Introduction to ultrafast lasers HHG, and relevant data analysis

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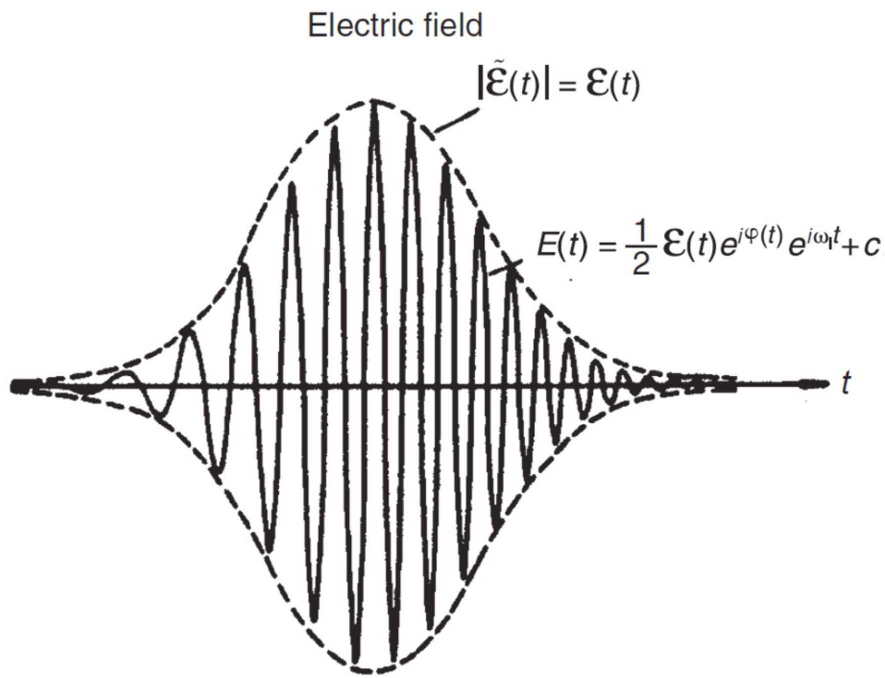
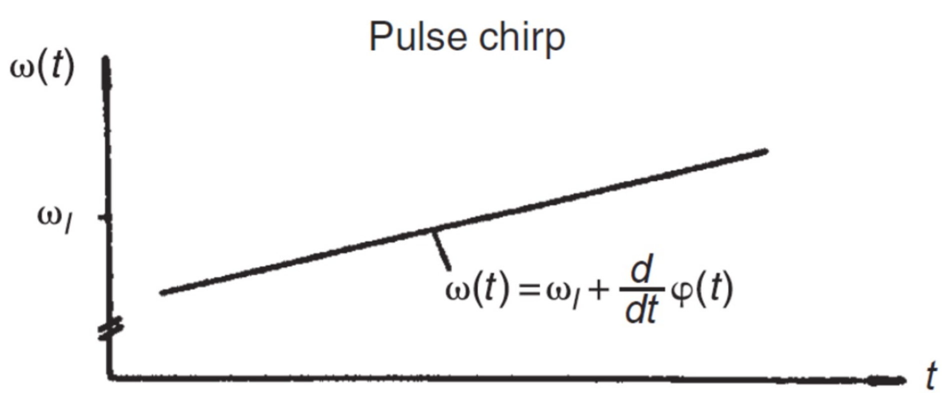
Basic ultrafast laser concepts



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:

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

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

Alternative description using only positive frequencies

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

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	$\mathrm{sech}^2(t/\tau_s)$	$1.763\tau_s$	$\mathrm{sech}^2 \frac{\pi\Omega\tau_s}{2}$	$1.122/\tau_s$	0.315	0.525
Lorentz	$[1+(t/\tau_L)^2]^{-2}$	$1.287\tau_L$	$e^{-2\vert\Omega\vert\tau_L}$	$0.693/\tau_L$	0.142	0.7
Asym. sech	$\left[e^{t/\tau_a} + e^{-3t/\tau_a}\right]^{-2}$	$1.043\tau_a$	$\mathrm{sech}\frac{\pi\Omega\tau_a}{2}$	$1.677/\tau_a$	0.278	
Square	1 for $\vert t/\tau_r\vert \leq 1$, 0 elsewhere	τ_r	$\mathrm{sinc}^2(\Omega\tau_r)$	$2.78/\tau_r$	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 φ(t) can lead to pulses longer than the Fourier-limit

carrier envelope phase **instantaneous frequency** **linear chirp** **N.B. positive chirp means dω/dt > 0**

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

group delay **group delay dispersion (GDD)**

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

N.B. positive GDD; red before blue

Phase velocity and group velocity

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

laser frequency

laser wavevector

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

Propagation through a medium with refractive index n(ω)

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

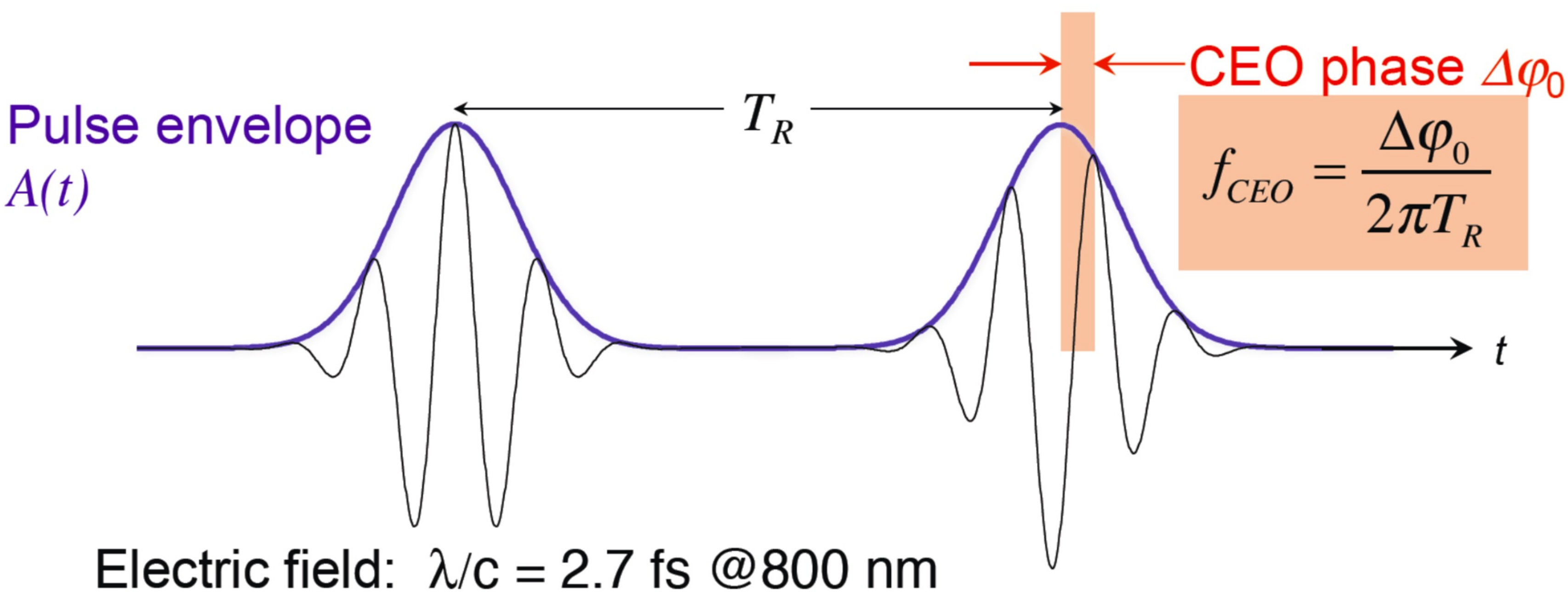
$$v_{group} = v_{phase}/(1 + \frac{\omega}{n}\frac{dn}{d\omega})$$

The phase and group velocity differ in a dispersive medium

Carrier-Envelope Phase

carrier envelope phase

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



Nobel Prize 2018

Gérard Mourou

Donna Strickland

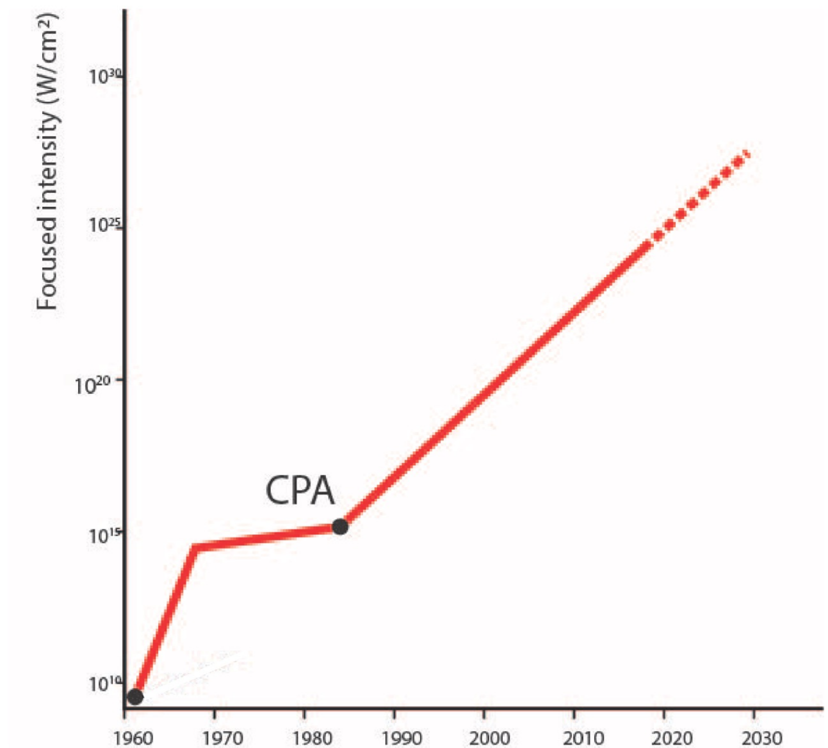
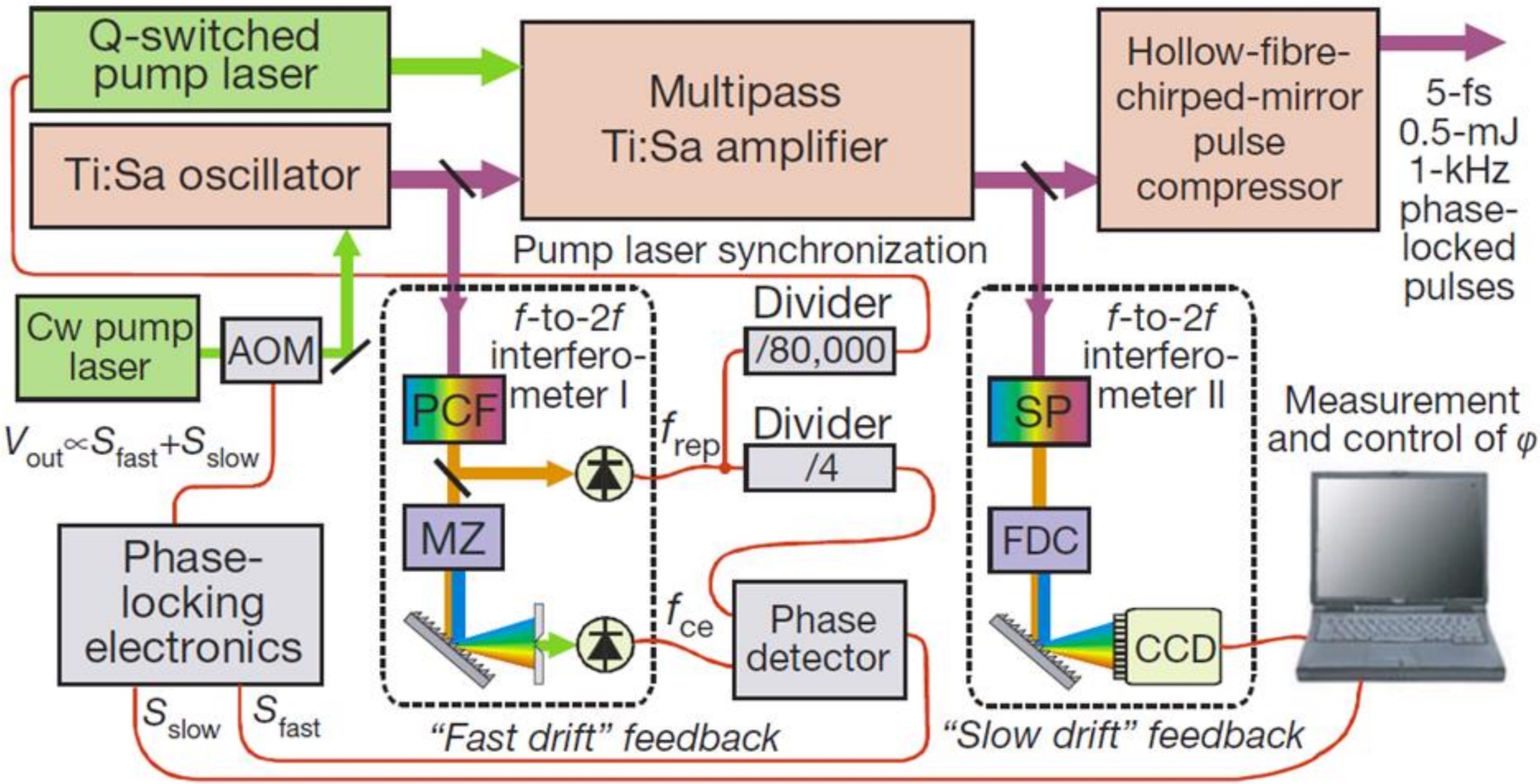


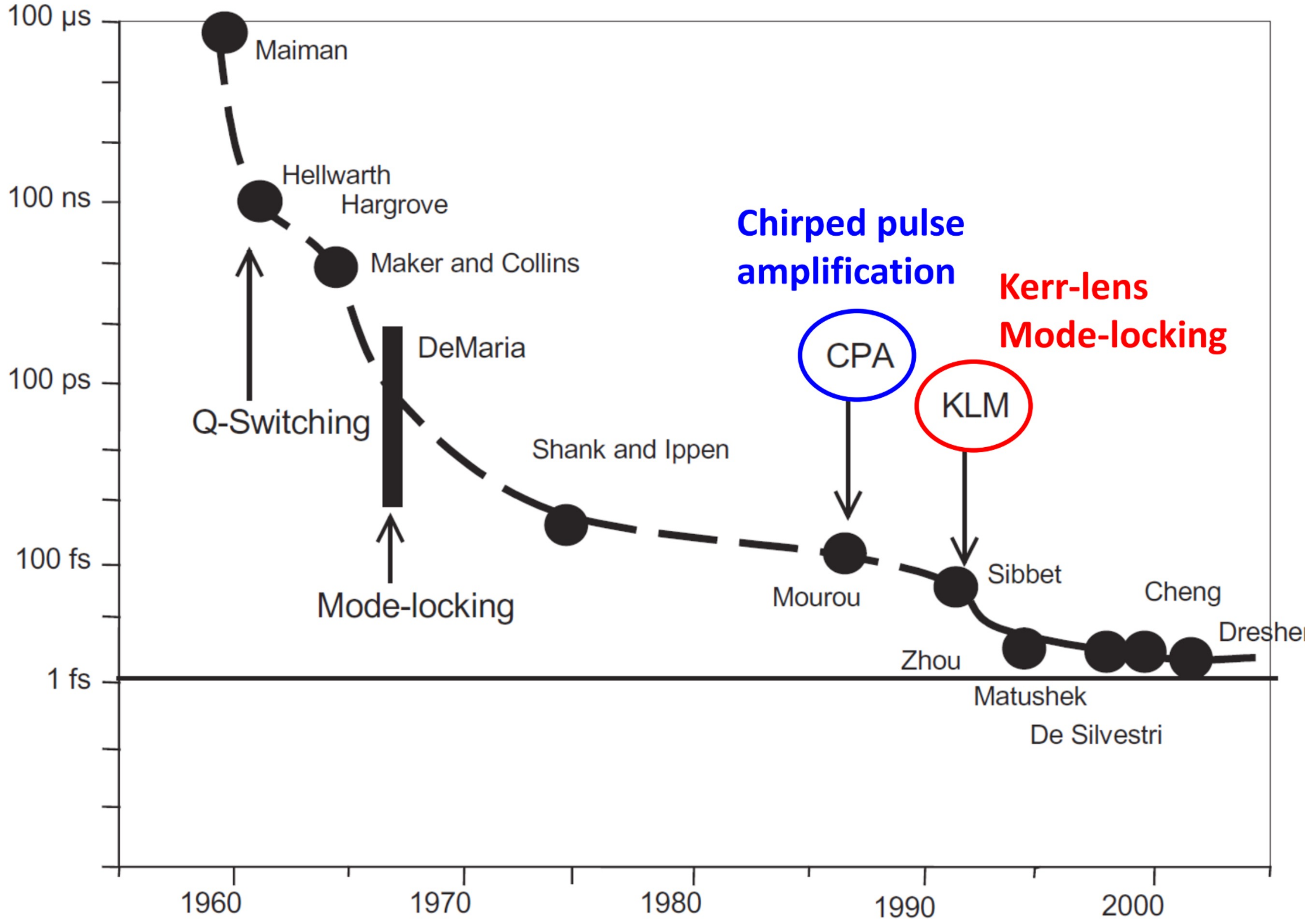
***Figure 3**. The history of pulsed laser technology in terms of peak intensity. After an initial increase in intensity during the 1960s, as described in the text, only an incremental increase occurred during more than a decade. The invention of the CPA technique for optical pulses by Strickland and 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.*

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

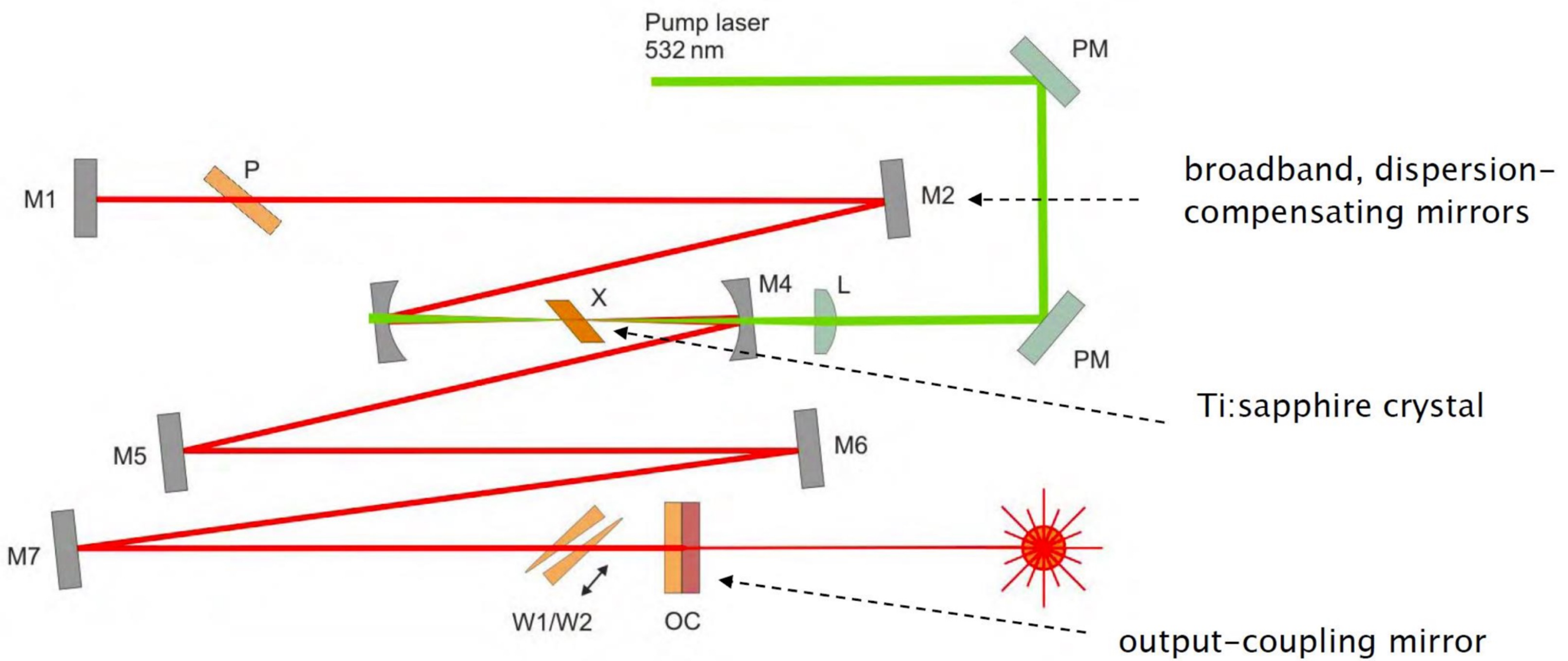


A. Baltuska et al., Nature 421, 611 (2003)

Basic operation of a mode-locked Ti:Sapphire oscillator

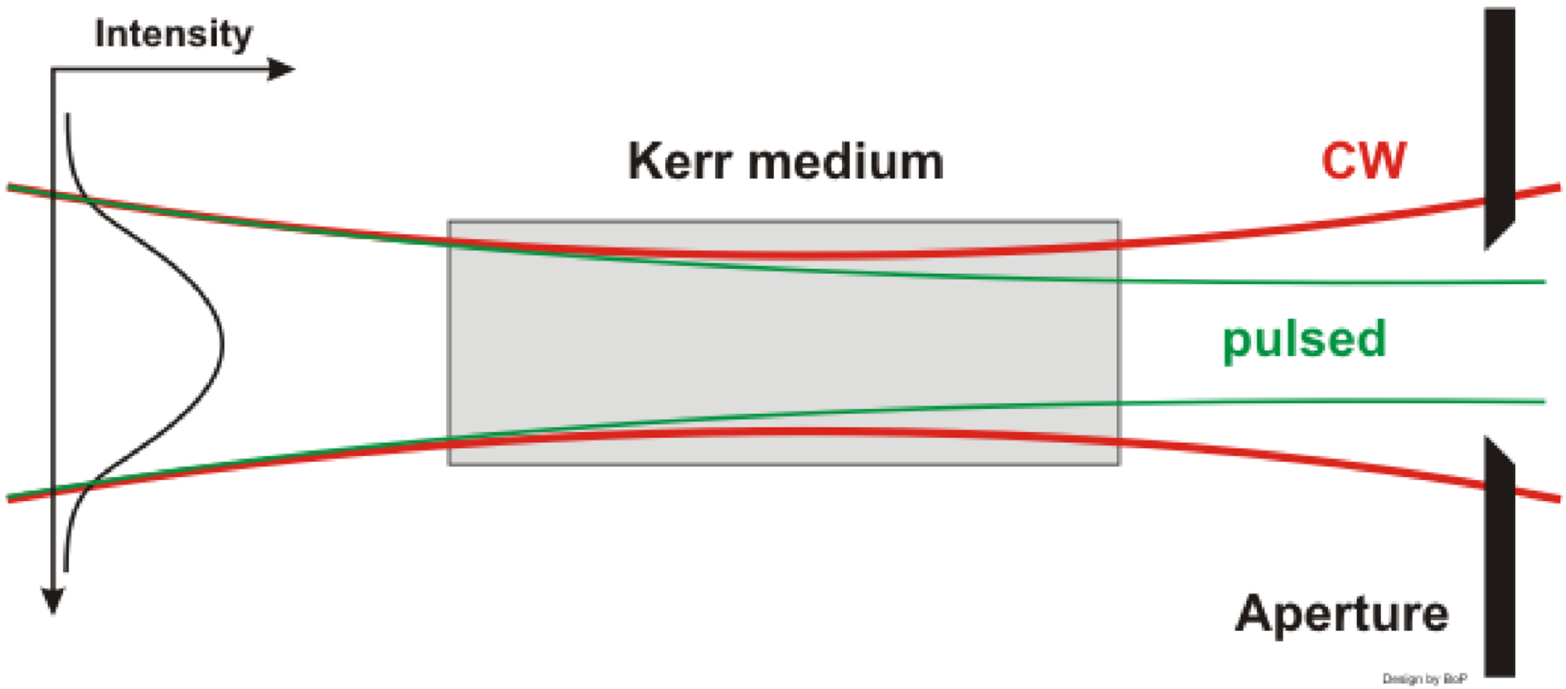


Basic operation of a mode-locked Ti:Sapphire oscillator



S. Rausch et al., Opt. Expr. 16, 9739 (2008)

Kerr-lens mode-locking



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

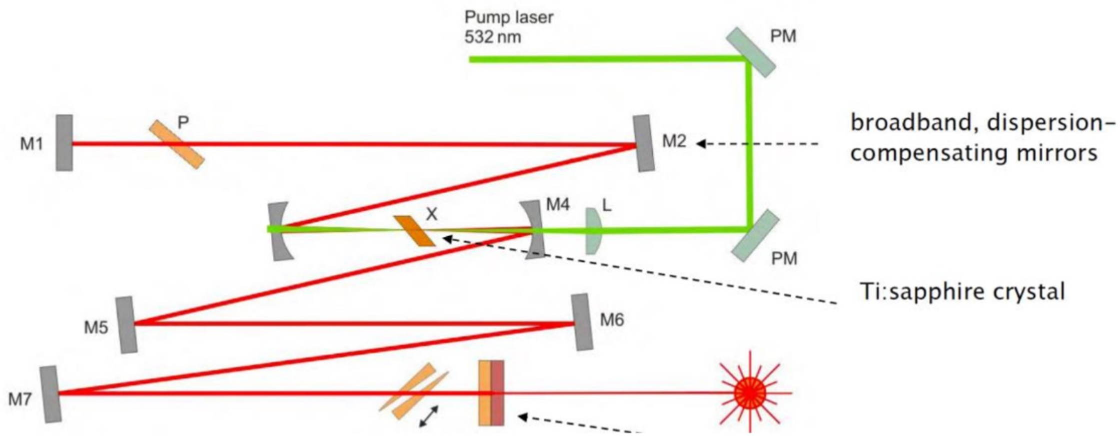
$$n(I) = n_0 + n_2 I$$

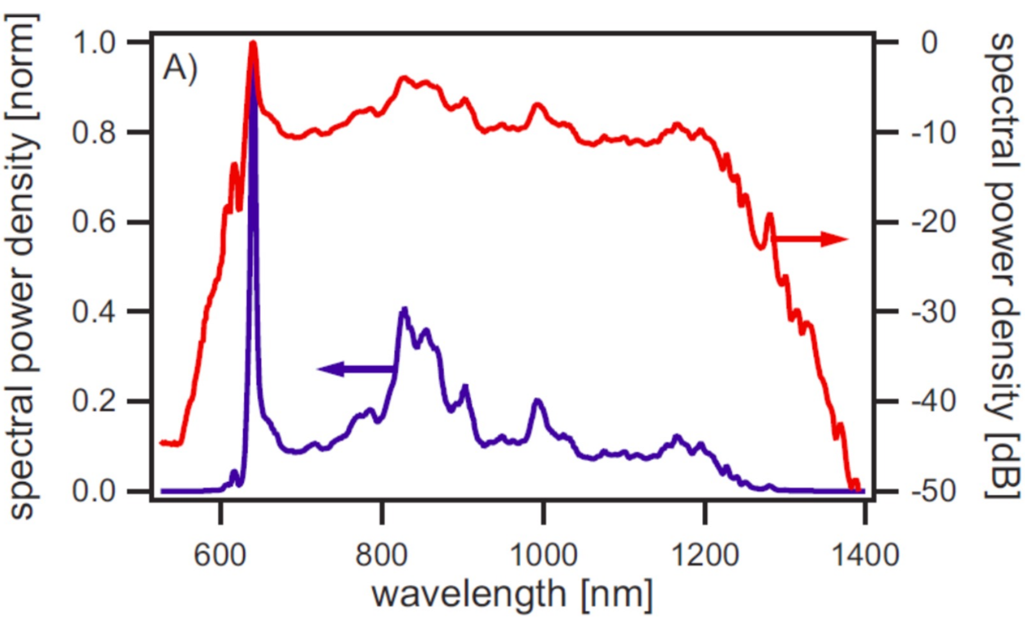
Focal length of the Kerr lens $f = \dfrac{w_0^2}{4n_2 d I_0}$

w_0 = beam radius
d = thickness

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

Example of an octave-spanning oscillator

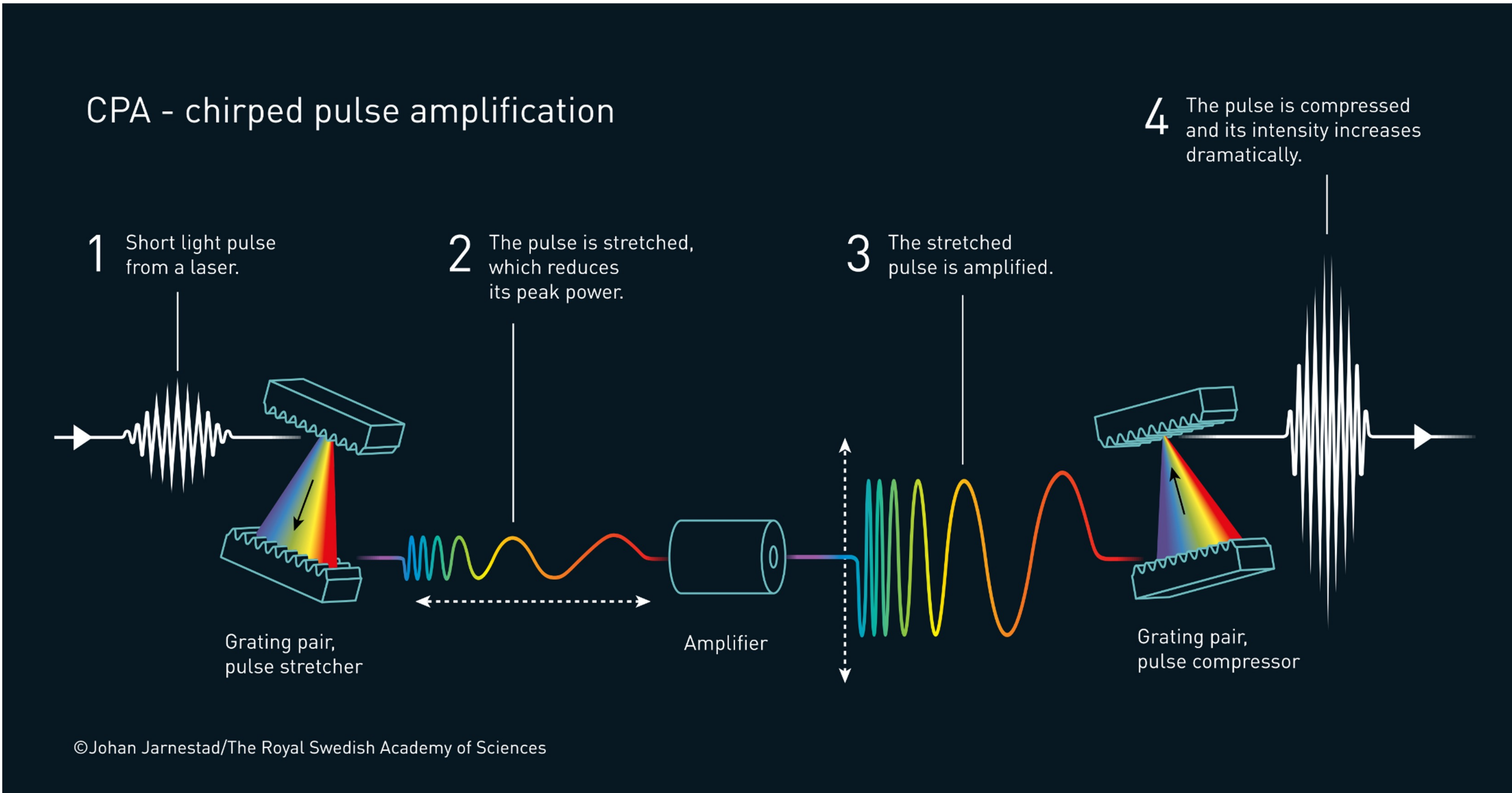




Using an octave-spanning oscillator 4-fs pulses can be generated and the stage is set for stabilization of the carrier envelope phase

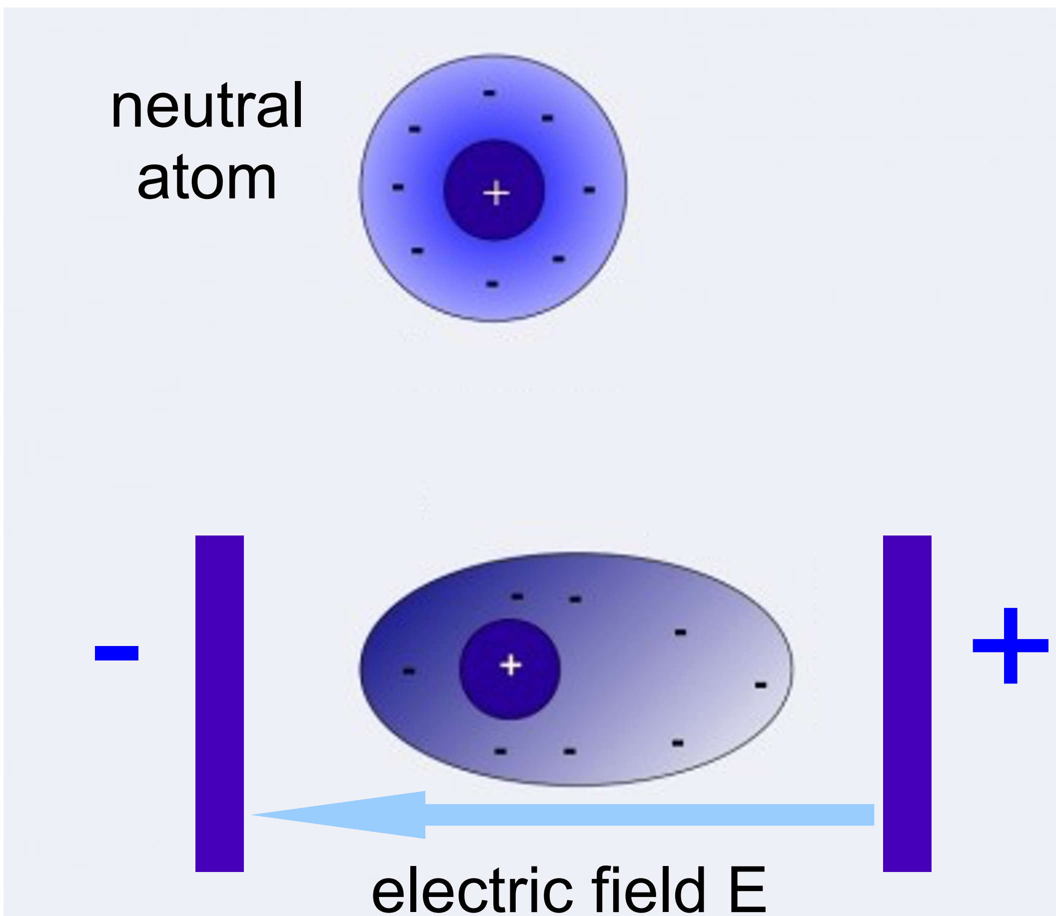
S. Rausch et al., Opt. Expr. 16, 9739 (2008)

Pulse energy: Chirped pulse amplification (CPA)



High-harmonic generation
and
production of attosecond pulses

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



P - polarization

E - electric field

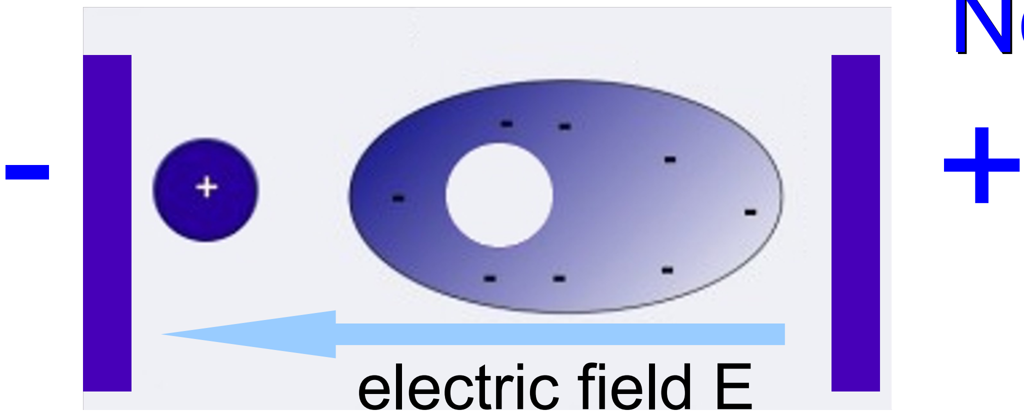
χ - susceptibility

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

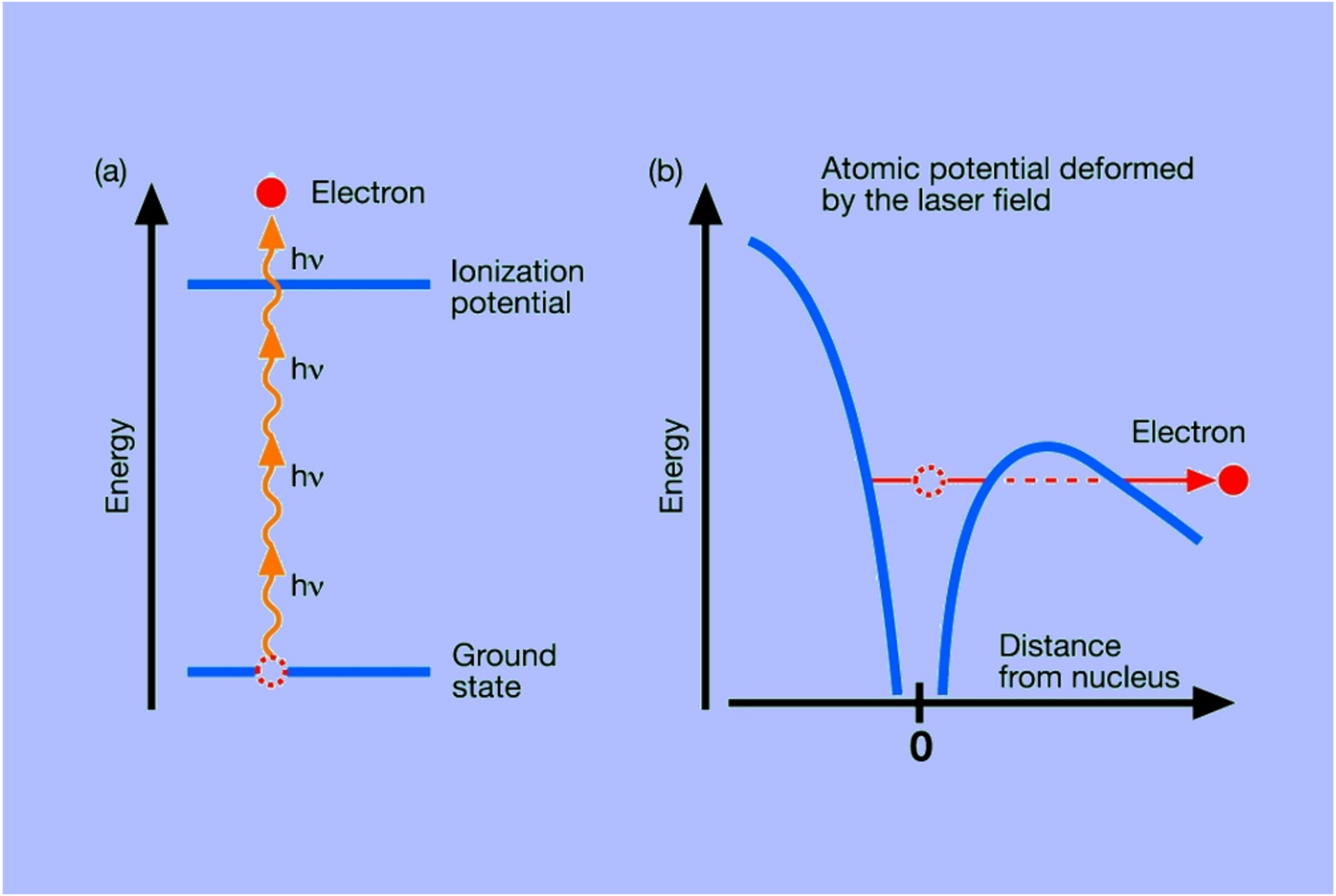
Contributions from high orders are small, typical nonlinear optics.

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

The series is divergent!
Not a valid description.



VERY nonlinear optics – STRONG FIELD!


(a)
Electron
hν
Ionization
potential
hν
hν
hν
Ground
state
Energy
(b)
Atomic potential deformed
by the laser field
Energy
Electron
Distance
from nucleus
0

(Keldysh parameter)

momentum in the field of ion

1/time force of electric field

⍰ - electric field frequency
E – electric field amplitude
V_{IP} – ionization potential

(Keldysh parameter)

multiphoton ionization
perturbative

tunneling ionization
non-perturbative

Tunneling ionization:

What laser do we need?

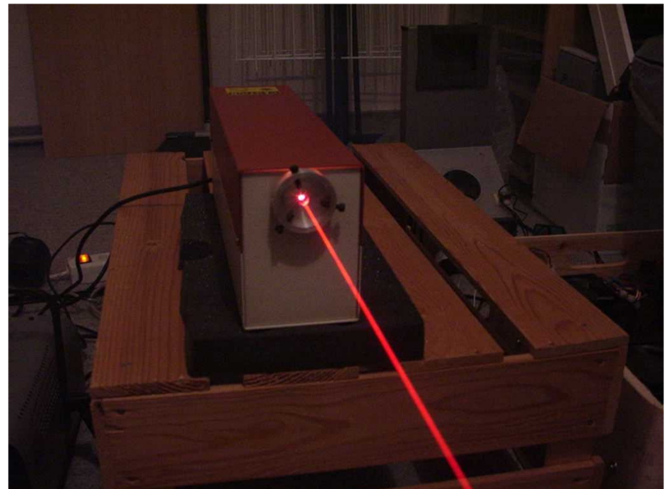
Typical:

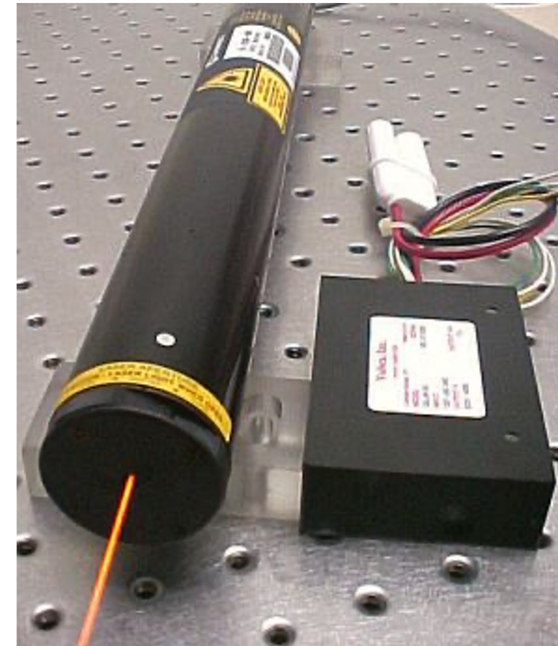
λ=633 nm → ω=3 10^{15} Hz

V_{IP} = 1 Ry = 13.6 eV

E ≈ 10^{11} V/m

Intensity
Poynting vector





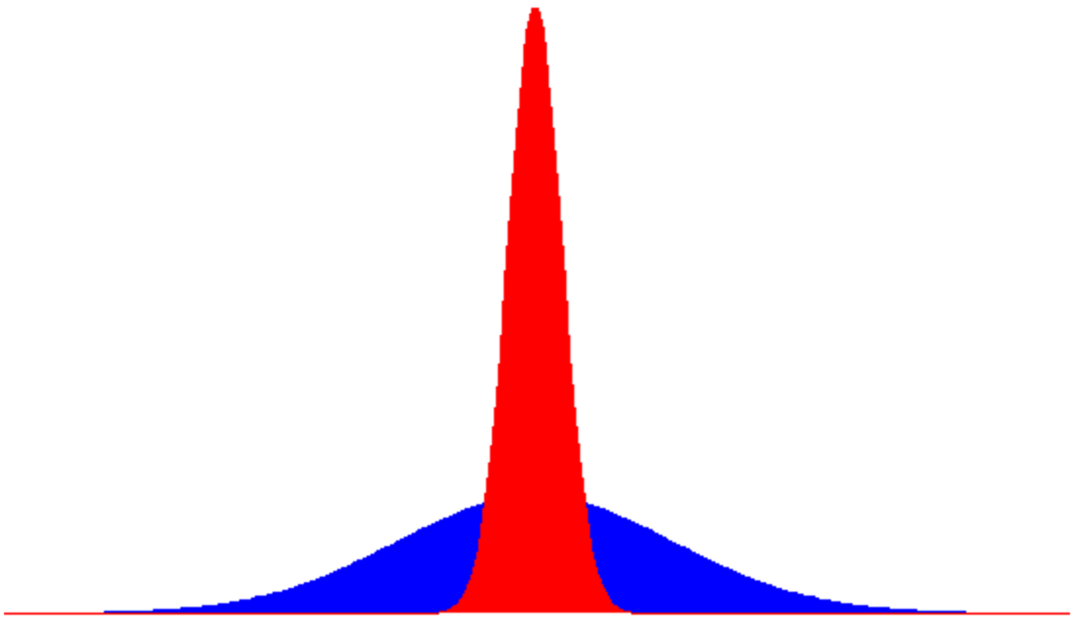
HeNe laser?

Typical:
Power: 5 mW
Focus: 20 μm
Intensity: 1.6×10^7 W/m^2

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

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

Pulsed lasers!

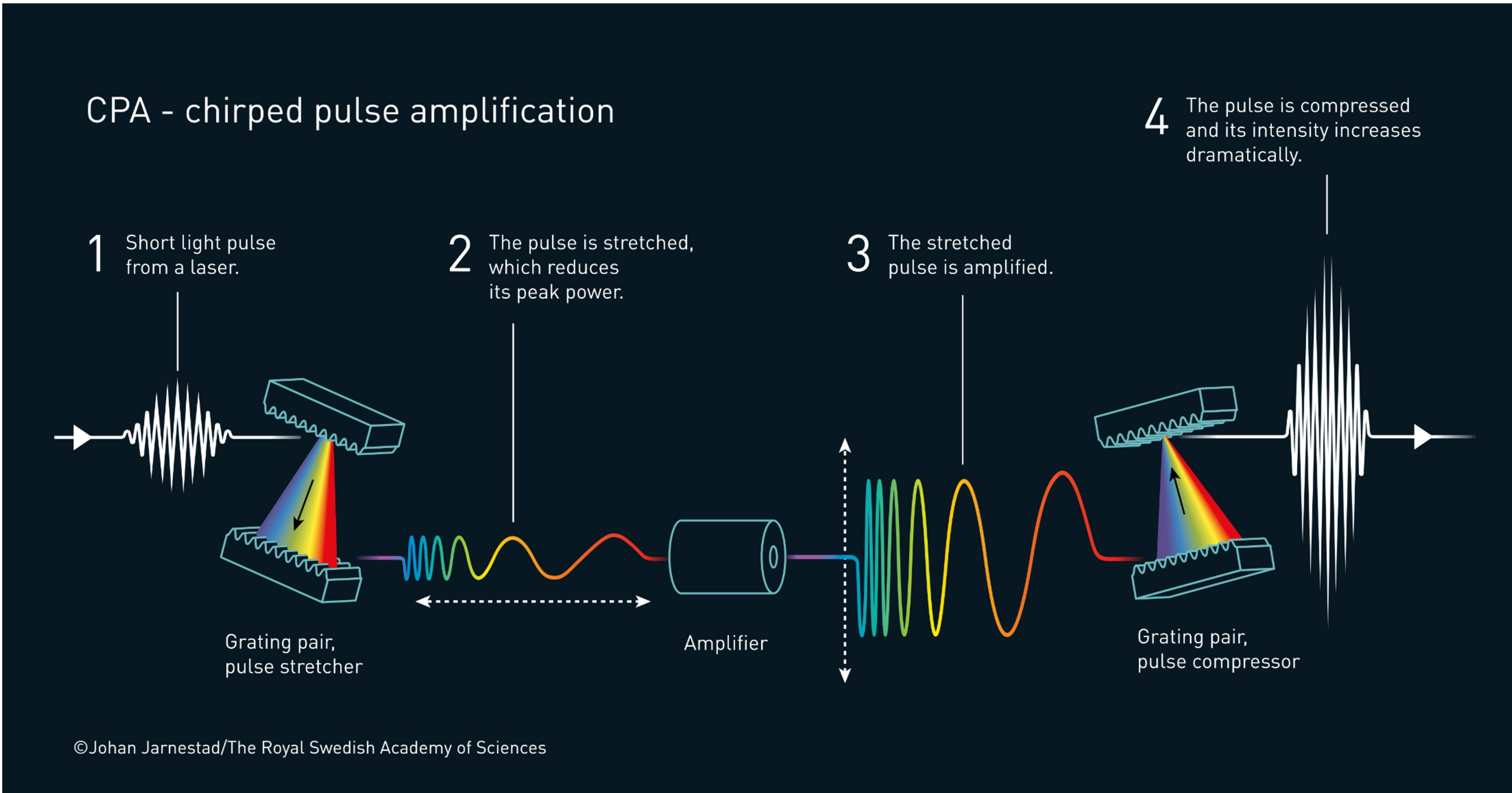


Typical:
Power: 1W
Rep. rate: 1kHz
Pulse energy: 1mJ

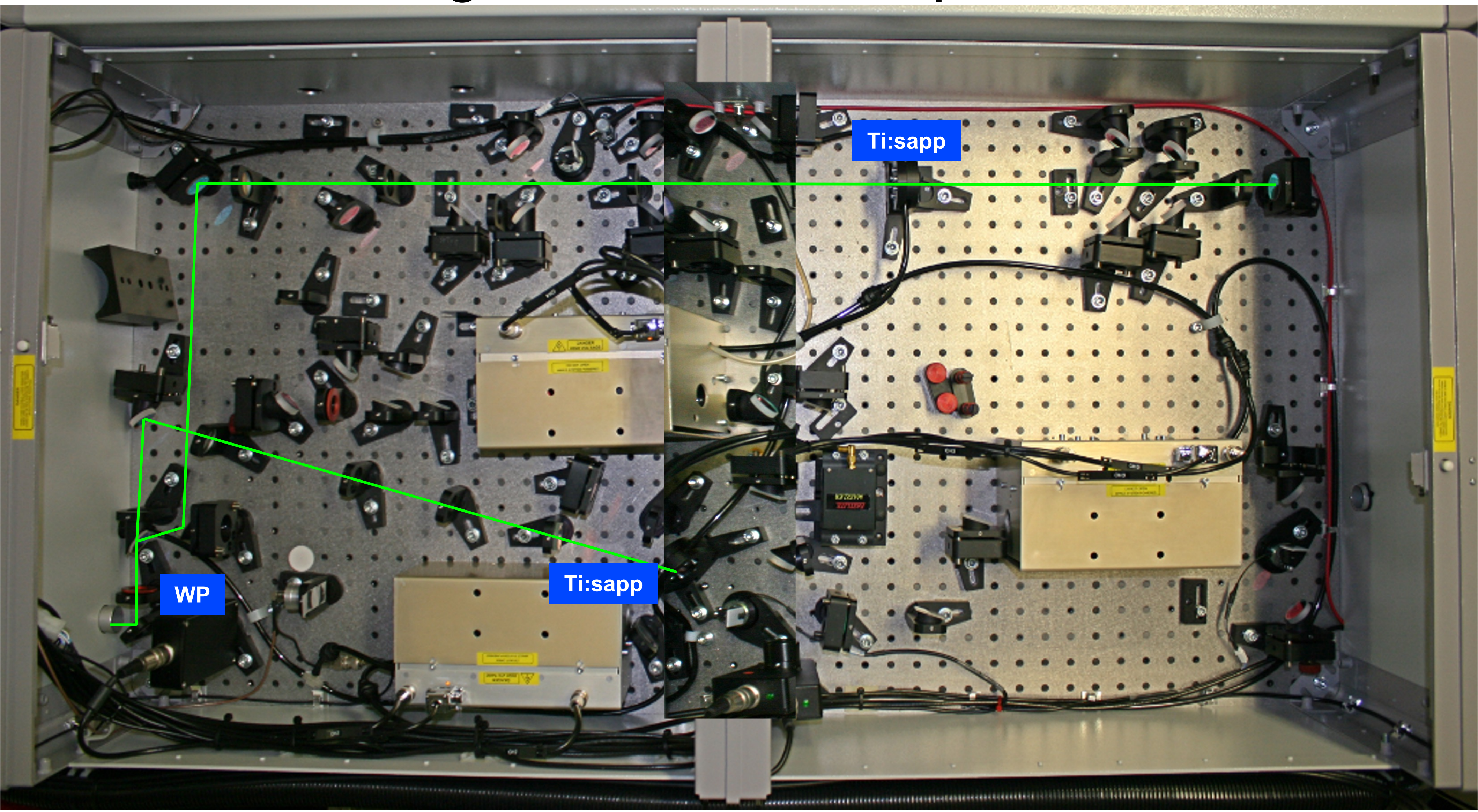
Need pulse duration of 200 fs to get 10^{11} V/m.

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

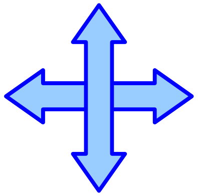
Pulse energy: Chirped pulse amplification (CPA)



regen+booster amplifier



polarization



rep. rate

10 kHz

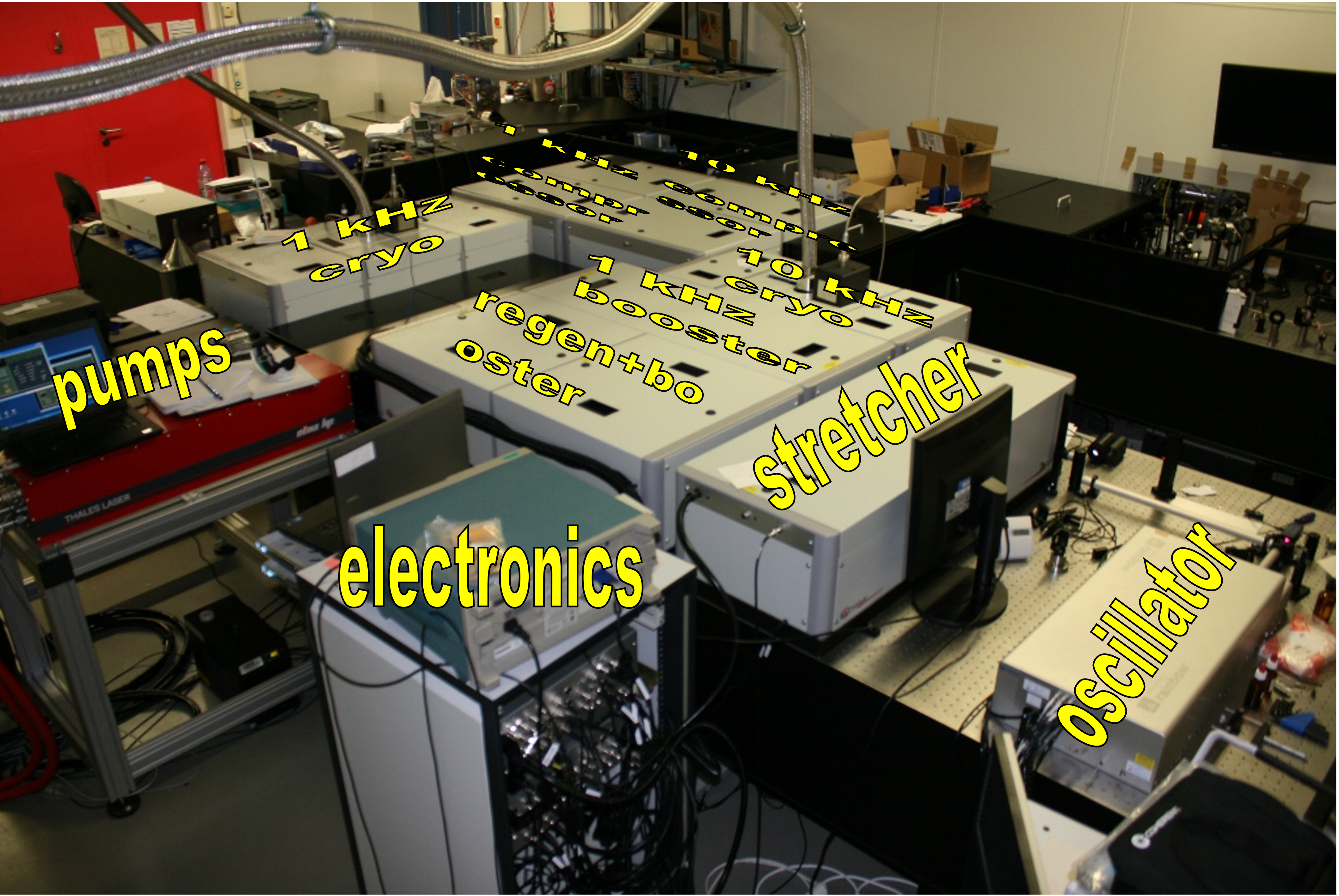
pulse duration

50 ns

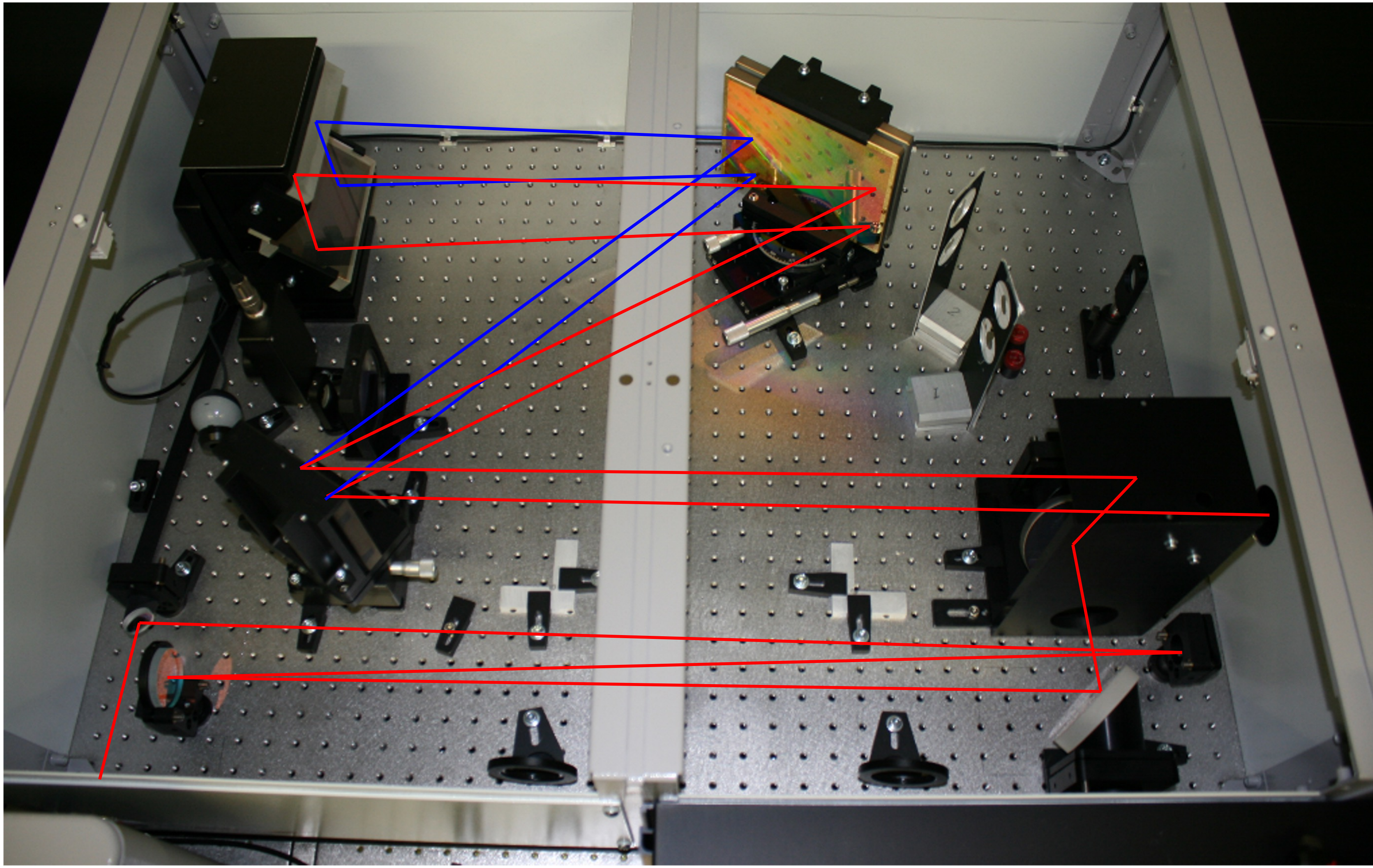
trigger source

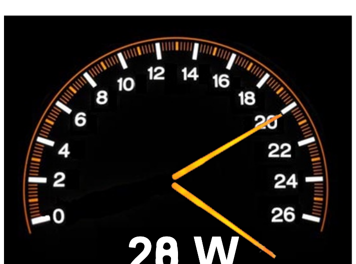
ETNA

Aurora laser

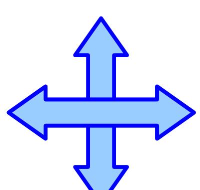


10 kHz compressor





polarization



rep. rate

10 kHz

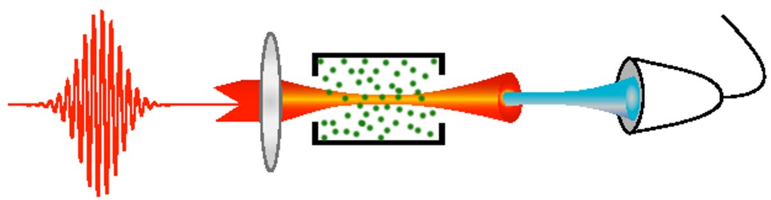
pulse duration

300 fs

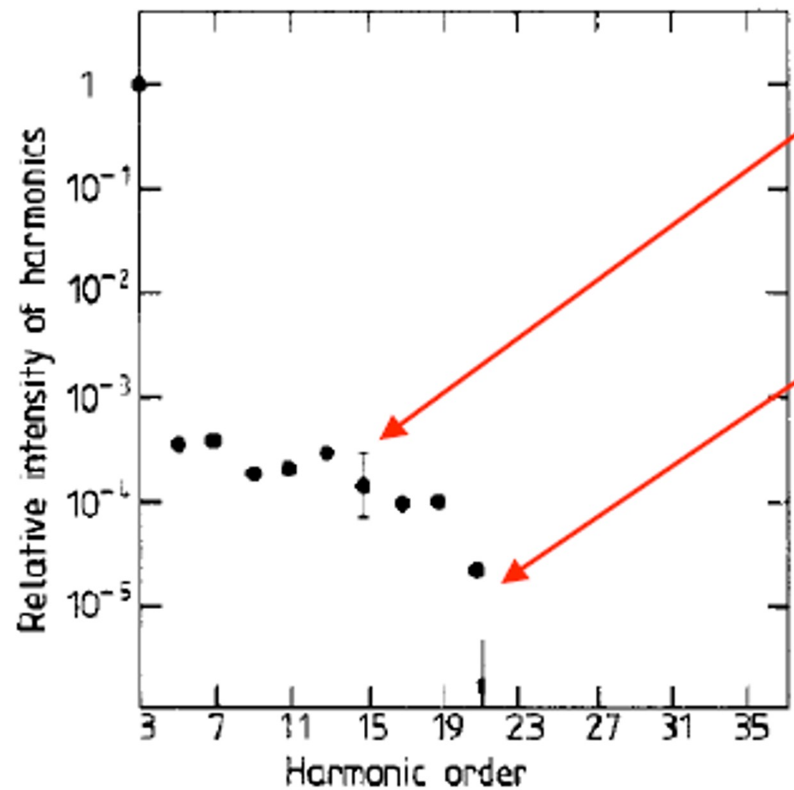
trigger source

PC 2

High-order harmonic generation



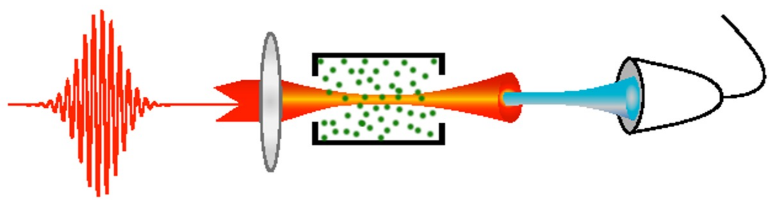
only odd harmonics



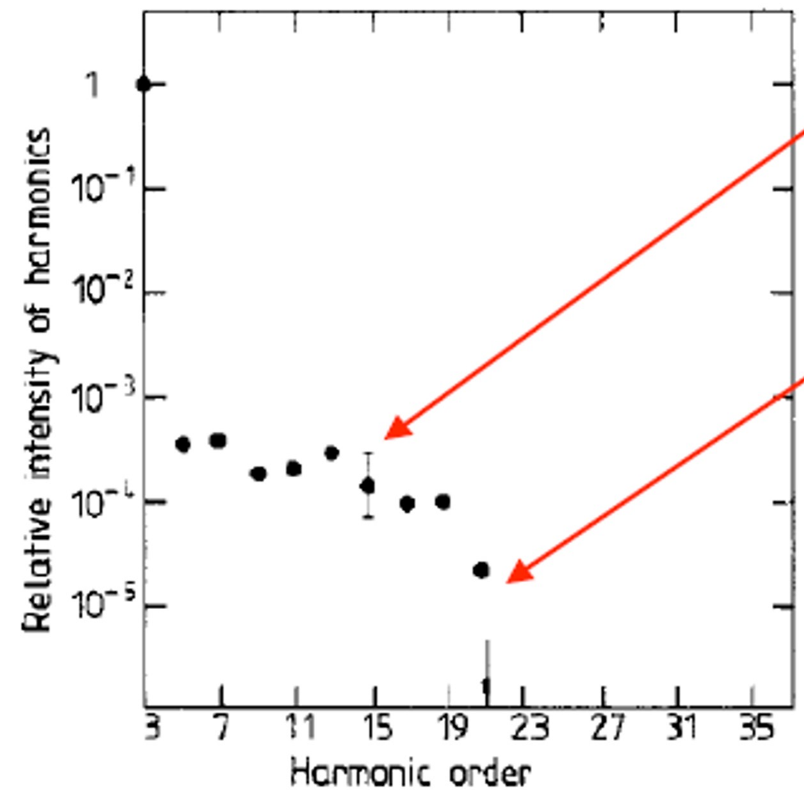
plateau

cut-off

Ferray *et al.* (1988)



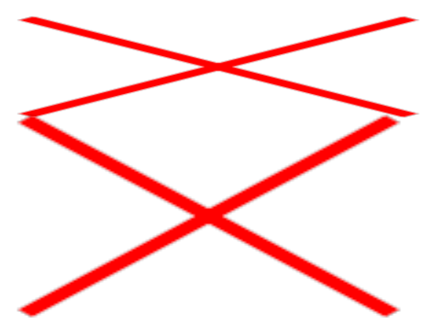
only odd harmonics



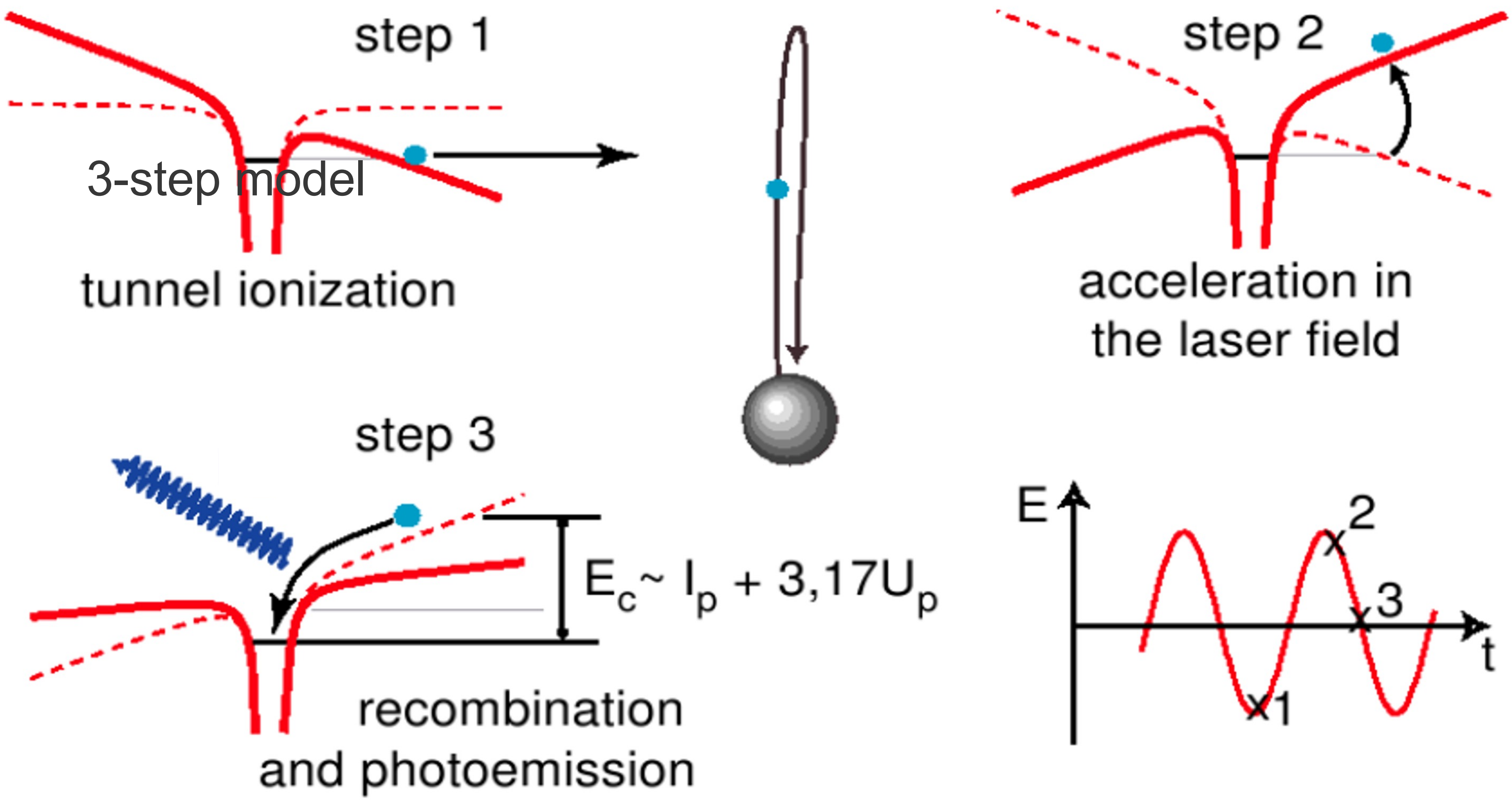
Ferray *et al.* (1988)

plateau

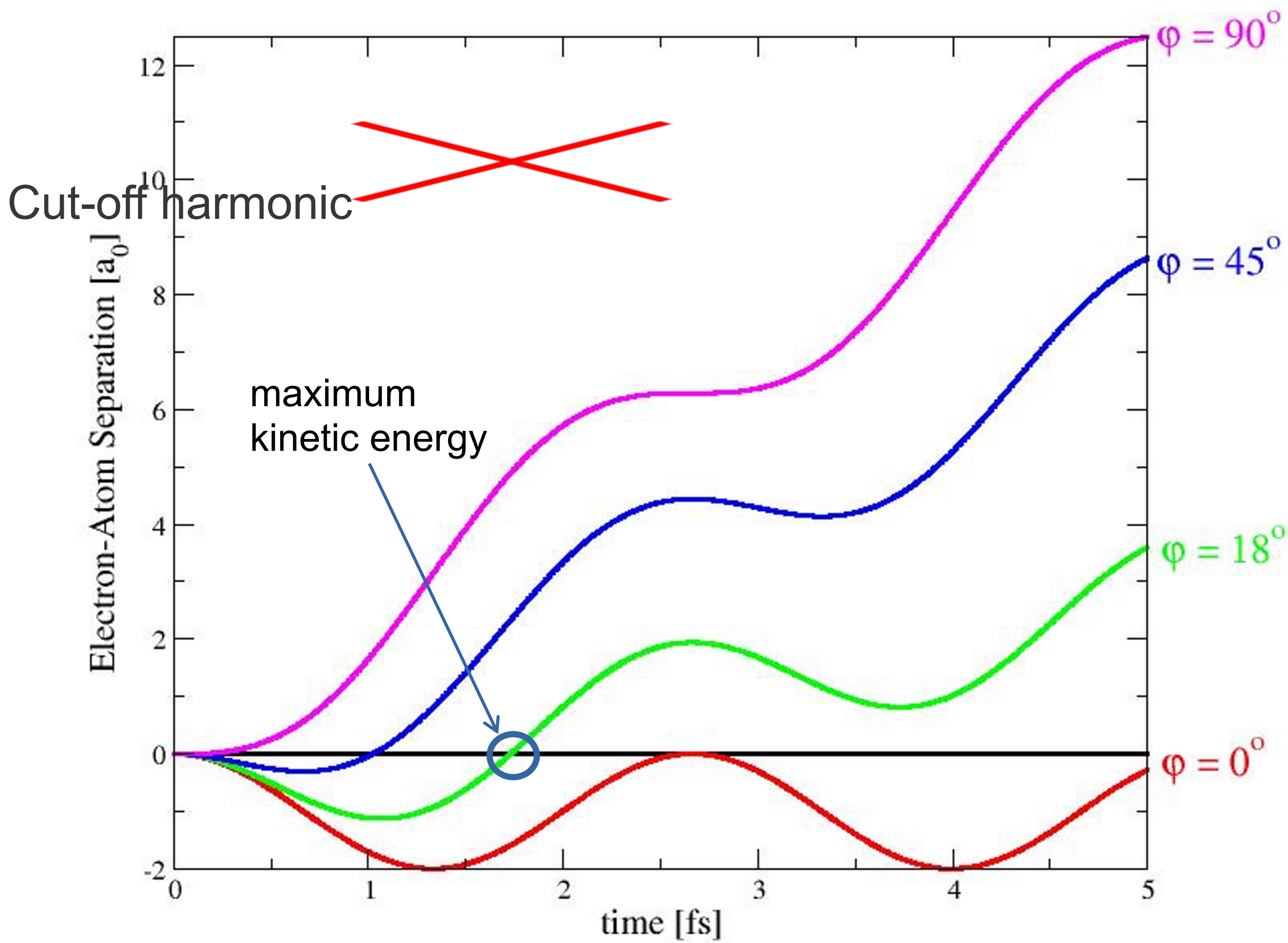
cut-off



Ponderomotive potential



Corkum, Kulander



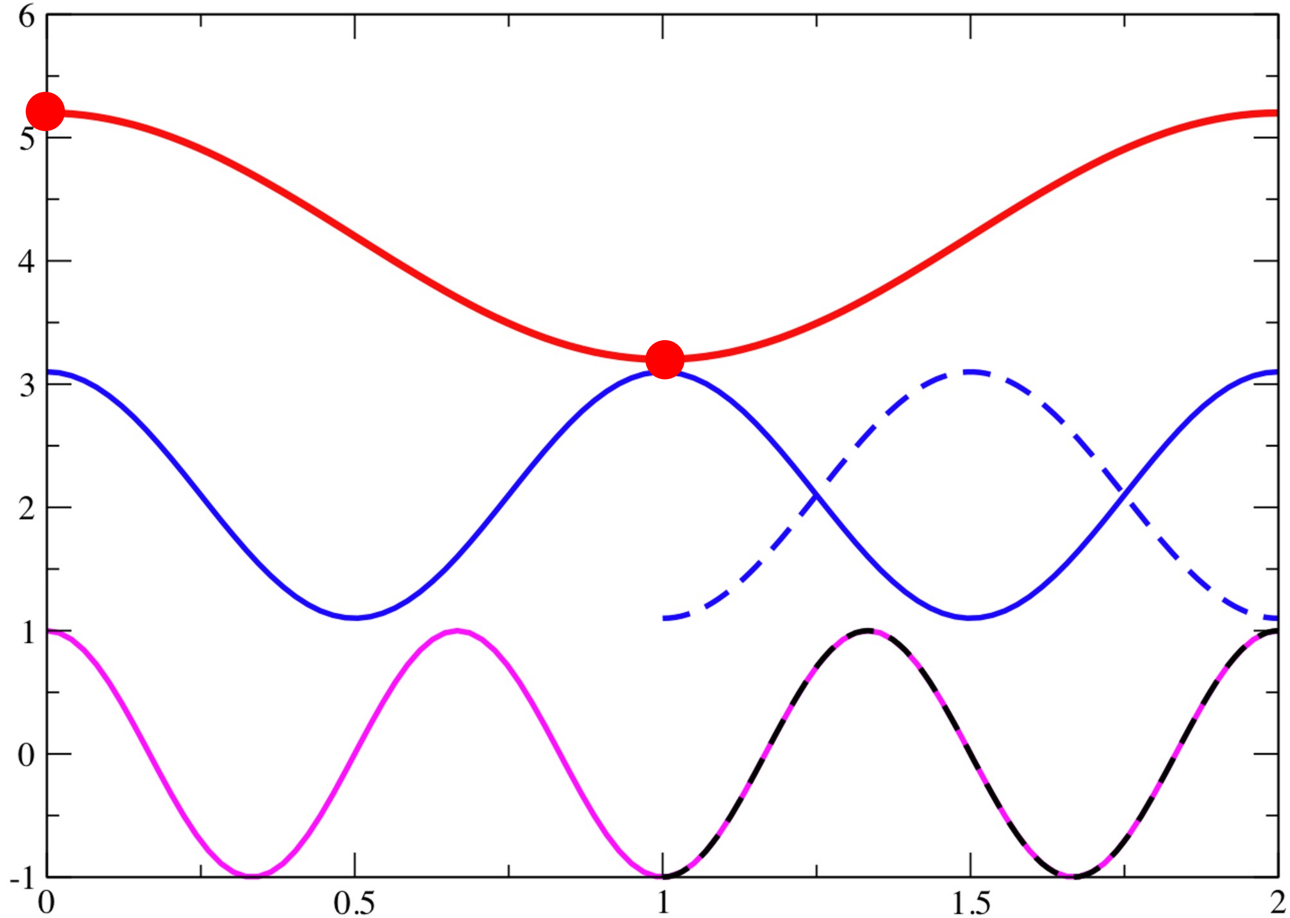
Cut-off harmonic
maximum
kinetic energy
Electron-Atom Separation [a0]
time [fs]
φ = 90°
φ = 45°
φ = 18°
φ = 0°
12
10
8
6
4
2
0
-2
0
1
2
3
4
5

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

fundamental

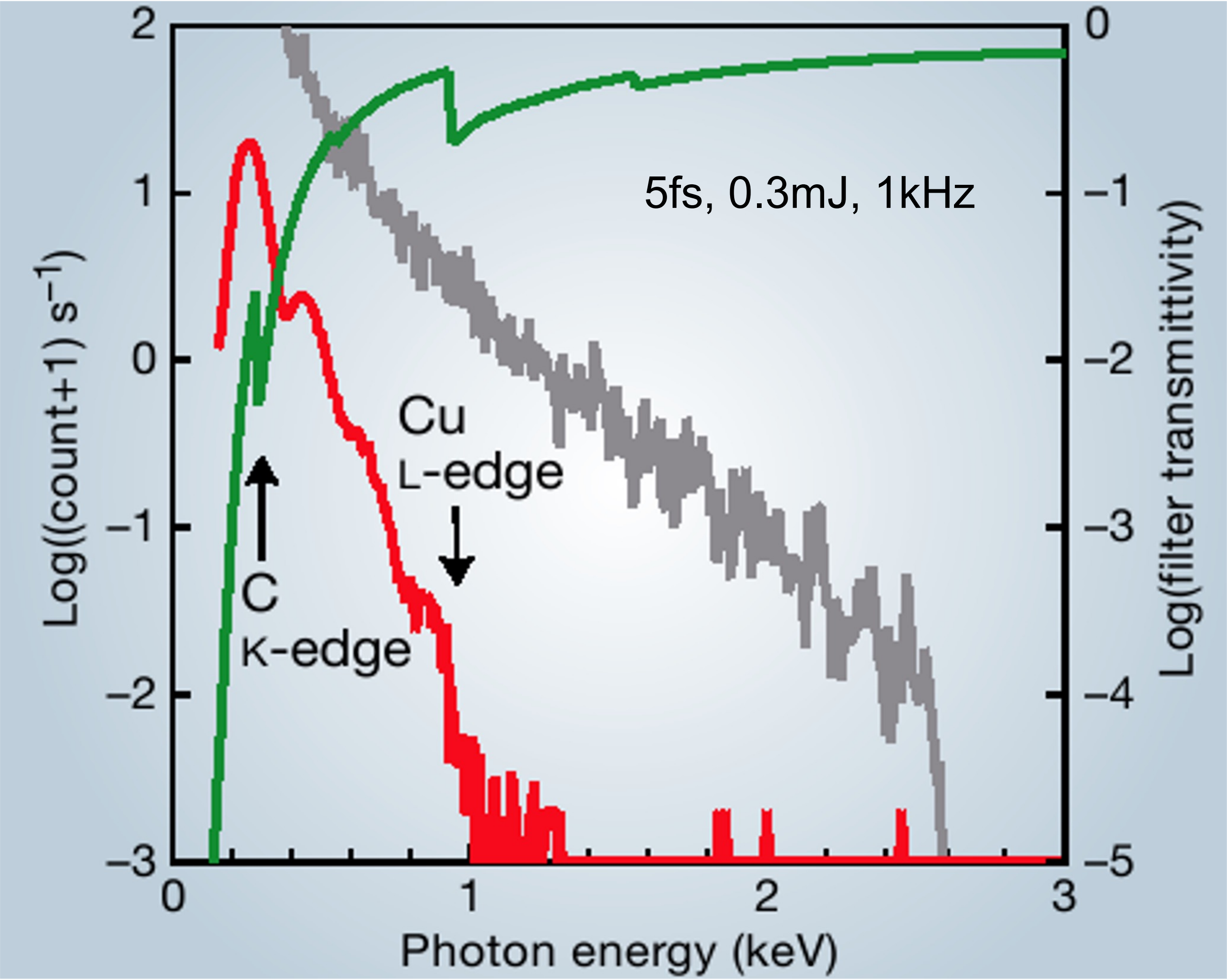
2nd harmonic

3rd harmonic



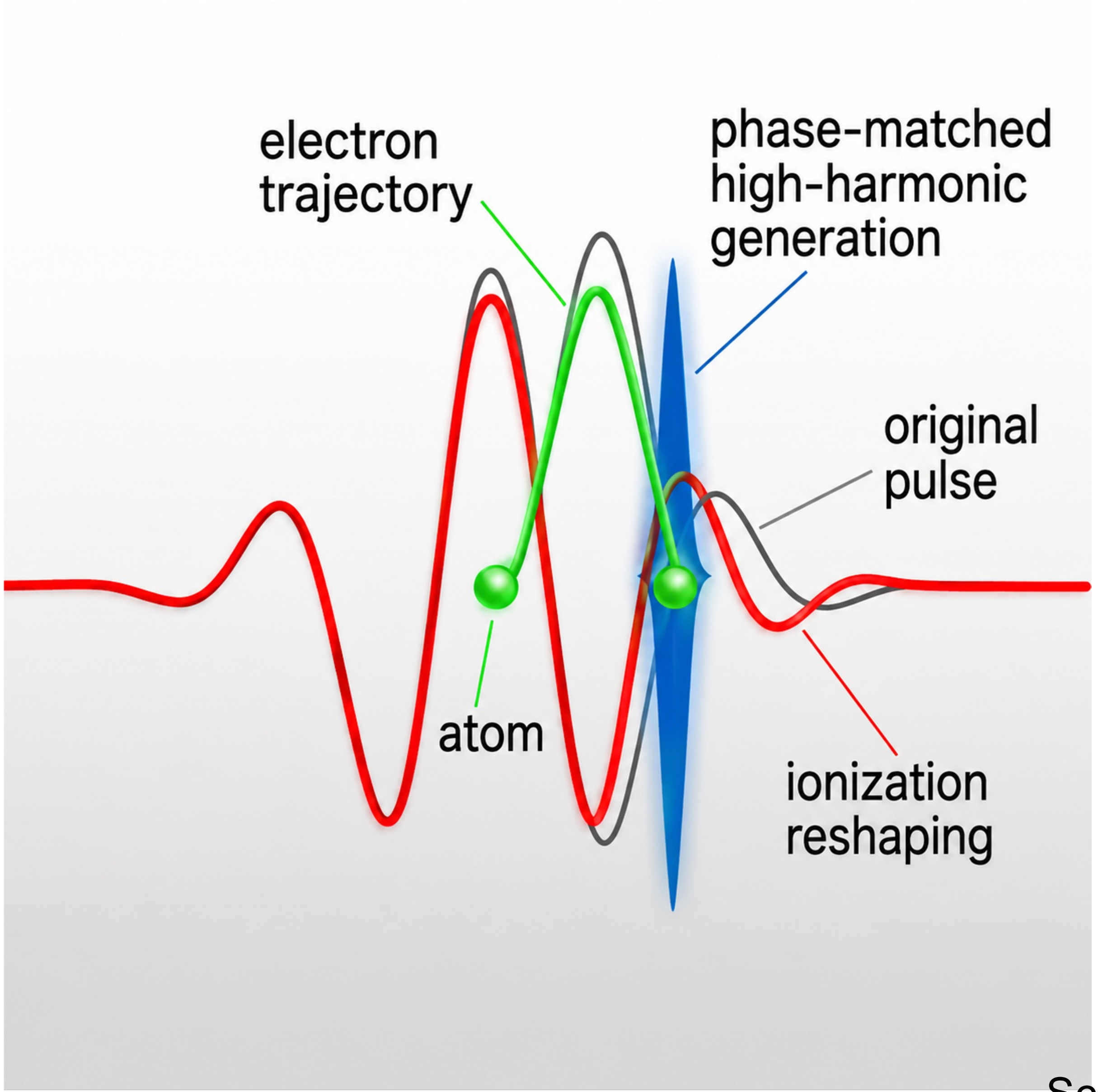
Only odd harmonics are generated! Phase matching is important!

Energies up to 2.5 keV! N>1500!

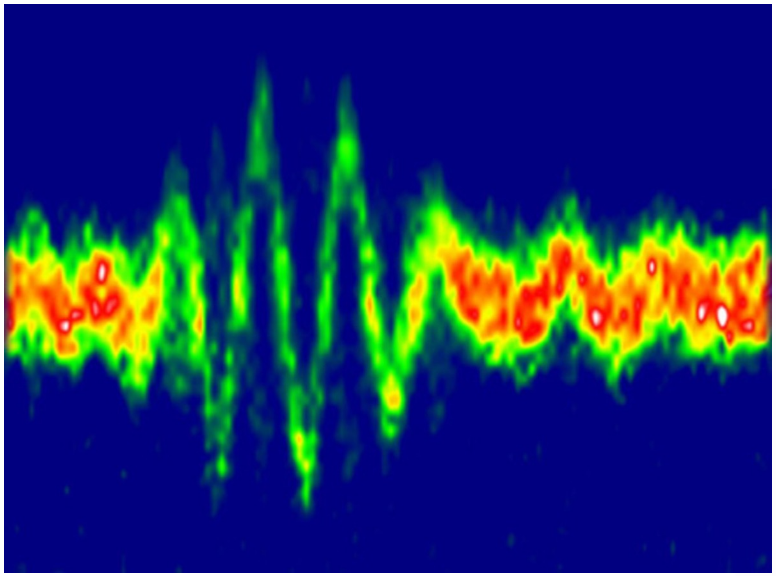


J. Seres et al, Nature 433, 596 (2006)

Attosecond pulse generation

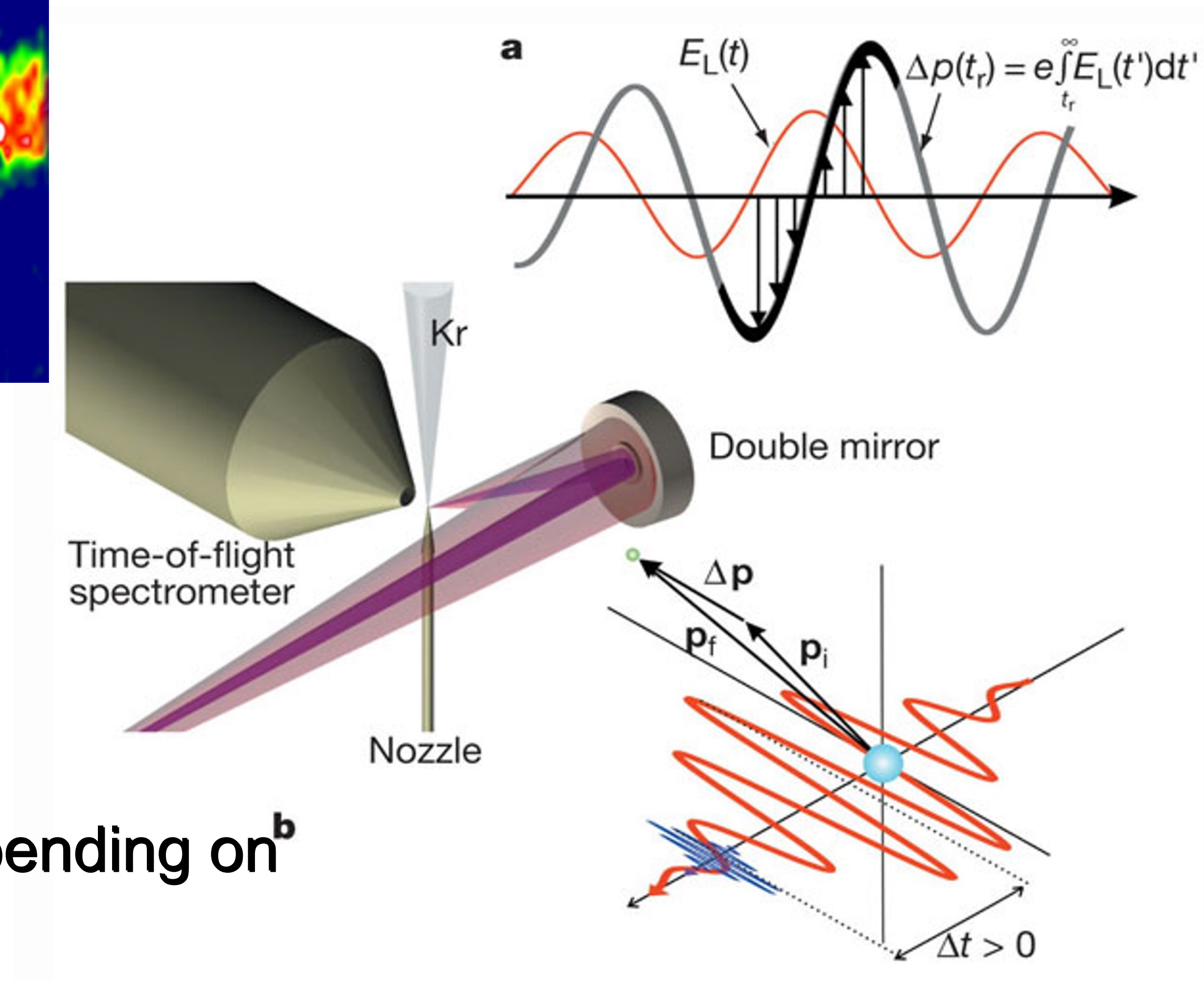


Schötz et al (2020)



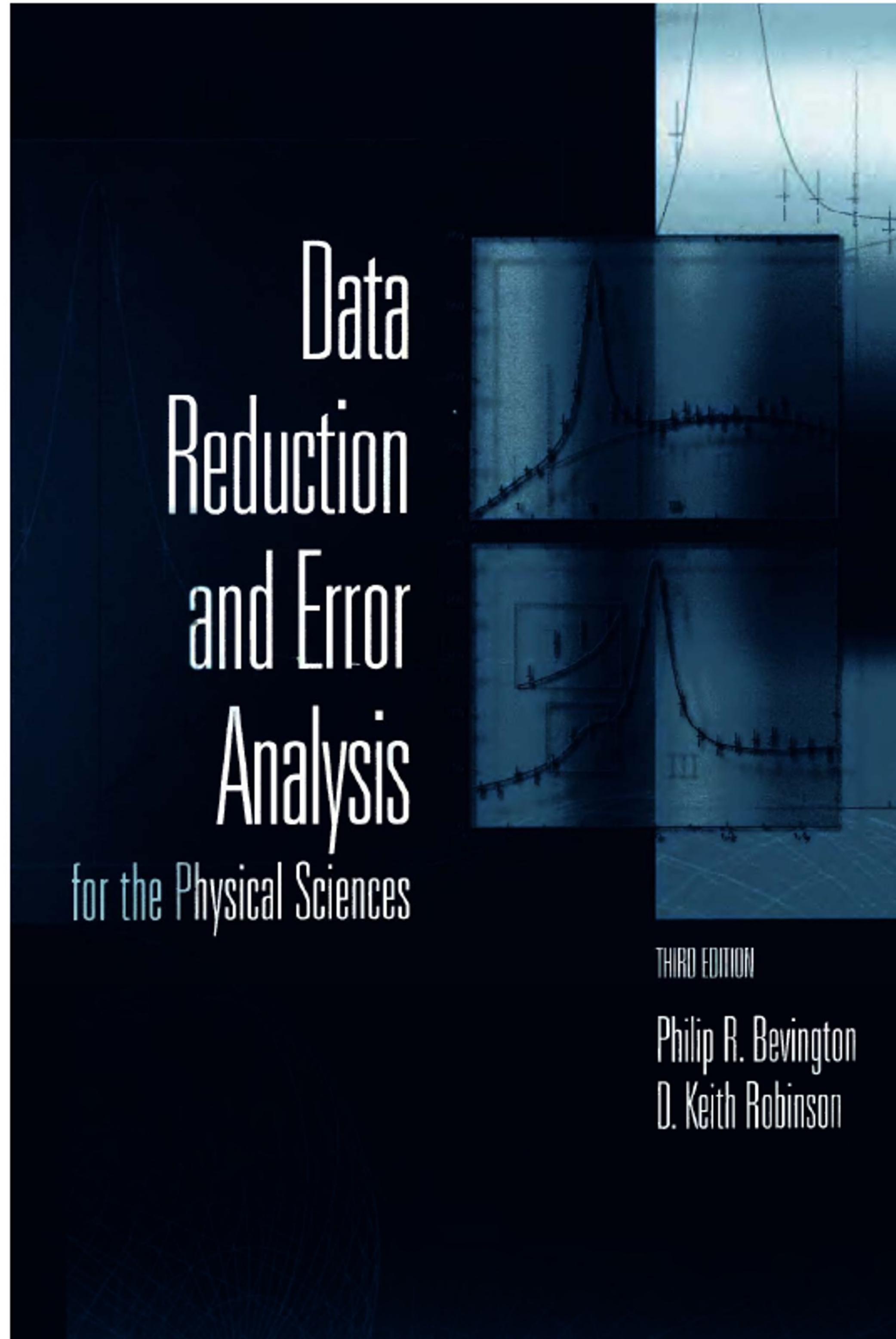
Energy

Time



Electrons gain energy depending on momentary IR field.

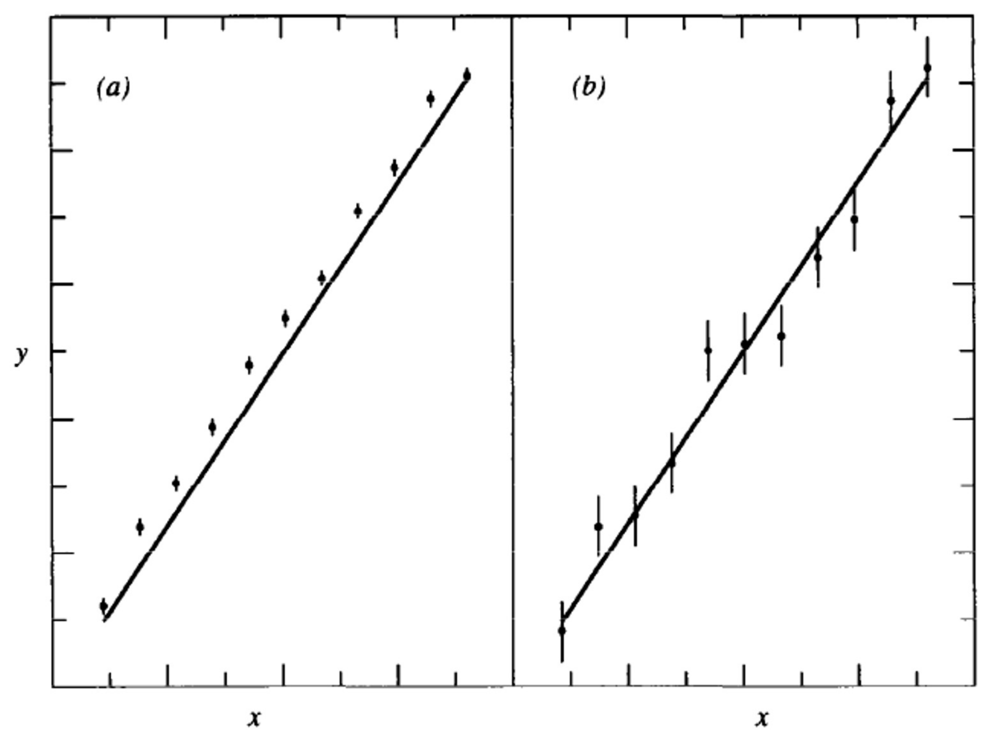
The best book ever!



Error is defined by Webster as "the difference between an observed or calculated value and the true value." Usually we do not know the "true" value; otherwise

Accuracy Versus Precision

It is important to distinguish between the terms *accuracy* and *precision*. The accuracy of an experiment is a measure of how close the result of the experiment is to the true value; the precision is a measure of how well the result has been determined, without reference to its agreement with the true value. The precision is also



The term *error* suggests a deviation of the result from some "true" value. Usually we cannot know what the true value is, and can only estimate the errors inherent in the experiment. If we repeat an experiment, the results may well differ from those of the first attempt. We express this difference as a *discrepancy* between the two results. Discrepancies arise because we can determine a result only with a given *uncertainty*. For example, when we compare different measurements of a standard physical constant, or compare our result with the accepted value, we should refer to the differences as *discrepancies*, not errors or uncertainties.

In order to determine the parameters of the parent distribution, we assume that the results of experiments asymptotically approach the parent quantities as the number of measurements approaches infinity; that is, the parameters of the experimental distribution equal the parameters of the parent distribution *in the limit of an infinite number of measurements.* If we specify that there are N observations in a given experiment, then we can denote this by

$$\text{(parent parameter)} = \lim_{N\to\infty} \text{(experimental parameter)}$$

$$\mu \equiv \lim_{N\to\infty}\left(\frac{1}{N}\sum x_i\right)$$

mean

$$\sigma^2 \equiv \lim_{N\to\infty}\left[\frac{1}{N}\sum (x_i-\mu)^2\right] = \lim_{N\to\infty}\left(\frac{1}{N}\sum x_i^2\right) - \mu^2$$

variance (square of standard deviation)

$$s^2 \equiv \frac{1}{N-1}\sum (x_i-\bar{x})^2$$

- estimate from data

2.1 BINOMIAL DISTRIBUTION

Suppose we toss a coin in the air and let it land.

With these definitions of p and q, the probability $P_B(x; n, p)$ for observing x of the n items to be in the state with probability p is given by the *binomial distribution*

$$P_B(x; n, p) = \binom{n}{x} p^x q^{n-x} = \frac{n!}{x!(n-x)!} p^x (1-p)^{n-x} \tag{2.4}$$

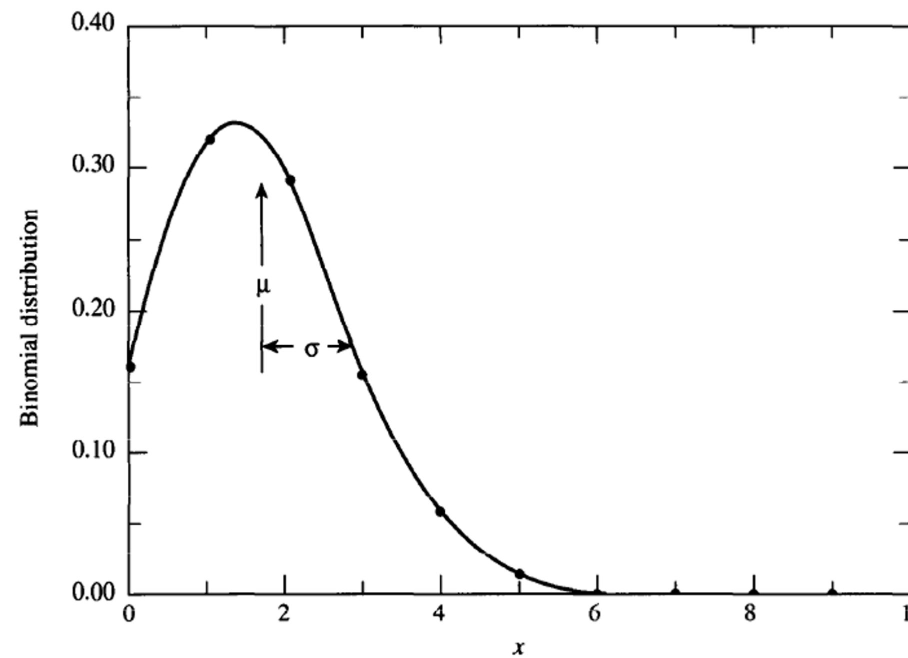


FIGURE 2.2
Binomial distribution for $\mu = 10/6$ and $p = 1/6$ shown as a continuous curve.

$$\mu = \sum_{x=0}^{n} \left[x \frac{n!}{x!(n-x)!} p^x (1-p)^{n-x} \right] = np$$

$$\sigma^2 = \sum_{x=0}^{n} \left[(x-\mu)^2 \frac{n!}{x!(n-x)!} p^x (1-p)^{n-x} \right] = np(1-p)$$

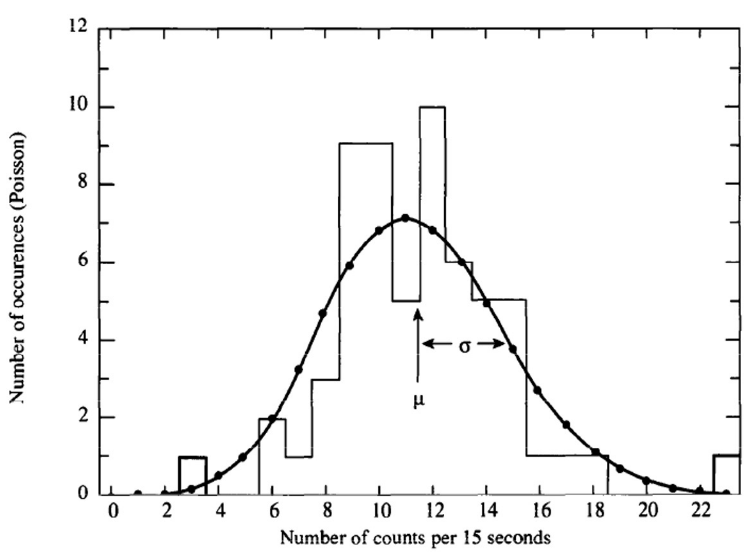
2.2 POISSON DISTRIBUTION

when $\mu \ll n$ because $p \ll 1$.

$$\lim_{p \to 0} P_B(x; n, p) = P_P(x; \mu) \equiv \frac{\mu^x}{x!} e^{-\mu}$$

$$\langle x \rangle = \sum_{x=0}^{\infty} \left(x \frac{\mu^x}{x!} e^{-\mu} \right) = \mu e^{-\mu} \sum_{x=1}^{\infty} \frac{\mu^{x-1}}{(x-1)!} = \mu e^{-\mu} \sum_{y=0}^{\infty} \frac{\mu^y}{y!} = \mu$$

$$\sigma^2 = \langle (x-\mu)^2 \rangle = \sum_{x=0}^{\infty} \left[(x-\mu)^2 \frac{\mu^x}{x!} e^{-\mu} \right] = \mu$$



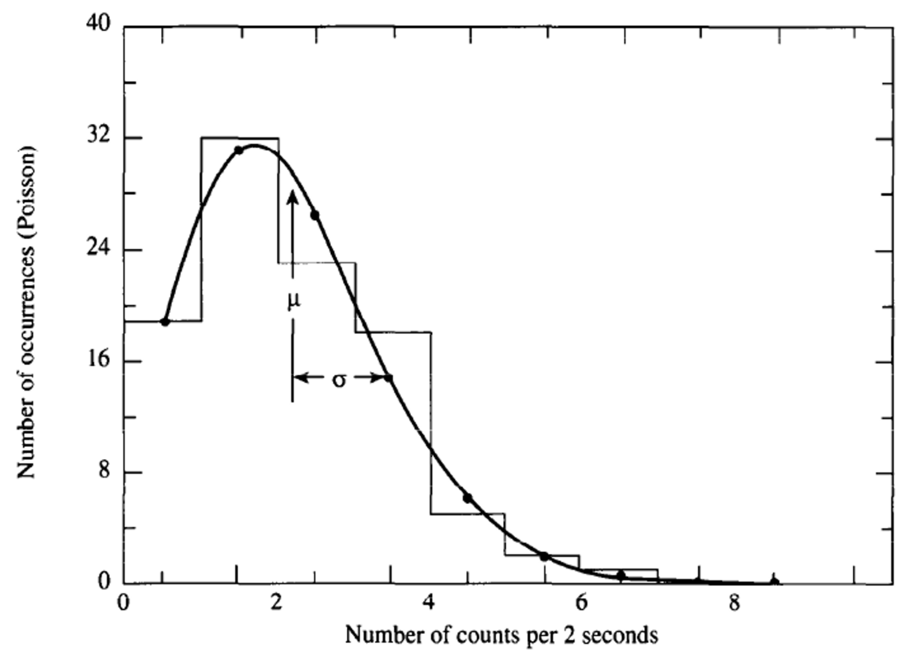


FIGURE 2.3
Histogram of counts in a cosmic ray detector. The Poisson distribution is an estimate of the parent distribution based on the measured mean $\bar{x} = 1.69$. It is shown as a continuous curve although the function is only defined at the discrete points indicated by the round dots.

2.3 GAUSSIAN OR NORMAL ERROR DISTRIBUTION

The Gaussian distribution is an approximation to the binomial distribution for the special limiting case where the number of possible different observations n becomes infinitely large and the probability of success for each is finitely large so $np \gg 1$. It is

There are several derivations of the Gaussian distribution from first principles, none of them as convincing as the fact that the distribution is reasonable, that it has a fairly simple analytic form, and that it is accepted by convention and experimentation to be the most likely distribution for most experiments. In addition, it has the satisfying characteristic that the most probable estimate of the mean μ from a random sample of observations x is the average of those observations $\bar{x}$.

sum of uniformly distributed random variables - Gaussian

$$p_G = \frac{1}{\sigma\sqrt{2\pi}} \exp\left[-\frac{1}{2}\left(\frac{x-\mu}{\sigma}\right)^2\right]$$

3.2 PROPAGATION OF ERRORS

Variance and Covariance

$$\sigma_u^2 = \lim_{N\to\infty}\left[\frac{1}{N}\sum (u_i - \bar{u}_i)^2\right] \qquad \sigma_v^2 = \lim_{N\to\infty}\left[\frac{1}{N}\sum (v_i - \bar{v}_i)^2\right]$$

$$\sigma_{uv}^2 \equiv \lim_{N\to\infty}\left[\frac{1}{N}\sum [(u_i - \bar{u})(v_i - \bar{v})]\right]$$

$$\sigma_x^2 \simeq \sigma_u^2\left(\frac{\partial x}{\partial u}\right)^2 + \sigma_v^2\left(\frac{\partial x}{\partial v}\right)^2 + \cdots + 2\sigma_{uv}^2\left(\frac{\partial x}{\partial u}\right)\left(\frac{\partial x}{\partial v}\right) + \cdots \tag{3.13}$$

Equation (3.13) is known as the *error propagation equation.*

3.4 APPLICATION OF ERROR EQUATIONS

Covariance: $\sigma_{uv}^2 = \langle (u - \bar{u})(v - \bar{v}) \rangle$.

Propagation of errors: Assume $x = f(u, v)$:

$$\sigma_x^2 = \sigma_u^2 \left(\frac{\partial x}{\partial u}\right)^2 + \sigma_v^2 \left(\frac{\partial x}{\partial v}\right)^2 + 2\sigma_{uv}^2 \left(\frac{\partial x}{\partial u}\right)\left(\frac{\partial x}{\partial v}\right)$$

For u and v uncorrelated, $\sigma_{uv}^2 = 0$.

Specific formulas:

$x = au + bv$	$\sigma_x^2 = a^2\sigma_u^2 + b^2\sigma_v^2 + 2ab\sigma_{uv}^2$
$x = auv$	$\dfrac{\sigma_x^2}{x^2} = \dfrac{\sigma_u^2}{u^2} + \dfrac{\sigma_v^2}{v^2} + 2\,\dfrac{\sigma_{uv}^2}{uv}$
$x = \dfrac{au}{v}$	$\dfrac{\sigma_x^2}{x^2} = \dfrac{\sigma_u^2}{u^2} + \dfrac{\sigma_v^2}{v^2} - 2\,\dfrac{\sigma_{uv}^2}{uv}$
$x = au^b$	$\dfrac{\sigma_x}{x} = b\,\dfrac{\sigma_u}{u}$
$x = ae^{bu}$	$\dfrac{\sigma_x}{x} = b\sigma_u$
$x = a^{bu}$	$\dfrac{\sigma_x}{x} = (b \ln a)\sigma_u$
$x = a \ln(bu)$	$\sigma_x = ab\,\dfrac{\sigma_u}{u}$
$x = a \cos(bu)$	$\sigma_x = -\sigma_u ab \sin(bu)$
$x = a \sin(bu)$	$\sigma_x = \sigma_u ab \cos(bu)$

4.1 METHOD OF LEAST SQUARES

$$P(\mu') = \left(\frac{1}{\sigma\sqrt{2\pi}}\right)^N \exp\left[-\frac{1}{2}\sum\left(\frac{x_i - \mu'}{\sigma}\right)^2\right] \qquad (4.5)$$

According to the *method of maximum likelihood,* if we compare the probabilities $P(\mu')$ of obtaining our set of observations from various parent populations with different means μ' but with the same standard deviation $\sigma' = \sigma$, the probability is greatest that the data were derived from a population with $\mu' = \mu$; that is, the most likely population from which such a set of data might have come is assumed to be the correct one.

The method of maximum likelihood states that the most probable value for μ' is the one that gives the maximum value for the probability $P(\mu')$ of Equation (4.5). Because this probability is the product of a constant times an exponential to a negative argument, maximizing the probability $P(\mu')$ is equivalent to minimizing the argument X of the exponential,

$$X = -\frac{1}{2}\sum\left(\frac{x_i - \mu'}{\sigma}\right)^2 \qquad (4.6)$$

$$\sigma_\mu^2 = \sum\left[\sigma_i^2\left(\frac{\partial\mu'}{\partial x_i}\right)^2\right]$$

$$\sigma_\mu^2 = \sum\left[\sigma_i^2\left(\frac{1}{N}\right)^2\right] = \frac{\sigma^2}{N}$$

$$\sigma_\mu = \frac{\sigma}{\sqrt{N}} \simeq \frac{s}{\sqrt{N}}$$

A Warning About Statistics

We may be able to make corrections for these problems, once we are aware of their existence, but there are always others. At some level, things will happen that we cannot understand, and for which we cannot make corrections, and these "things" will cause data to appear where statistically no data should exist, and data points to vanish that should have been there. The moral is, be aware and do not trust statistics in the tails of the distributions.