Lecture on:

Multiphoton Physics

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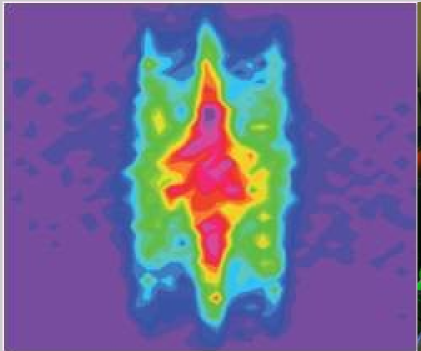
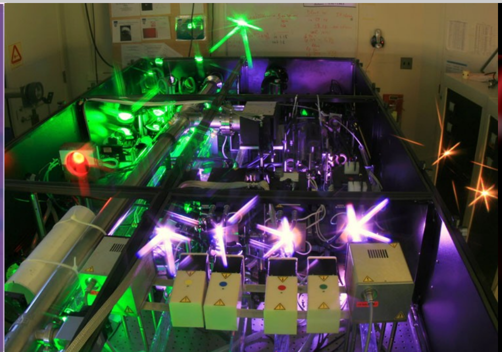
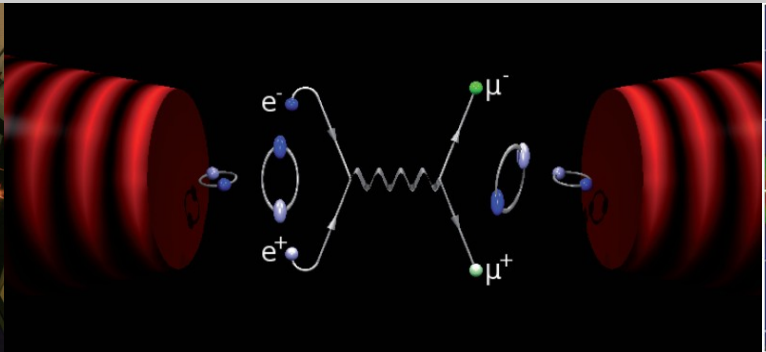
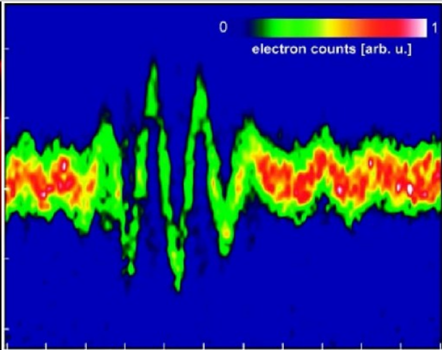
Outline

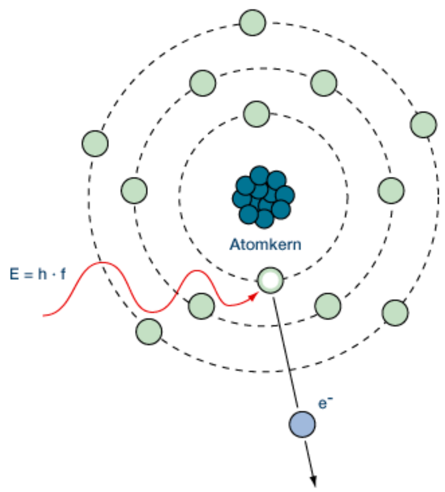
- Introduction
- Theory of multiphoton ionization
- Transforming many small photons into a big one
- History of multiphoton physics
- Further multiphoton effects

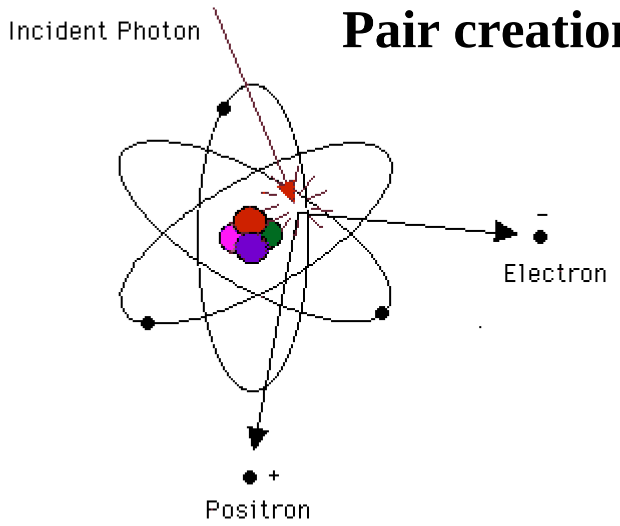


Interaction of electrons with photons

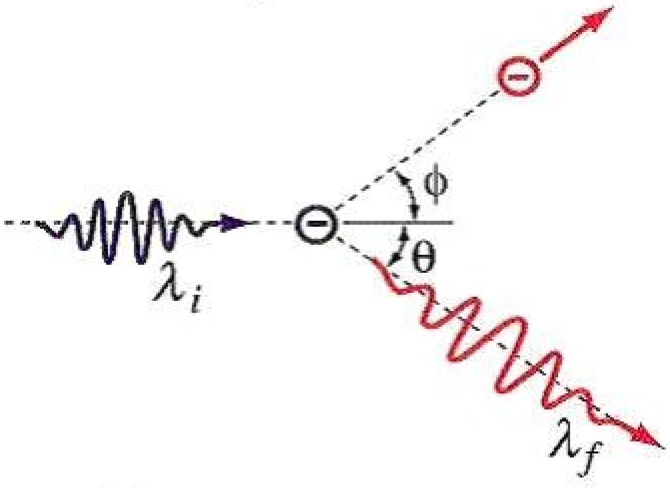
Photoeffect



Pair creation



Compton Effect



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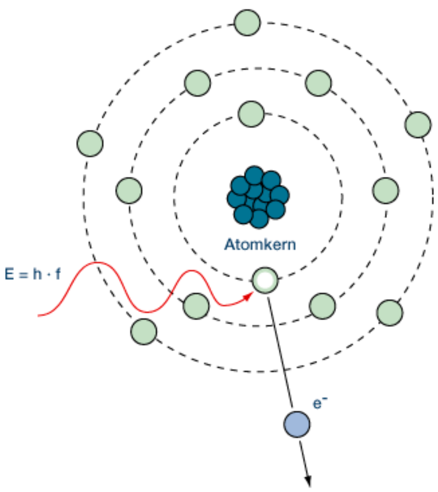
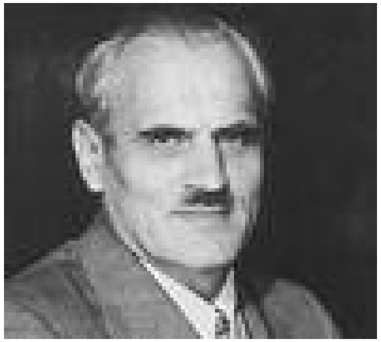
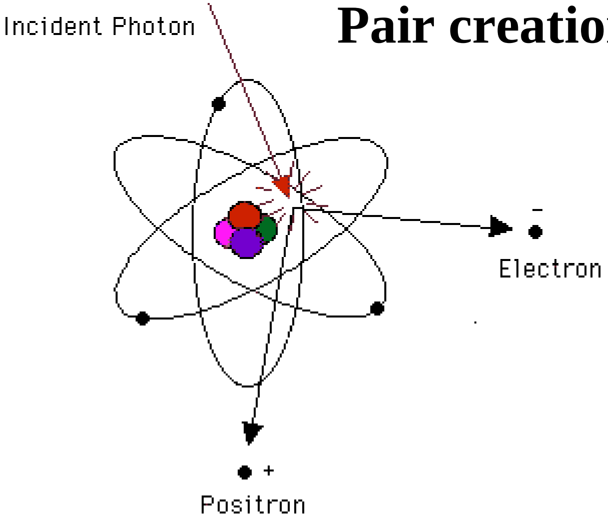




A. H. Compton

Pair creation



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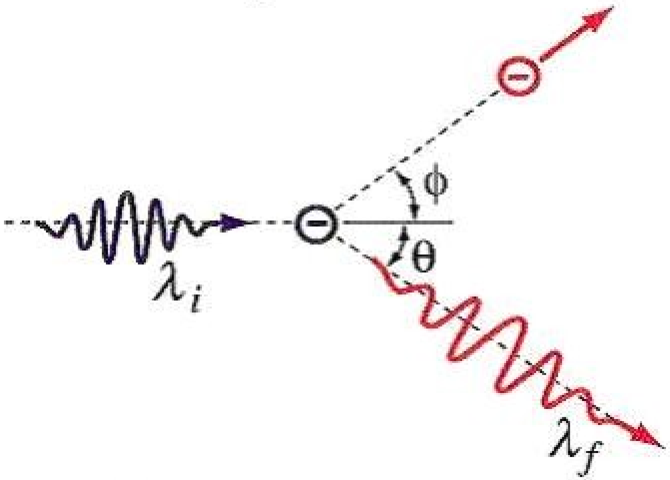
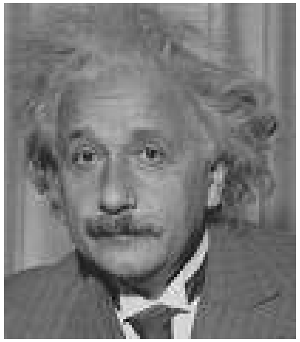
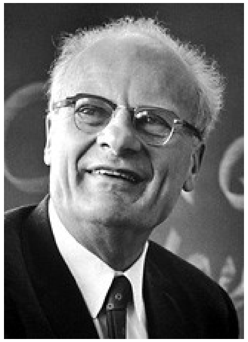


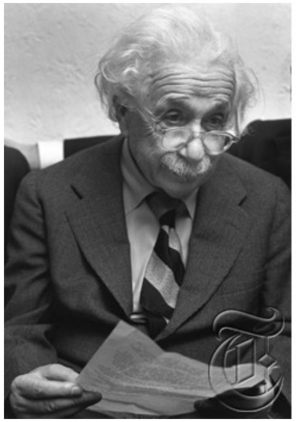


A. Einstein

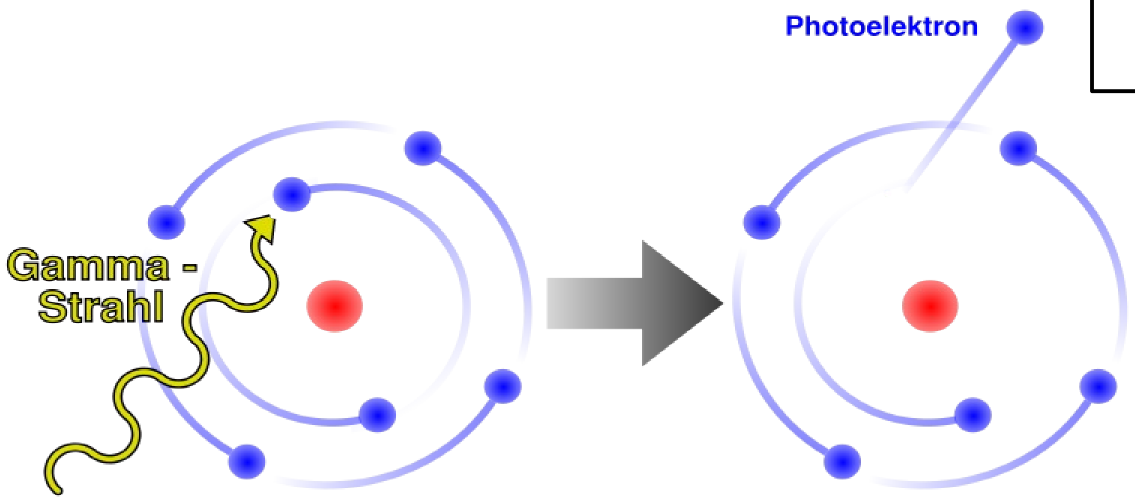


H. A. Bethe

W. Heitler



Einstein's photoelectric effect



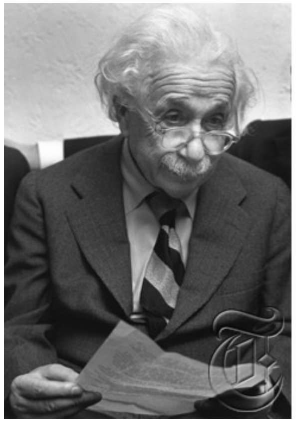
$$E_{kin} = \hbar\omega - \varepsilon_{bind}$$

Laws of photoelectric emission

1) For a given atom, there exists a certain minimum frequency of incident radiation below which no photoelectrons can be emitted.

2) For a given atom and frequency of incident radiation, the rate at which photoelectrons are ejected is proportional to the intensity of the incident light.

3) Above the threshold frequency, the maximum kinetic energy of the emitted photoelectron is independent of the intensity of the incident light.



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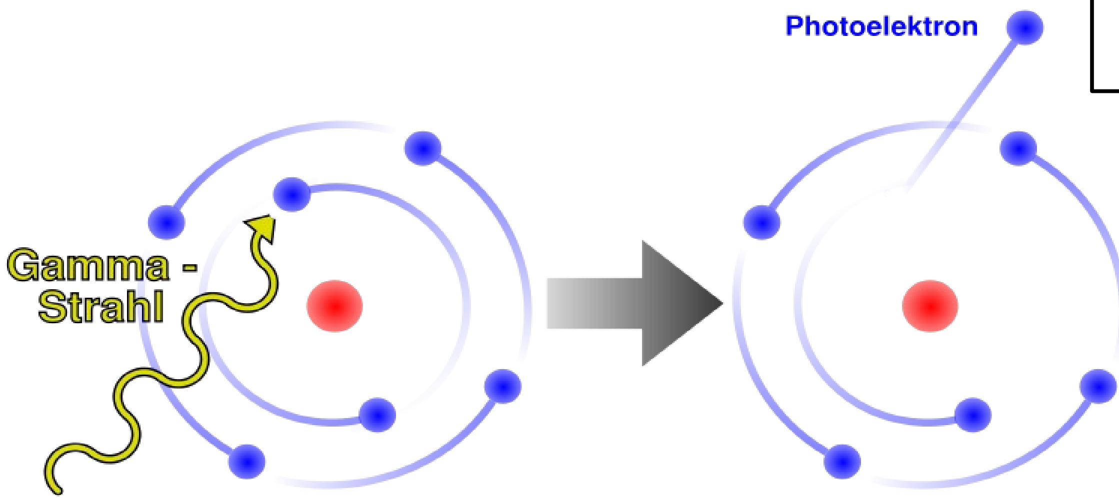


$$E_{kin} = \hbar\omega - \varepsilon_{bind}$$

Laws of photoelectric emission

1) For a given atom, there exists a certain <u>minimum frequency</u> of incident radiation below which no photoelectrons can be emitted.

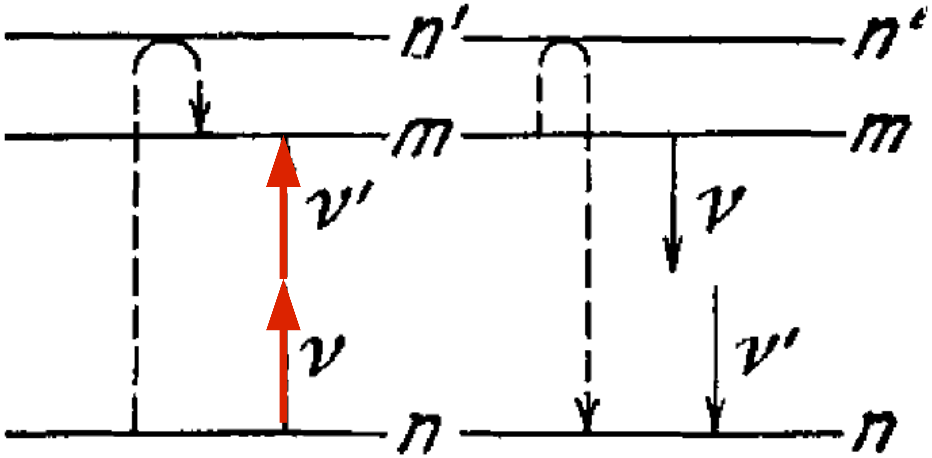
2) For a given atom and frequency of incident radiation, the rate at which photoelectrons are ejected is <u>proportional to the intensity</u> of the incident light.

3) Above the threshold frequency, the maximum kinetic energy of the emitted photoelectron is <u>independent of the intensity</u> of the incident light.

These laws become wrong at high intensity of the incident light!

Processes with two photons

Maria Göppert-Mayer (1931):
“Über Elementarakte mit zwei Quantensprüngen”



Physical unit for two-photon absorption: **1 GM = 10^{-50} cm^{4}s**

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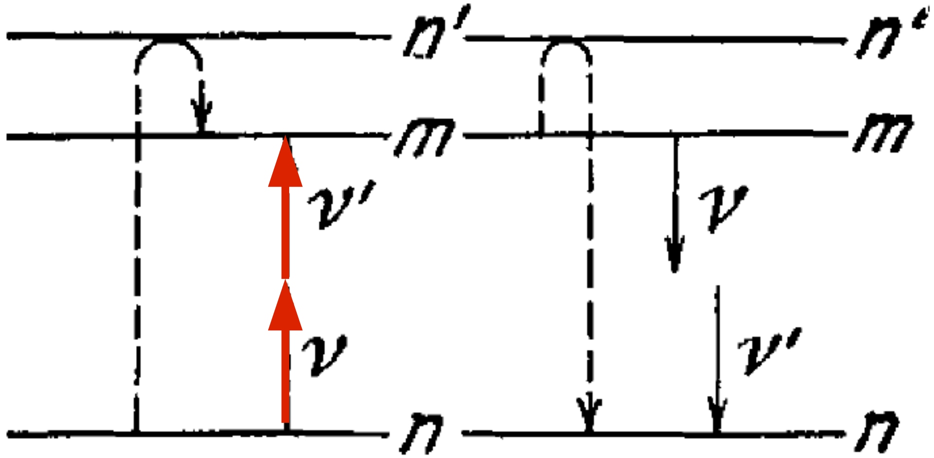


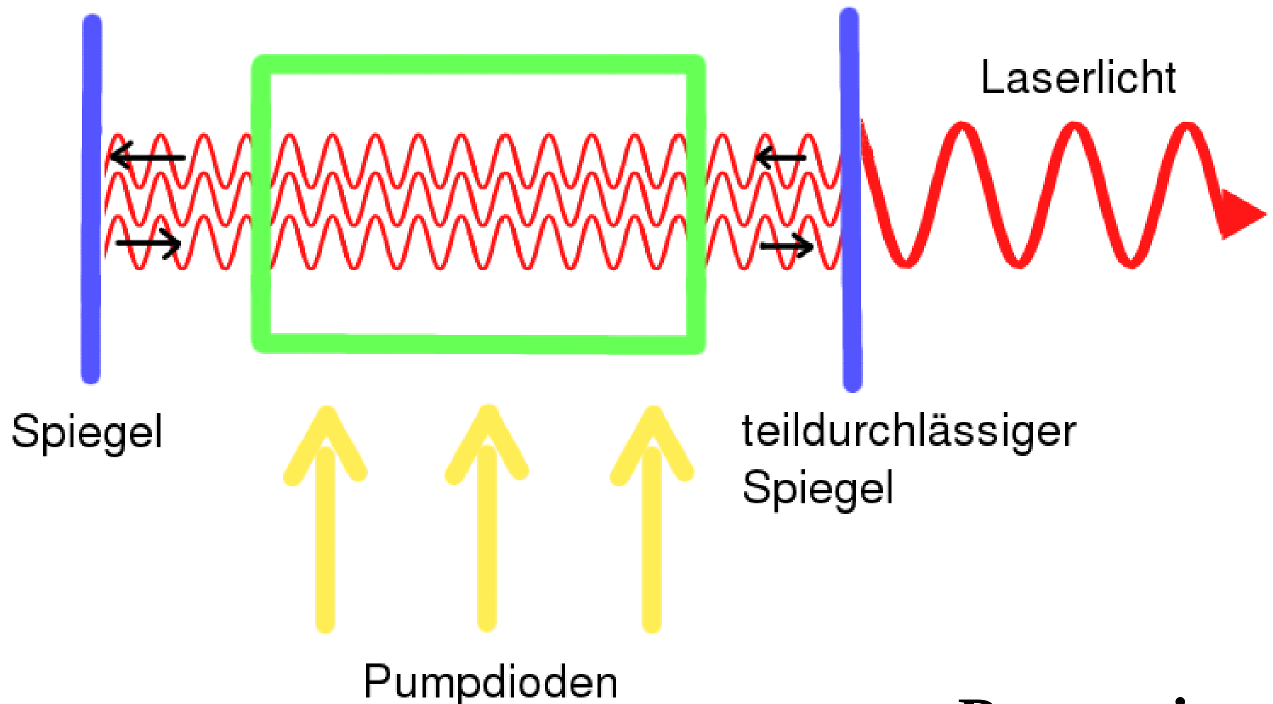
Physical unit for two-photon absorption: **1 GM = 10^{-50} cm^{4}s**

Experimental observation of two-photon absorption processes:
Franken et al., 1961; Kaiser & Garrett, 1961; Abella, 1962; ......

What had happened in the mean time?

Construction of the first laser

Basic principle:

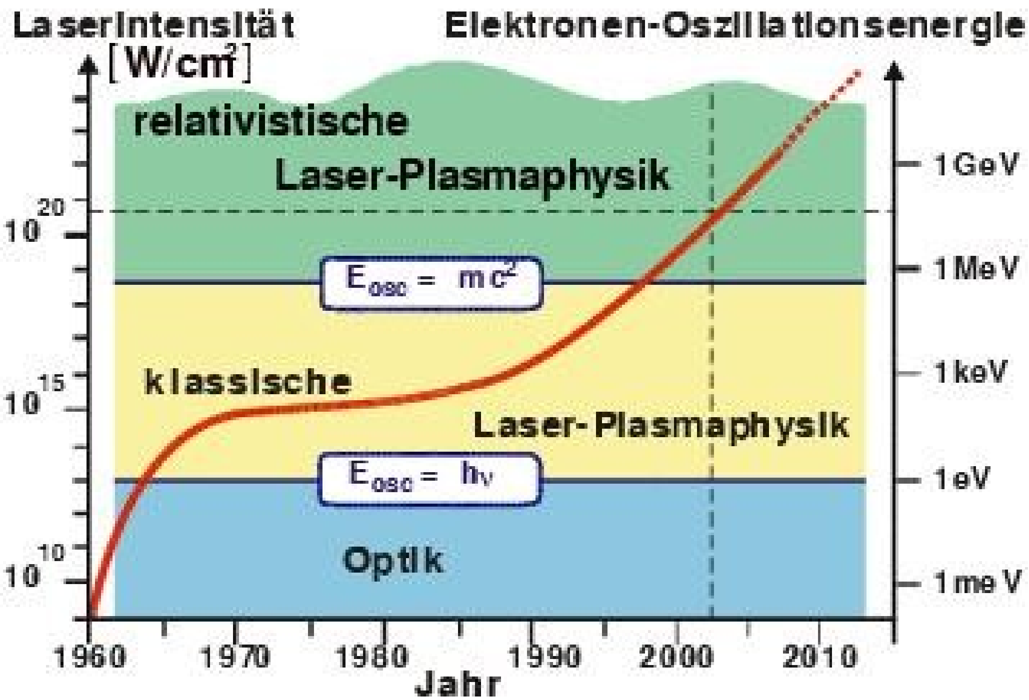


Theodore Maiman, *Nature* (1960):
"Stimulated optical radiation in ruby"

Properties:

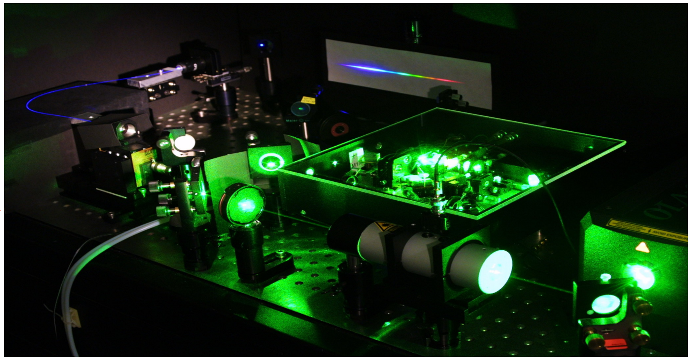
- monochromatic
- coherent
- <u>intense</u>

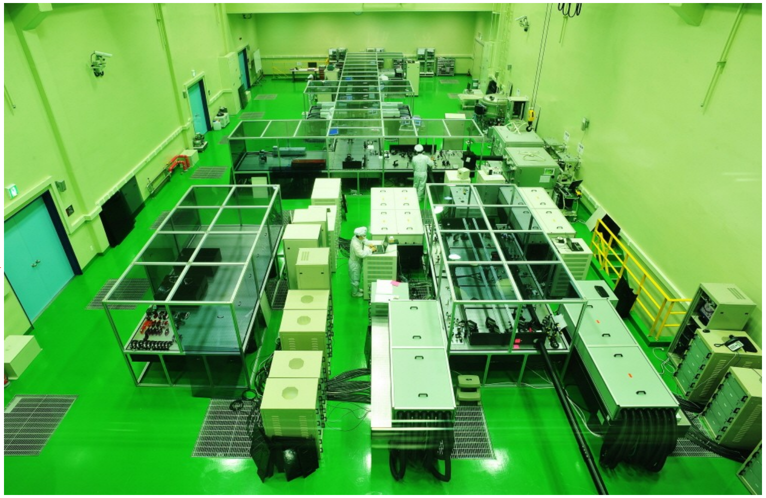
Progress in laser technology



Temporal development of available field intensities

(© Prof. Willi)





Some illustrative numbers

Source	λ	Intensity	Photon density
	$[\mu m]$	$[W cm^{-2}]$	$[cm^{-3}]$
Light bulb	0.58	10^{-3}	10^{5}
Sun (on Earth)	0.1-1	10^{-1}	10^{7}
cw CO_2-Laser	10	10^{10}	10^{19}
Ti-Saphir-Laser	0.8	10^{18}	10^{26}
Petawatt Livermore	1.06	10^{21}	10^{29}

10^{15} W/cm^2 ≙ 1 photon in the volume of an atom

10^{21} W/cm^2 ≙ Sun light on earth focussed down to cross section of a human hair

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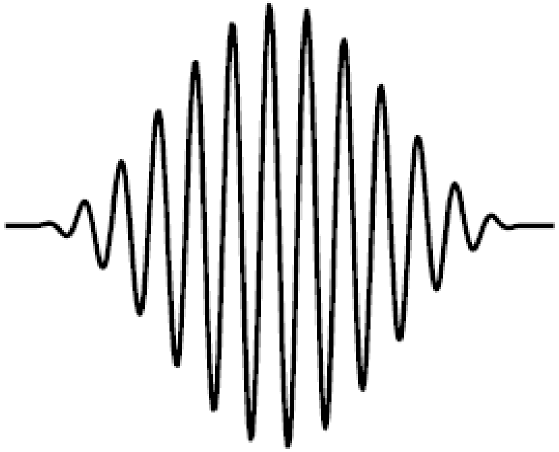
Time scale:

Femtoseconds

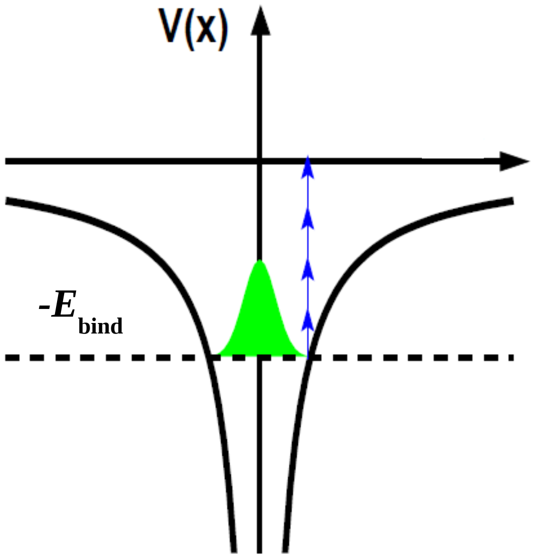


1 fs = 0.000000000000001 s

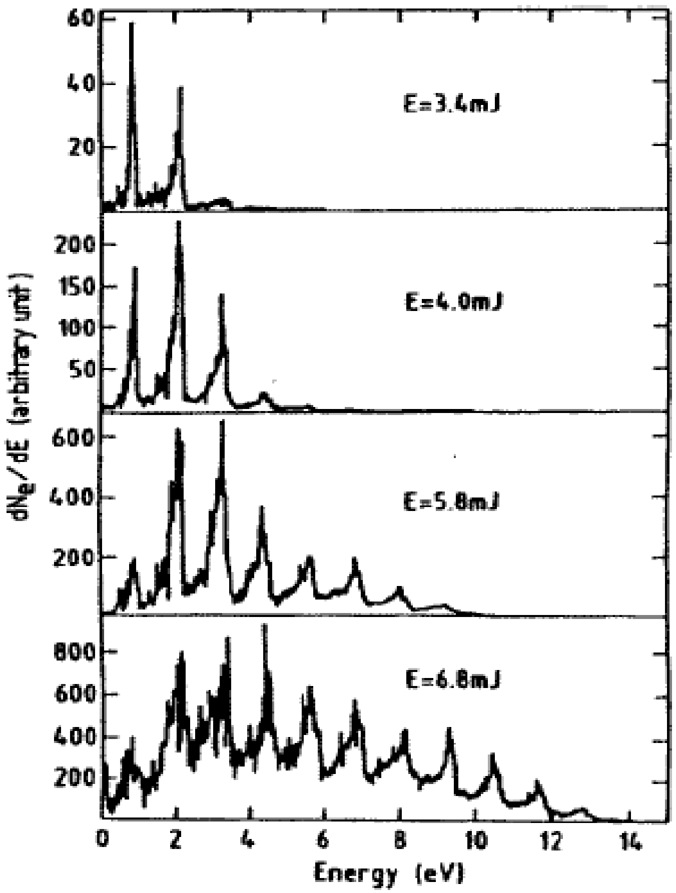
$$\frac{1\,\text{fs}}{1\,\text{s}} \simeq \frac{8\,\text{min}}{\text{age of the universe}}$$

Photo-effect in an intense laser field

Peaks at $E_{kin} = \boldsymbol{n} \cdot \hbar\omega - \varepsilon_{bind}$



Multi-photon ionisation



Yergeau *et al.*, JPB (1986)

Theory of multiphoton ionization

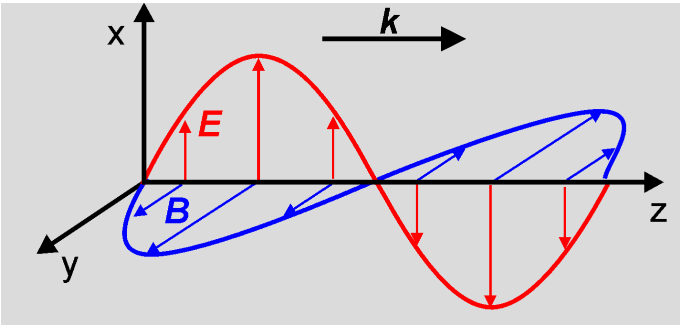
Theoretical description of laser wave

Maxwell's wave equation:

$$\left(\frac{\partial^2}{\partial t^2} - c^2\vec{\nabla}^2\right)\vec{A} = 0$$

Solved by plane waves:

$$\vec{A}(t,\vec{r}) = A_0 \cos(\omega t - \vec{k}\cdot\vec{r}\,)\,\vec{e}_x$$



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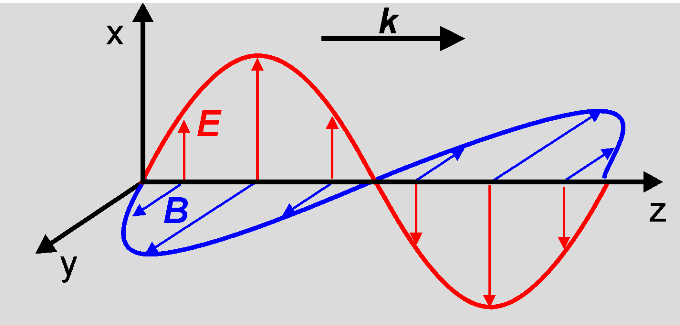

Very often, dipole approximation applies:

$$\vec{A}(t) = A_0\cos(\omega t)\vec{e}_x$$

- linear pol.

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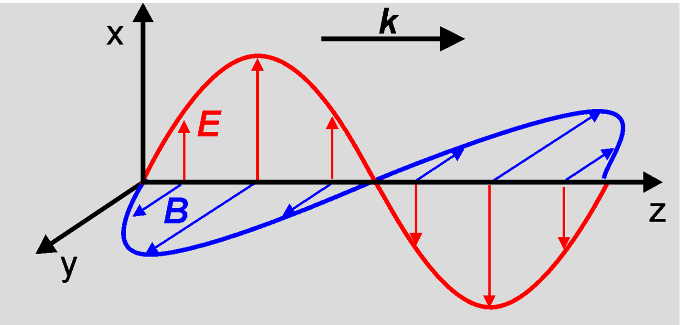

Very often, dipole approximation applies:

$$\vec{A}(t) = A_0 \cos(\omega t)\vec{e}_x$$ - linear pol.

$$\vec{A}(t) = A_0(\cos(\omega t)\vec{e}_x + \sin(\omega t)\vec{e}_y)$$ - circular pol.

Electron states in laser field

Time-dependent Schrödinger equation

$$i\hbar\frac{\partial\Psi}{\partial t} = \frac{1}{2m}\left(\hat{\vec{p}} + \frac{e}{c}\vec{A}\right)^2 \Psi$$

Ansatz: $\Psi = e^{\frac{i}{\hbar}\vec{p}\vec{r}}\phi(t)$

$$\Rightarrow \qquad \dot{\phi} = \frac{1}{2mi\hbar}\left(\vec{p} + \frac{e}{c}\vec{A}\right)^2 \phi$$

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Volkov state:

$$\Psi_p^V(\vec{r},t)=\frac{1}{\sqrt{V}}e^{\frac{i}{\hbar}\vec{p}\vec{r}}e^{-\frac{i}{2m\hbar}\int_{t_0}^{t}\left(\vec{p}+\frac{e}{c}\vec{A}(t')\right)^2dt'}$$

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free solution

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modulations due to field

Volkov states for circular polarization

$$\frac{1}{2m}\int_{t_0}^{t}\left(p^2+\frac{2e}{c}\vec{p}\cdot\vec{A}(t')+\frac{e^2}{c^2}A_0^2\right)dt'$$

$$=\epsilon_p t+\alpha_0[p_x\sin(\omega t)-p_y\cos(\omega t)]+Ut$$

with $\epsilon_p=\frac{p^2}{2m}$ $\alpha_0=\frac{eA_0}{\omega mc}$ $U=\frac{e^2A_0^2}{2mc^2}$

kinetic energy **excursion amplitude** **"Stark shift"**

Volkov state in closed form:

$$\Psi_p^V(\vec{r},t)=\frac{1}{\sqrt{V}}e^{\frac{i}{\hbar}(\vec{p}\vec{r}-\epsilon_p t)}e^{-\frac{i}{\hbar}(\alpha_0(p_x\sin(\omega t)-p_y\cos(\omega t))+Ut)}$$

Ionisation in laser field

Transition amplitude: $$S = \frac{1}{i\hbar}\int_{-\infty}^{\infty} dt \langle \Psi_p | \hat{W} | \Psi_a \rangle$$

Initial state: $$|\Psi_a\rangle = \frac{e^{-r/a_0}}{\sqrt{\pi a_0^3}} e^{-\frac{i}{\hbar}\epsilon_{100} t}$$

Interaction Hamiltonian: $$\hat{W} = \frac{e}{mc}\vec{A}\hat{\vec{p}} + \frac{e^2}{2mc^2}\vec{A}^2$$

Final state: should actually be associated with full Hamiltonian

$$\hat{H} = \frac{1}{2m}\hat{\vec{p}}^2 + \hat{W} + \hat{V}_C$$

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Ignoring $\hat{V}_C$ is called **strong-field approximation (SFA)**

Ionisation in laser field

$$S = \frac{1}{i\hbar}\int_{-\infty}^{\infty} dt \int d^3r \, \frac{1}{\sqrt{V}} e^{-\frac{i}{\hbar}(\vec{p}\vec{r}-\epsilon_p t)} e^{\frac{i}{\hbar}[\alpha_0(p_x \sin(\omega t)-p_y\cos(\omega t))+Ut]}$$

$$\times\left(\frac{e}{mc}\vec{A}\vec{p}+U\right)\frac{e^{-r/a_0}}{\sqrt{\pi a_0^3}} e^{-\frac{i}{\hbar}\epsilon_{100}t}$$

Spatial integral: $\frac{1}{\sqrt{V\pi a_0^3}}\int d^3r \, e^{-\frac{i}{\hbar}\vec{p}\vec{r}} e^{-r/a_0} = \frac{1}{\sqrt{V}}\frac{8\sqrt{\pi}a_0^{3/2}}{\left(1+\left(\frac{pa_0}{\hbar}\right)^2\right)^2} =: \tilde{\phi}_{100}(p)$

Ionisation in laser field

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Tools for time integral:

$$p_x \sin(\omega t) - p_y \cos(\omega t) = p_\perp \sin(\omega t - \varphi_p)$$

$$e^{-iz\sin(\omega t)} = \sum_{n=-\infty}^{\infty} J_n(z) e^{-in\omega t}$$

Ionisation in laser field

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Ionisation in laser field

$$S = -2\pi i \sum_{n=-\infty}^{\infty} J_n\left(\frac{-\alpha_0 p_\perp}{\hbar}\right)\tilde{\phi}_{100}(p)e^{in\varphi_p}(U - n\hbar\omega)\delta(\epsilon_p + U - \epsilon_{100} - n\hbar\omega)$$

number of photons!

Discrete photo-electron momenta:

$$p_n = \sqrt{2m(\epsilon_{100} + n\hbar\omega - U)}$$

Ionisation in laser field

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Differential ionisation rate:

$$\frac{d\dot{W}}{d\Omega} = \frac{1}{T}\int \frac{Vp^2dp}{(2\pi\hbar)^3}\left|S\right|^2$$

$$= \sum_{n=n_{\min}}^{\infty} \frac{2\pi m p_n}{\hbar(2\pi\hbar)^3}(U - n\hbar\omega)^2 V\left|\tilde{\phi}_{100}(p_n)\right|^2 J_n^2\left(\frac{-\alpha_0 p_n \sin(\theta)}{\hbar}\right)$$

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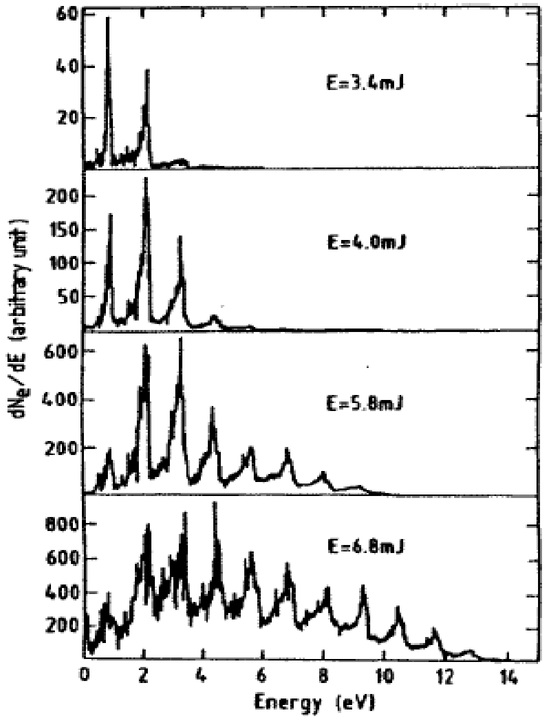

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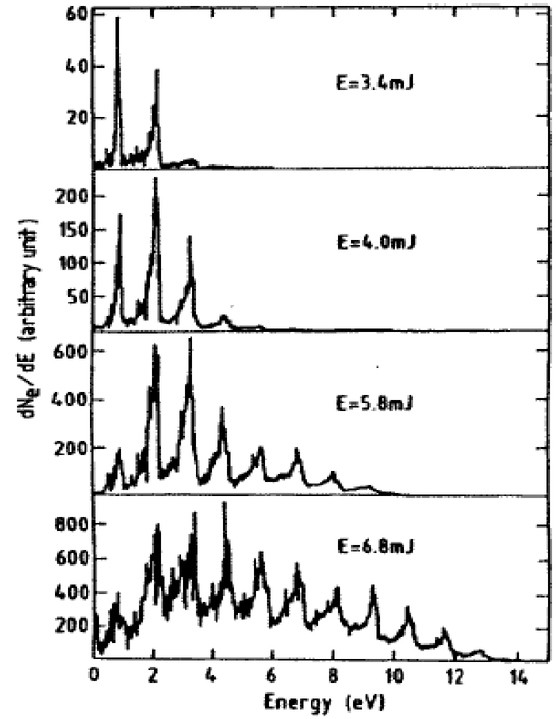


Differential ionisation rate:

$$\frac{d\dot{W}}{d\Omega} = \frac{1}{T}\int \frac{Vp^2dp}{(2\pi\hbar)^3}|S|^2$$

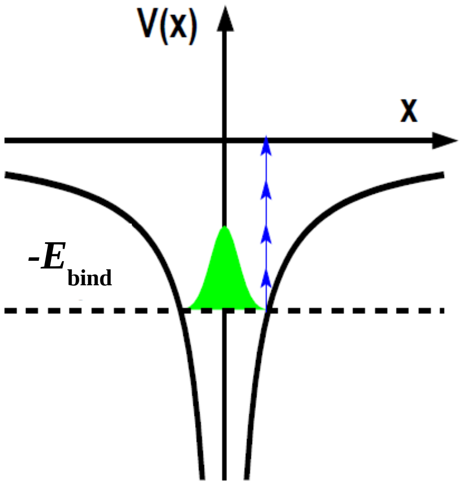
generalizes rules of usual photo-effect

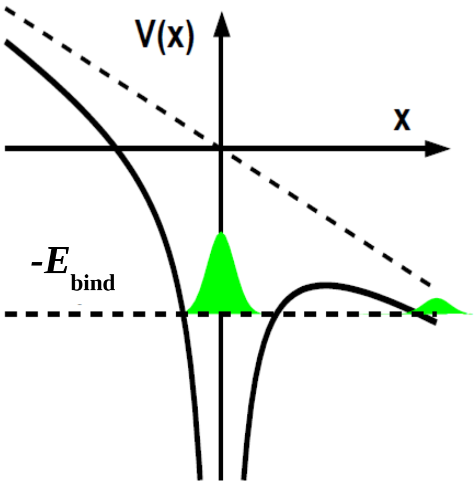
$$= \sum_{n=n_{\min}}^{\infty} \frac{2\pi m p_n}{\hbar(2\pi\hbar)^3}(U - n\hbar\omega)^2 V\left|\tilde{\phi}_{100}(p_n)\right|^2 J_n^2\left(\frac{-\alpha_0 p_n \sin(\theta)}{\hbar}\right)$$

$\sim A^{2n}$

Transforming many small photons into a big one

Multiphoton versus tunneling ionization





multiphoton ionization ($\kappa \gg 1$)

rate $\sim I^n$

photon aspect dominates

tunneling ionization ($\kappa \ll 1$)

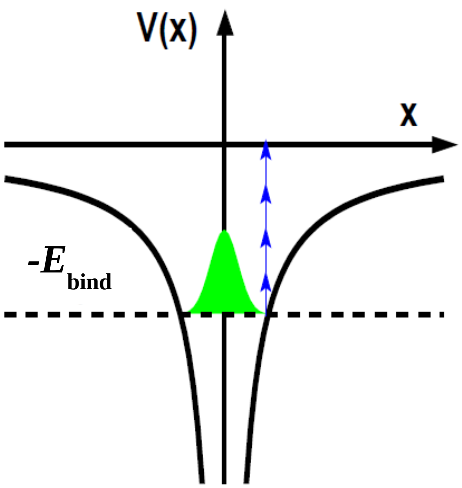
rate $\sim \exp(-E_{at}/E_L)$

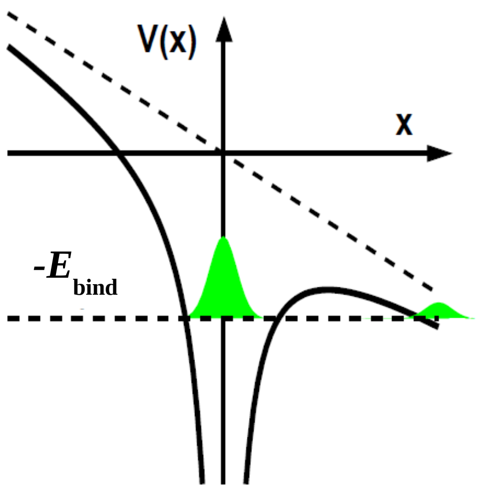
field aspect dominates

Multiphoton versus tunneling ionization

Keldysh parameter :

$$\kappa = \omega_L / \omega_{\text{tun}} \sim \omega_L / E_L$$





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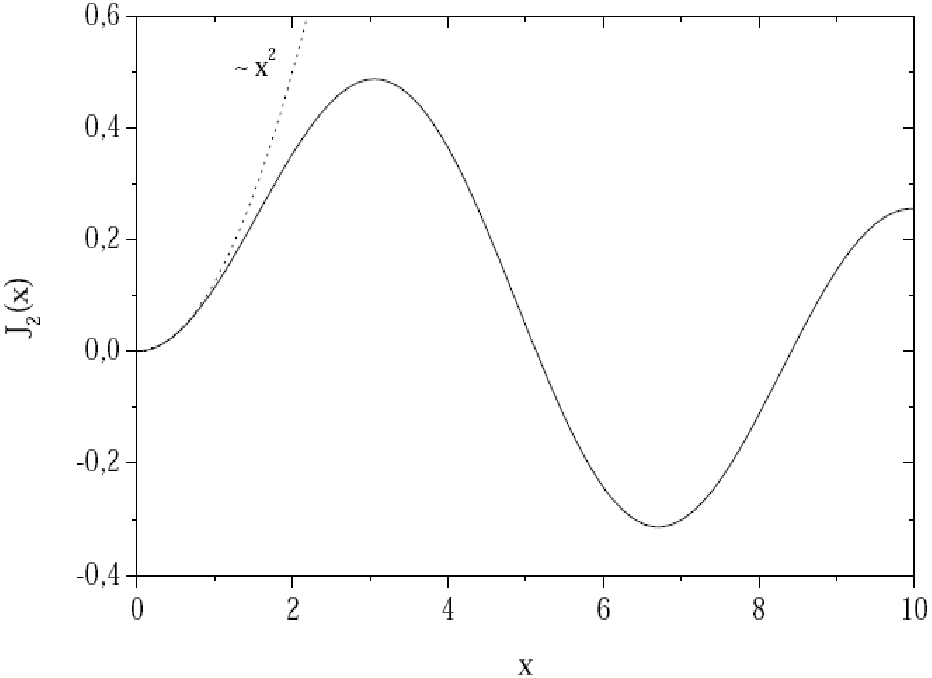
tunneling ionization ($\kappa \ll 1$)

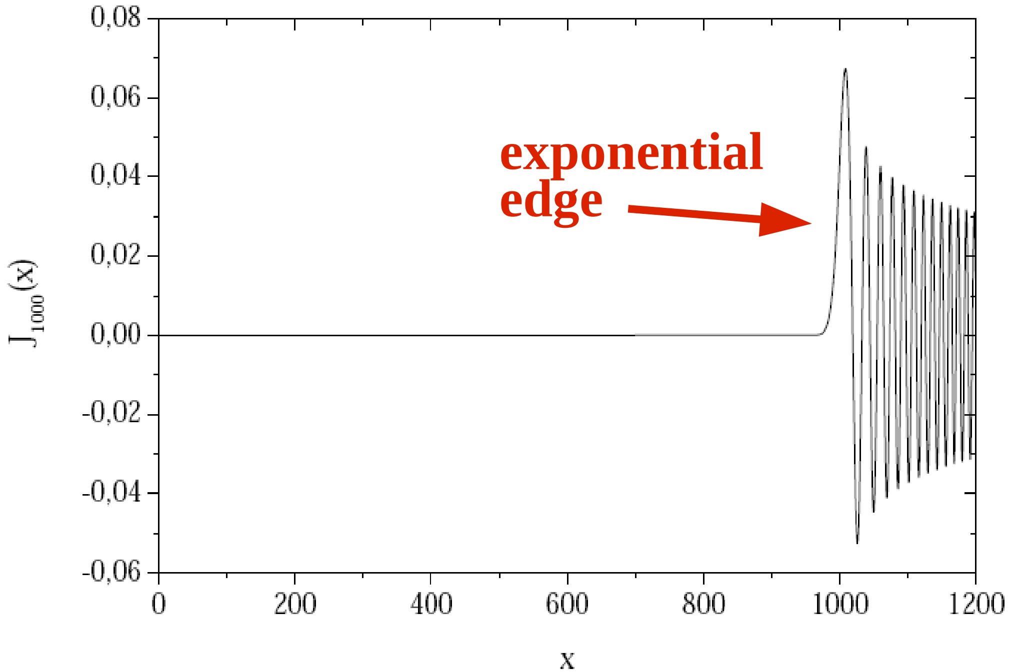
rate $\sim \exp(-E_{\text{at}} / E_L)$

field aspect dominates

Bessel functions

$$f(t) = \sum_{n=-\infty}^{+\infty} J_n(x)\, e^{-in\omega_0 t}$$





multiphoton ionization

$$\text{rate} \sim I^n$$

tunneling ionization

$$\text{rate} \sim \exp(-E_{at}/E_L)$$

Motion of a free electron in a (weak) laser field

$$m\vec{a} = \vec{F} = q\vec{E} = eE_0 sin\,(\omega\,t)\vec{x}$$

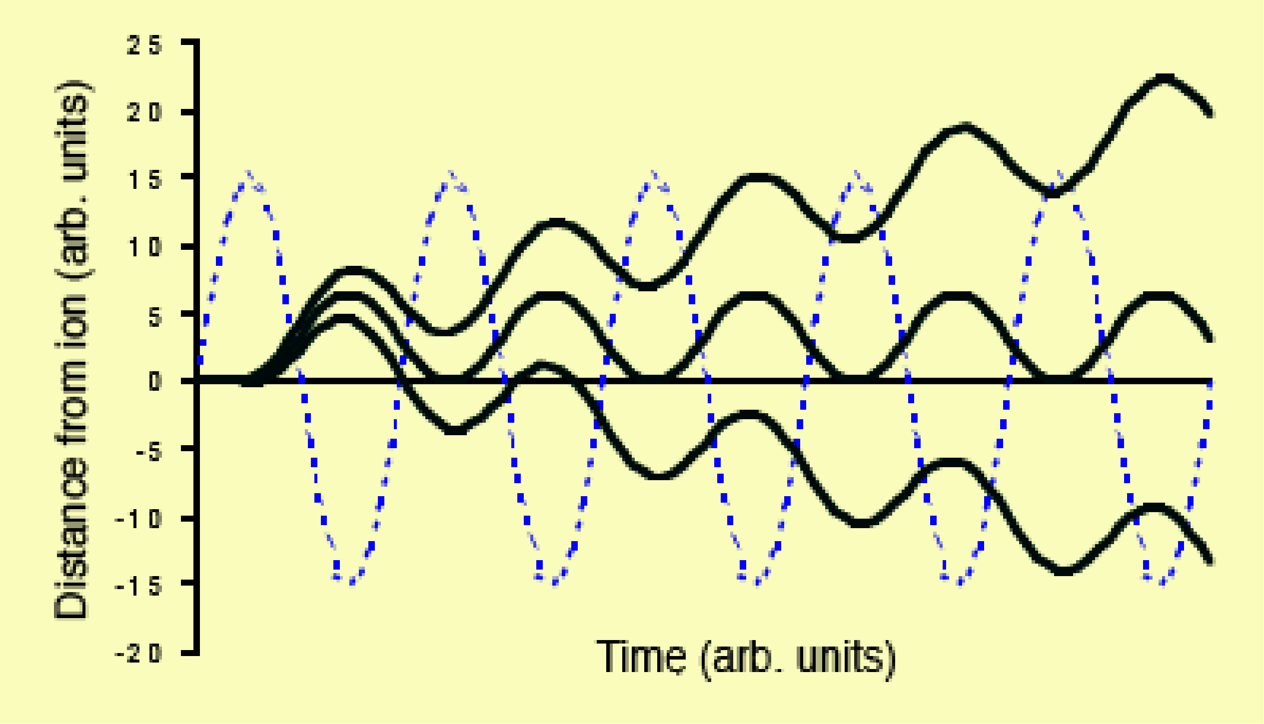
$$v_x = -\frac{eE_0}{m\omega}cos\,(\omega\,t) + v_0$$

$$x = -\frac{eE_0}{m\omega^2}sin\,(\omega\,t) + v_0 t + x_0$$

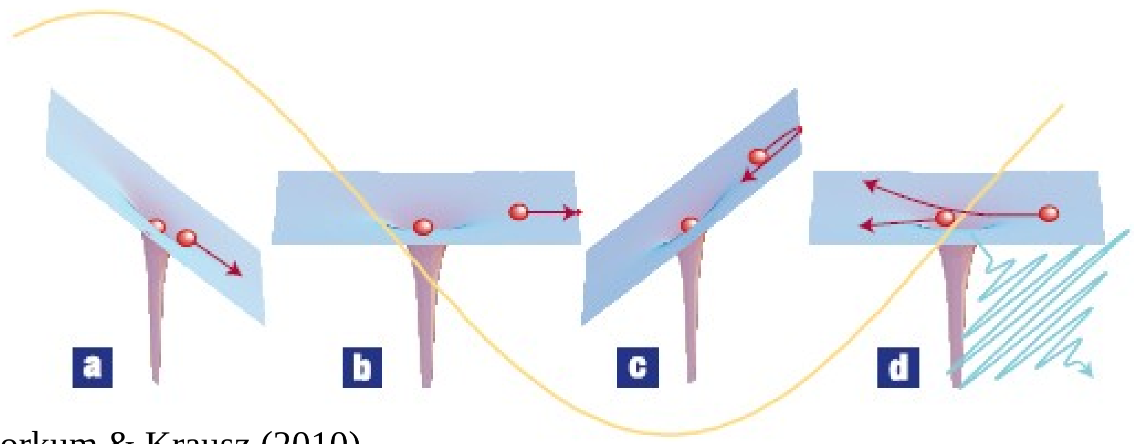
Classical EoM

Recall: $\alpha_0 = \frac{eA_0}{\omega mc}$

For v_0=0 and x_0=0:
multiple periodic returns to the origin (e.g. the ionic core)



Laser-driven electron-ion recollisions



Corkum & Krausz (2010)

Three-step model:

1) tunnel ionization

2) field propagation

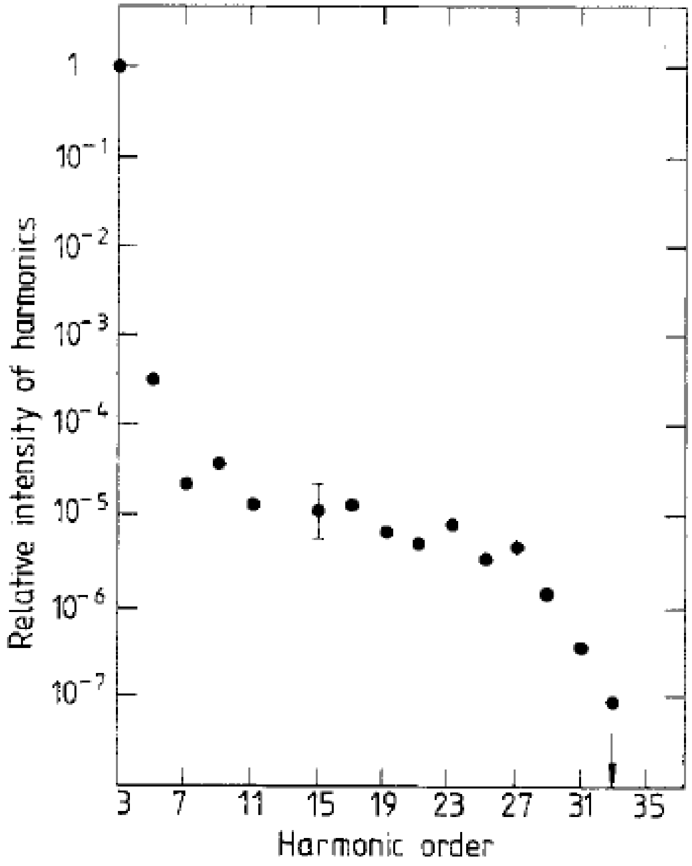
3) recollision

Recollision can lead to...

... scattering
... double ionization
... recombination

$\rightarrow$ *High-harmonic generation*: $\Omega = N\omega$

High-harmonic spectra

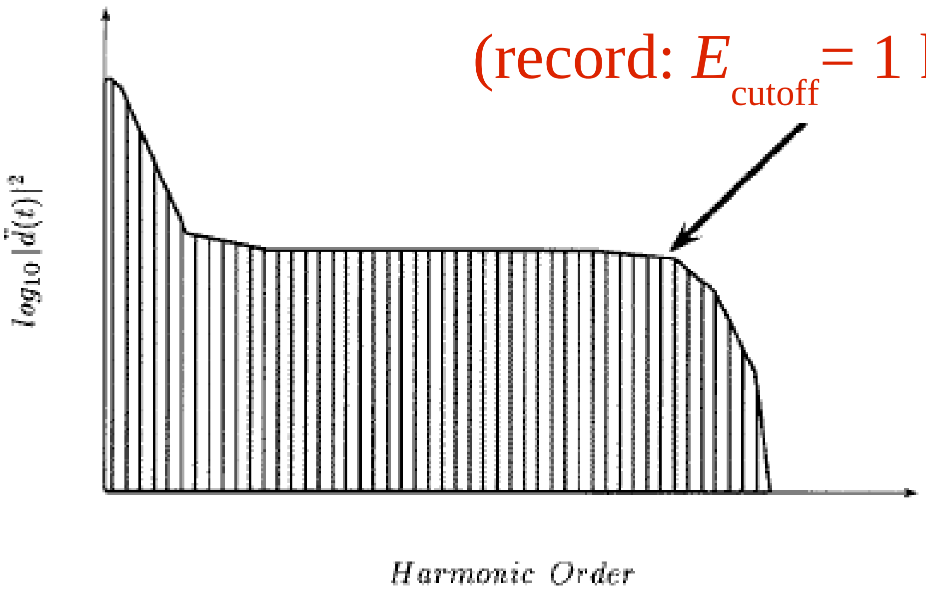


Ferray *et al.*, JPB **21** (1988):

10^{13} W/cm^2 at 1000 nm in Ar

$E_{cutoff} = I_p + 3.17\ U_p$

(record: E_{cutoff} = 1 keV!)



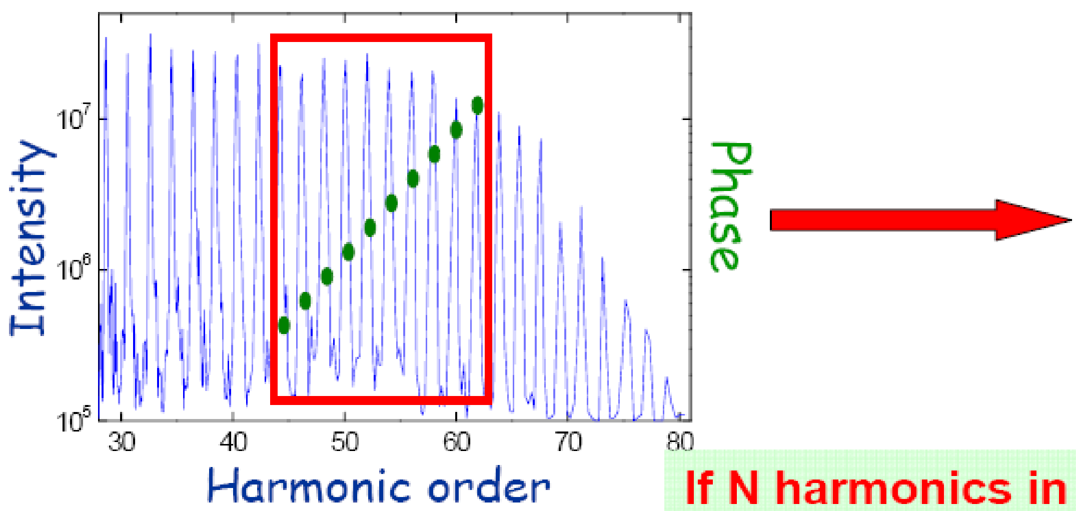
Important application:

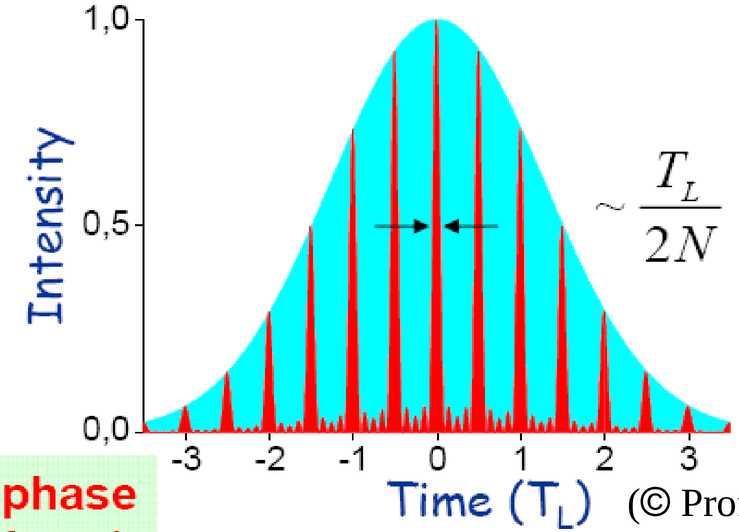
Generation of “attosecond pulses”

(T < 1 fs)

Attosecond laser pulses

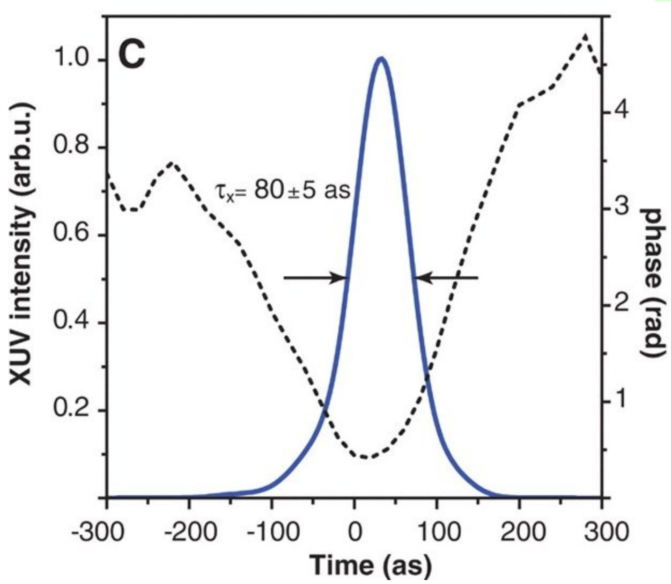
Novel application of HHG: Generation of attosecond pulses (T<1fs)





(© Prof. Saliere)

If N harmonics in phase (linear spectral phase)

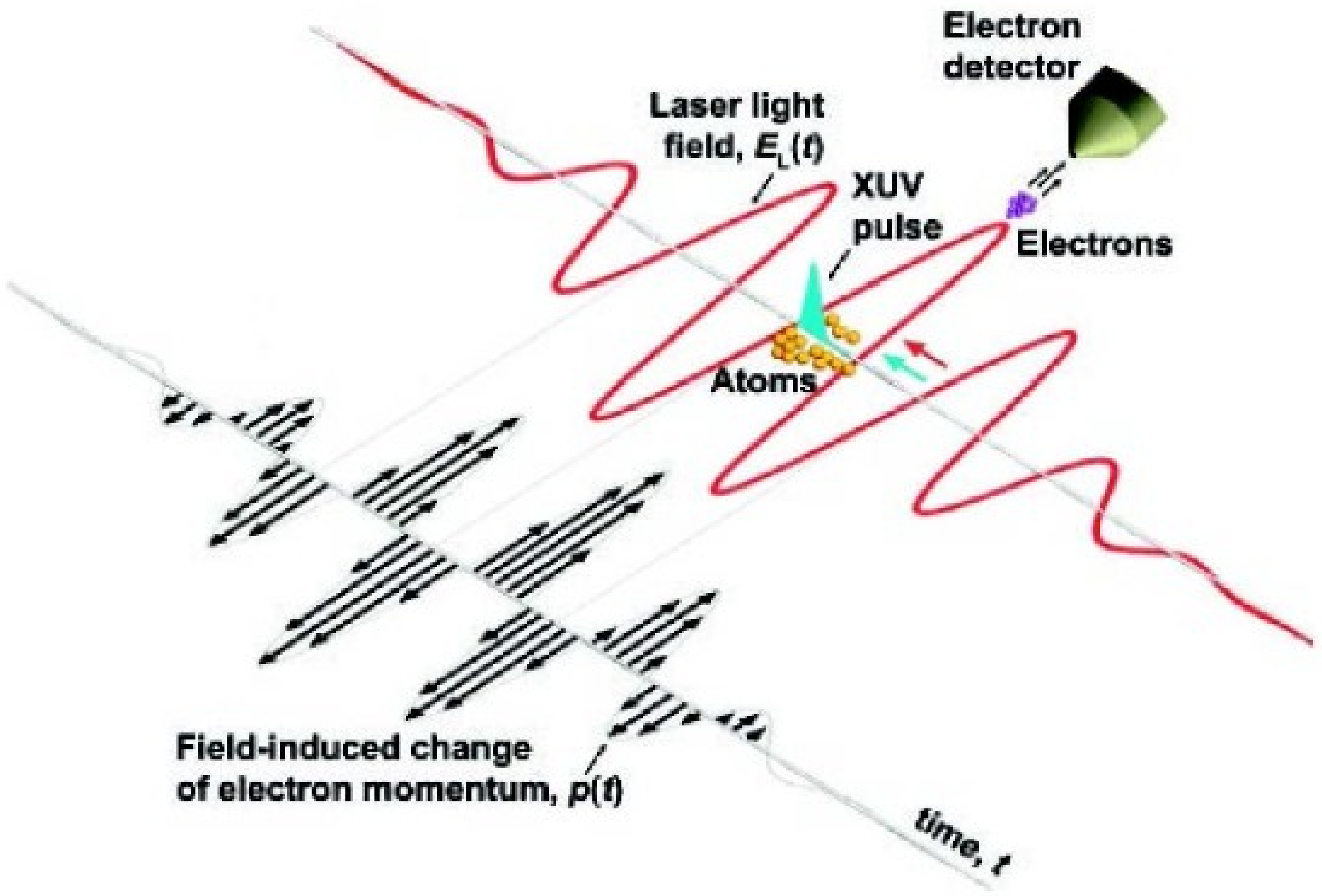


Goulielmakis *et al.*, Science (2008)

Why is it interesting?

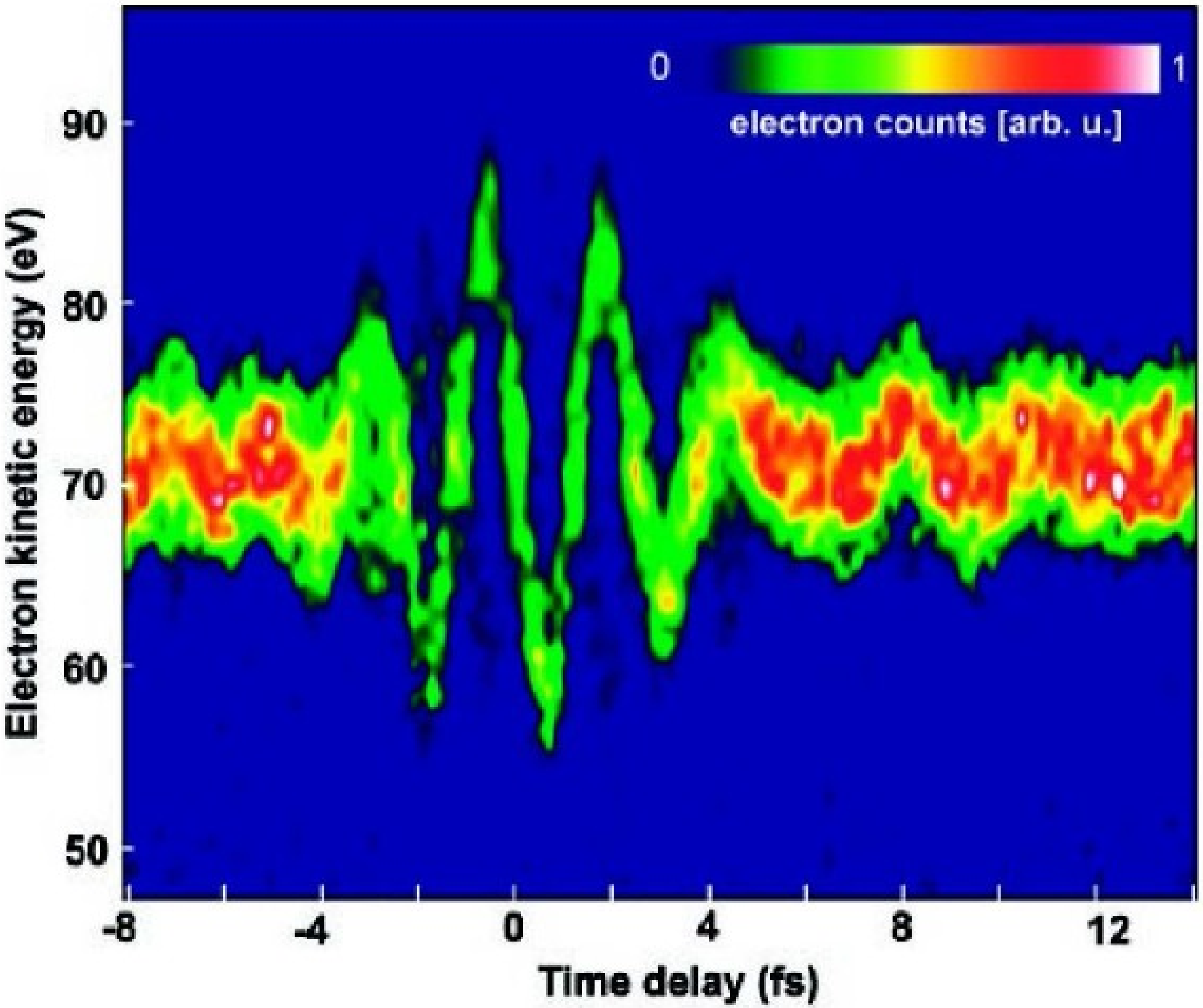
Time scale of electron motion in atoms is ~ 100 as!

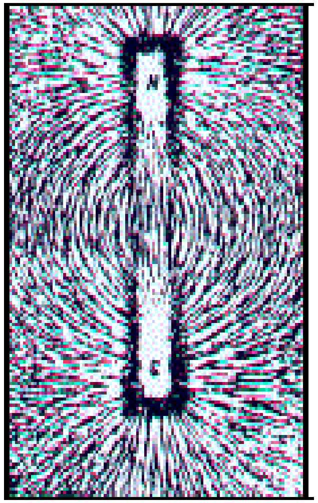
Attosecond streak camera



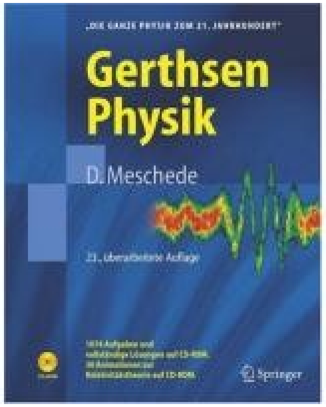
Goulielmakis *et al.*, Science (2004)

“Photograph” of femtosecond laser pulse





static magnetic field



Goulielmakis *et al.*, Science (2004)

History of multiphoton physics

A short history of multiphoton physics

Perturbative few-photon processes:

1931: Theory of two-photon absorption (M. Göppert-Mayer)

1961: Second harmonic generation in laser-crystal interaction

Two-photon ionization of atoms

Nonperturbative multiphoton era:

1979: Discovery of above-threshold ionization

1988: Observation of high-order harmonic generation

Towards high-energy multiphoton physics:

1997: Laser-induced electron-positron pair creation

1999: Nuclear fusion in laser-heated deuterium clusters

2006: Generation of 1 GeV electron beams by laser acceleration

10^6 W/cm^2

Mode locking

10^{14} W/cm^2

CPA

$>10^{18}$ W/cm^2

First evidence for a second harmonic?

"...exploiting extraordinary ruby laser intensities of 10^6 W/cm^2 "

VOLUME 7, NUMBER 4 PHYSICAL REVIEW LETTERS AUGUST 15, 1961

GENERATION OF OPTICAL HARMONICS*

P. A. Franken, A. E. Hill, C. W. Peters, and G. Weinreich
The Harrison M. Randall Laboratory of Physics, The University of Michigan, Ann Arbor, Michigan
(Received July 21, 1961)

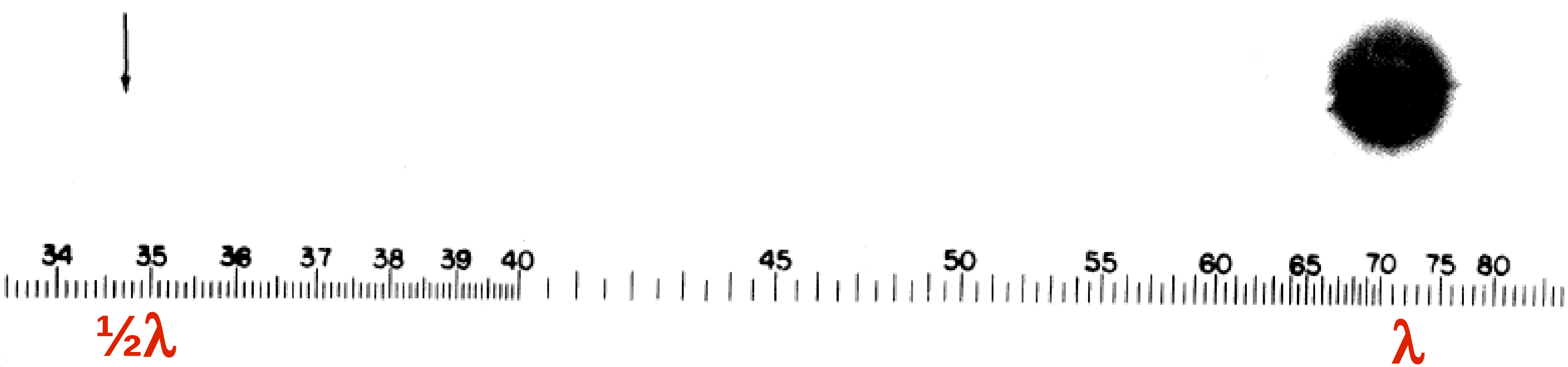


FIG. 1. A direct reproduction of the first plate in which there was an indication of second harmonic. The wavelength scale is in units of 100 A. The arrow at 3472 A indicates the small but dense image produced by the second harmonic. The image of the primary beam at 6943 A is very large due to halation.

The production editors accidentally removed the "small piece of dirt"...

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Further multiphoton effects

Multiphoton Thomson scattering

Transition amplitude:

$$S = \frac{1}{i\hbar} \int_{-\infty}^{+\infty} dt \ \langle \Psi_{p'}^{V} | H_{\text{int}} | \Psi_{p}^{V} \rangle$$

Interaction Hamiltonian:

$$H_{\text{int}} = \frac{e}{mc} \left(\hat{\vec{p}} + \frac{e}{c} \vec{A}_L \right) \cdot \hat{\vec{A}}'$$

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Scattered photon: $$\hat{\vec{A}}' = \sqrt{\frac{2\pi\hbar c^2}{V\omega'}}\, e^{i(\omega' t - \vec{k}'\cdot\vec{r})}\, \boldsymbol{\epsilon}_{k'}\, \hat{c}_{k'}^{\dagger}$$

Rate: $$\frac{d\dot{W}}{d\Omega_{k'}} = \int \frac{V\,d^3p'}{(2\pi\hbar)^3}\int \frac{V\omega'^2\,d\omega'}{(2\pi c)^3}\frac{|S|^2}{T} = \sum_{n=1}^{\infty}\frac{d\dot{W}_n}{d\Omega_{k'}}$$

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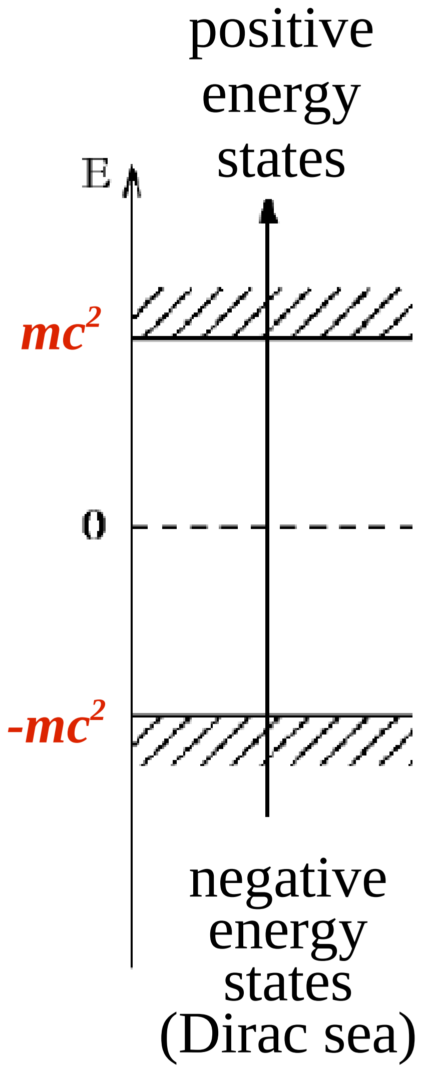
$$\varepsilon_{p'} + \hbar\omega' = \varepsilon_p + n\hbar\omega_L$$

"Photo-effect" from the quantum vacuum



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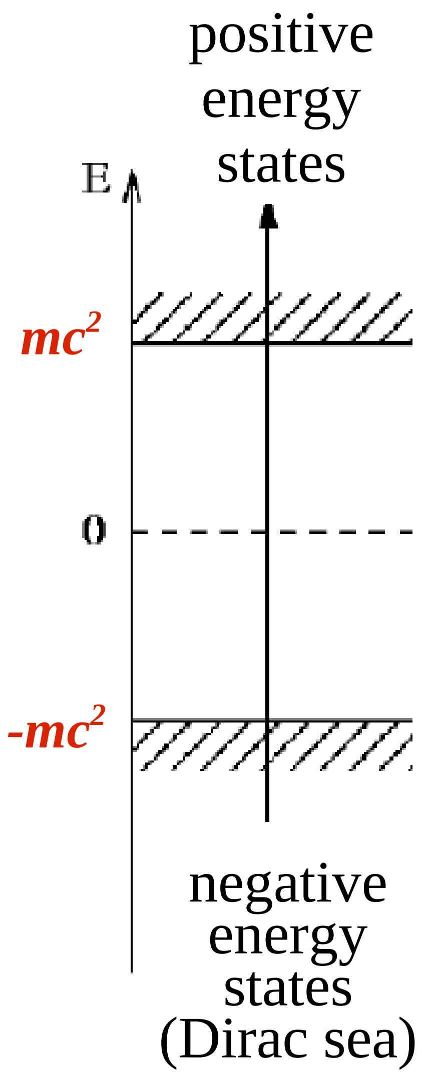
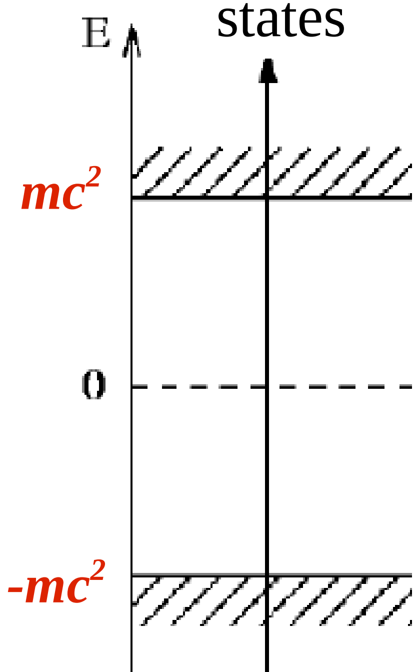


Production of electron-positron pairs according to $E=mc^2$ from laser photons possible, if $\hbar\omega \approx mc^2$ or $eE_L\lambda_C \approx mc^2$ ($I_{cr} \sim 10^{29}$ W/cm^2)

"Photo-effect" from the quantum vacuum

positive
energy
states



negative
energy
states
(Dirac sea)

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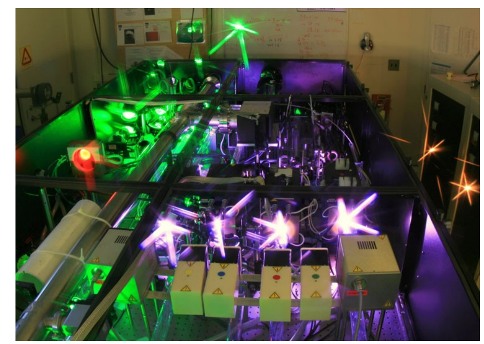
Available frequencies & intensities much smaller:

Free-electron laser:

$\hbar\omega \sim 10^{-4} mc^2$

Multiphoton regime:

$W \sim 10^{-100000}$



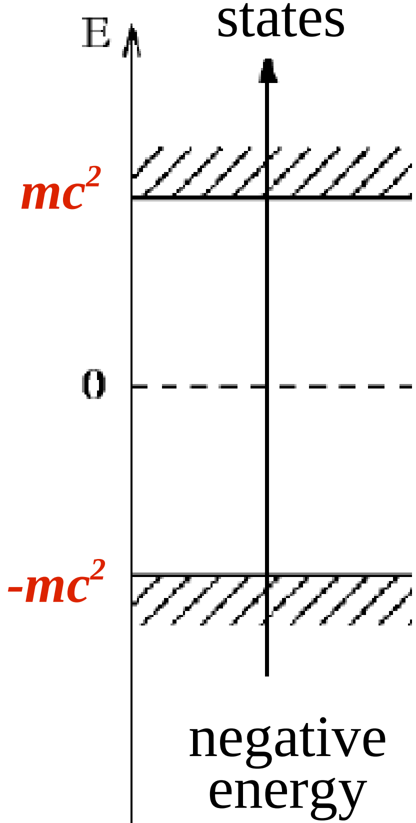
Petawatt laser:

$E_L \sim 10^{-4} E_{cr}$

Tunneling regime:

$W \sim 10^{-5000}$

"Photo-effect" from the quantum vacuum



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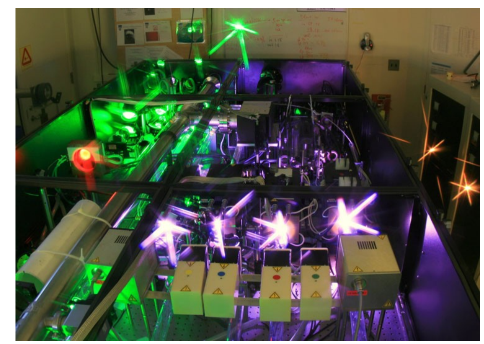


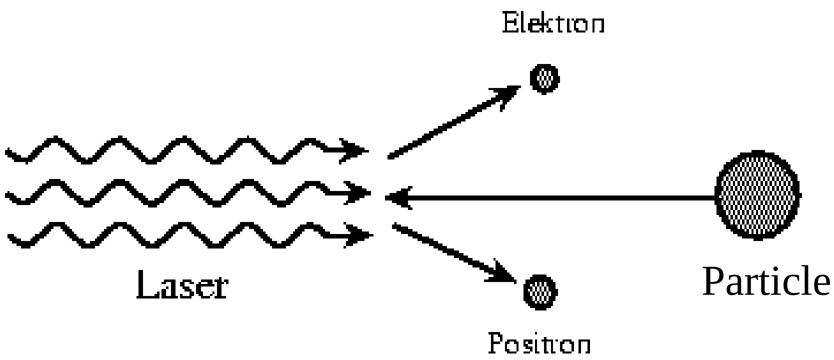
Petawatt laser:

$E_L \sim 10^{-4}\, E_{cr}$

"The cross section for this process at optical frequencies or below is so small at any laser intensity as to make it completely negligible. It may be the smallest (nonzero) cross section on record."

(M. Mittleman, 1987)

Relativistic particle beam colliding with laser pulse

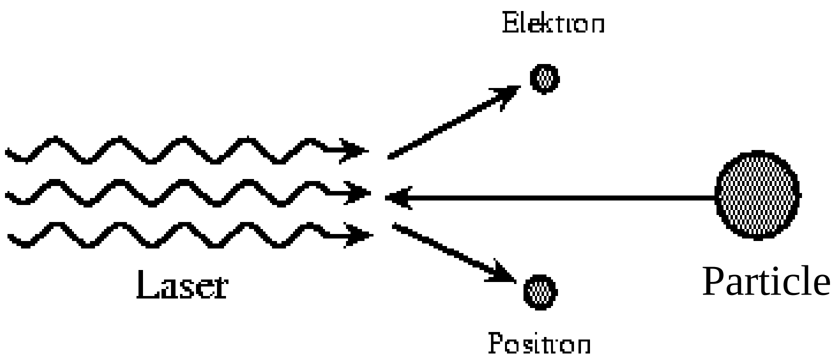


Exploit relativistic Doppler shift

lab frame: $\hbar\omega \approx 100$ eV , $E \approx 10^{12}$ V/cm

rest frame: $\hbar\omega'$ and E' enhanced by 2γ

Relativistic particle beam colliding with laser pulse



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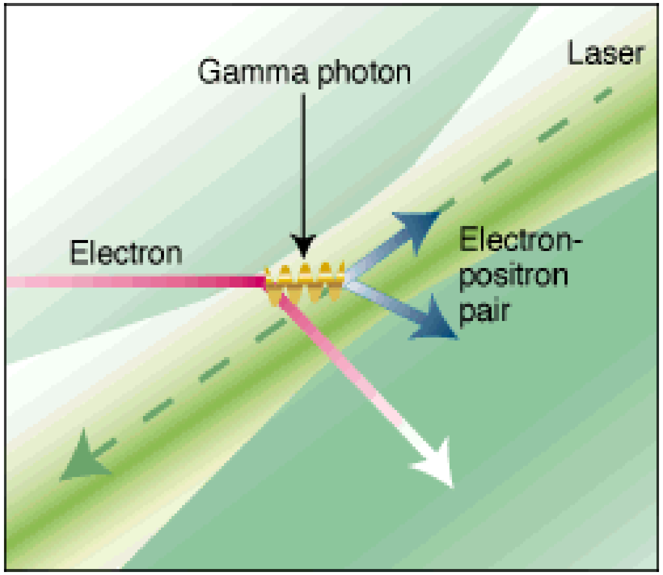
SLAC experiment:
46 GeV electron + optical laser pulse
(D. Burke et al., PRL 1997)

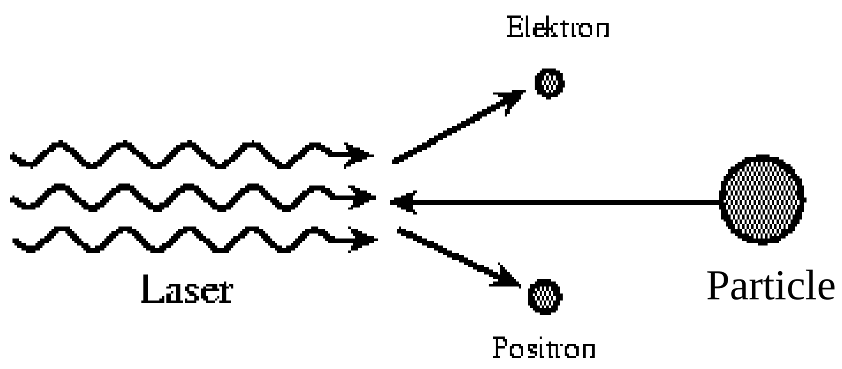


Pairs were produced in two-step process through an intermediate high-energy Compton photon:

$$\Omega_c + n\omega \rightarrow e^+e^-$$

(nonlinear Breit-Wheeler process)

Relativistic particle beam colliding with laser pulse



Exploit relativistic Doppler shift

lab frame: $\hbar\omega \approx 100$ eV , $E \approx 10^{12}$ V/cm

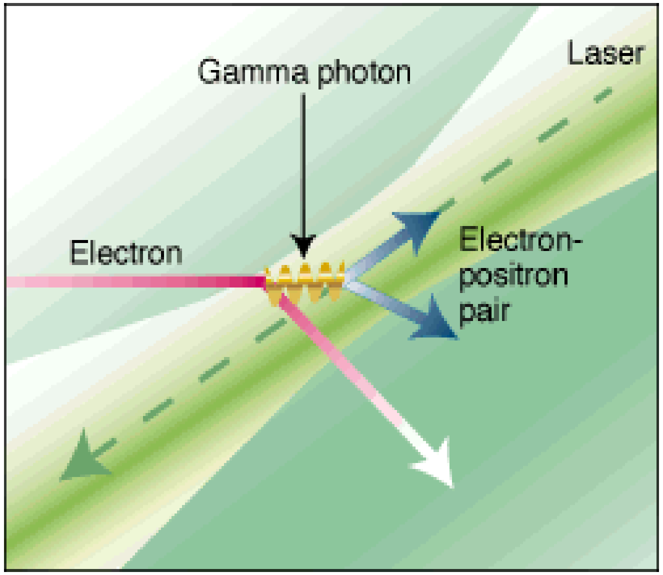
rest frame: $\hbar\omega'$ and E' enhanced by 2γ

For heavy projectiles such as nuclei
Compton channel strongly suppressed:
pairs would be produced directly
by nuclear Coulomb field:

$$\mathbf{Z + n\omega \rightarrow Z + e^+e^-}$$

(nonlinear Bethe-Heitler process)

SLAC experiment:
46 GeV electron + optical laser pulse
(D. Burke et al., PRL 1997)



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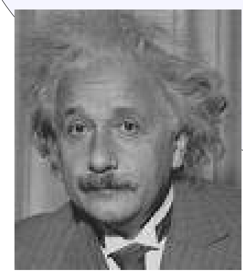
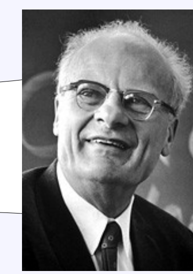
Summary

Atomic
Physics



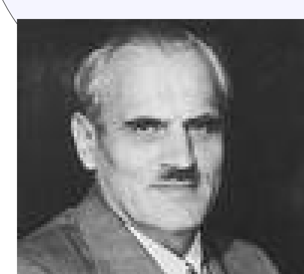
Nuclear
&
Particle Physics



**Multiphoton
Physics**

Attosecond
Physics

QED and Relativistic
Physics



Take-home message:

Unity is strength!

(Gemeinsam sind wir stark!)