2019/10/23

Advanced Radiation Engineering

放射線応用工学E

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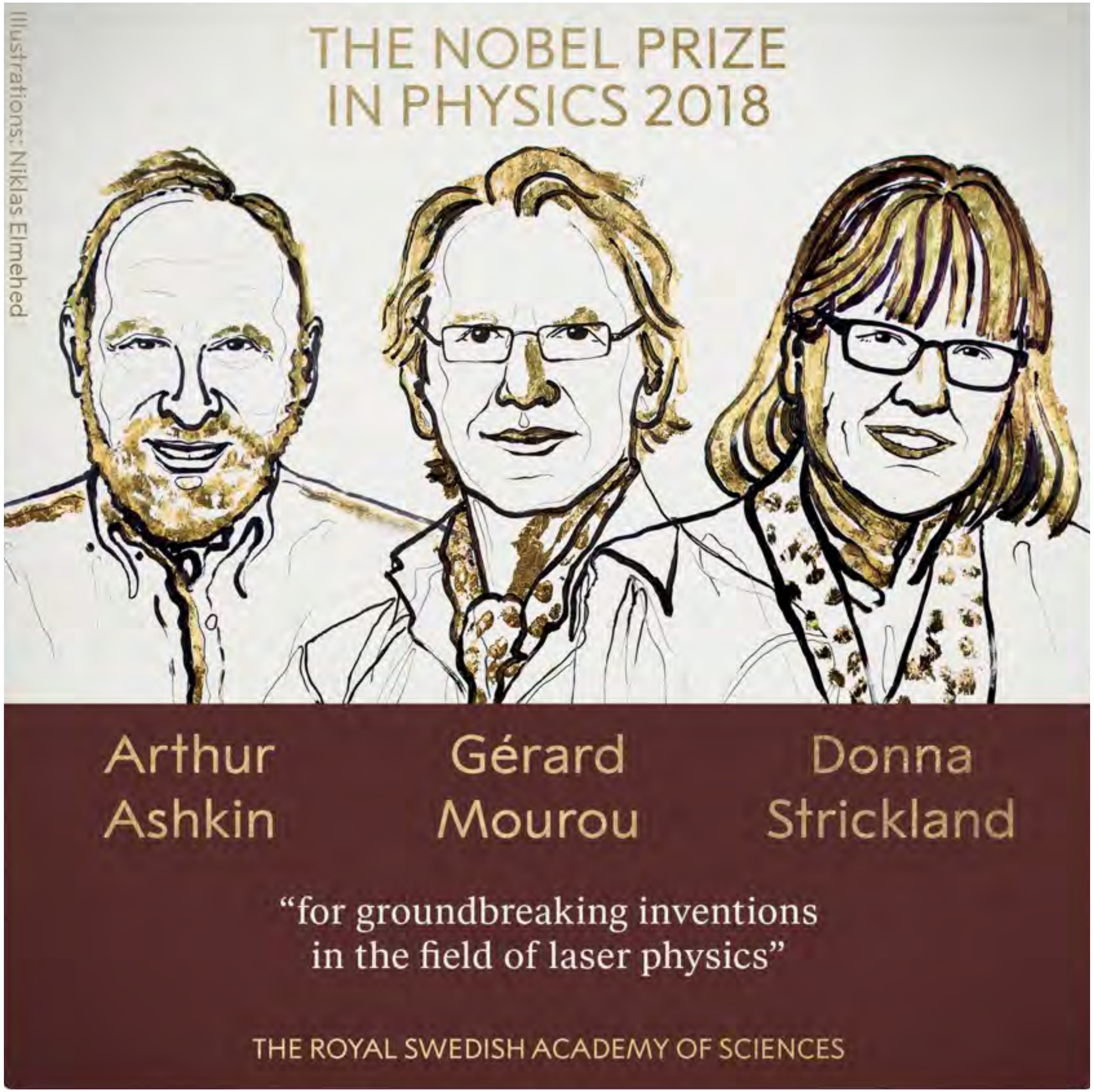
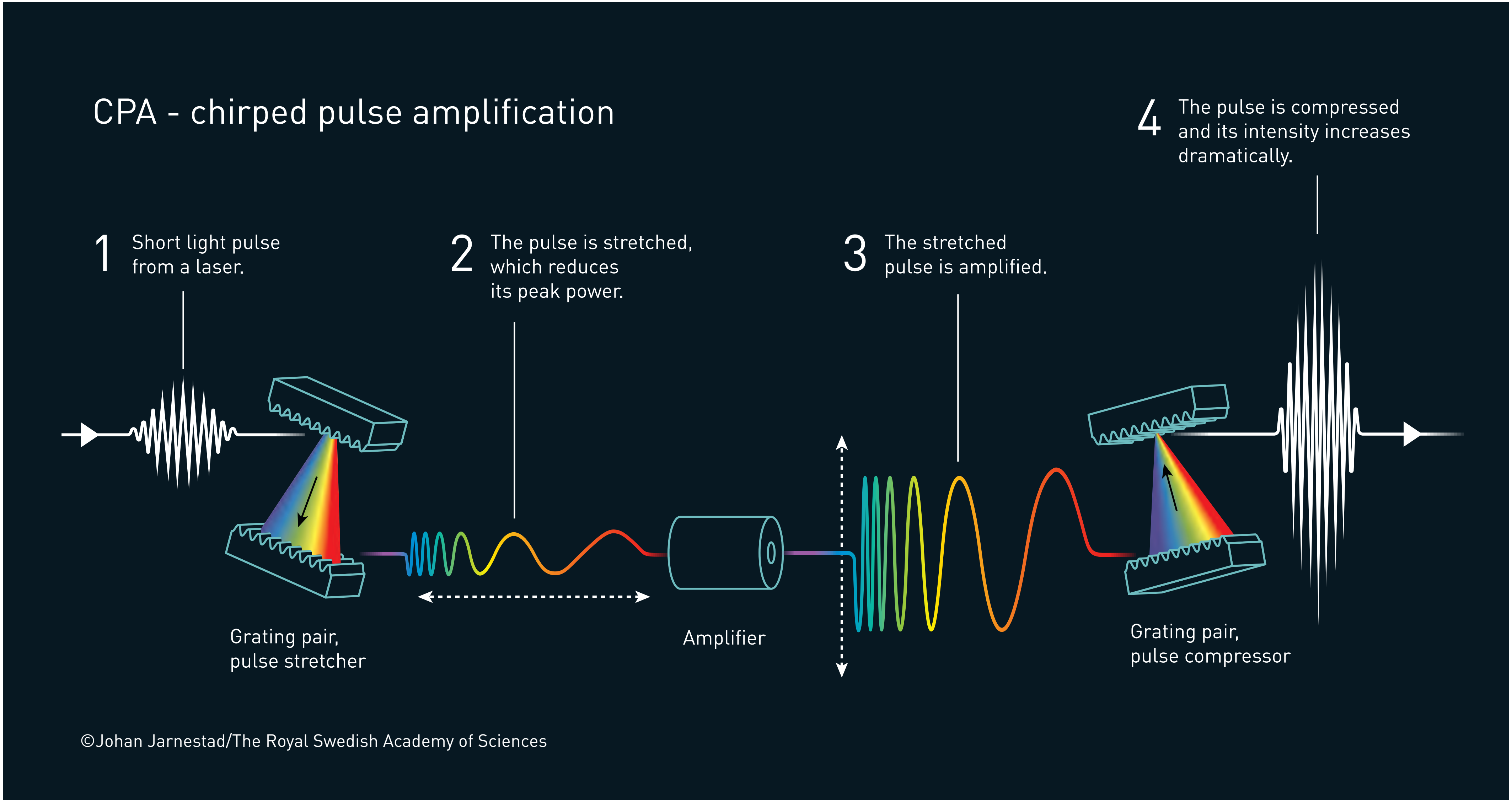
高次高調波発生と アト秒科学

High harmonic generation & Attosecond science



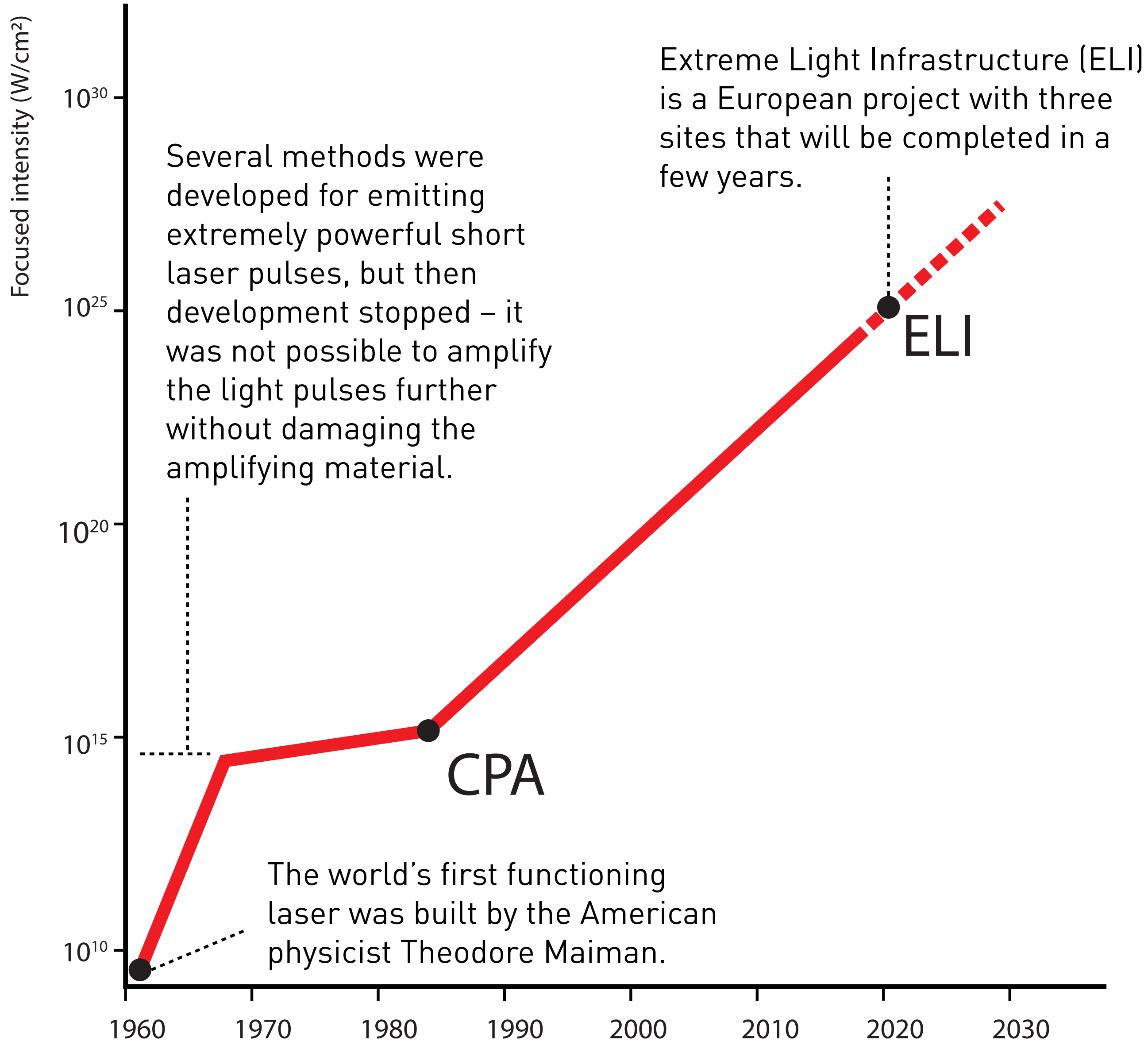
Optical tweezers

method of generating high-intensity, ultra-short optical pulses



https://6702d.https.cdn.softlayer.net/2019/10/pop_fy_en_18.pdf

history of pulsed laser technology in terms of peak intensity



https://6702d.https.cdn.softlayer.net/2019/10/pop_fy_en_18.pdf

"Scientific Background on the Nobel Prize in Physics 2018" lists applications of CPA technology

https://www.nobelprize.org/uploads/2018/10/advanced-physicsprize2018.pdf

- Strong-field physics and attosecond science ← today's topic
- Laser-plasma acceleration
- High-intensity lasers in industry and medicine

High-harmonic generation

(= one of the strong-field phenomena)

高次高調波発生

References 参考文献

- The lecture material is downloadable from:
  http://ishiken.free.fr/english/lecture.html
- M. Protopapas, C.H. Keitel and P.L. Knight, “Atomic physics with super-high intensity lasers”, Rep. Prog. Phys. **60**, 389–486 (1997)
- F. Krausz and M. Ivanov, “Attosecond Physics”, Rev. Mod. Phys. **81**, 163-234 (2009)
- K. L. Ishikawa, High-harmonic generation, in Advances in Solid-State Lasers, ed. by M. Grishin (INTEH, 2010), pp. 439-464
- 大森賢治編「アト秒科学: 1京分の1秒スケールの超高速現象を光で観測・制御する」（化学同人、2015/8/10）

高調波発生 (Harmonic generation)

結晶、ガス等(crystal, gas)

Linear optical effect
線形光学効果（弱い光）

Material response is linear in light intensity 物質の応答が、入射光強度に比例

非線形光学効果（強い光）
Nonlinear optical effect

Nonlinear material response
物質の応答が、入射光強度に非線形に依存

波長変換
(frequency conversion)

$$D = \varepsilon_0 E + P$$

$$P = \varepsilon_0 \left[\chi^{(1)} E + \chi^{(2)} E^2 + \chi^{(3)} E^3 + \cdots \right]$$

非線形分極 (nonlinear)

線形分極 linear polarization

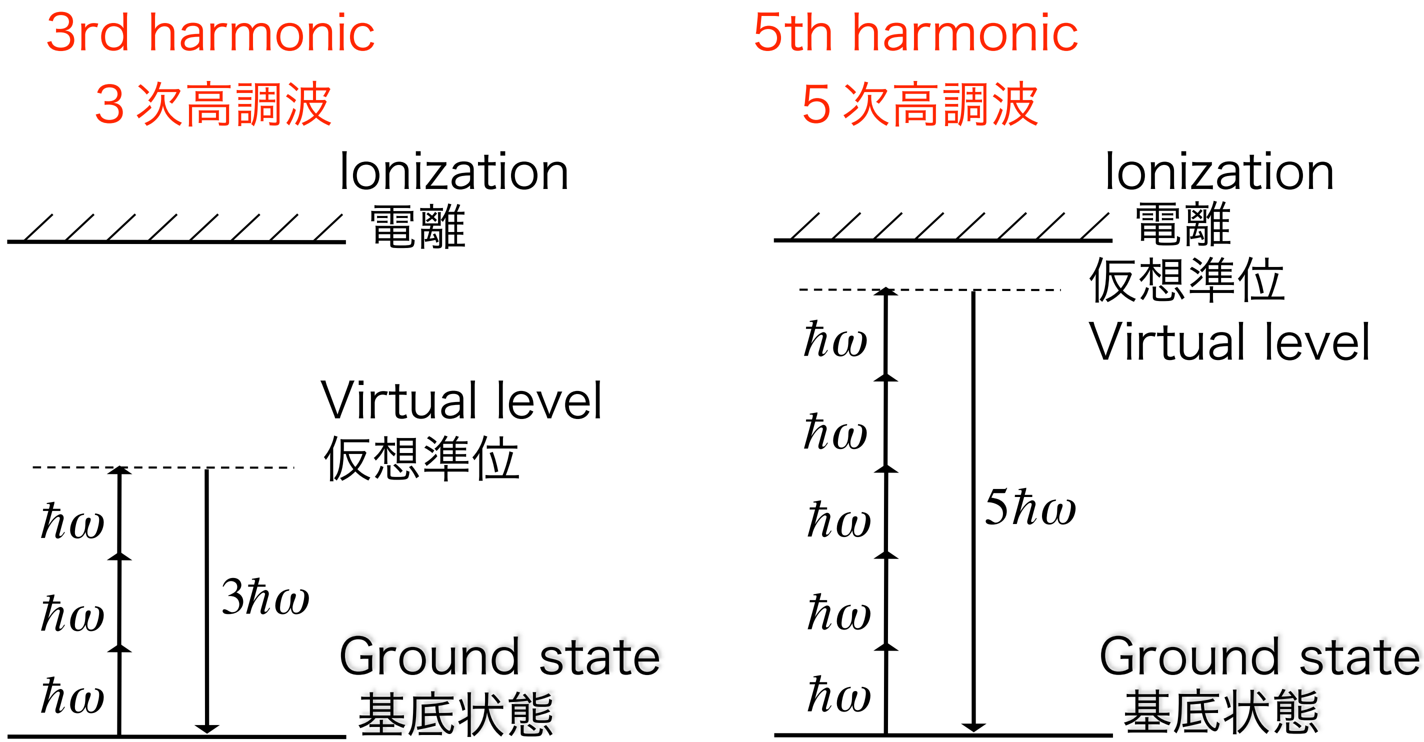
反転対称な媒質では、 $\chi^{(2)} = 0$
for a medium with inversion symmetry

$$\nabla \times \nabla \times \mathbf{E} = -\mu_0 \frac{\partial^2 \mathbf{D}}{\partial t^2}$$

3ω： 3 次高調波(3rd harmonic)

5ω： 5 次高調波(5th harmonic)

摂動論的高調波発生
(perturbative harmonic generation)



次数が高くなるほど、発生効率は減少。

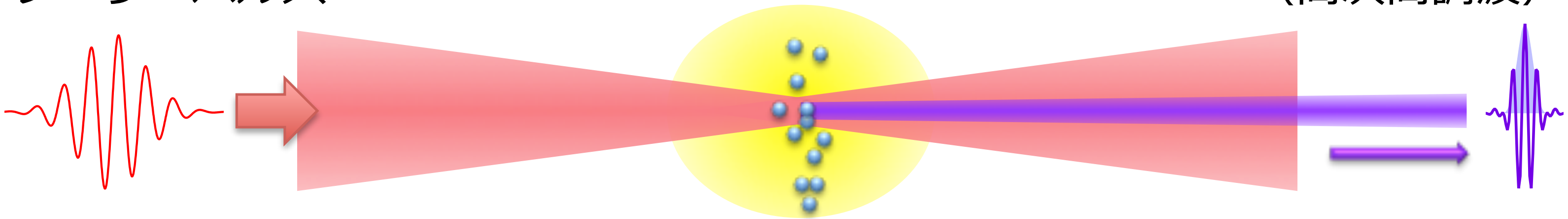
Harmonic order ↑ ⇨ Efficiency ↓

高次高調波発生
High-harmonic generation (HHG)

discovered in 1987

Intense femtosecond laser pulse
高強度フェムト秒レーザーパルス

High-order short-wavelength pulse
高次の短波長光（高次高調波）



ガスジェット
gas jet

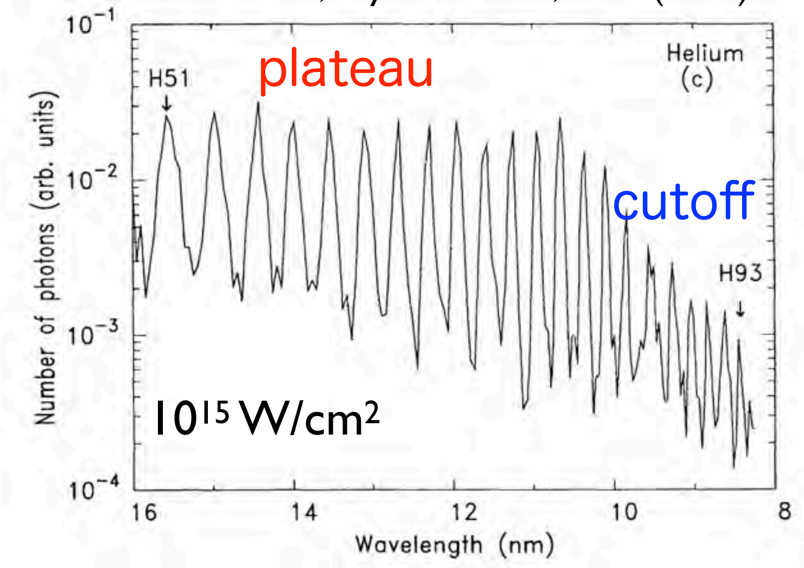
Highly nonlinear optical process in which the frequency of laser light is converted into its integer multiples. Harmonics of very high orders are generated.

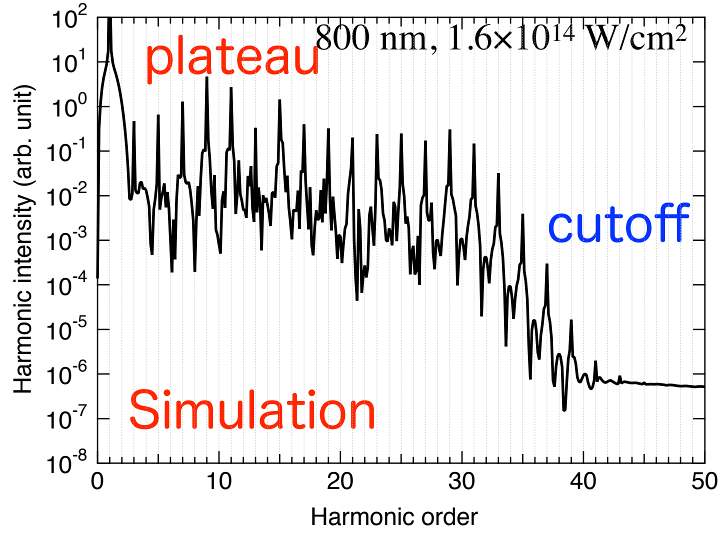
新しい極端紫外・軟エックス線光源として注目される。

New extreme ultraviolet (XUV) and soft X-ray source

Plateau（プラトー） - remarkable feature of high-harmonic generation

Wahlström et al., Phys. Rev. A 48, 4709 (1993)





プラトー(plateau)：Efficiency does NOT decrease with increasing harmonic order. 次数が上がっても強度が落ちない。

カットオフ(cutoff)：Maximum energy of harmonic photons

$$E_c \approx I_p + 3U_p \qquad U_p(\mathrm{eV}) = \frac{e^2 E_0^2}{4m\omega^2} = 9.3 \times 10^{-14} I(\mathrm{W/cm^2})\lambda^2(\mu\mathrm{m})$$

ponderomotive energy

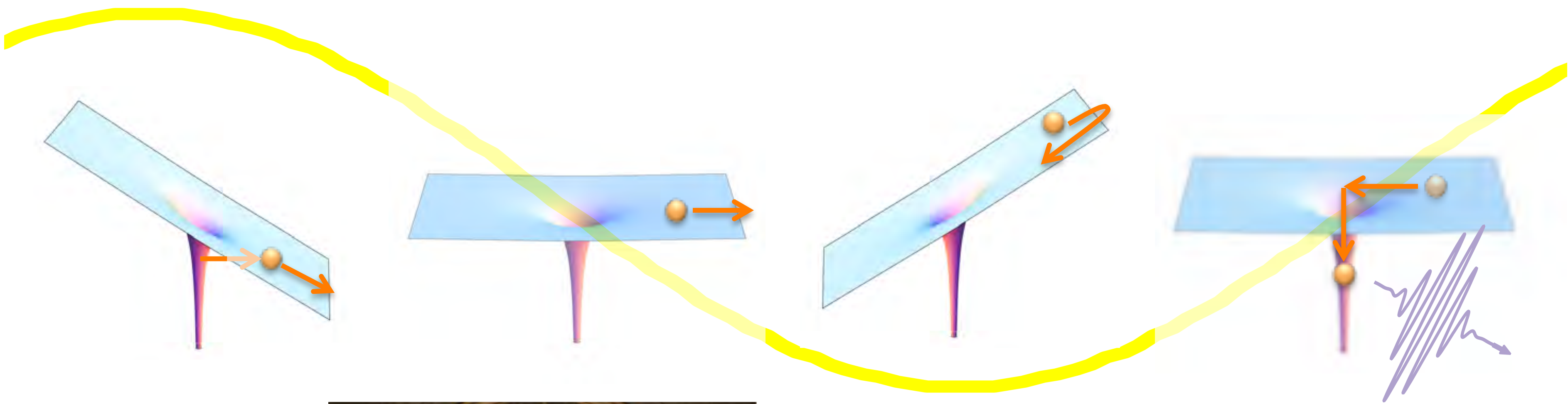
- 摂動論的には解釈できない(non-perturbative)

= strong-field phenomena

高次高調波発生のメカニズム = 3 step model
Mechanism of HHG = 3 step model

Paul B. Corkum, Phys. Rev. Lett. 71, 1994 (1993)

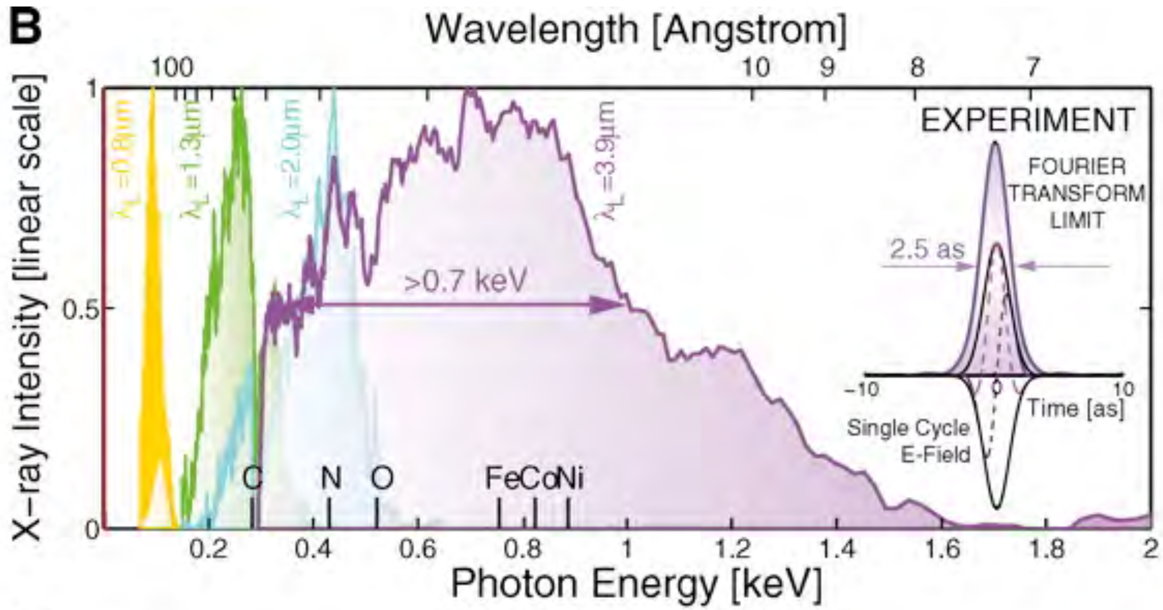
K. C. Kulander *et al.*, in *Super-Intense Laser-Atom Physics*, NATO ASI Ser. B, Vol. 316, p. 95 (1993)



Paul B. Corkum

Even up to 1.6 keV, > 5000 orders

(almost) x-ray!

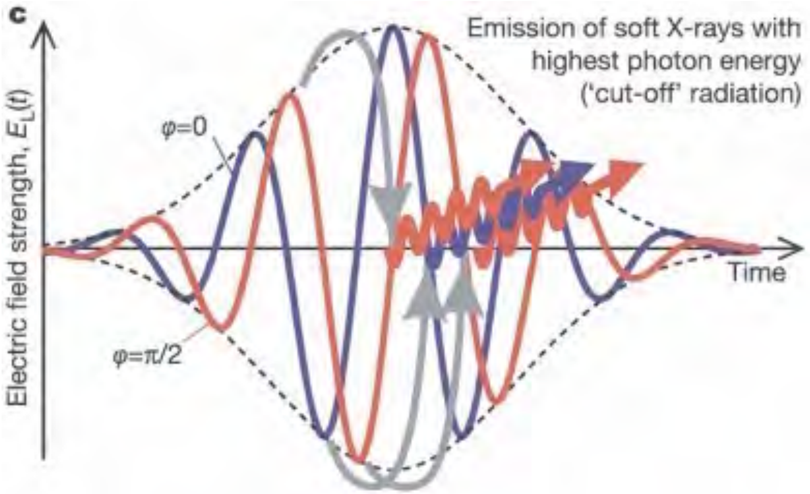


Popmintchev et al., Science 336, 1287 (2012)

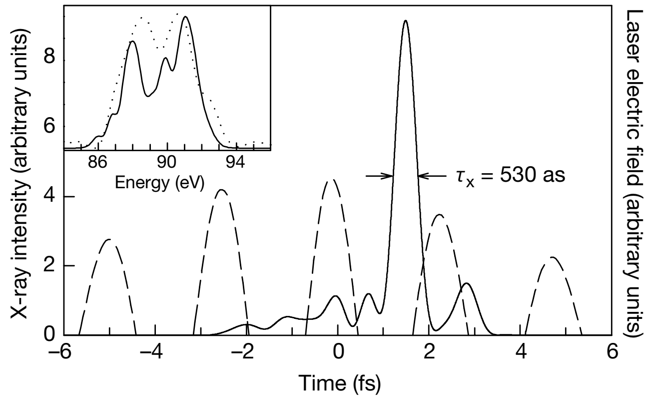
a new type of laser-based radiation source

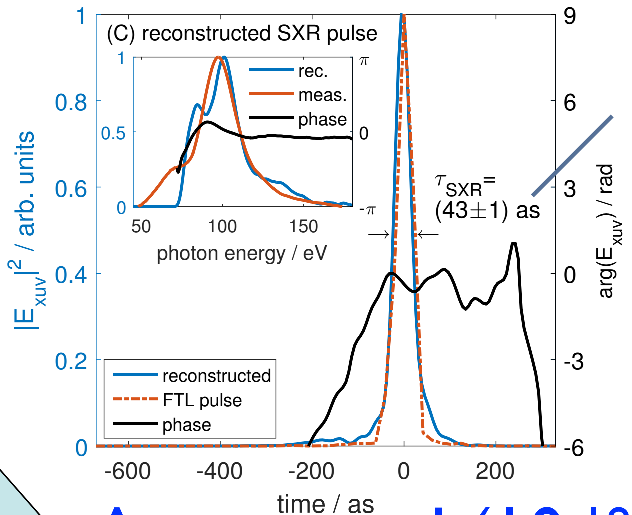
レーザーをベースにした新しいタイプの放射線源

What happens if the fundamental laser pulse is very short? では、超短パルスレーザーによる高次高調波はどんな感じ？



Hentschel et al. (2001)





attosecond

Gaumnitz *et al.* (2017)

Light emission takes place only once.

光の放出は１回だけ

Attosecond (10[-18] sec) pulse

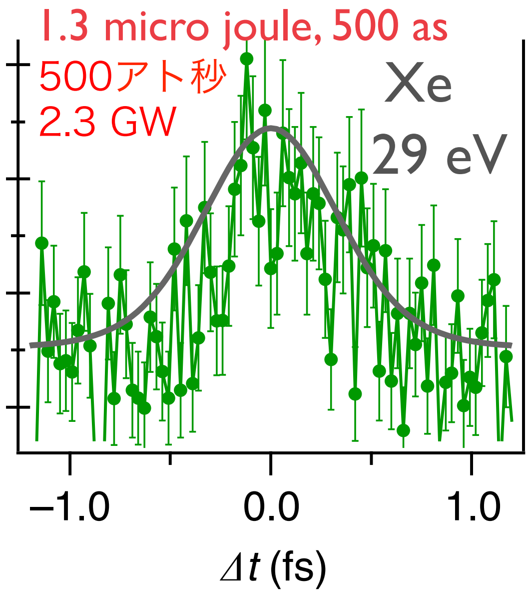
アト秒パルス

Light propagates only 16 nm within 53 attoseconds.

Attosecond pulses アト秒パルス

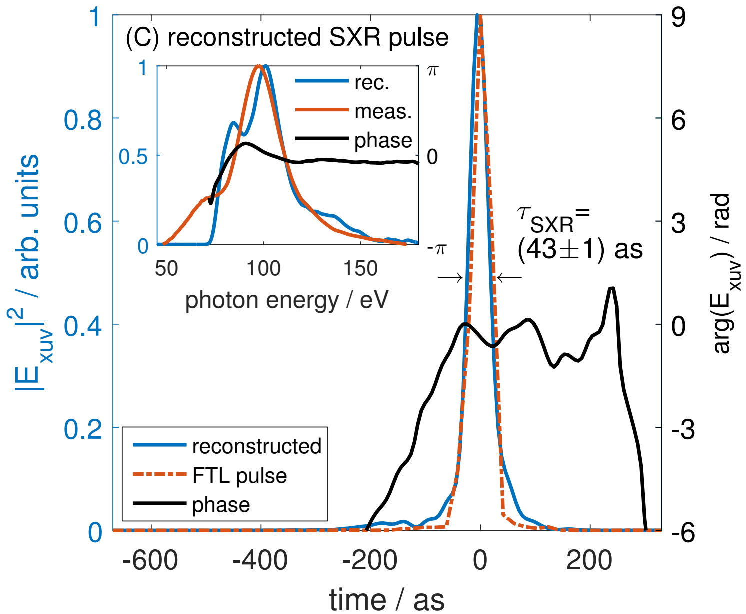
highest power

Takahashi et al., Nat. Commun. 4, 2691 (2013)

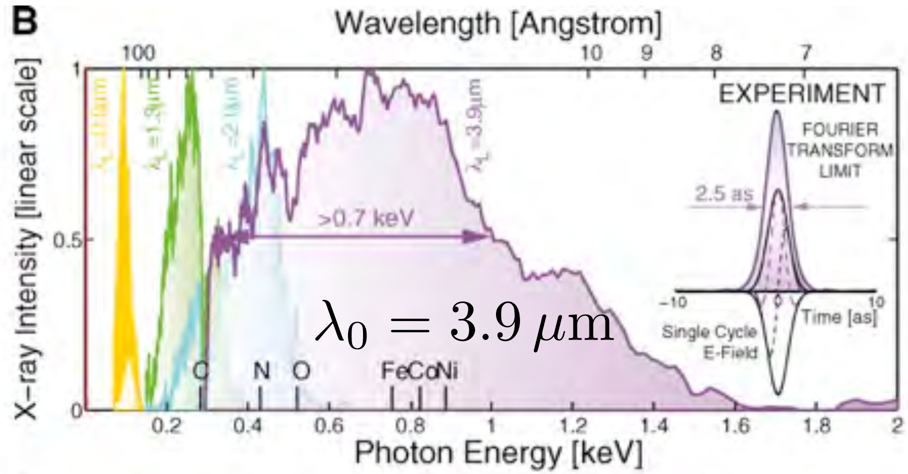


shortest pulse

Gaumnitz et al., Opt. Express 25, 27506 (2017)



highest photon energy

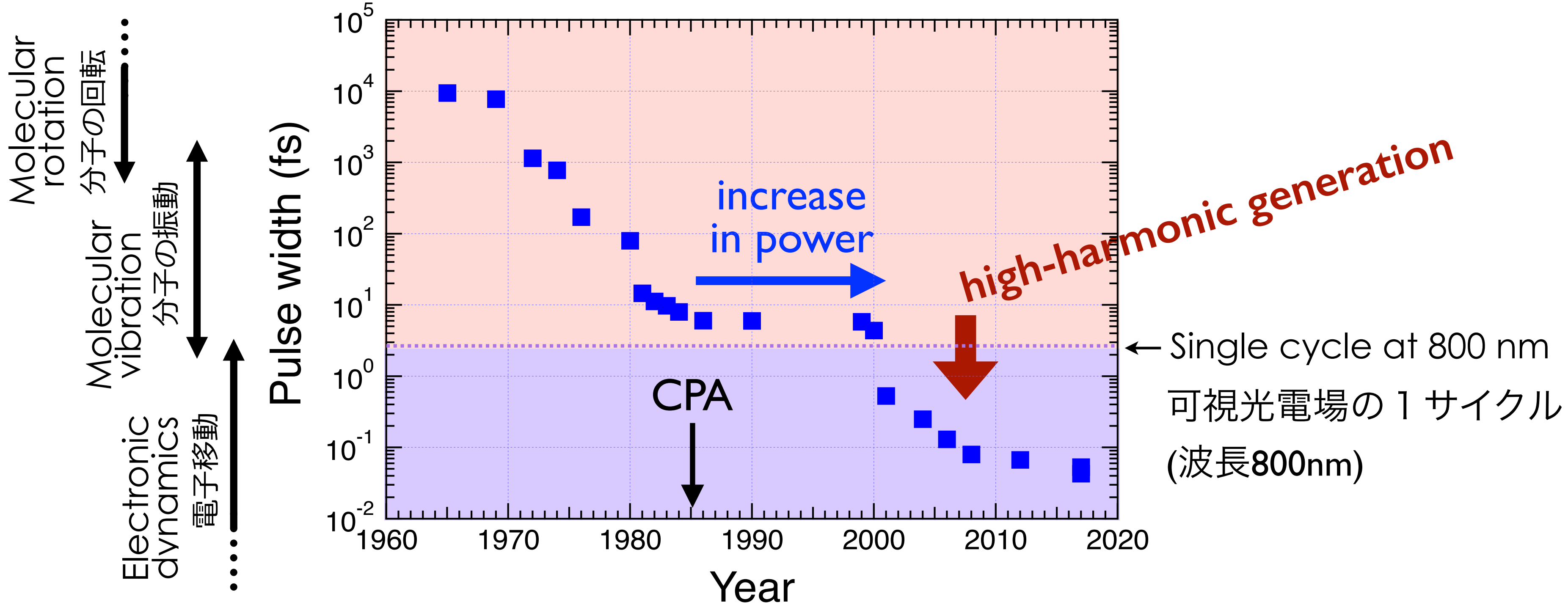


1.6 keV

(> 5000 orders!)

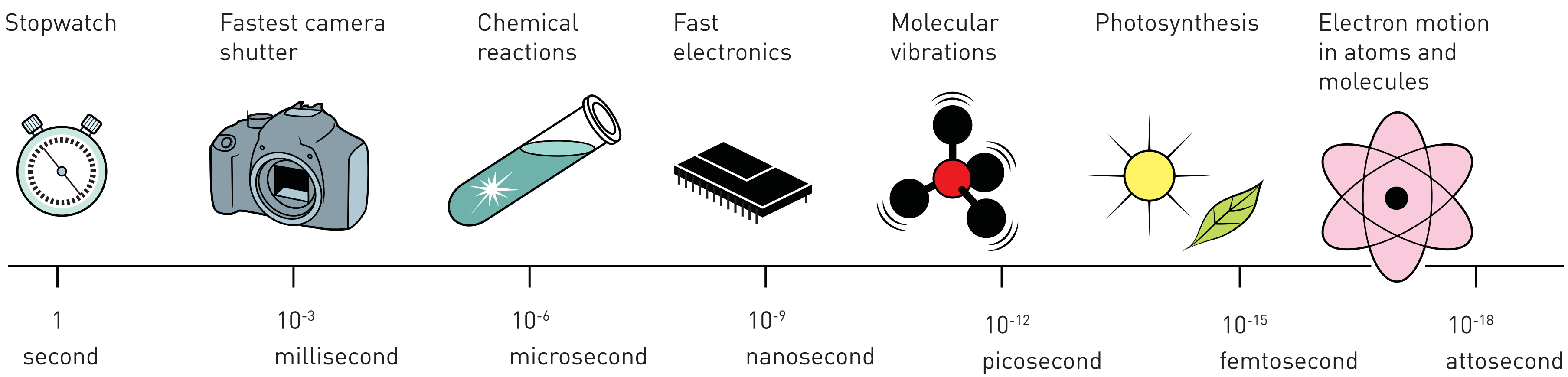
From femtosecond to attosecond

10^{-15} sec 10^{-18} sec



(based on the figure by Prof. J. Itatani)

Timescale



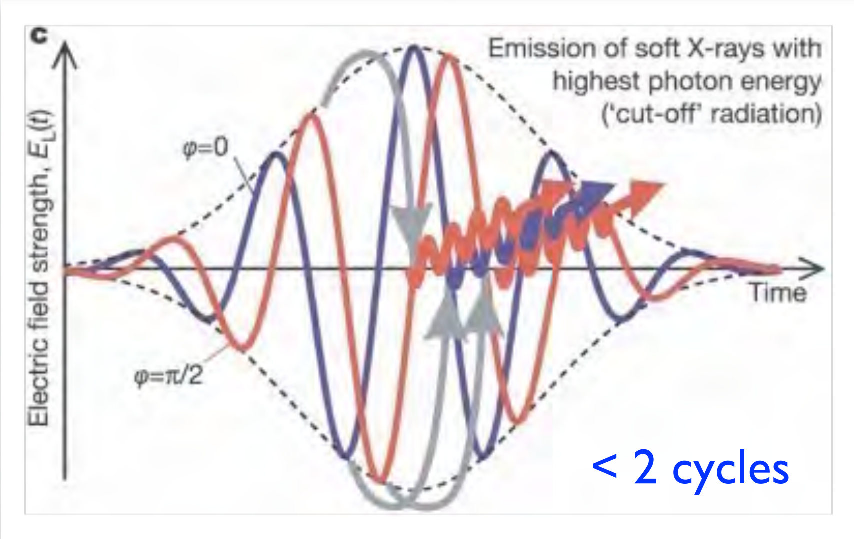
https://6702d.https.cdn.softlayer.net/2019/10/pop_fy_en_18.pdf

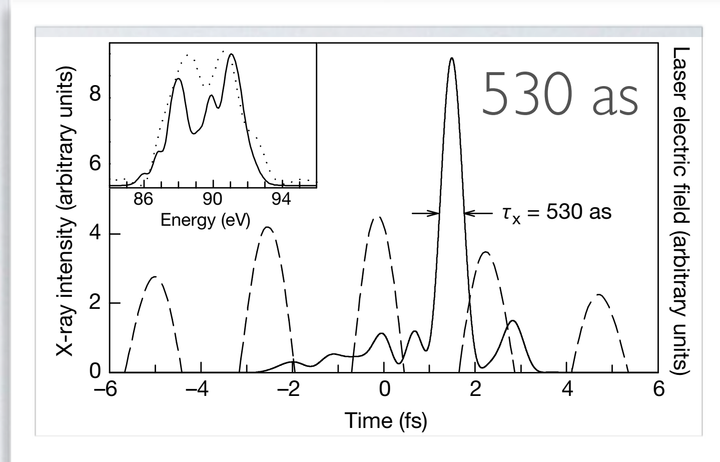
How to generate an isolated attosecond pulse (IAP)

AMPLITUDE GATING

Hentschel et al. Nature 414, 509 (2001)

Baltuska et al. Nature 421, 611 (2003)

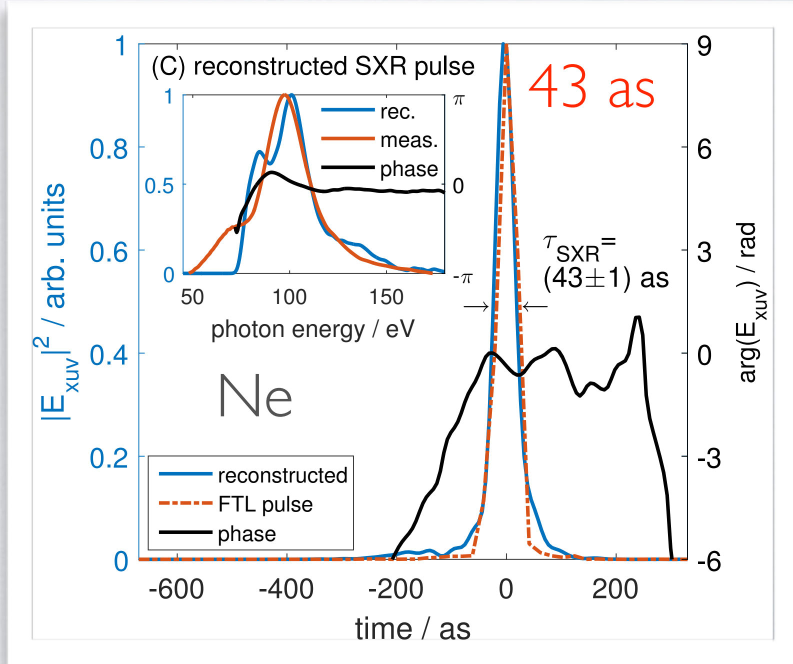




Light emission takes place only once.

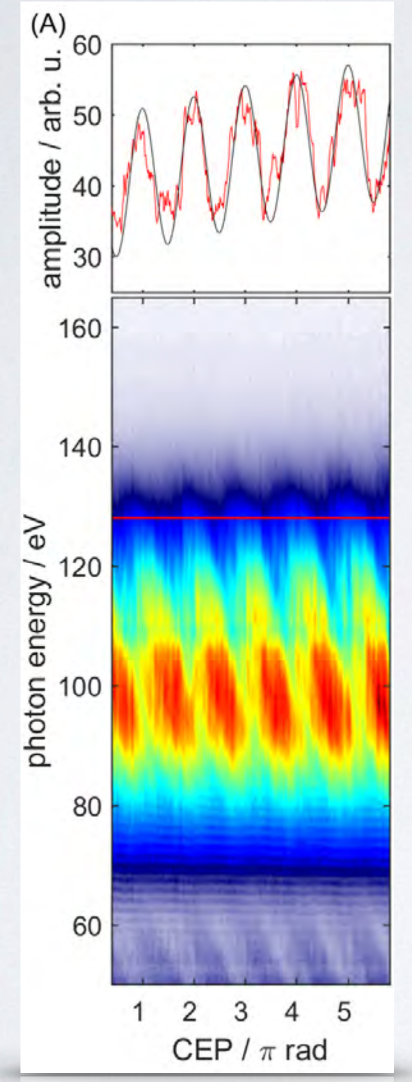
Attosecond (10^{-18} sec) pulse

Gaumnitz et al., Opt. Express 25, 27506 (2017)

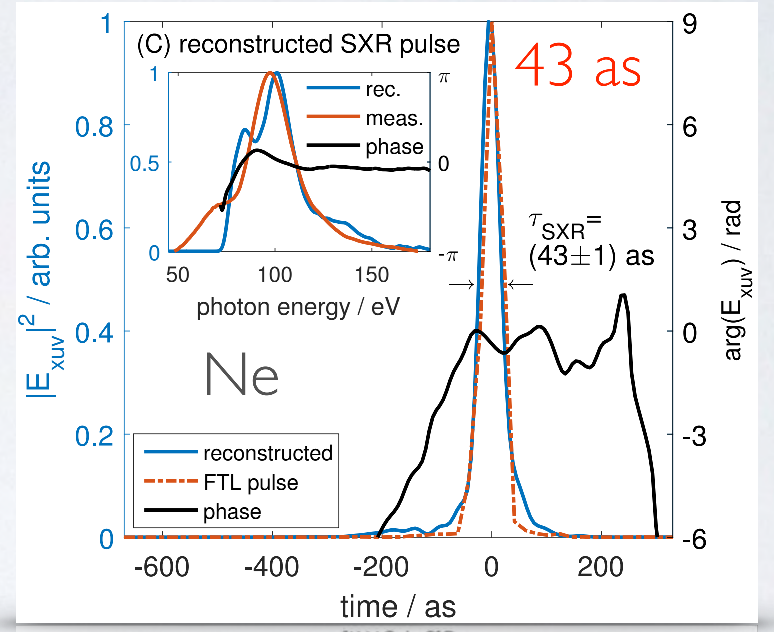


K. L. Ishikawa

AMPLITUDE GATING

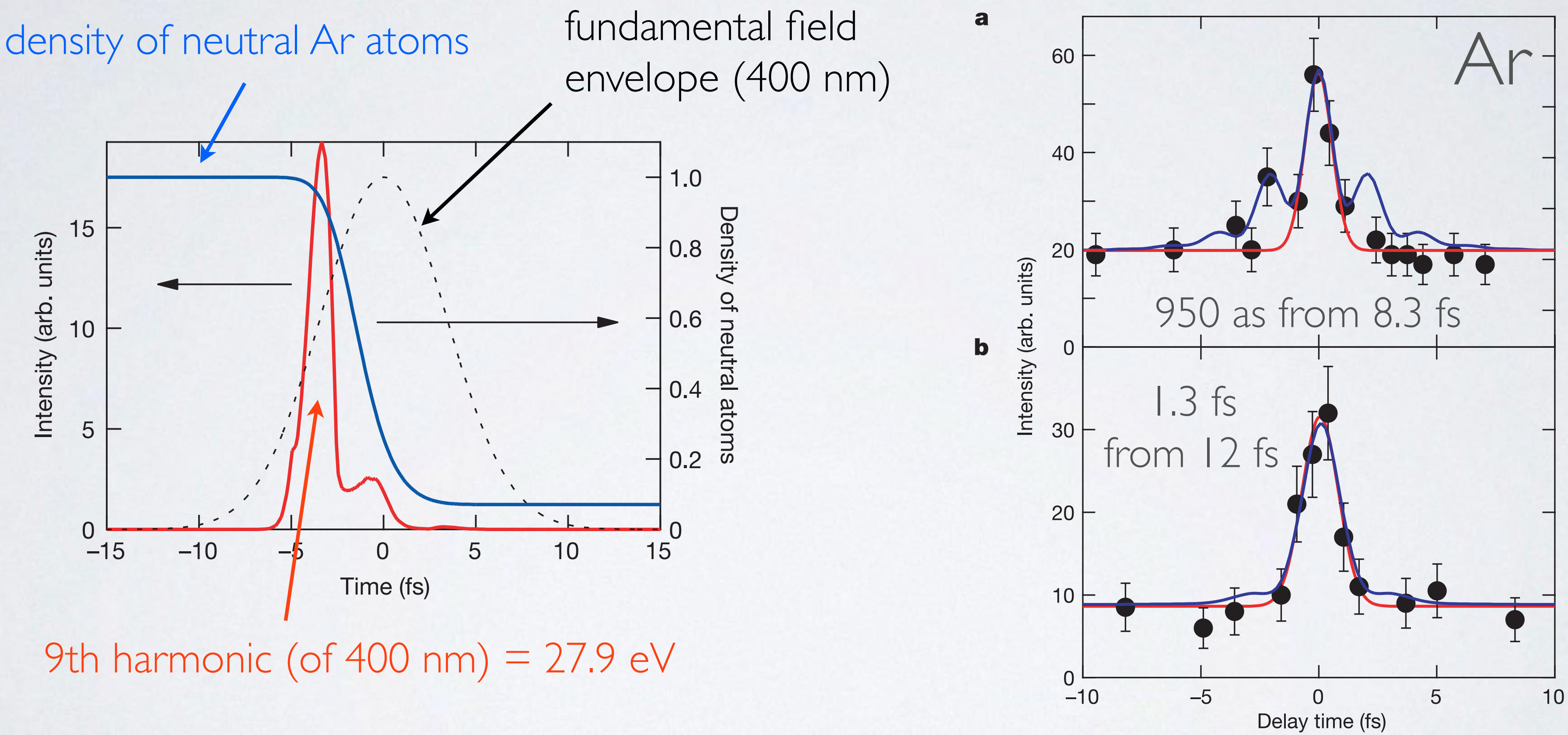


Gaumnitz et al., Opt. Express 25, 27506 (2017)



IONIZATION SHUTTER

HHG is suppressed when neutral atoms are depleted



Isolated sub-fs pulse generation from a ~10 fs pulse

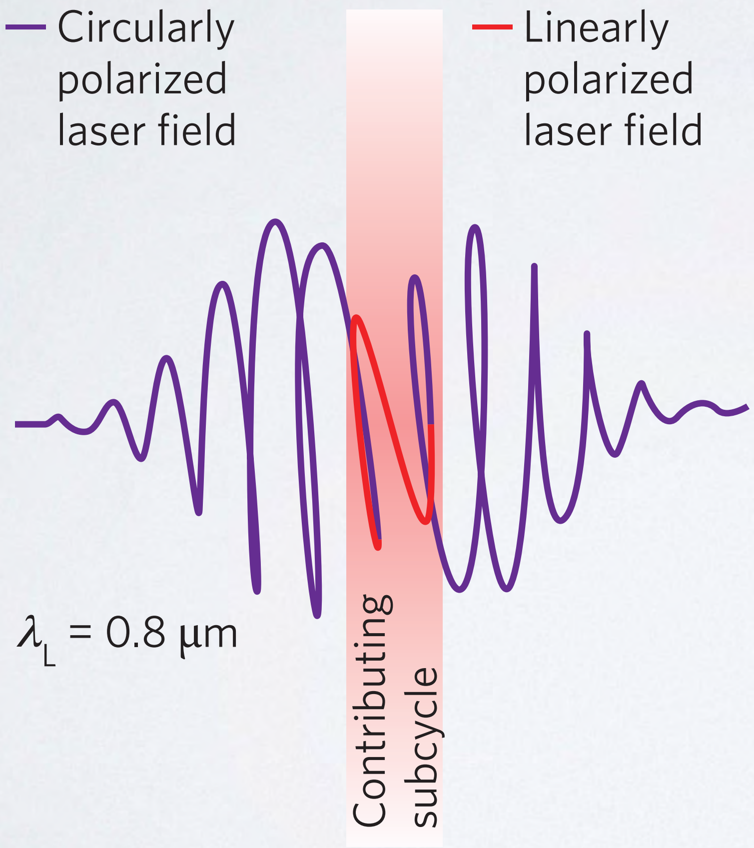
Sekikawa *et al.*, Nature 432, 605 (2004)

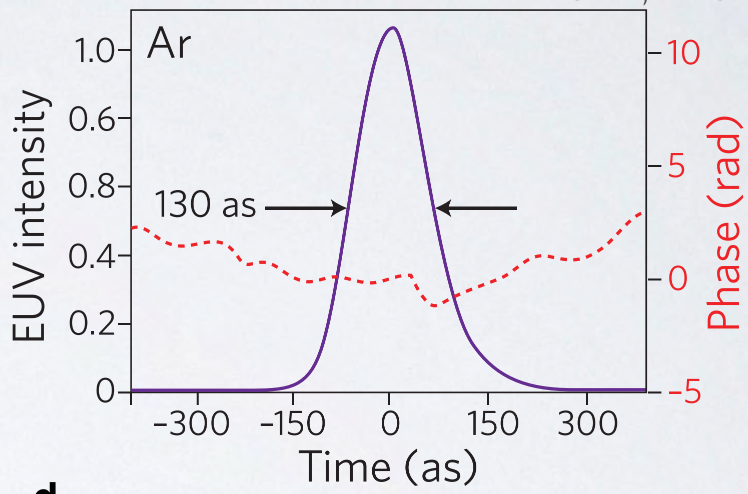
POLARIZATION GATING (PG)

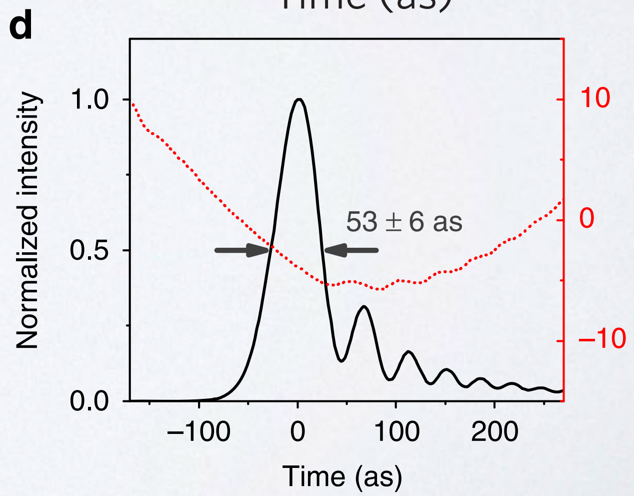
HHG is suppressed when circular polarization is used

counter-rotating circularly polarized pulses with a delay

Sansone *et al.*, Science 314, 443 (2006)







Lin et al. (2017)

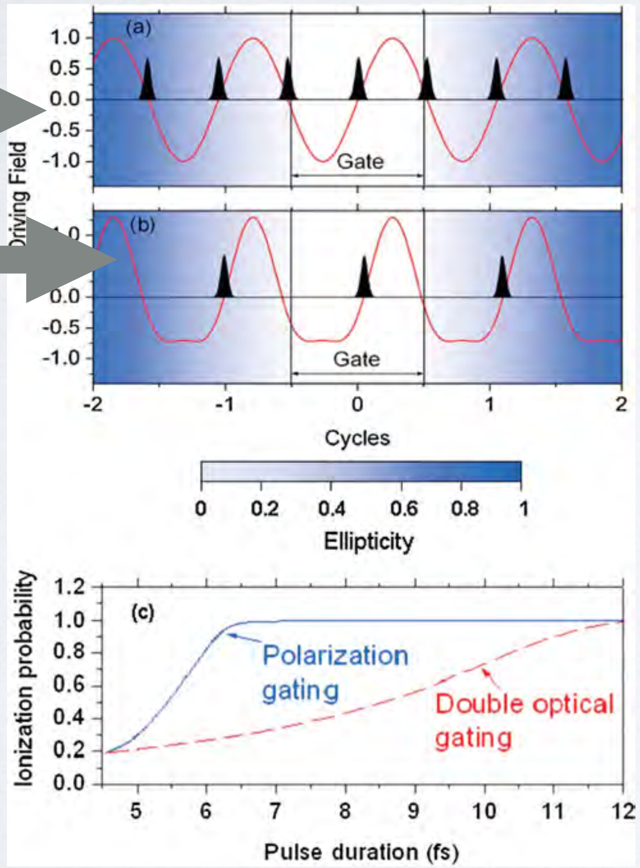
DOUBLE OPTICAL GATING (DOG)

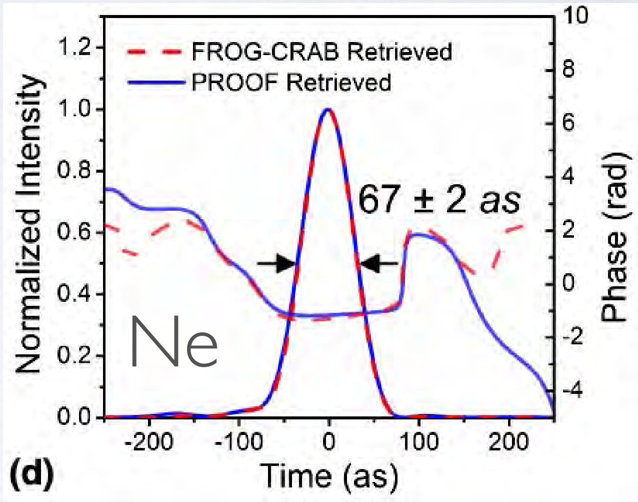
Polarization gating + two-color gating

ω

$\omega + 2\omega$

with second-harmonic field





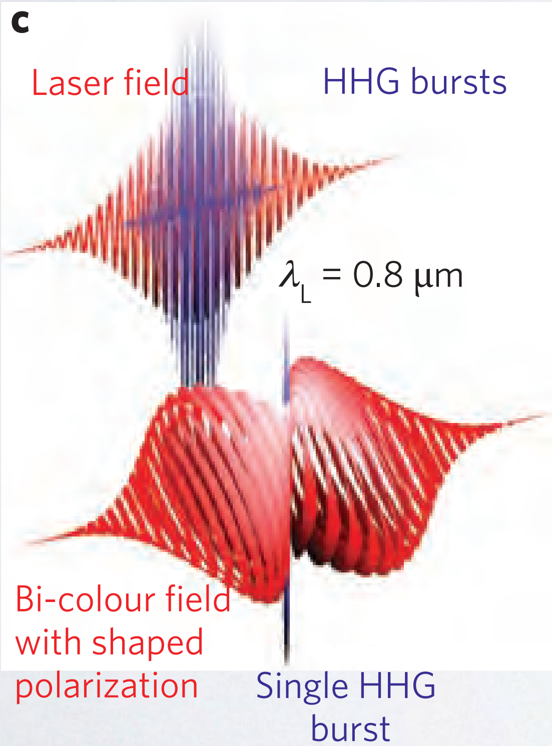
IAP generation from a ~10 fs pulse

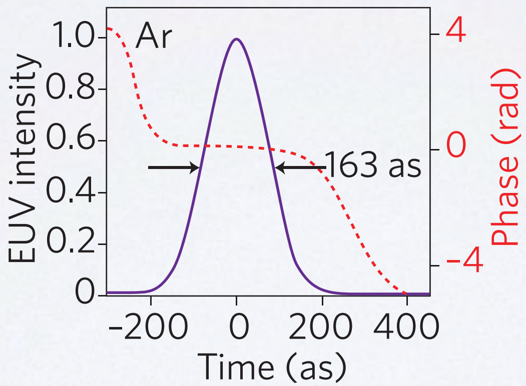
Mashiko et al., PRL 2008, 103906 (2008)

Zhao et al., Opt. Lett. 37, 3891 (2012)

GENERALIZED DOUBLE OPTICAL GATING (GDOG)

Elliptical instead of circular polarization





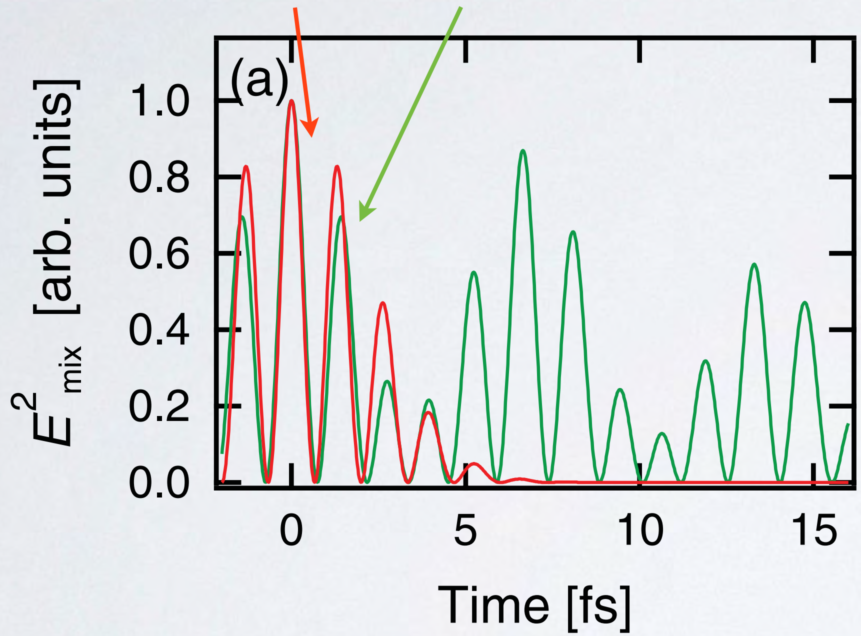
IAP generation from a > 20 fs pulse without need of carrier-envelope stabilization

Gilbertson et al., PRL 105, 093902 (2010)
Gilbertson et al., PRA 81, 043810 (2010)

INFRARED TWO-COLOR SYNTHESIS

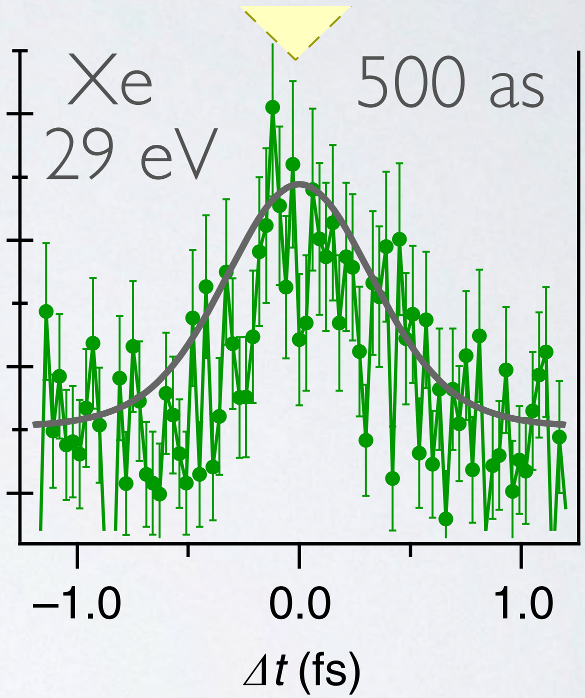
800 nm + 1300 nm two-color driving field

800 nm   800 nm + 1300 nm



Takahashi et al., PRL 104, 233901 (2010)

autocorrelation trace



Takahashi et al., Nat. Commun. 4, 2691 (2013)

High-energy (1.3 micro J), high-power (2.6 GW) IAP

more than 100 times more energetic than previously reported

Quest for higher photon energy (shorter wavelength)

cutoff $$E_c = I_p + 3.17U_p$$

$$U_p(\mathrm{eV}) = \frac{e^2E_0^2}{4m\omega^2} = 9.3 \times 10^{-14} I(\mathrm{W/cm^2})\lambda^2(\mu\mathrm{m})$$

Longer fundamental wavelength is advantageous

Optical parametric chirped-pulse amplification (OPCPA)

WATER-WINDOW HHG

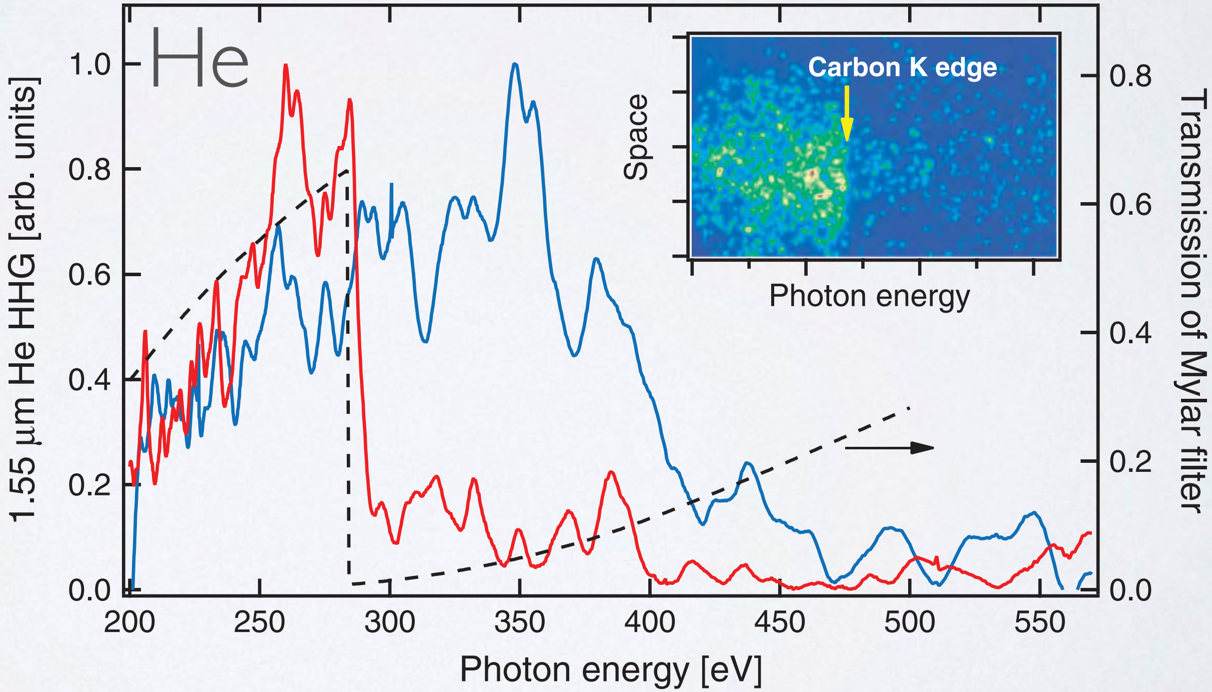
spectral range between the *K*-absorption edges of C (284 eV) and O (543 eV)

absorbed by biological samples but not by water

attractive for high-contrast biological imaging

$\lambda_0 = 1.55\,\mu\mathrm{m}$

$I = 5.5 \times 10^{14}\ \mathrm{W/cm^2}$

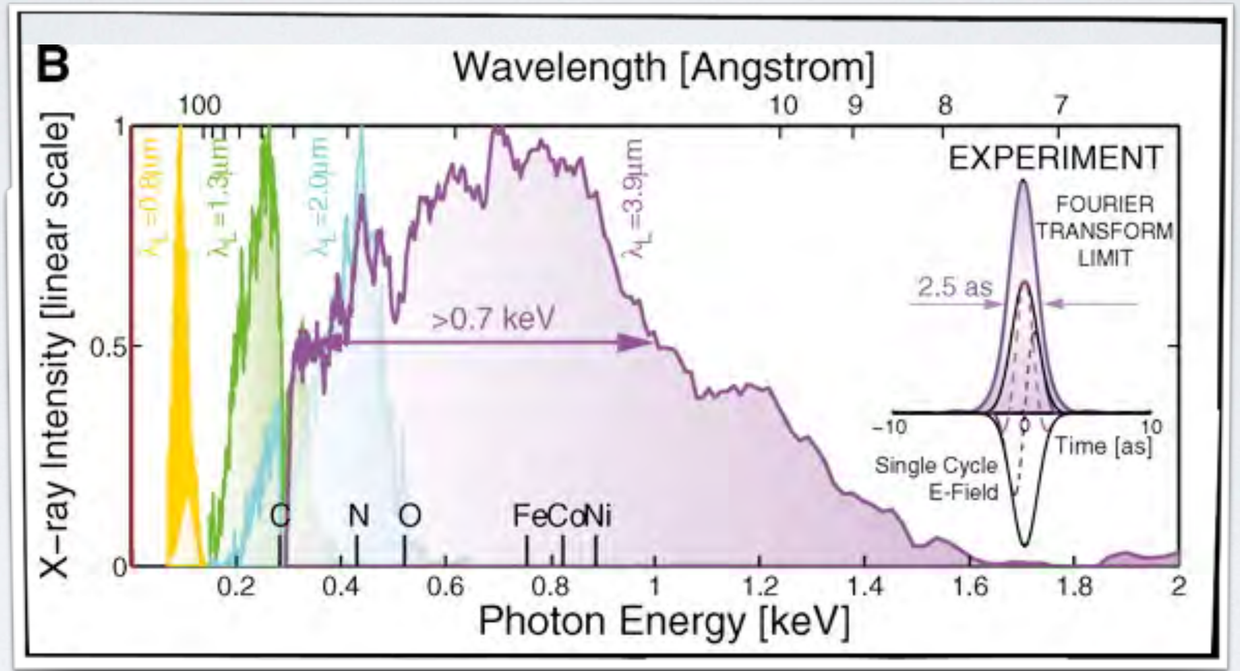


Takahashi et al., PRL 101, 253901 (2008)

keV HHG

Even up to 1.6 keV, > 5000 orders
almost x-ray!

$\lambda_0 = 3.9\,\mu\text{m}$



Popmintchev et al., Science 336, 1287 (2012)

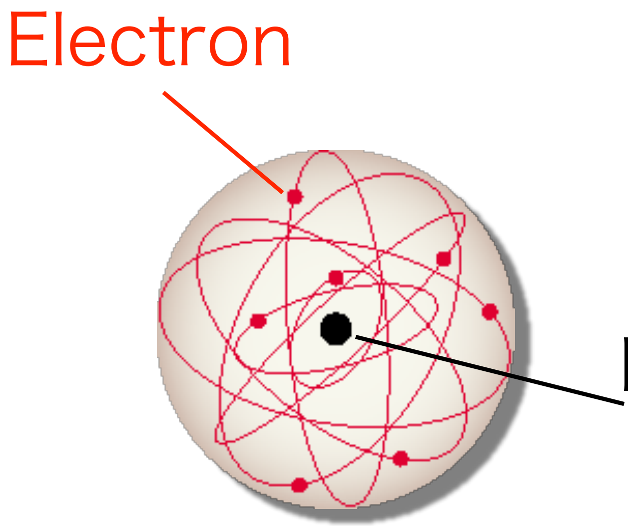
a new type of laser-based radiation source

Attosecond Science
アト秒科学

femtosecond, attosecond

ミリ	m	10^{-3}
マイクロ	μ	10^{-6}
ナノ	n	10^{-9}
ピコ	p	10^{-12}
フェムト	f	10^{-15}
アト	a	10^{-18}

Electrons moving around the nucleus



Orbital period of the electron inside an atom

$$m\omega^2 r = \frac{1}{4\pi\epsilon_0}\frac{e^2}{r^2}$$

$$T = \frac{2\pi}{\omega} = 2\pi\sqrt{\frac{4\pi\epsilon_0 m r^3}{e^2}} = 152 \times 10^{-18}\ \mathrm{s} = 152\ \mathrm{as}$$

Need for attosecond shutter

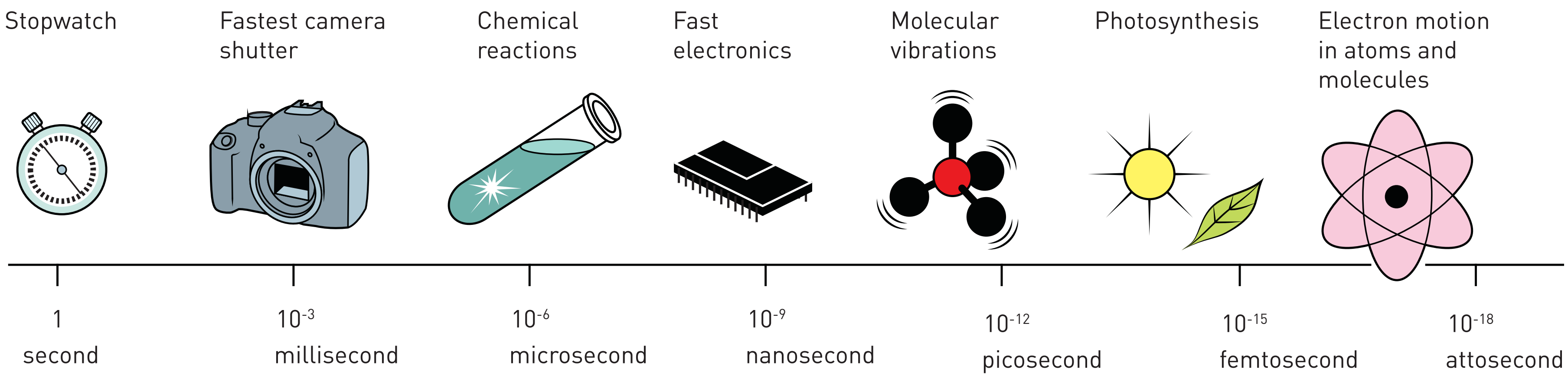
Attosecond science studies electron motion in atoms, molecules, and solids.

★ radiation-matter interaction

放射線と物質の相互作用

★ control of chemical reactions

化学反応の制御



https://6702d.https.cdn.softlayer.net/2019/10/pop_fy_en_18.pdf

Dynamics of the Auger effect
オージェ効果のダイナミクス

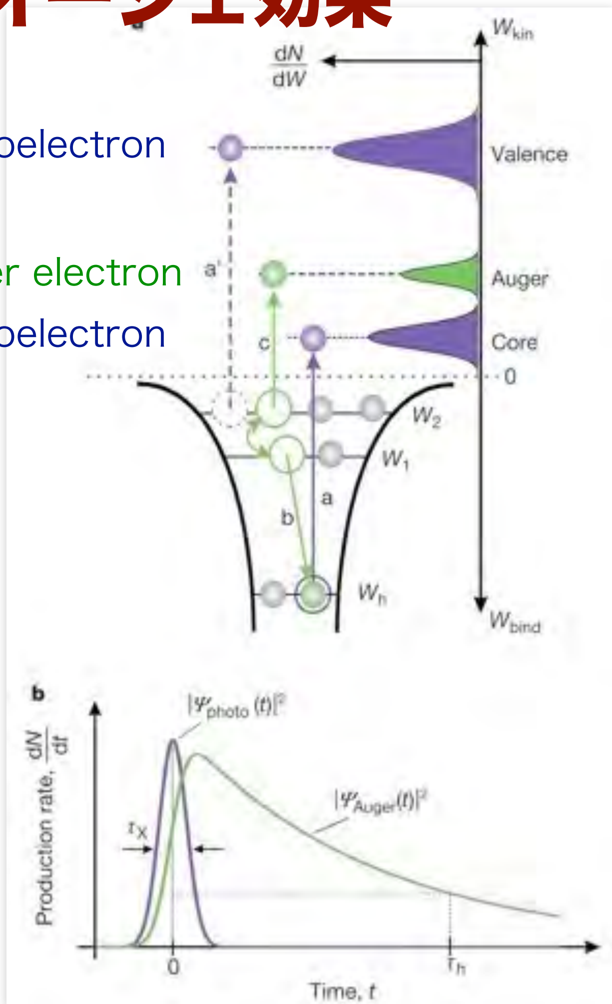
Auger effect

オージェ効果

Photoelectron

Auger electron

Photoelectron



Ejection of a core electron

内殻電子が電離（光電効果）

Instantaneous

Core-excited ion

内殻励起状態のイオン

~ a few fs

Ejection of a valence electron

特性X線を放出するかわり
に軌道電子を放出

Observation of the ejection of Auger electrons

→Ionizing X rays < a few fs

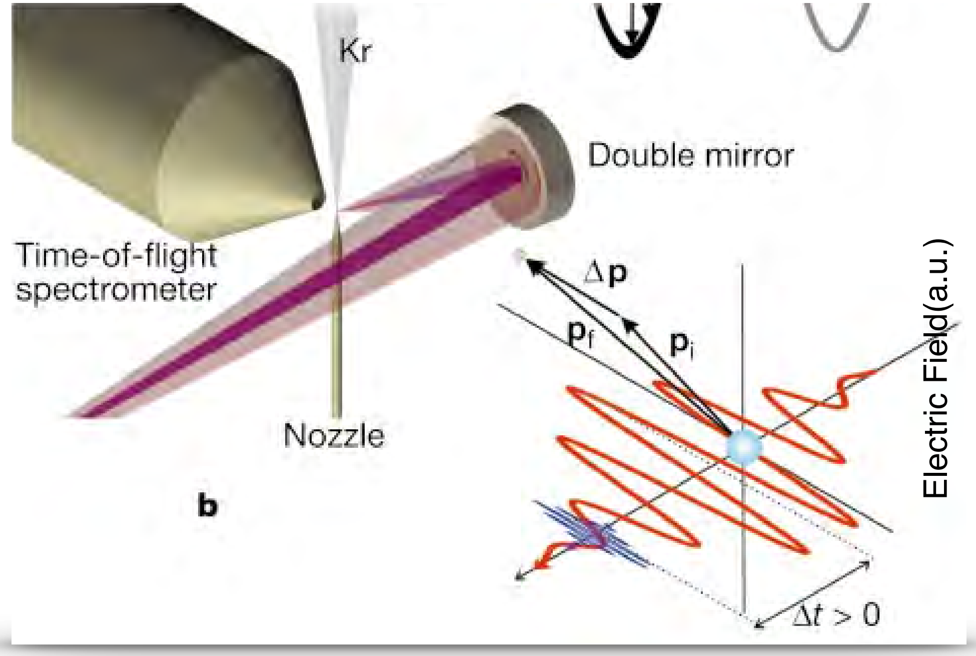
→Attosecond pulse

How to measure the electron ejection time?

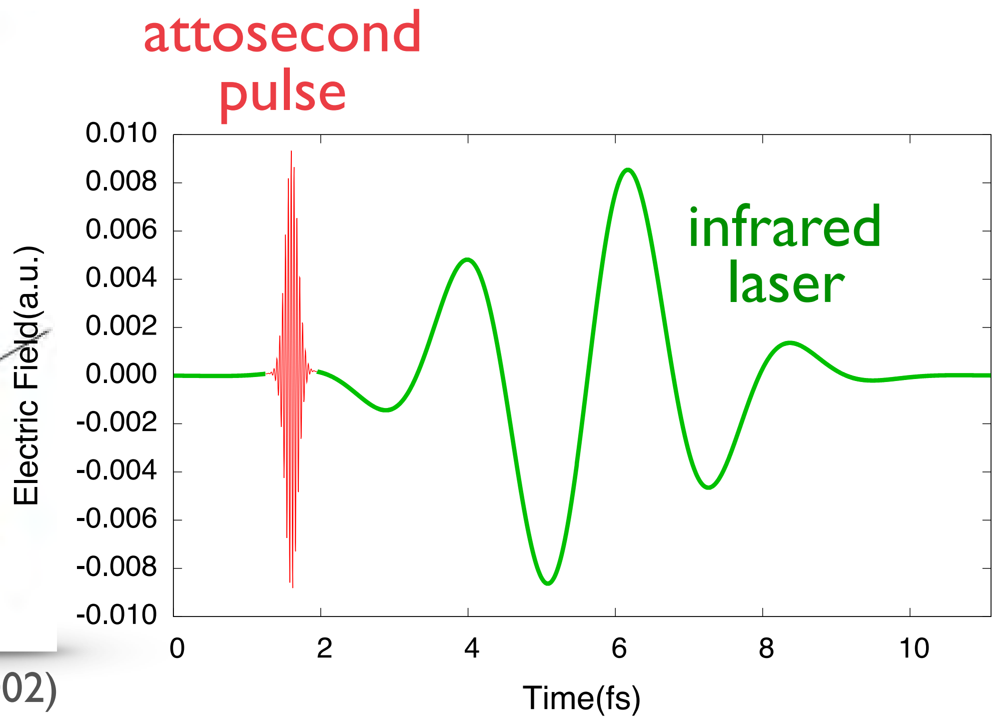
Pump（イオン化を引き起こす）	高調波(HHG)
Probe（電子の放出時刻を測る）	レーザー光（laser）

How to measure the electron ejection time? Itatani *et al.*, Phys. Rev. Lett. 88, 173903 (2002)

Attosecond streaking



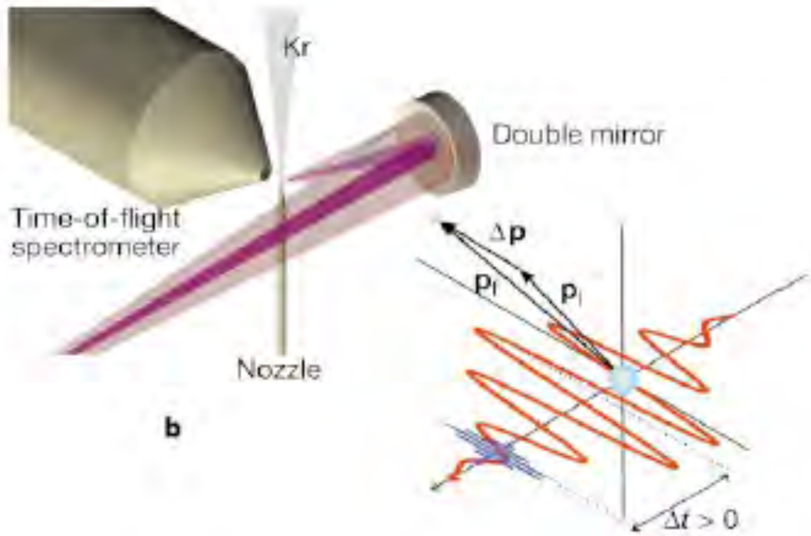
Drescher et al., Nature 419, 803 (2002)



高調波とレーザー光を遅延時間を持たせて照射

Irradiate an atom with an attosecond pulse and laser pulse with delay

How to measure the electron ejection time?



$$E(t) = E_0(t)\cos(\omega t + \phi)$$

$$\frac{dp}{dt} = m\frac{dv}{dt} = -eE(t)$$

ionization at $t = t_r$ で電離

Initial momentum 初速度（運動量）

$$p_0 = \sqrt{2m(\hbar\omega_X - I_p)}$$

検出器での運動量 Momentum at the detector

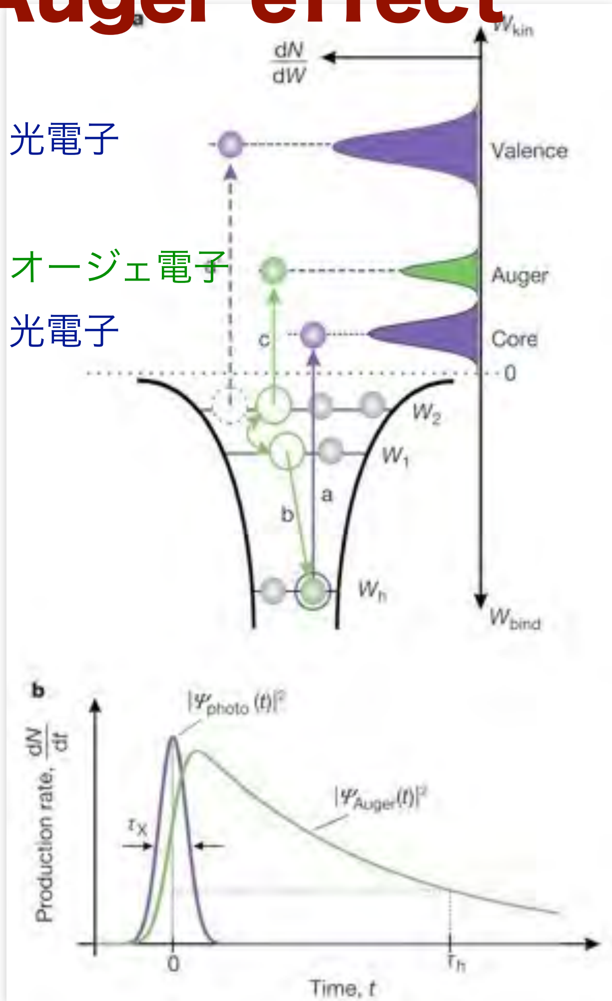
vector potential

$$\mathbf{p} = \mathbf{p_0} - \int_{t_0}^{\infty} e\mathbf{E}(t)dt = \mathbf{p_0} + e\mathbf{A}(t_0)$$

electron ejection time ⇔ momentum (energy) change

Life time of the Auger decay～8 fs

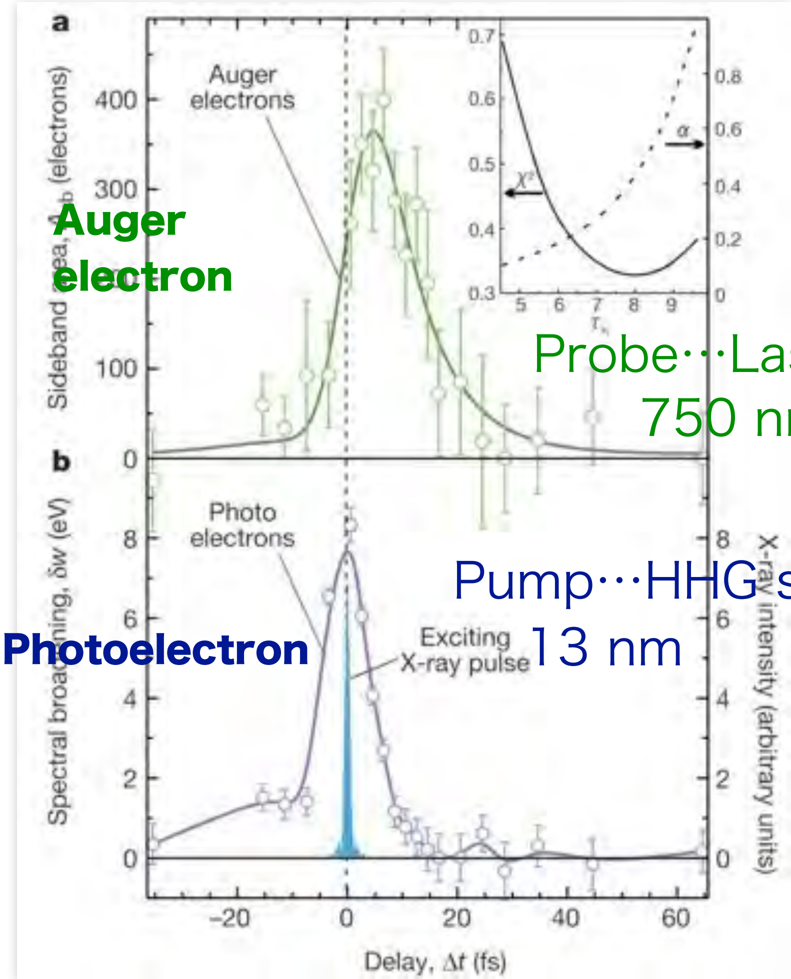
Auger effect



光電子

オージェ電子

光電子



Auger electron

Photoelectron

Probe…Laser 750 nm

Pump…HHG soft x rays 13 nm

数フェムト秒程度の超高速過程が見える！

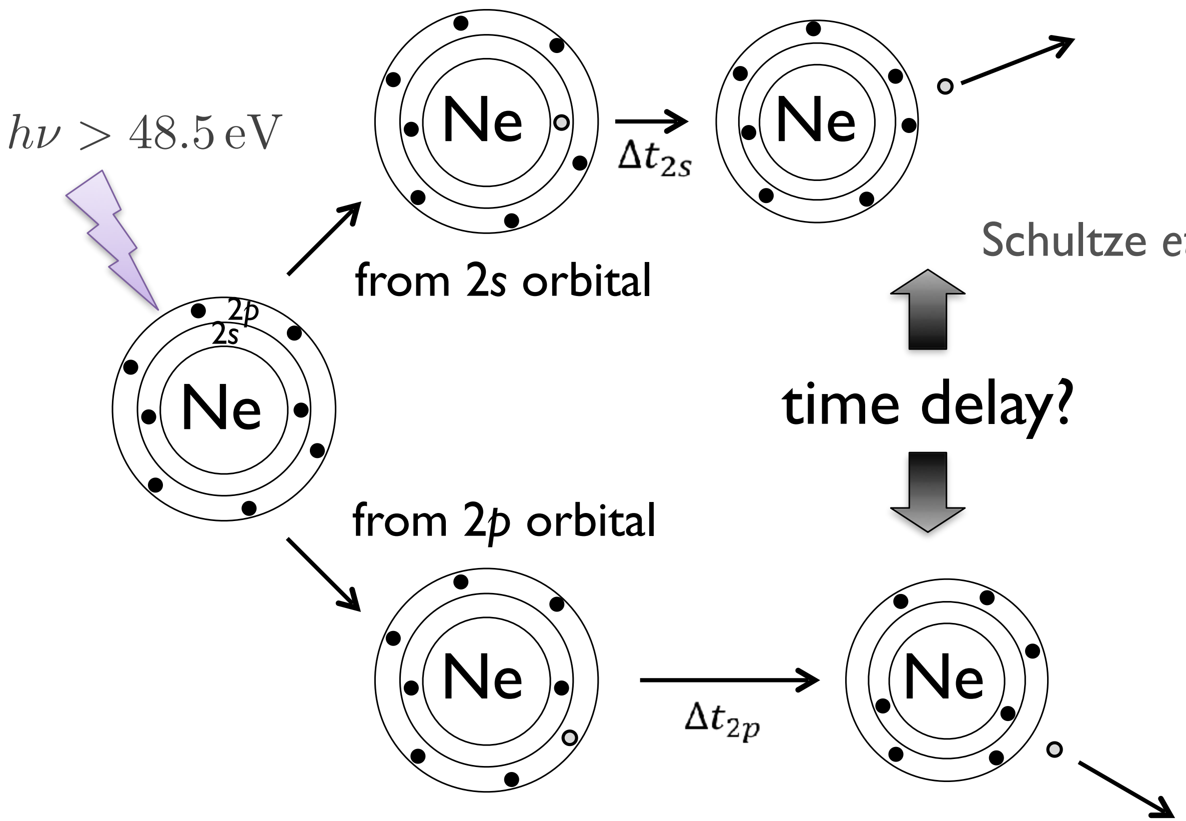
Ultrafast process ~ a few fs

Delay in photoemission
(How long does the photoelectric effect take?)

光電効果には何アト秒かかるか？

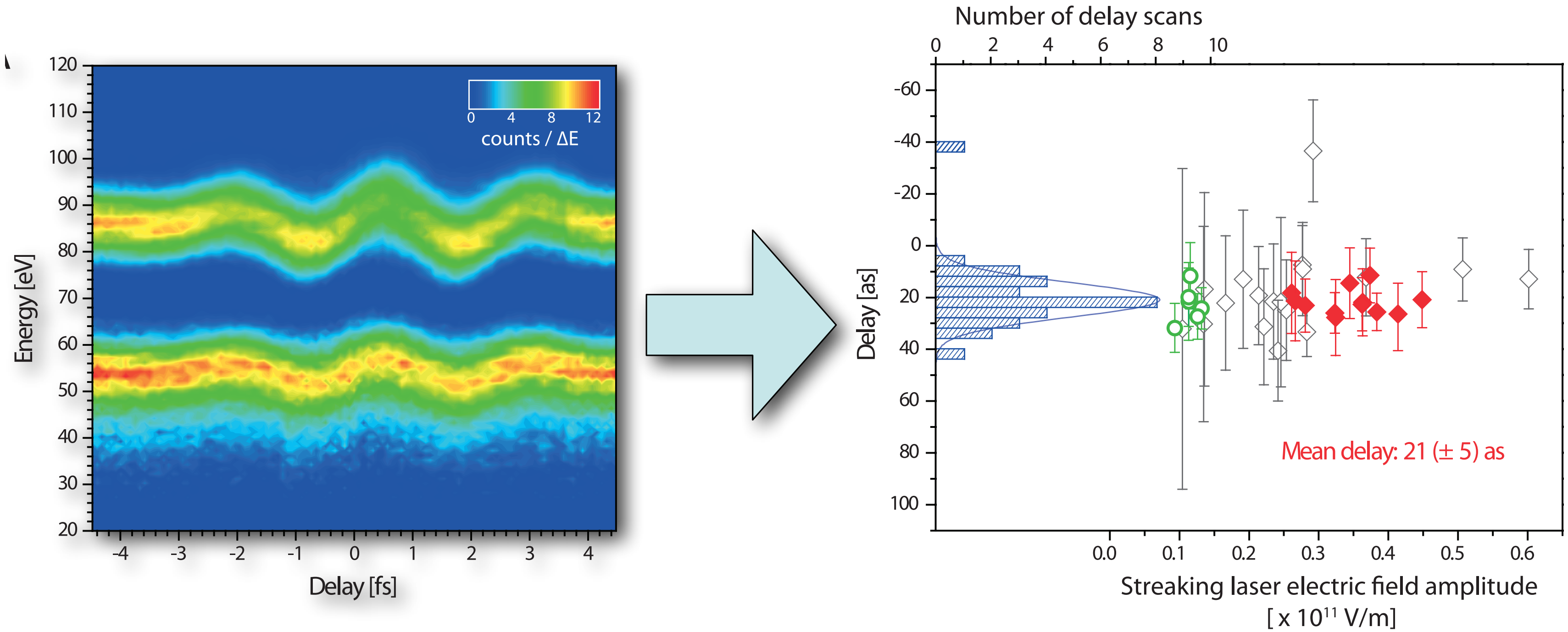
When Does Photoemission Begin?

The photoelectric effect is usually considered instantaneous.



Schultze *et al.*, Science 328, 1658 (2010)

The 2s electron appears to come out 21 attoseconds earlier than the 2p electron!



Schultze *et al.*, Science 328, 1658 (2010)

フェニルアラニン

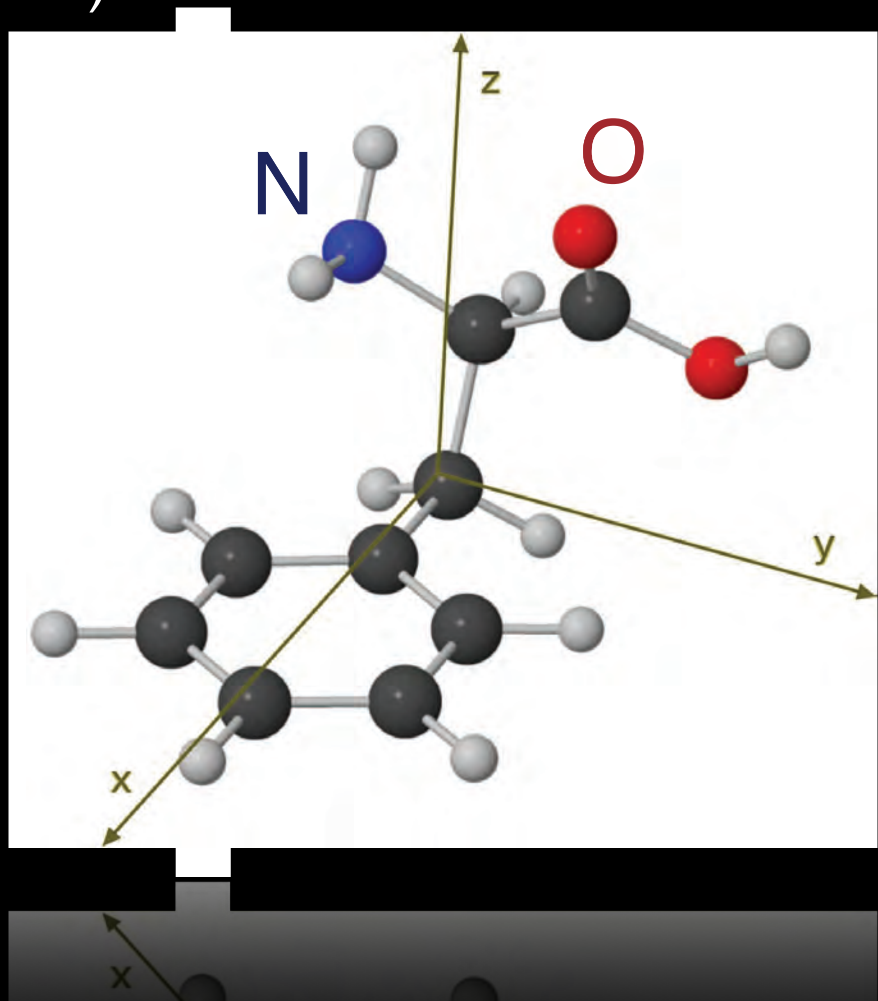
Ultrafast electron dynamics in phenylalanine initiated through ionization by attosecond pulses

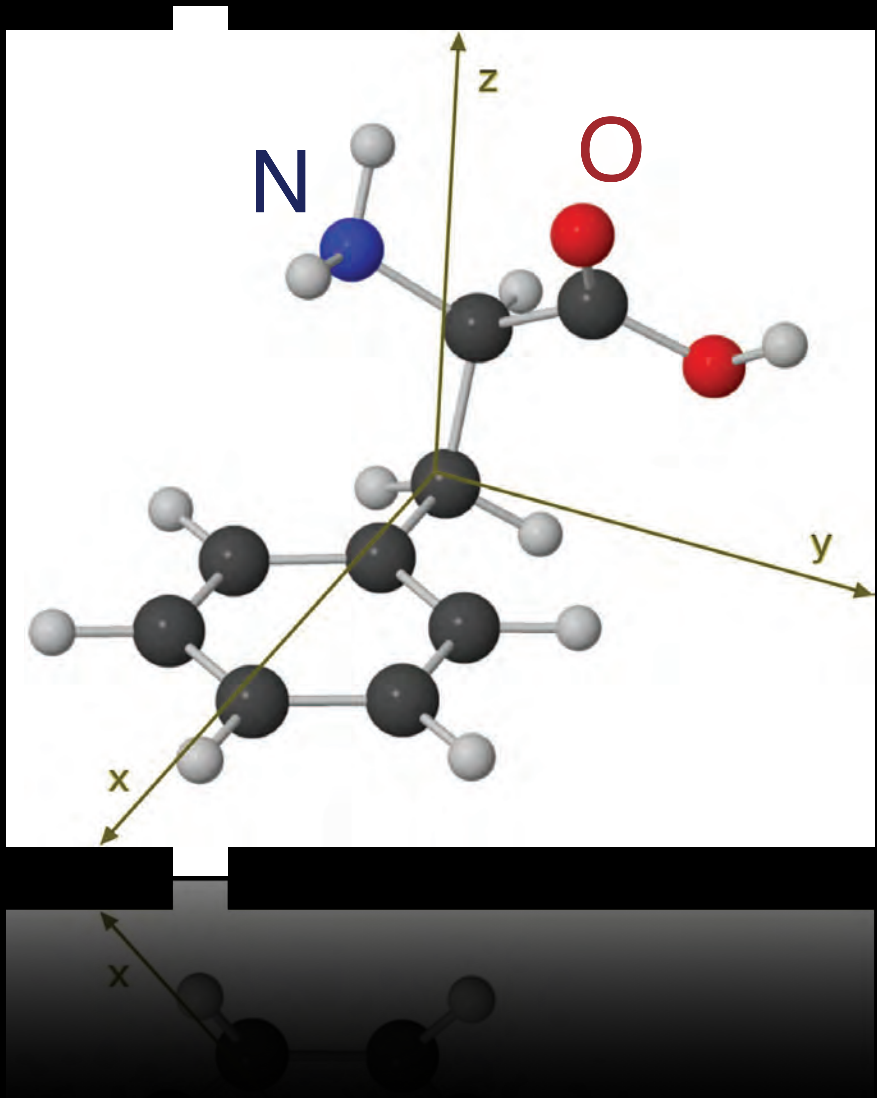
amino acid
アミノ酸

Calegari *et al.*, Science 346, 336-339 (2014)

radiation damage of biomolecules

生体分子の放射線損傷





pump

sub-300 as XUV

15-35 eV

probe

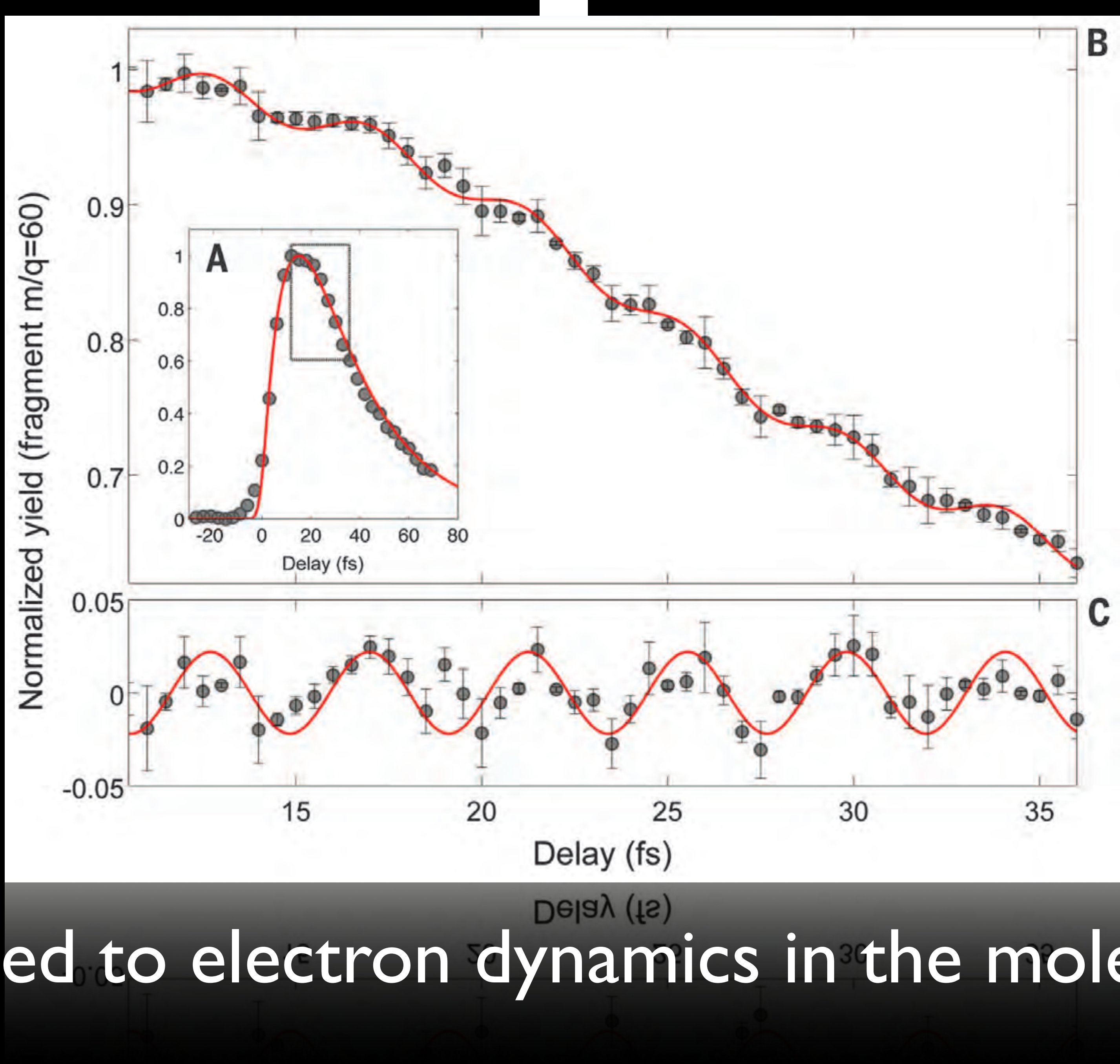
4 fs VIS/NIR

1.77 eV / 700 nm

detect

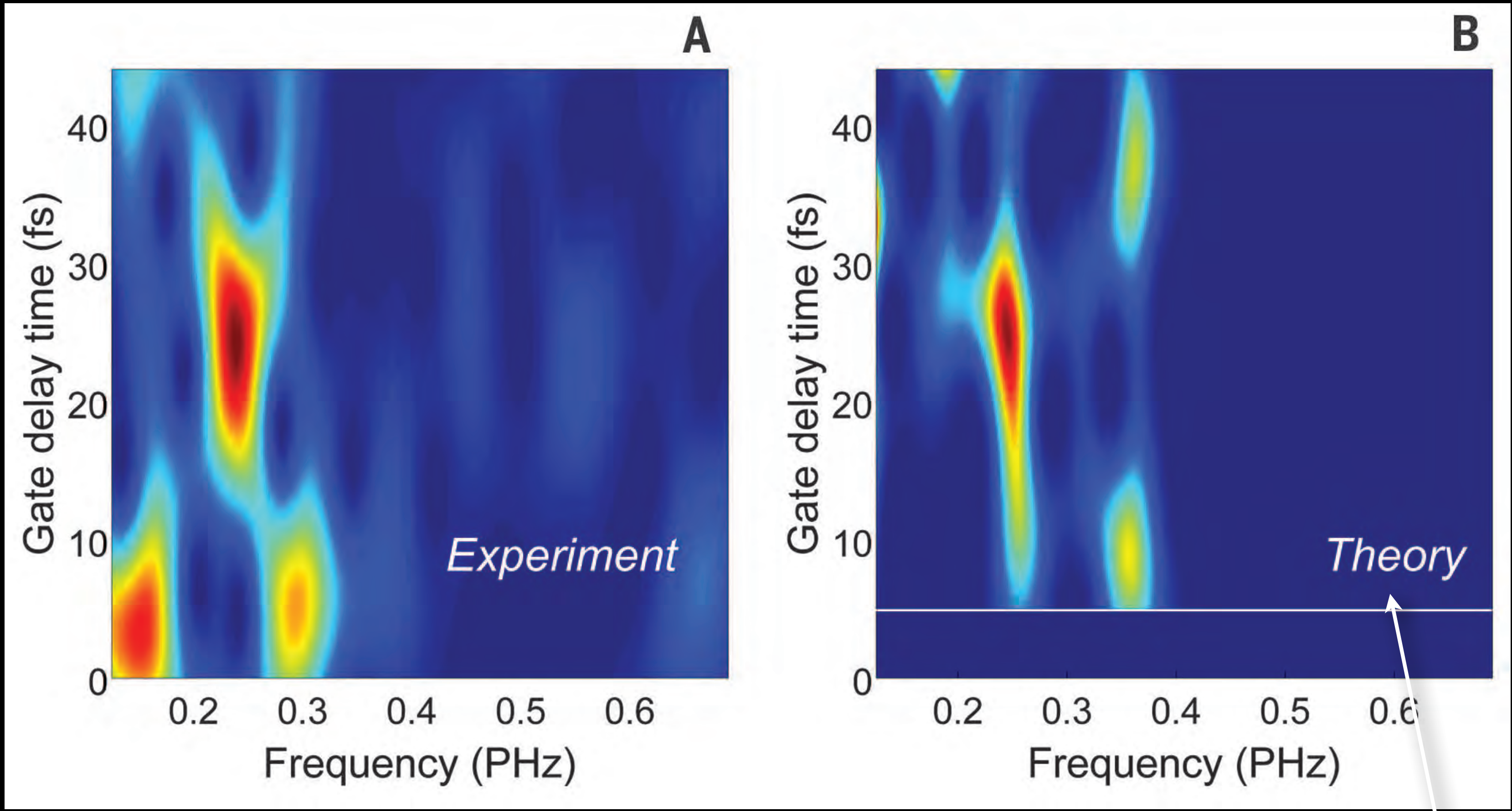
$^{++}NH_2$-CH-R dication

dication yield oscillates with period ~ 4.3 fs

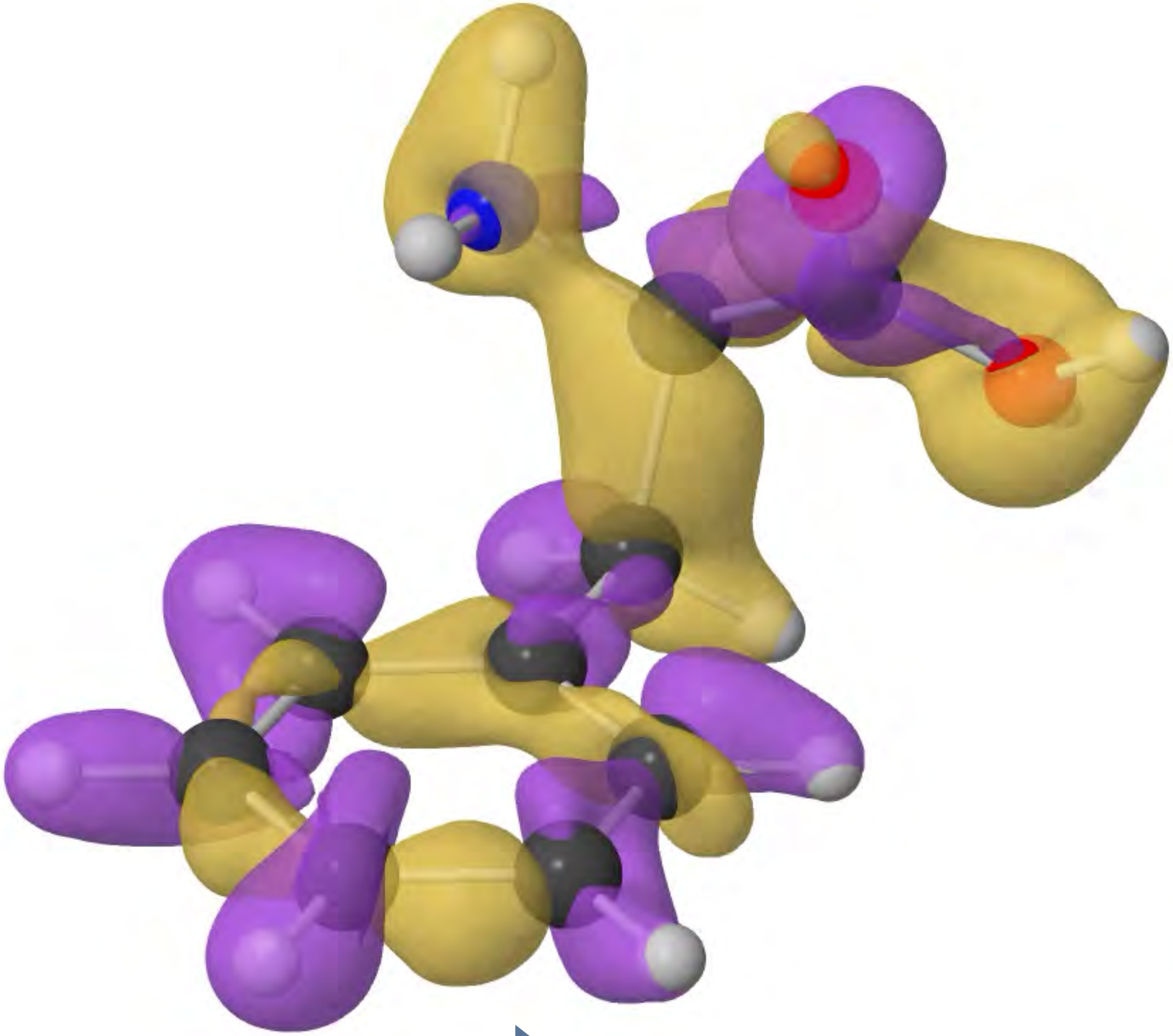


assigned to electron dynamics in the molecule

time-frequency analysis



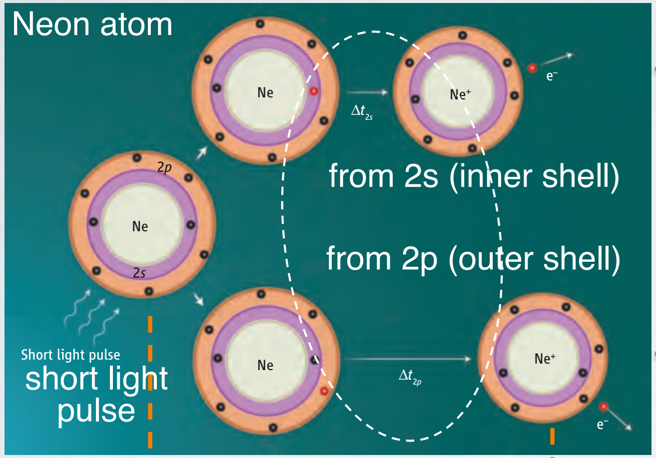
static-exchange DFT
+ first order time-dependent perturbation theory



0.00 fs

report assignment

Delay in photoemission



delay 21 as

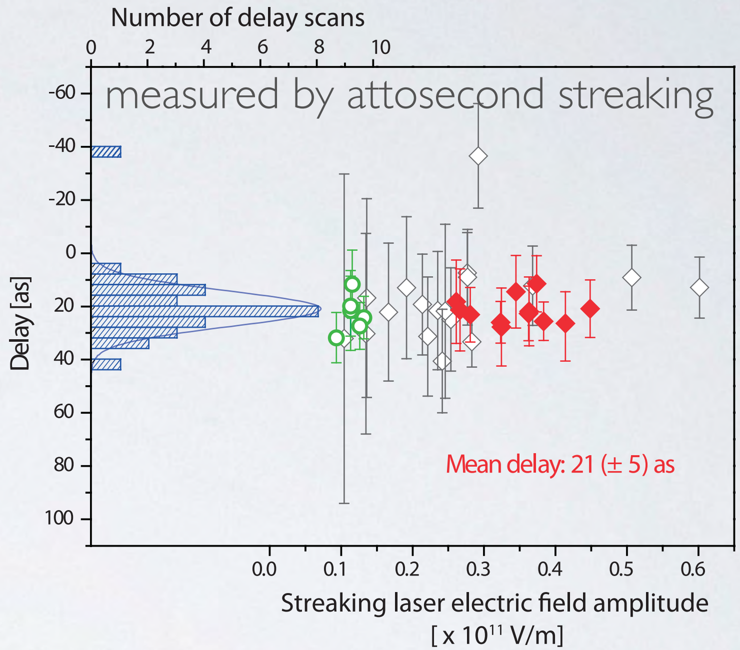
how long does it take?

photon absorption

electron emission

What is happening?

Dynamic multi-electron correlation?

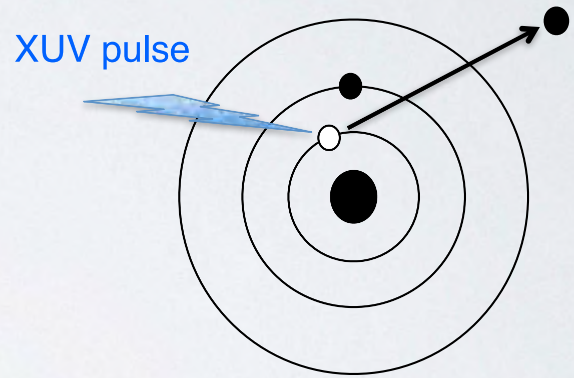


Schultze *et al.*, Science 328, 1658 (2010)

Mechanism

- Eisenbud-Wigner-Smith delay ← *stationary-state correlation*
- Coulomb-laser coupling ← *laser effect*
- laser-induced state distortion
- unknown mechanisms ...

Time-dependent ab-initio simulation of inner-shell photoionization of an excited He atom (e.g., 1s2p)



Method: Time-dependent Schrödinger equation (TDSE)

$$i\frac{\partial}{\partial t}\psi(\mathbf{r}_1,\mathbf{r}_2,t) = \left[H_{\text{atom}} + \left(z_1 + z_2\right)E(t)\right]\psi(\mathbf{r}_1,\mathbf{r}_2,t)$$

$$H_{\text{atom}} = -\frac{1}{2}\nabla_{r_1}^2 - \frac{1}{2}\nabla_{r_2}^2 - \frac{2}{r_1} - \frac{2}{r_2} + \frac{1}{r_{12}} \qquad \frac{1}{r_{12}} = \sum_{\lambda=0}^{\infty}\frac{4\pi}{2\lambda+1}\frac{r_<^{\lambda}}{r_>^{\lambda+1}}\sum_{q=-\lambda}^{\lambda}Y_{\lambda q}^{*}(\hat{\mathbf{r}}_1)Y_{\lambda q}(\hat{\mathbf{r}}_2)$$

$$\psi(\mathbf{r}_1,\mathbf{r}_2,t) = \sum_{L}\sum_{l_1,l_2}\frac{P_{l_1l_2}^{L}(r_1,r_2,t)}{r_1r_2}\Lambda_{l_1l_2}^{L}(\hat{\mathbf{r}}_1,\hat{\mathbf{r}}_2)$$

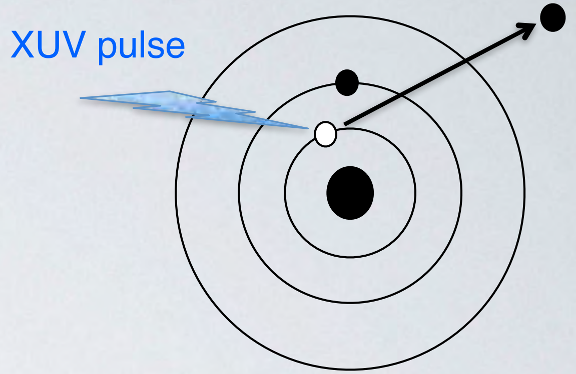
$$\Lambda_{l_1l_2}^{L}(\hat{\mathbf{r}}_1,\hat{\mathbf{r}}_2) = \sum_{m}\langle l_1 m l_2 - m | L0\rangle Y_{l_1m}(\hat{\mathbf{r}}_1)Y_{l_2,-m}(\hat{\mathbf{r}}_2)$$ Coupled spherical harmonics

Discretization of $P_{l_1l_2}^{L}(r_1,r_2,t)$ on (r_1,r_2) grid

$$r_1 \rightarrow \left(j_1 - \frac{1}{2}\right)\Delta r \qquad r_2 \rightarrow \left(j_2 - \frac{1}{2}\right)\Delta r \qquad \Rightarrow \qquad P_{l_1l_2}^{L}(r_1,r_2,t) \Rightarrow P_{l_1l_2j_1j_2}^{L}(t)$$

Ishikawa *et al*., Phys. Rev. A 72, 013407 (2005), Phys. Rev. Lett. 108, 033003 (2012), Phys. Rev. Lett. 108, 093001 (2012)

inner-shell photoionization of an excited helium atom



3D TDSE

$$\hat{H}_0 = \sum_{i=1}^{2}\left[\frac{\hat{\mathbf{p}}_i^2}{2} - \frac{2}{|\mathbf{r}_i|}\right] + \frac{1}{|\mathbf{r}_1 - \mathbf{r}_2|}$$

1D TDSE

$$\hat{H}_0 = \sum_{i=1}^{2}\left[\frac{\hat{p}_i^2}{2} - \frac{2}{\sqrt{z_i^2 + a^2}}\right] + \frac{1}{\sqrt{(z_1 - z_2)^2 + b^2}}$$

$$a = b = 0.8\,\mathrm{a.u.}$$

temporal evolution of the ionic state

1. remove the bound states of the neutral below the first ionization threshold
2. remove doubly excited (autoionizing) states
3. project on each ionic state

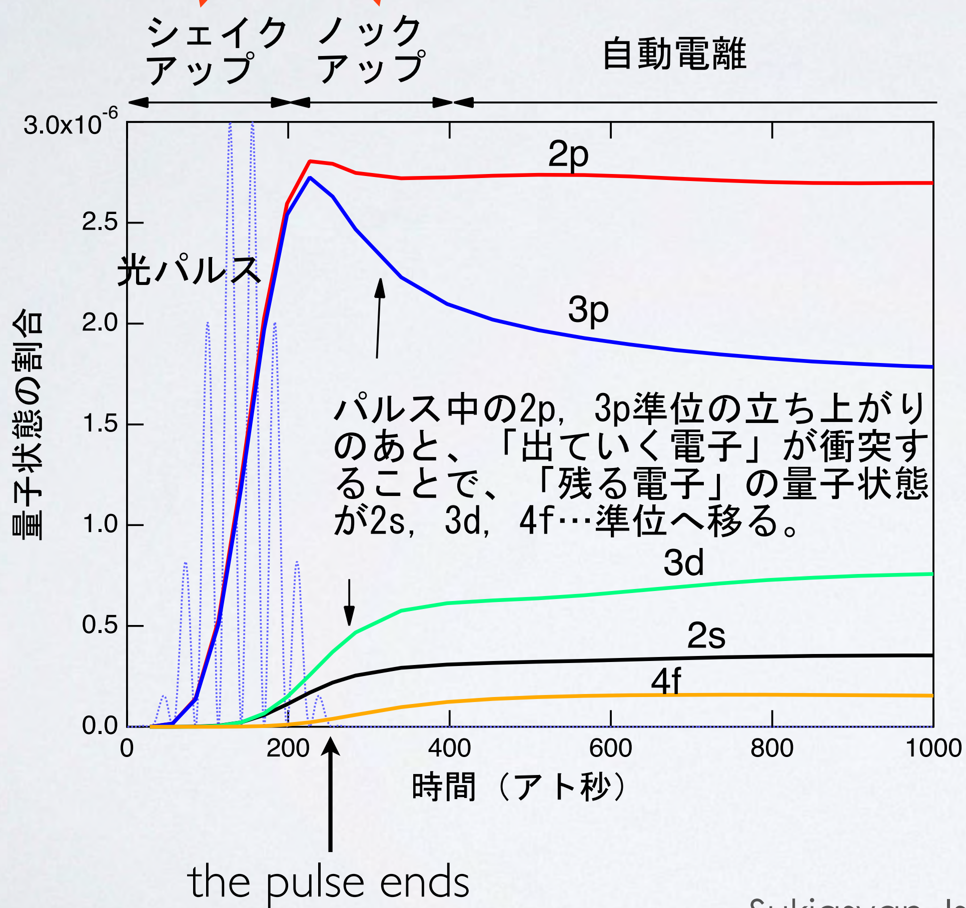
3D simulation

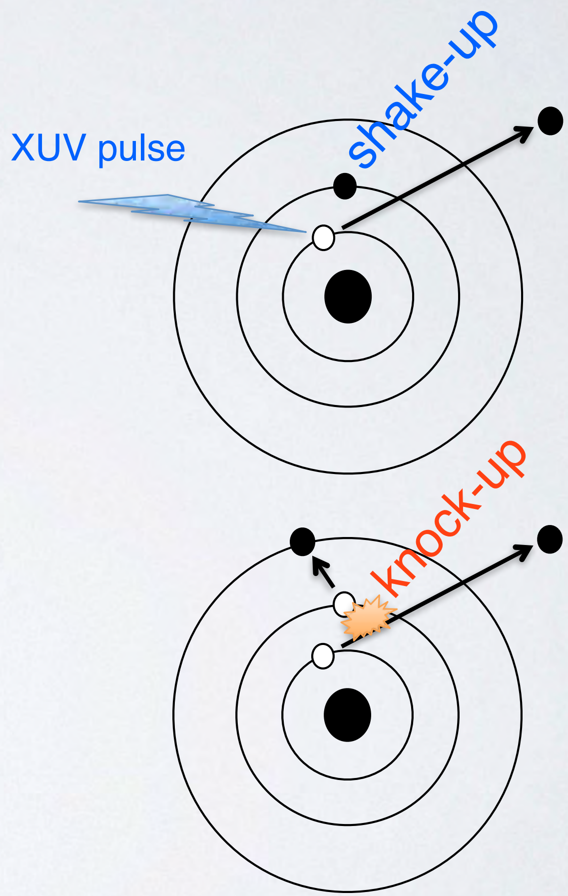
Photoionization of $1s2p\,^1P$ He

temporal evolution of the ionic state

two distinct time scales

72.9 eV, 5 cycles, 10^{12} W/cm^2

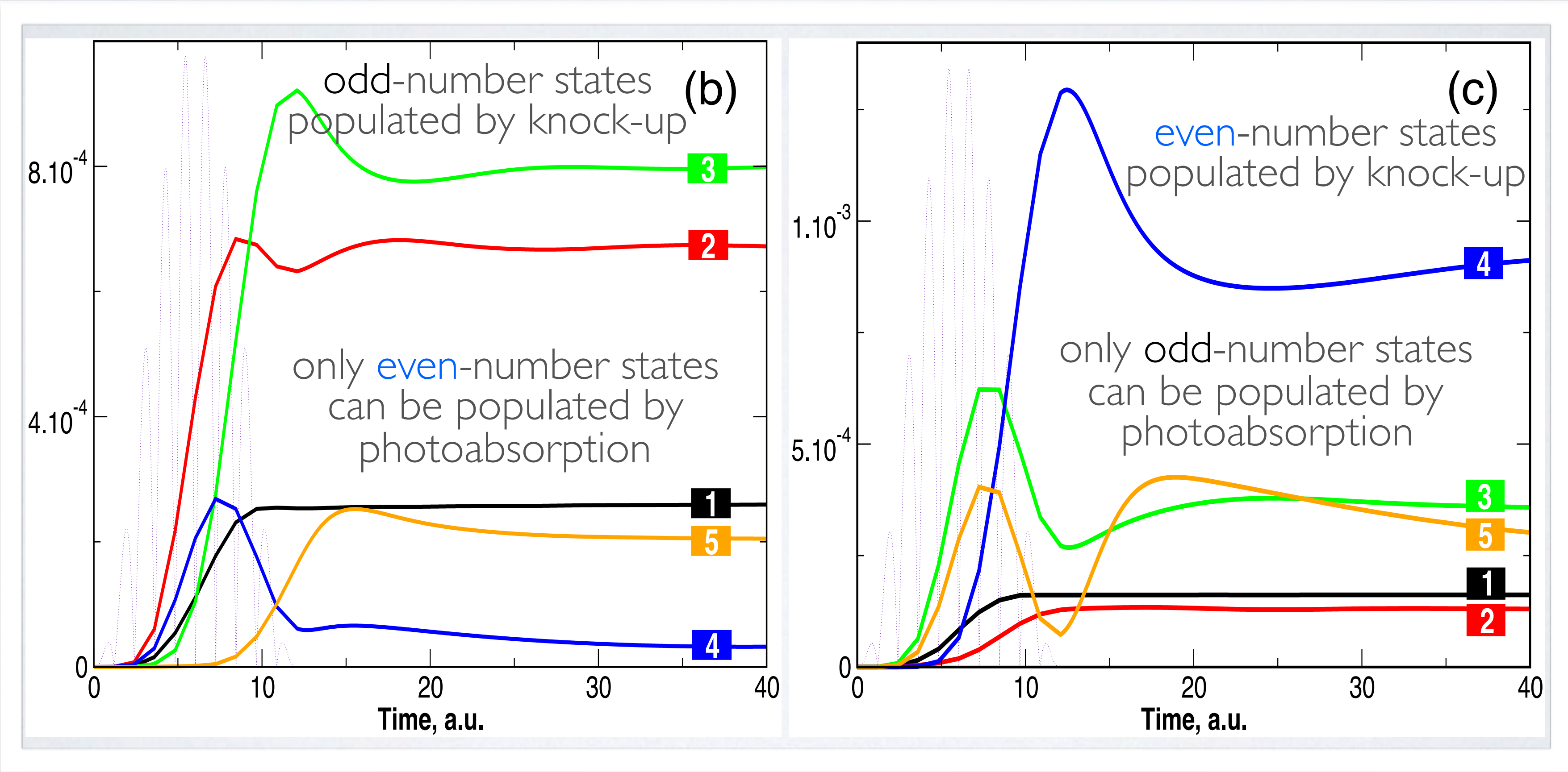




Sukiasyan, Ishikawa, Ivanov, Phys. Rev. A 86, 033423 (2012)

Similar dynamics is seen for 1D simulations

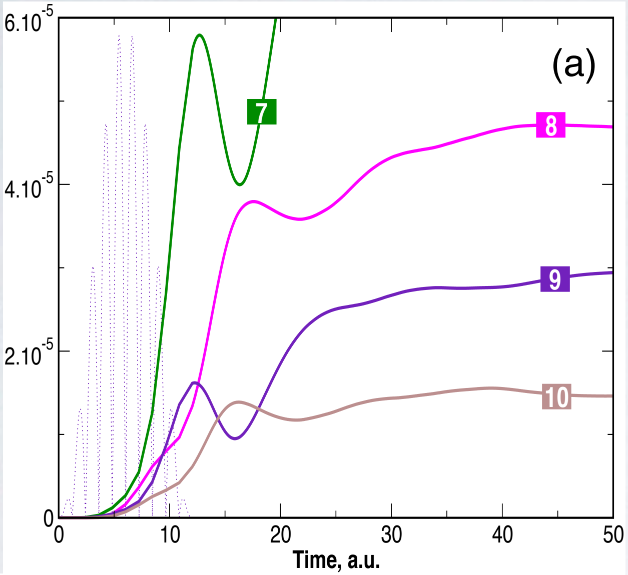
from the 1st excited atom   from the 2nd excited atom

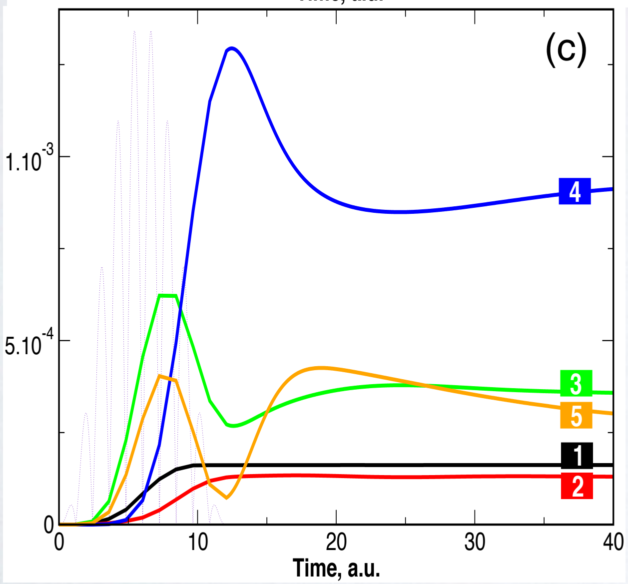


Sukiasyan, Ishikawa, Ivanov, Phys. Rev. A 86, 033423 (2012)

1D

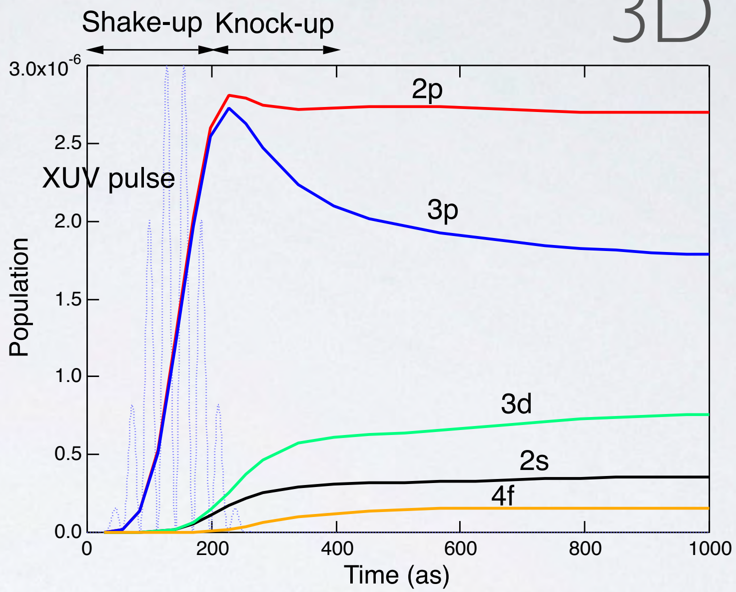
from the 2nd excited atom

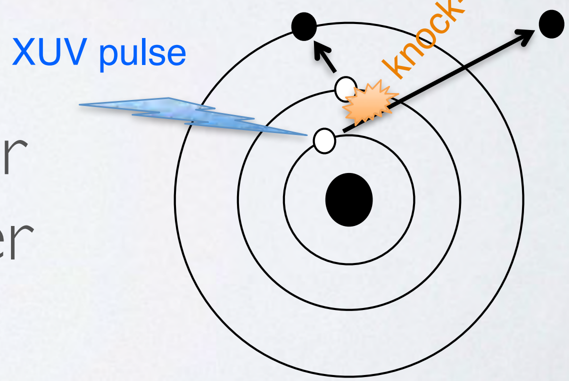




knock-up lasts longer for higher ionic channels.

3D



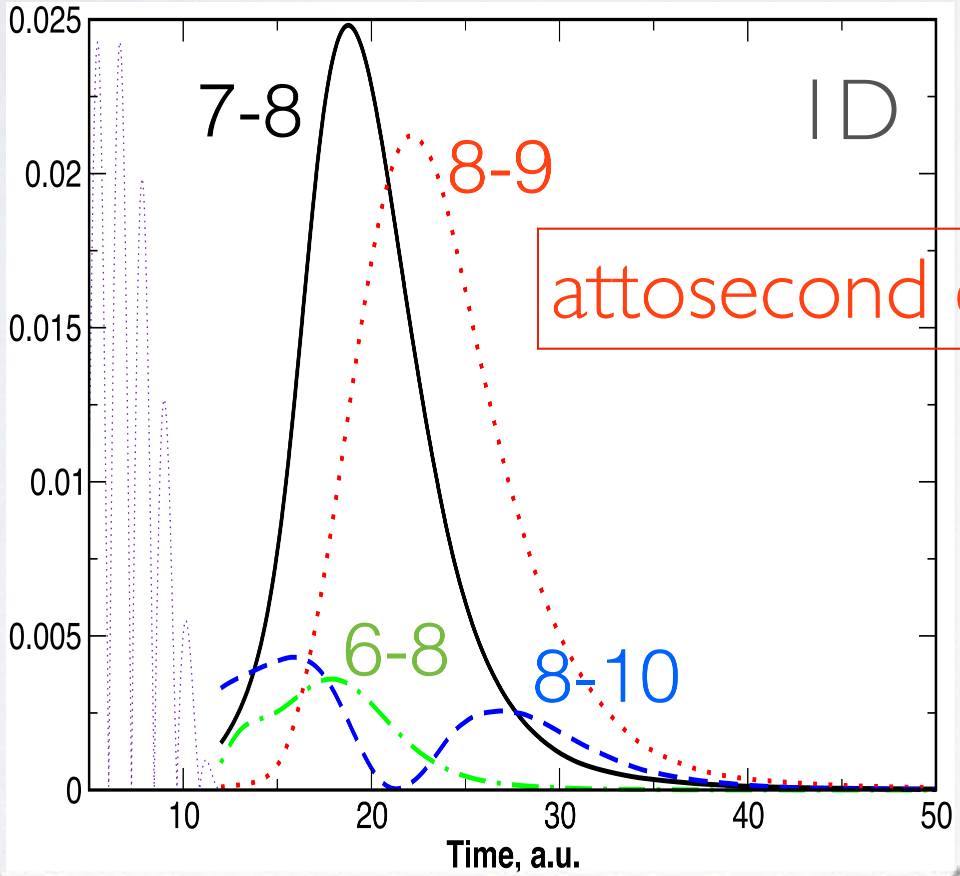


reflects the larger radii of the higher excited states

Sukiasyan, Ishikawa, Ivanov, Phys. Rev. A 86, 033423 (2012)

Time-dependent transition matrix element by the e-e interaction

$$\left|\langle \Phi_i(z_1, z_2, t)\,|\,1/\sqrt{(z_1 - z_2)^2 + b^2}\,|\,\Phi_j(z_1, z_2, t)\rangle\right|^2$$



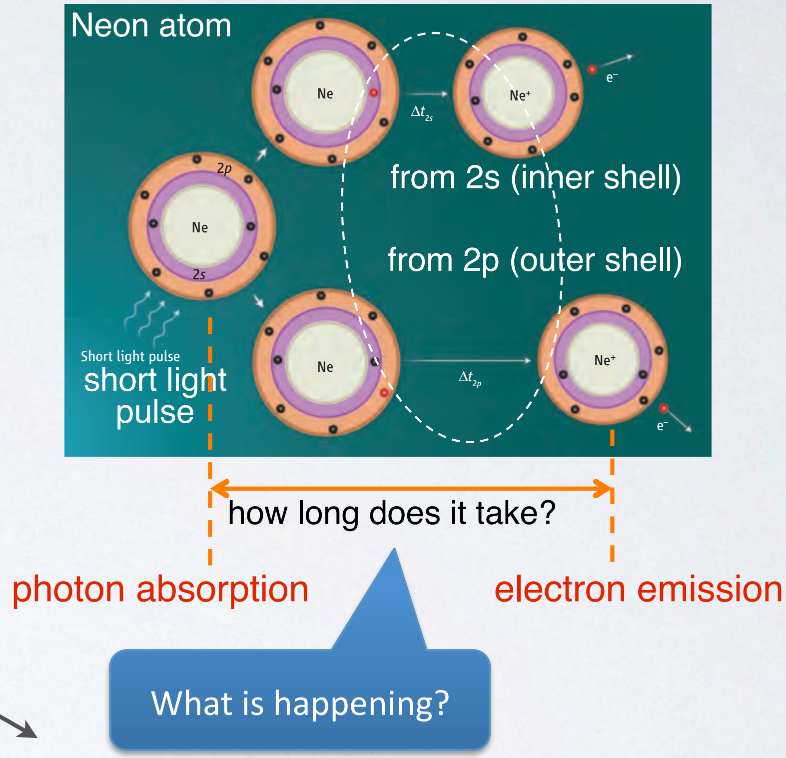
attosecond cascades

increasing delays reflect the larger radii of the excited states involved

Sukiasyan, Ishikawa, Ivanov, Phys. Rev. A 86, 033423 (2012)

knock-up in attosecond photoionization of an excited helium atom

Post-ionization interaction of the outgoing core electron with the outer spectator electron



Dynamic multi-electron correlation

Sukiasyan, Ishikawa, Ivanov, Phys. Rev. A 86, 033423 (2012)