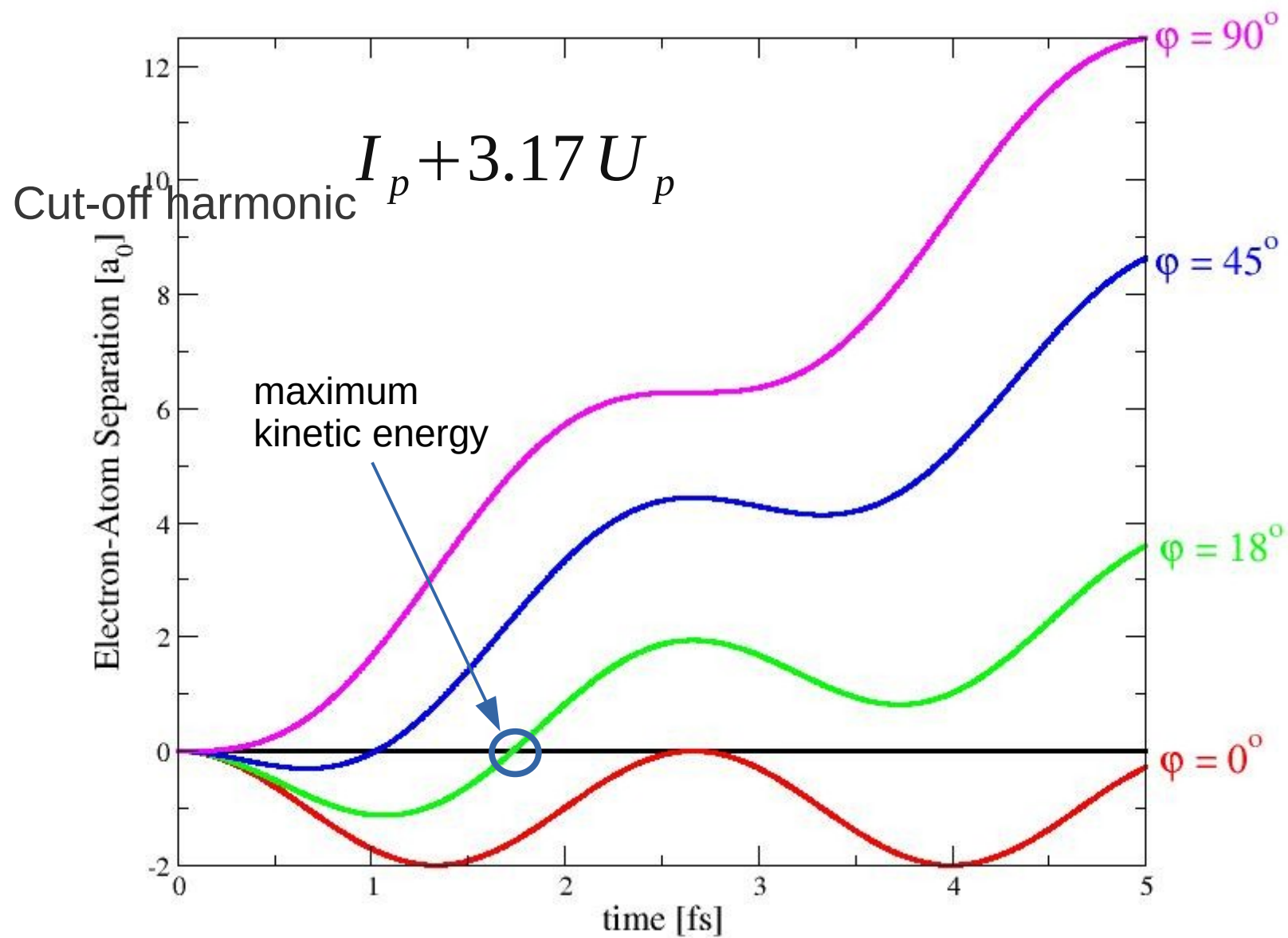
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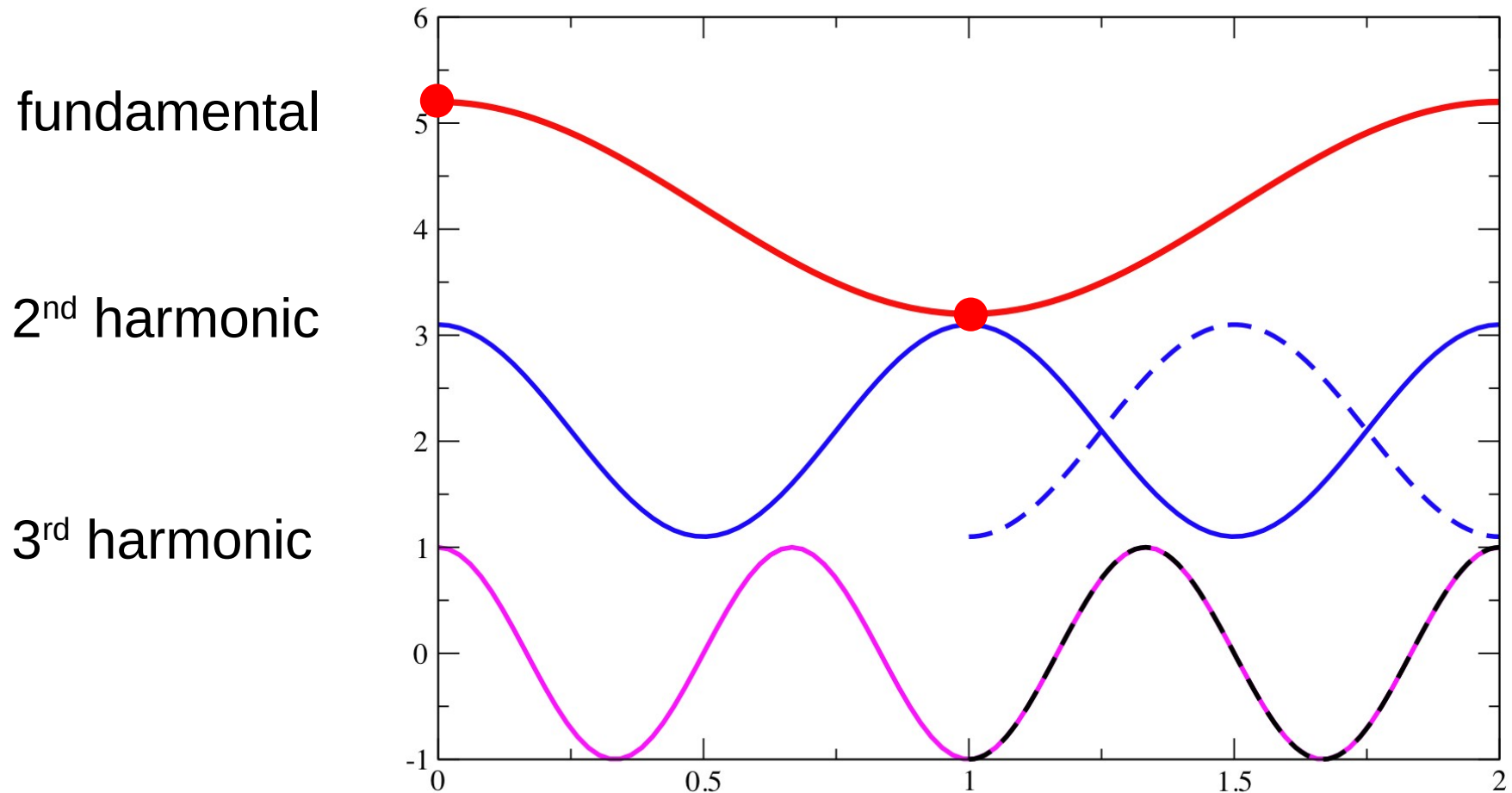


Corkum, Kulander



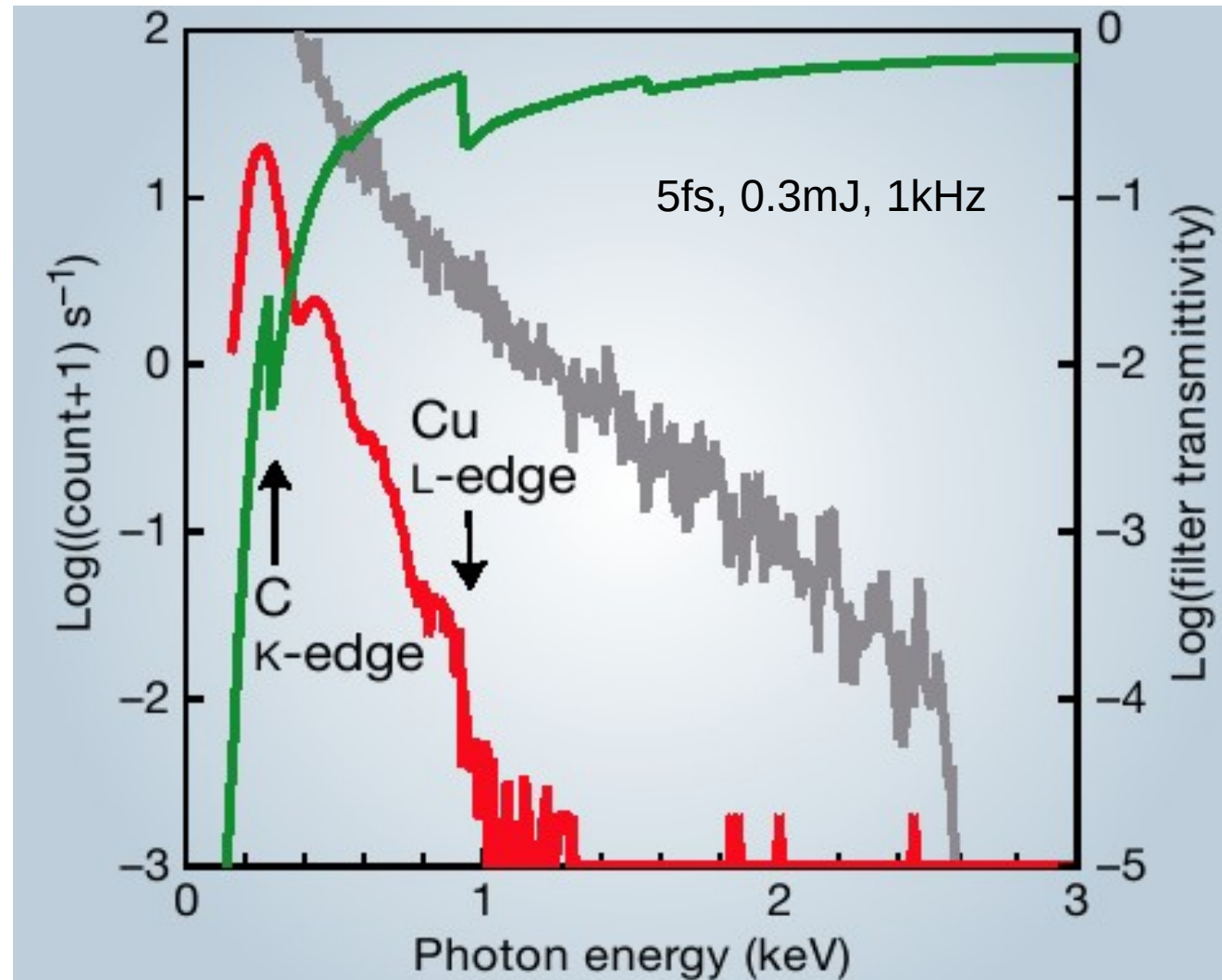


Only odd harmonics are emitted.
Even harmonics from two half-cycles interfere destructively



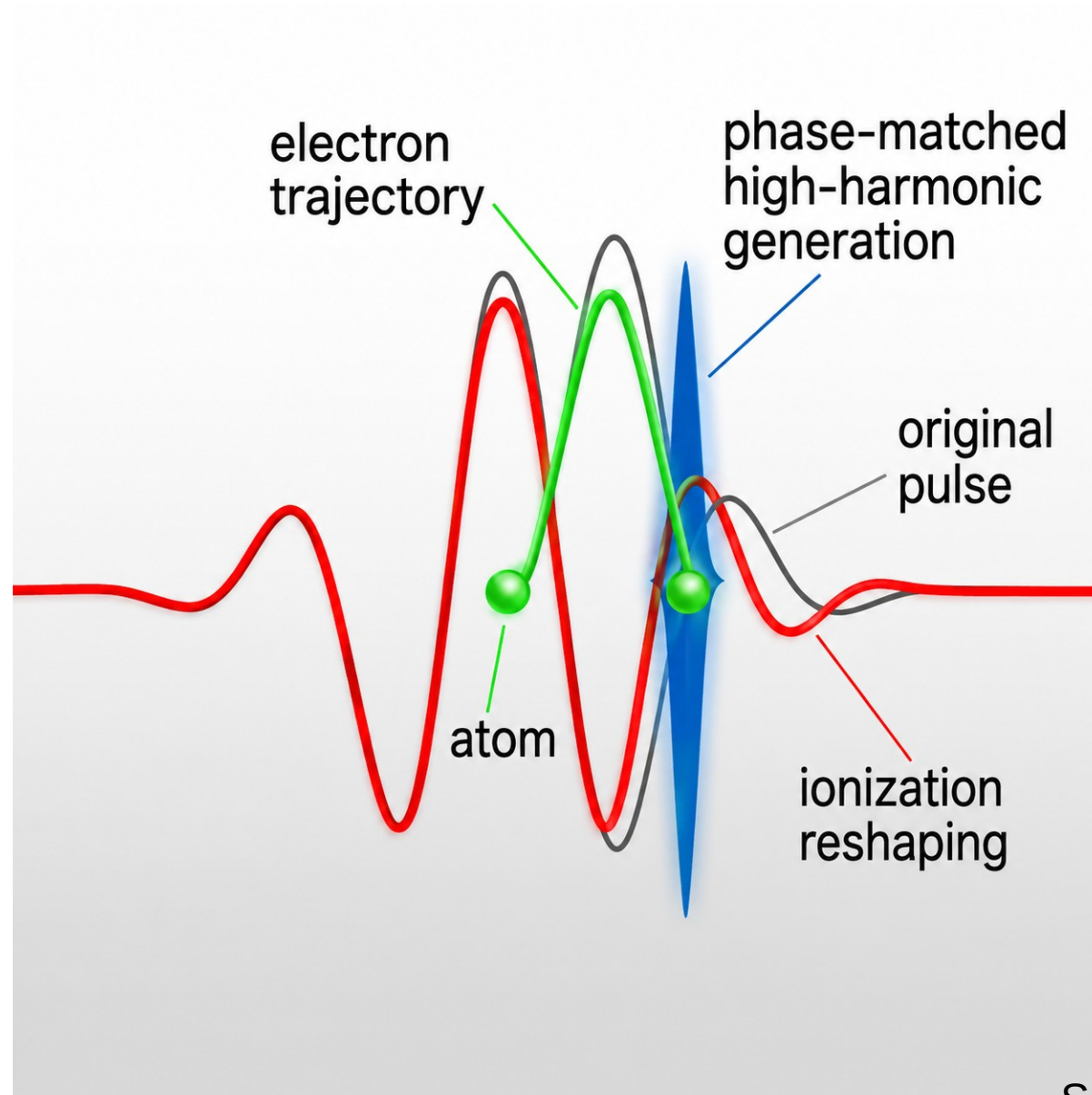
Only odd harmonics are generated! Phase matching is important!

Energies up to 2.5 keV! $N > 1500$!



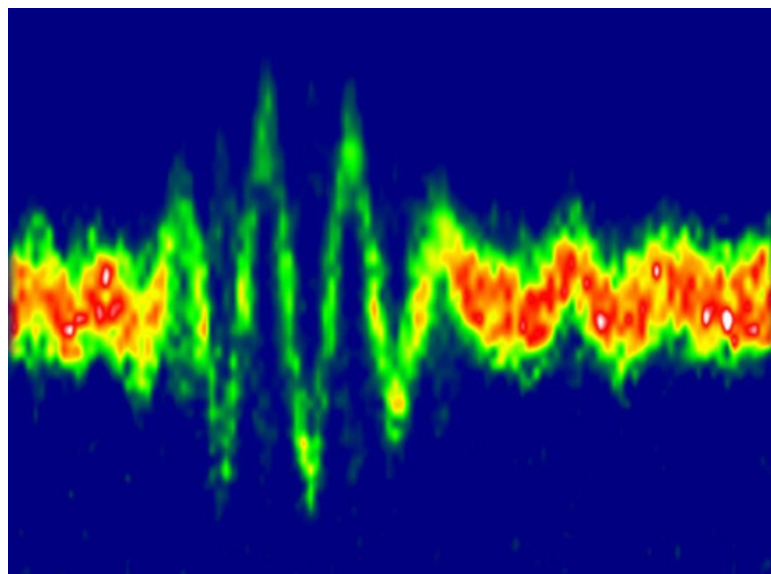
J. Seres et al, Nature 433, 596 (2006)

Attosecond pulse generation



Schötz et al (2020)

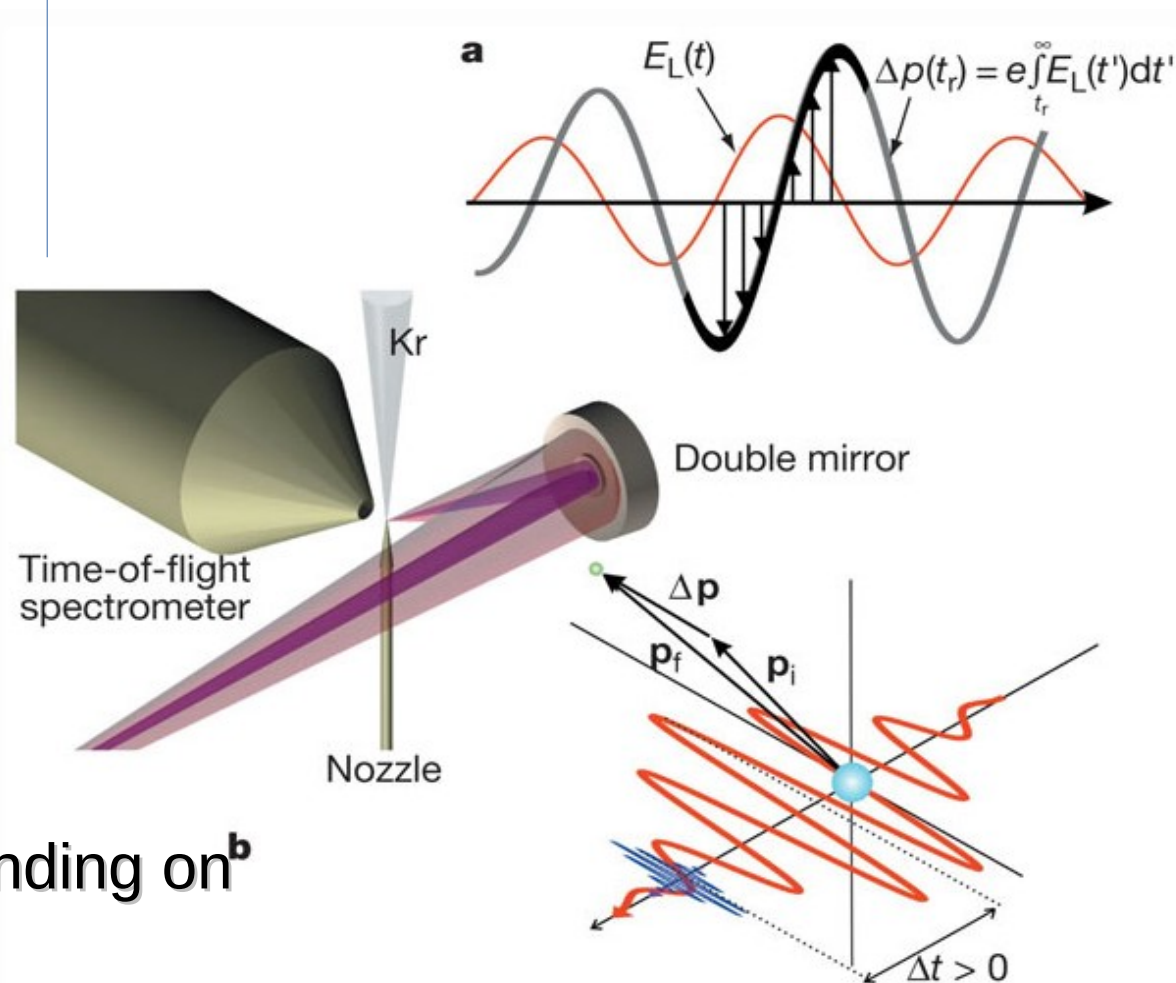




Time

Energy

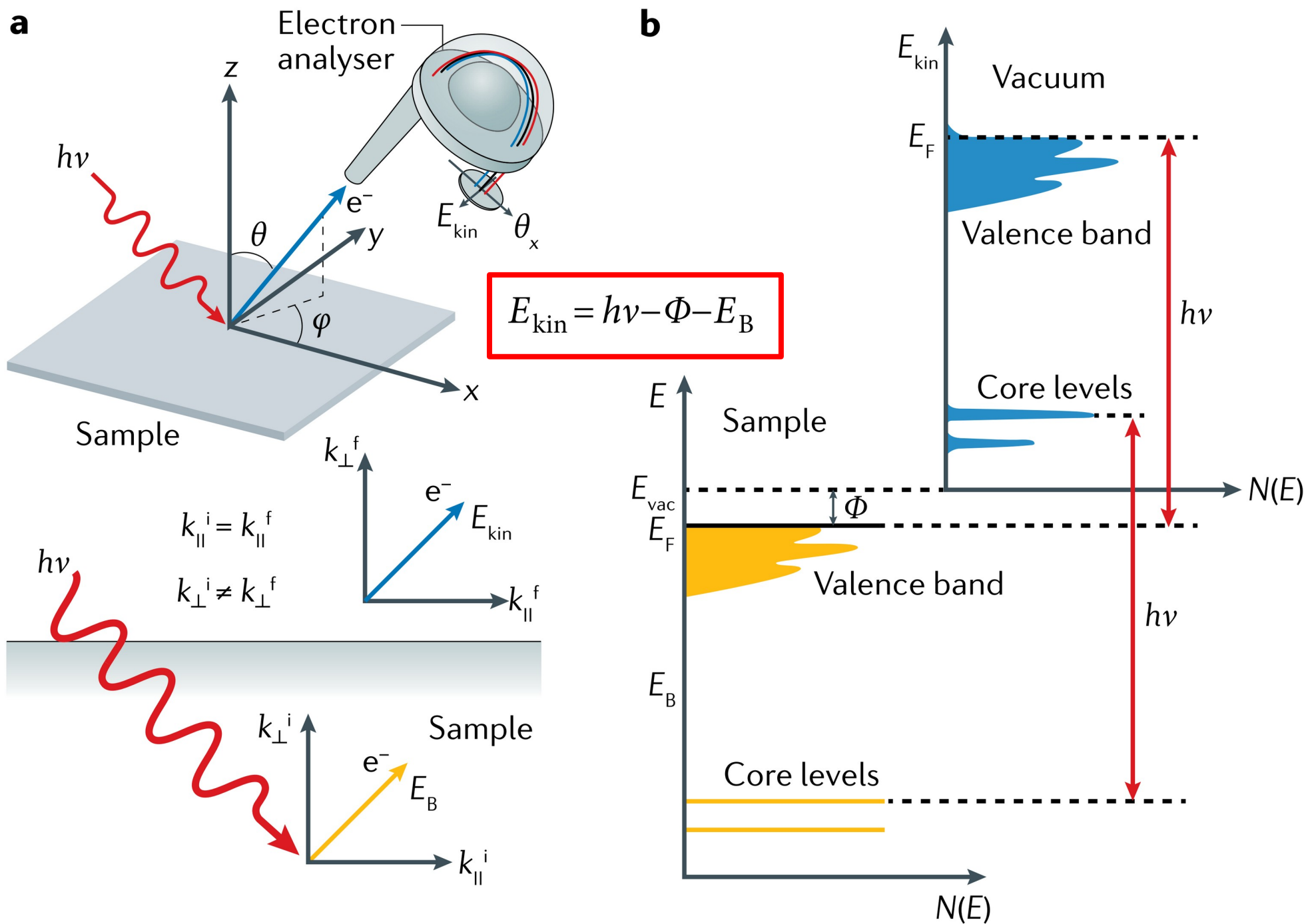
Electrons gain energy depending on^b momentary IR field.



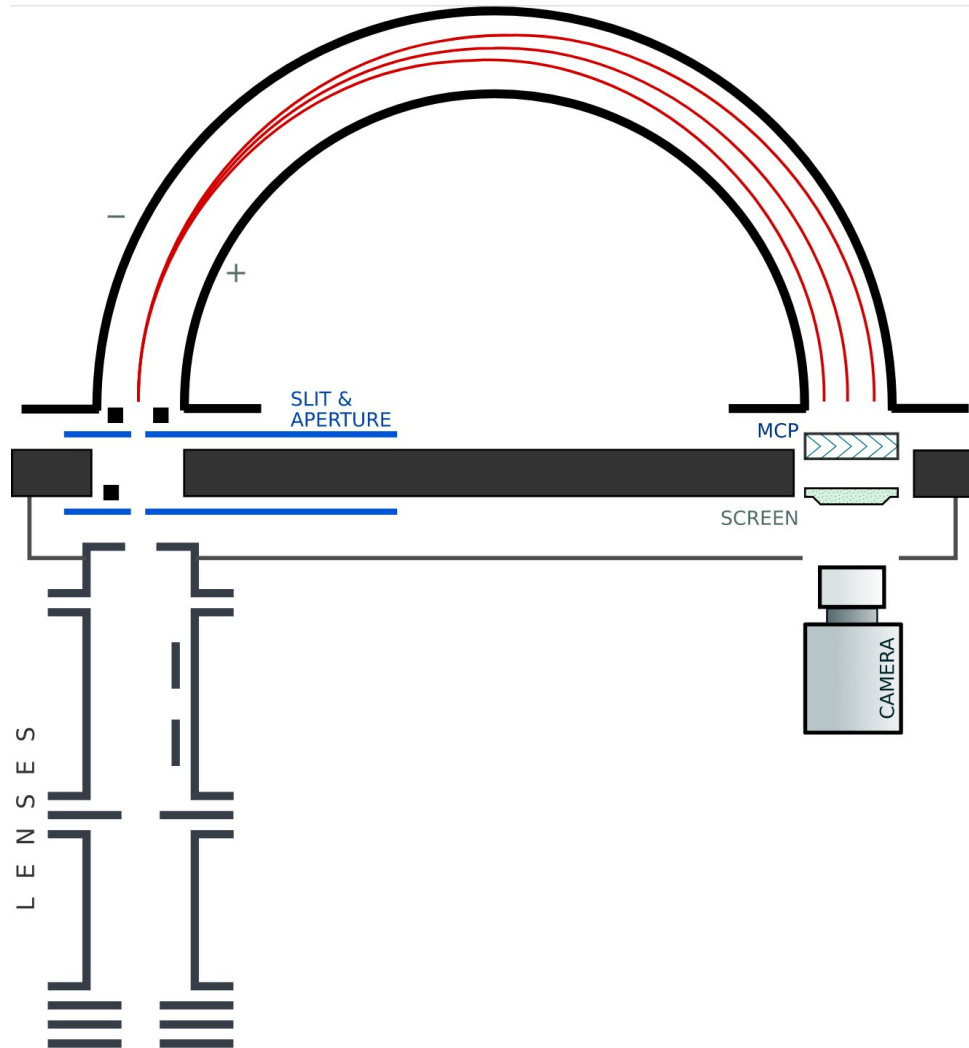
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Angle-Resolved PhotoEmission Spectroscopy



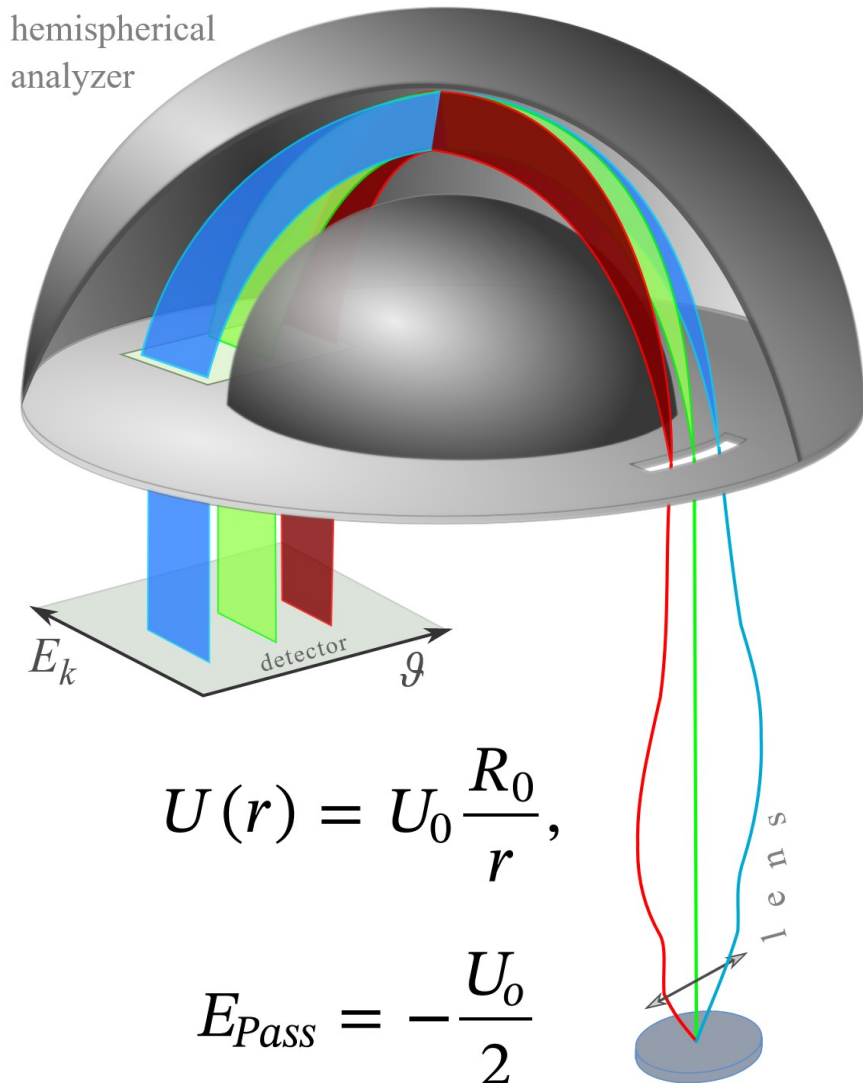
Hemispherical analyzer



$$\hbar k_{\parallel}^f = \hbar k_{\parallel}^i \quad \text{Momentum conversion}$$

$$= \sqrt{2mE_{\text{kin}}} \left(\sin\theta \cos\varphi \hat{k}_x + \sin\theta \sin\varphi \hat{k}_y \right)$$

hemispherical
analyzer



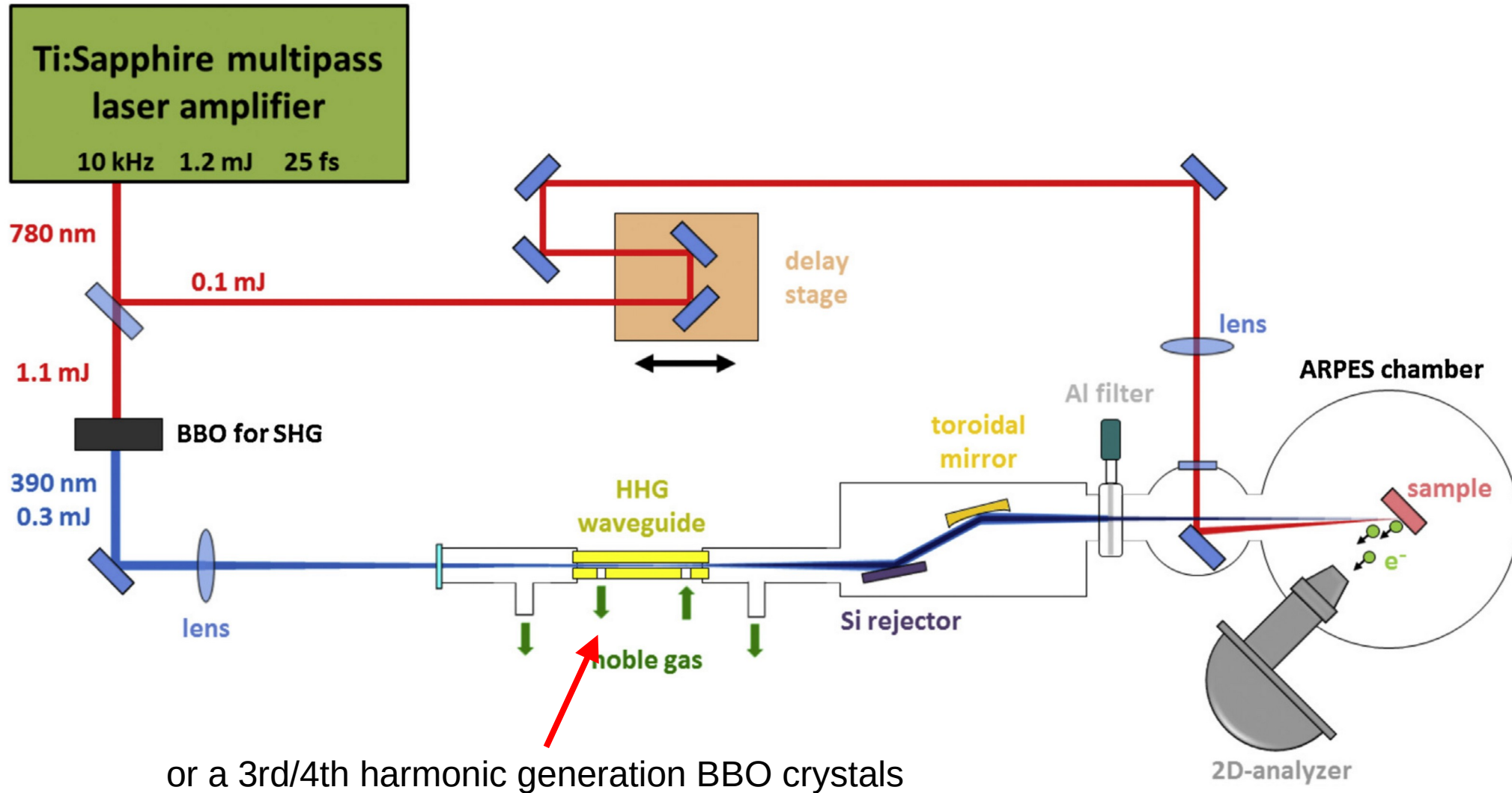
$$U(r) = U_0 \frac{R_0}{r},$$

$$E_{Pass} = -\frac{U_0}{2}$$

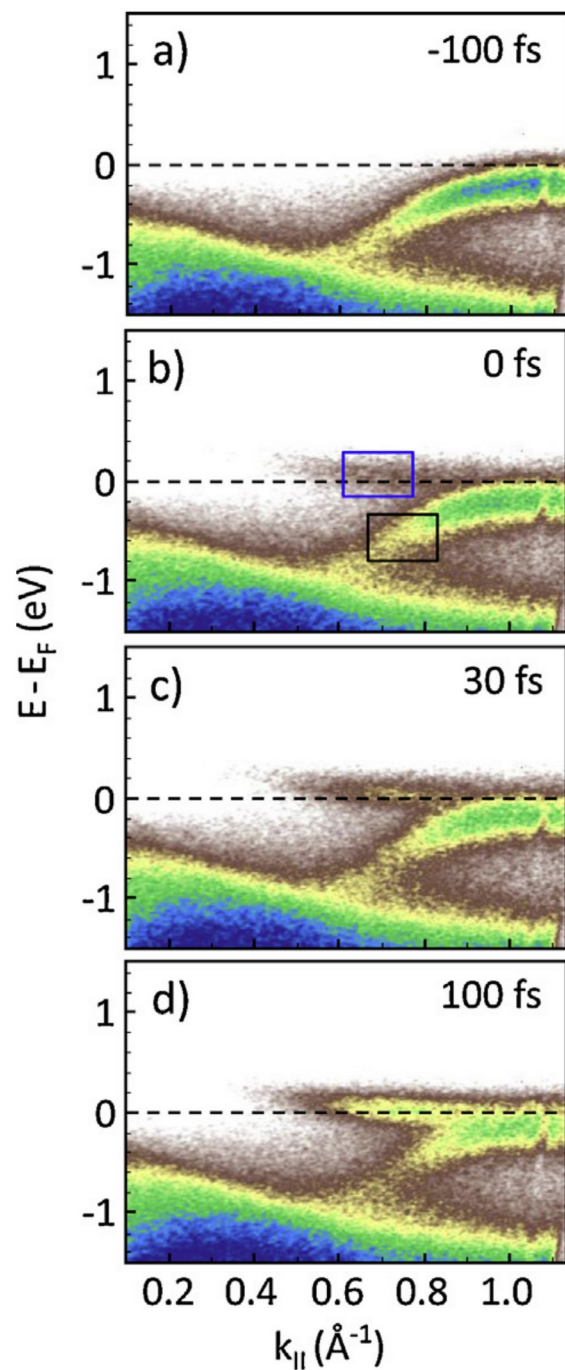
Lens focus is at the sample: emission angles are converted to x-position at the detector. deviations are small and approximately linear.

Bias voltage helps to choose energy range

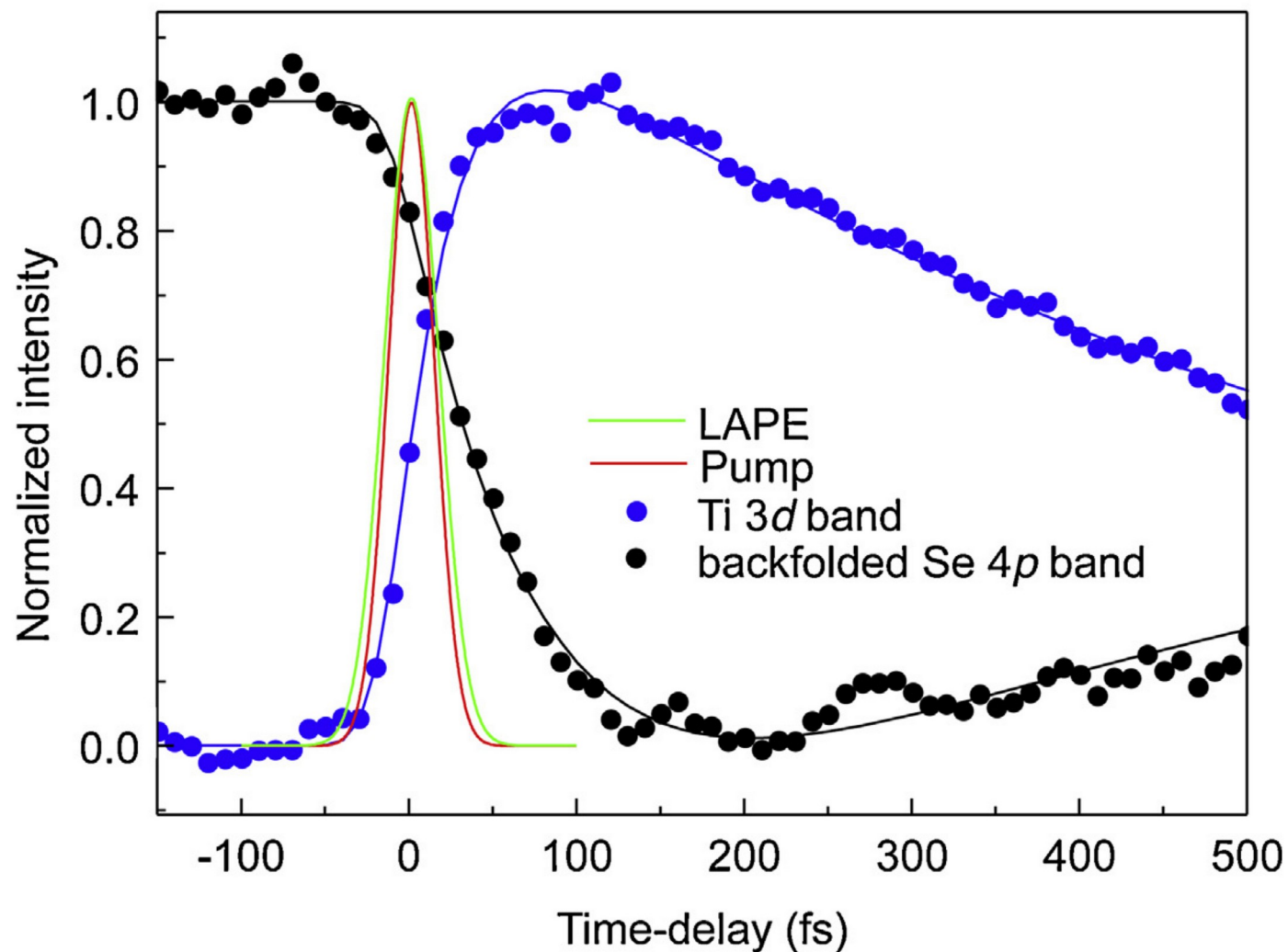
HHG-based ARPES



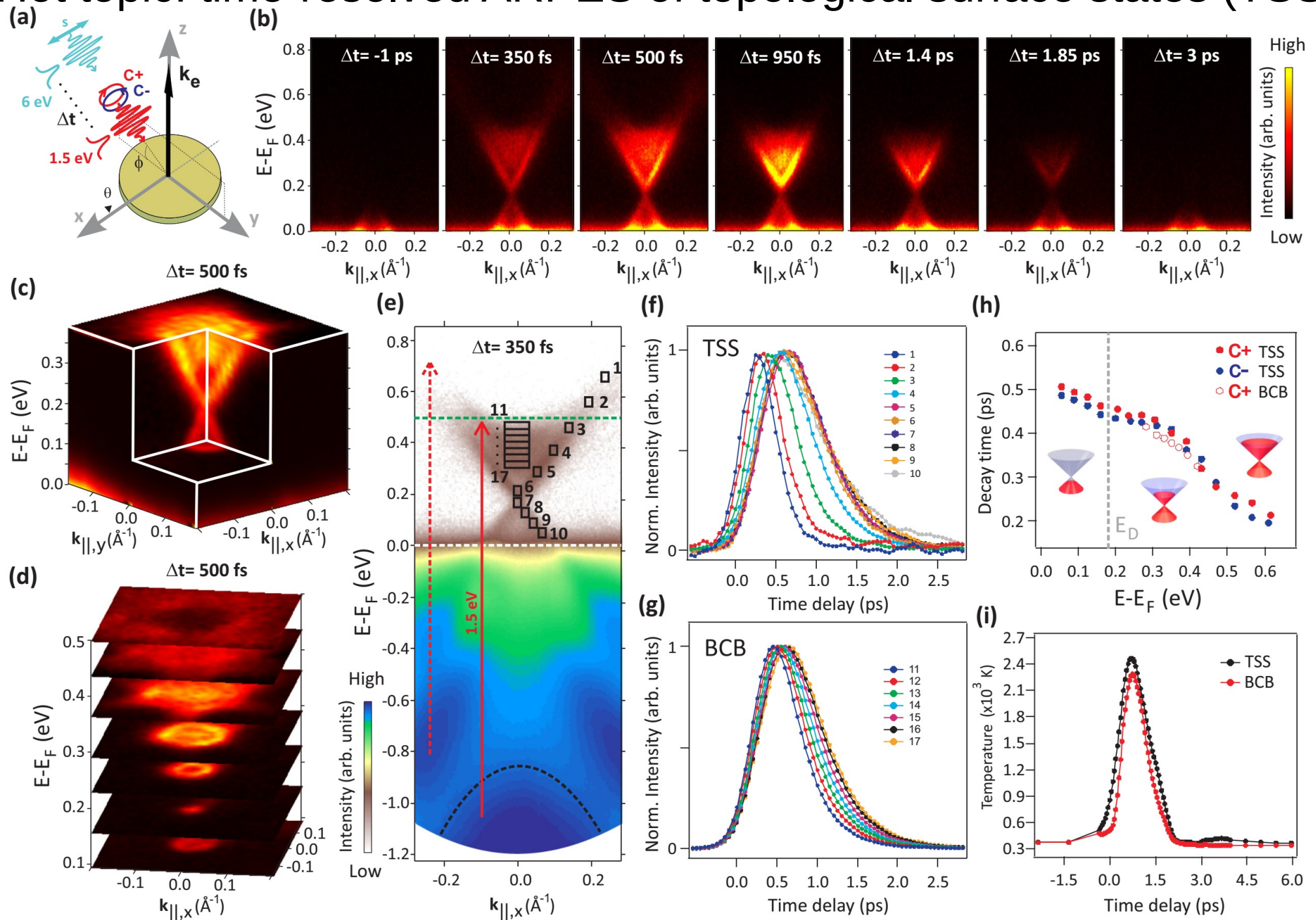
Ti:Se2 dynamics



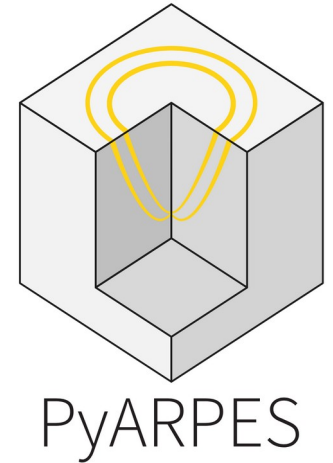
e)



Hot topic: time-resolved ARPES of topological surface states (TSS)



ARPES software: PyARPES



Applying corrections to ARPES data

Doing gap analysis

Performing sophisticated band analysis

Performing rapid and automated curve fitting, even over several dataset dimensions

Background subtraction

Dataset collation and combination

Producing common types of ARPES figures and reference figures

Converting to momentum space

Interactively masking, selecting, laying fits, and exploring data

Plotting data onto Brillouin zones

<http://arpes.netlify.app>