Quantum Beam Engineering E

量子ビーム発生工学特論E

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高次高調波発生と
アト秒レーザーパルス

high-order harmonic generation & attosecond laser pulse

High-harmonic generation
高次高調波発生

more details at **Advanced Laser and Photon Science** on May 22

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)

$$D = \varepsilon_0 E + P$$

$$P = \varepsilon_0 \left[\chi^{(1)} E + \underbrace{\chi^{(2)} E^2 + \chi^{(3)} E^3 + \cdots}_{\text{非線形分極 (nonlinear)}} \right]$$

線形分極 linear polarization

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

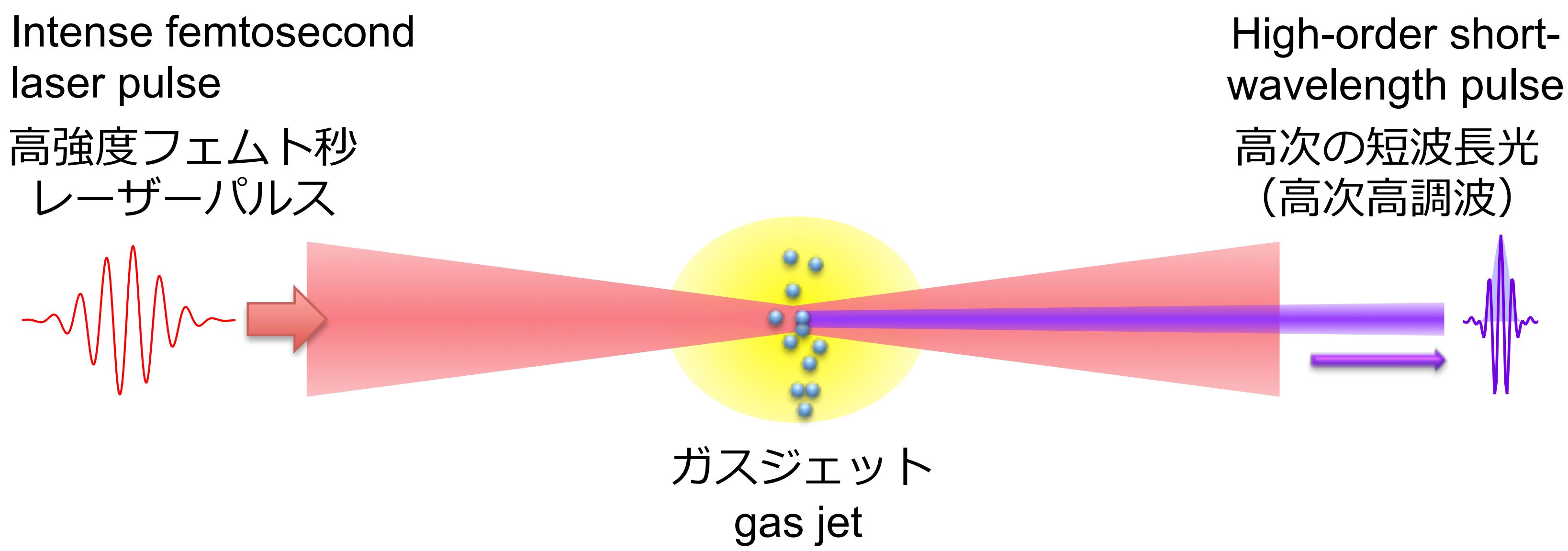
$$\nabla \times \nabla \times \mathbf{E} = -\mu_0 \frac{\partial^2 \mathbf{D}}{\partial t^2}$$

3ω： 3次高調波(3rd harmonic)

5ω： 5次高調波(5th harmonic)

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

discovered in 1987



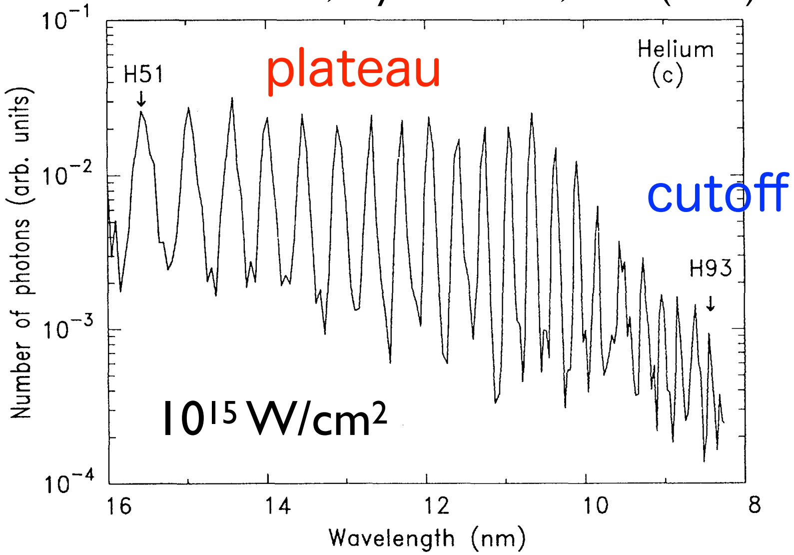
Highly nonlinear optical process in which the frequency of laser light is converted into its integer multiples. Harmonics of very high orders are generated.

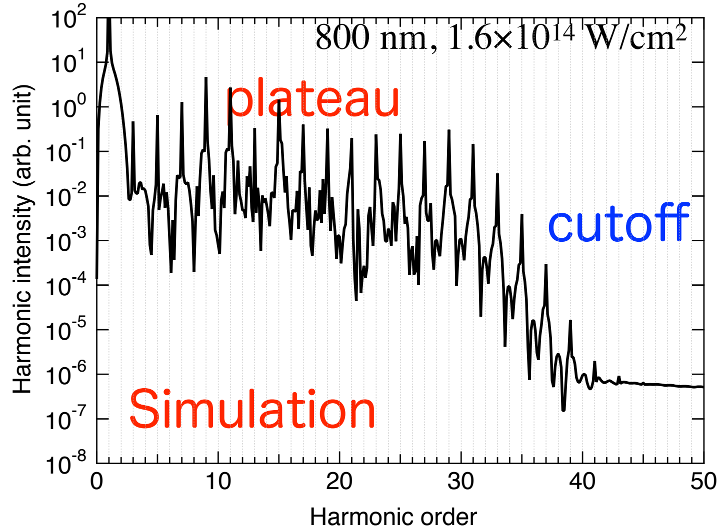
新しい極端紫外・軟エックス線光源として注目される。

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

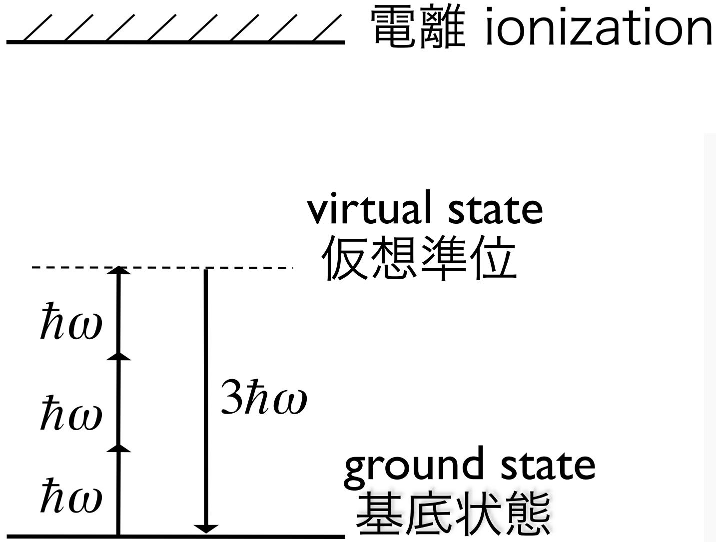
$$E_c \approx I_p + 3U_p \qquad U_p(\mathrm{eV}) = \frac{e^2 E_0^2}{4m\omega^2} = 9.3 \times 10^{-14} I(\mathrm{W/cm^2})\lambda^2(\mu\mathrm{m})$$

ponderomotive energy

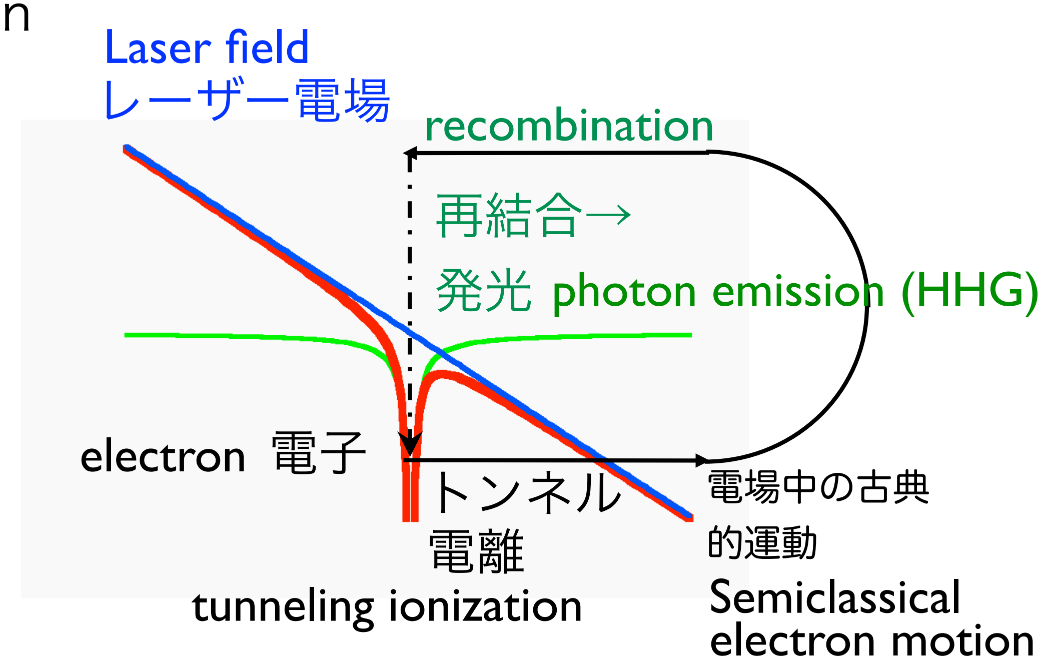
- 摂動論的には解釈できない(non-perturbative)

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

摂動論的高調波
perturbative



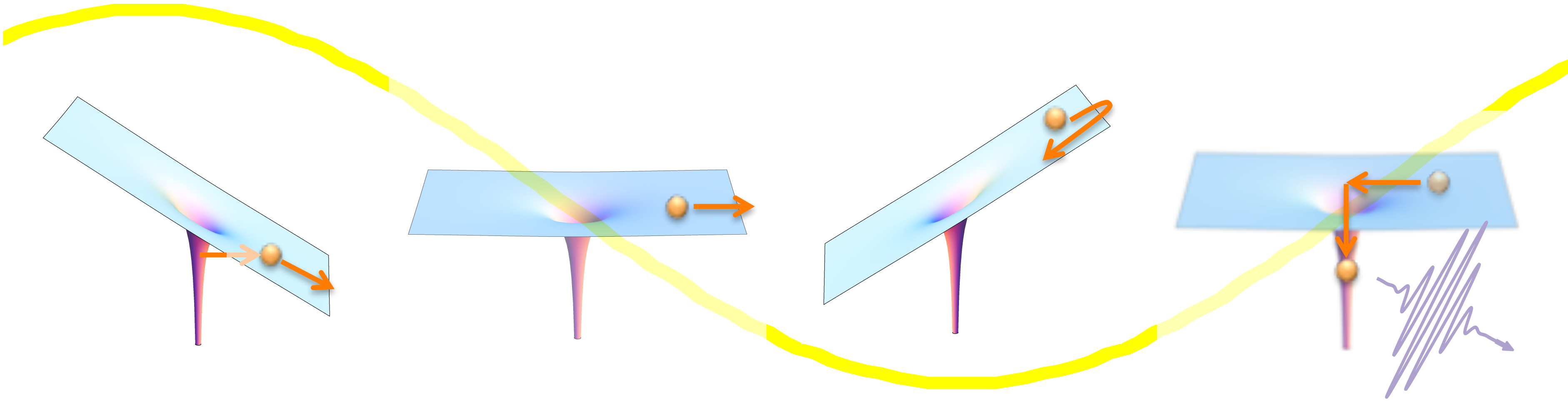
高次高調波（非摂動論的）
HHG(non-perturbative)



3-step model

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

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



高次高調波発生の３ステップモデル 3-step model of HHG

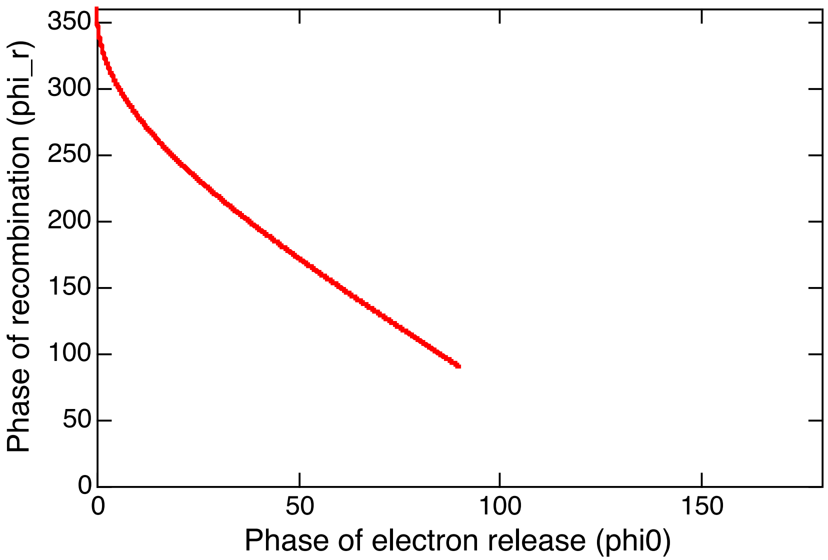
Paul B. Corkum, Phys. Rev. Lett. 71, 1994 (1993)

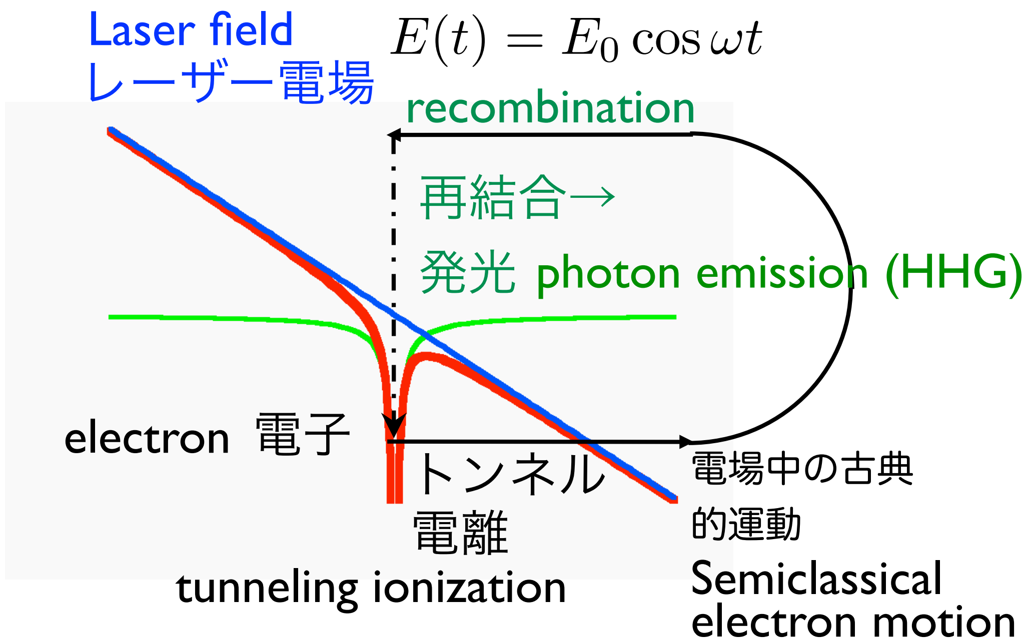
Ionization at $\omega t_0 = \phi_0$

$$z = \frac{E_0}{\omega^2}\left[(\cos\phi - \cos\phi_0) + (\phi - \phi_0)\sin\phi_0\right]$$

$$E_{\rm kin} = 2U_p(\sin\phi - \sin\phi_0)^2$$

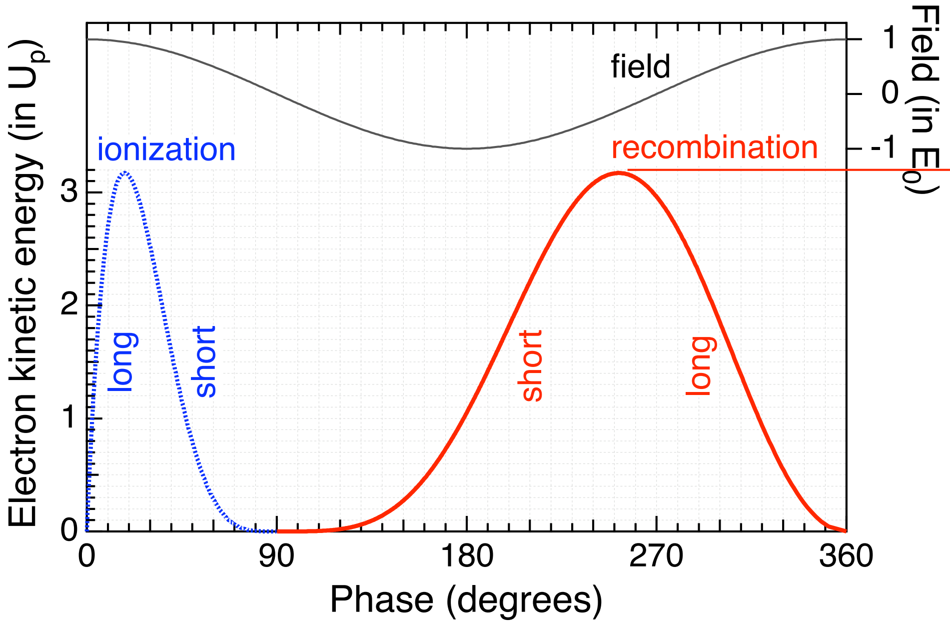
Recombination at $\phi = \phi_{\rm ret}(\phi_0)$, which satisfies $z = 0$





高次高調波発生の３ステップモデル
3-step model of HHG

Simple explanation of the cutoff law
カットオフ則のシンプルな説明



There is the maximum kinetic energy which is classically allowed.

$$E_c = I_p + 3.17U_p$$

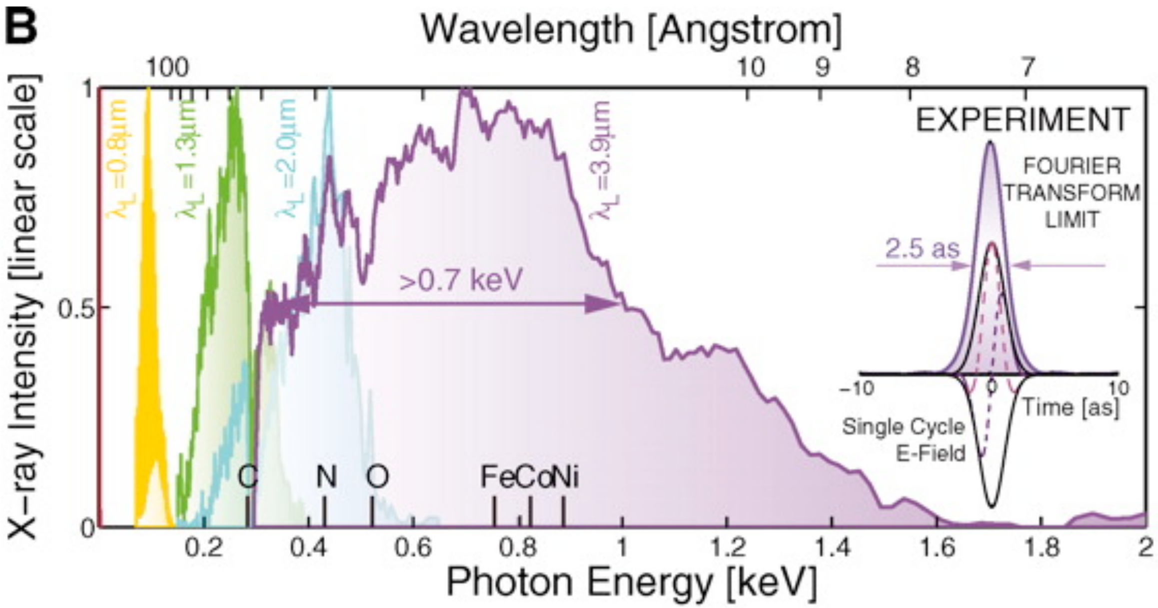
There are two pairs of ionization and recombination times which contribute to the same harmonic energy.

Short and long trajectories

more details in *Advanced Laser and Photon Science*

Even up to 1.6 keV, > 5000 orders

(almost) x-ray!



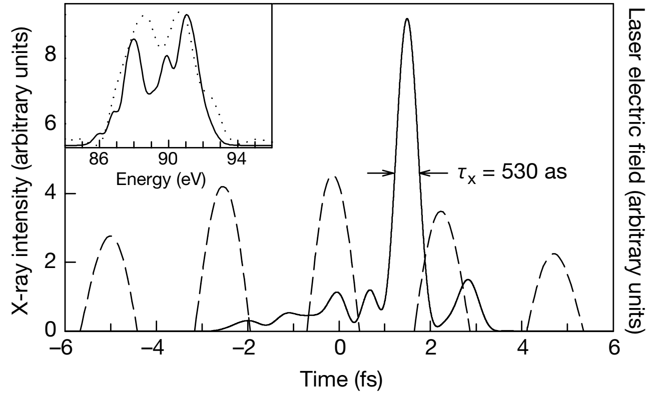
Popmintchev et al., Science 336, 1287 (2012)

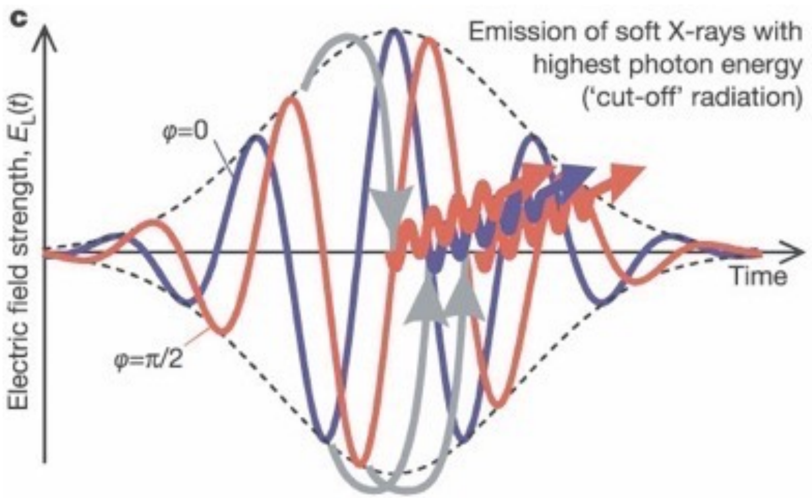
a new type of laser-based radiation source

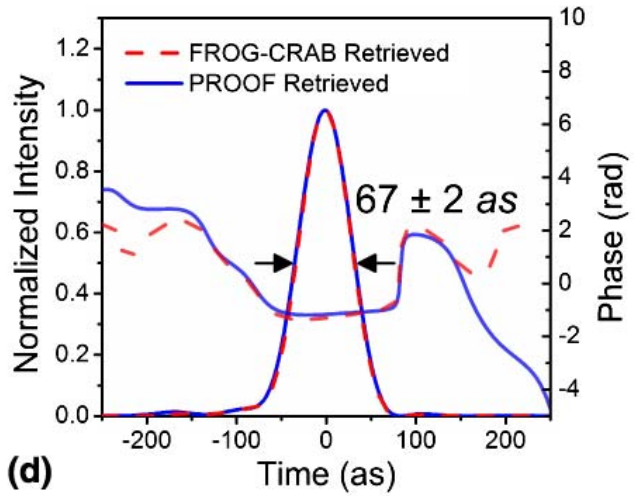
レーザーをベースにした新しいタイプの放射線源

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)







Zhao et al. (2012)

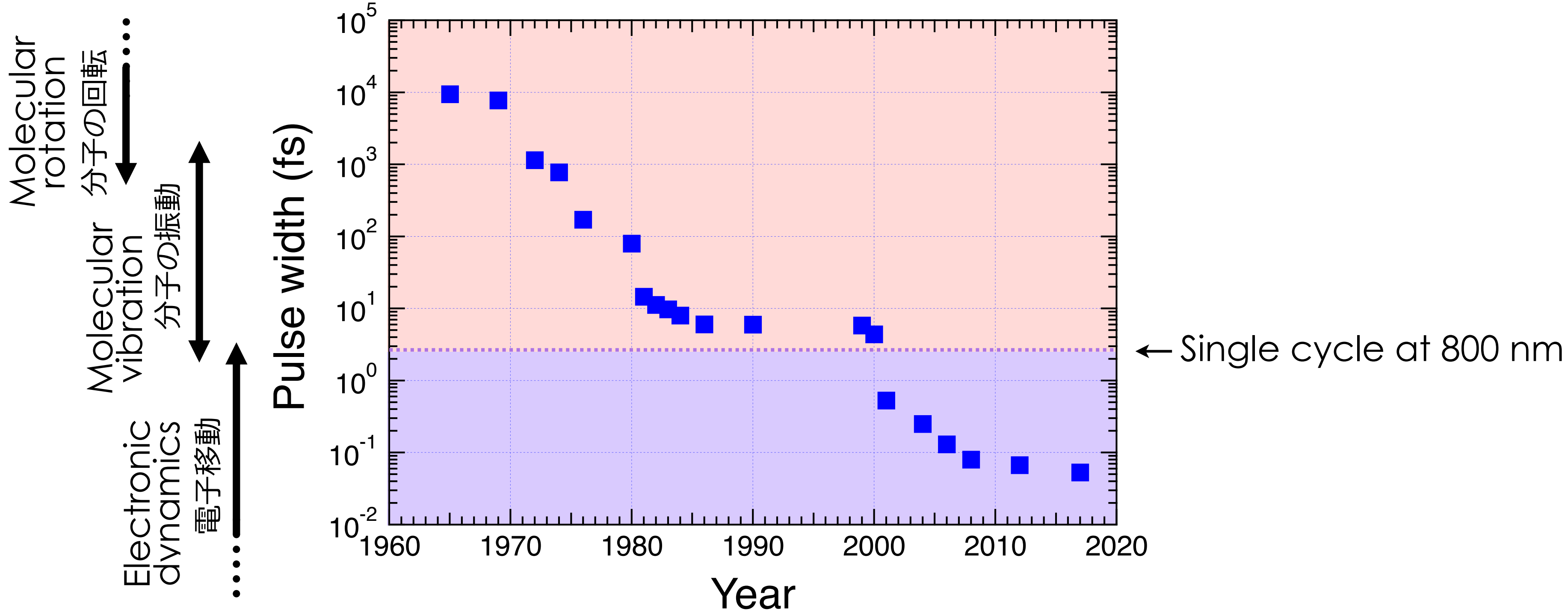
Light emission takes place only once.

光の放出は１回だけ

Attosecond (10^{-18} sec) pulse
アト秒パルス

From femtosecond to attosecond

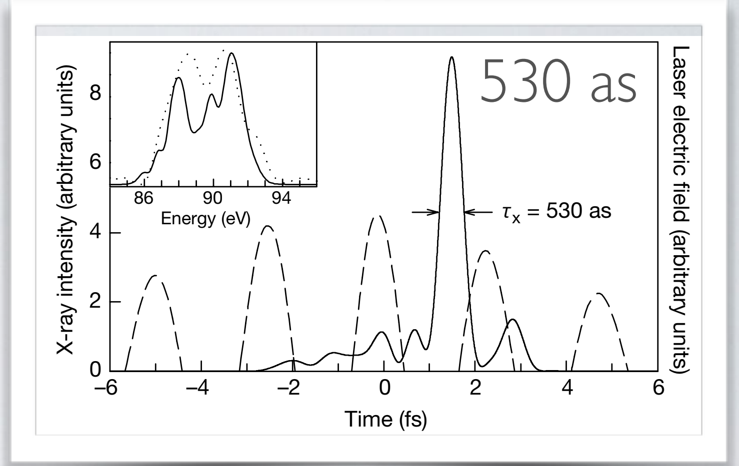
10^{-15} sec 10^{-18} sec



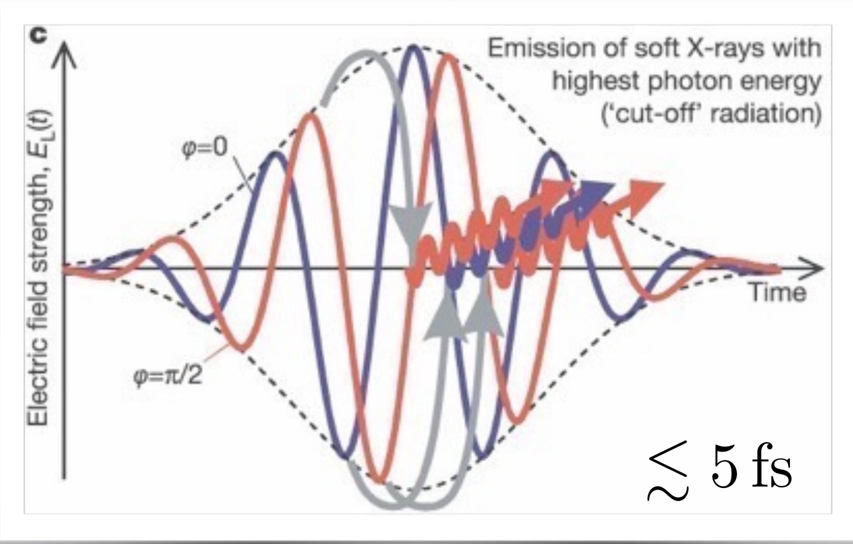
How to generate an isolated attosecond pulse (IAP)

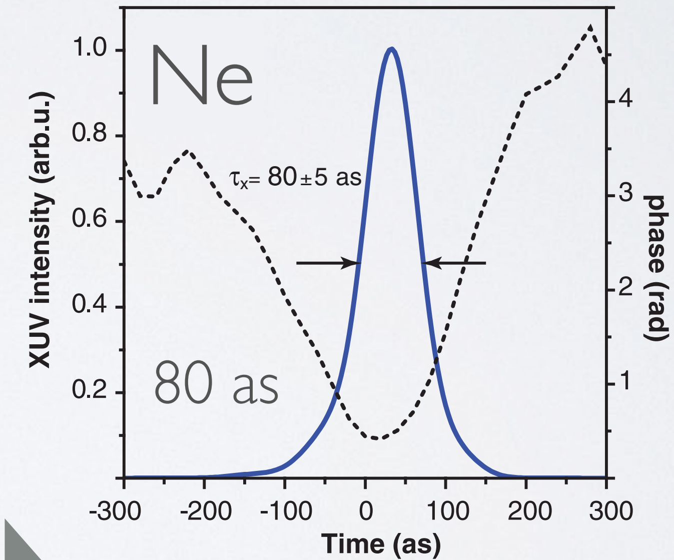
Isolated attosecond pulse generation by a few-cycle laser pulse

Hentschel et al. Nature 414, 509 (2001)



Baltuska et al. Nature 421, 611 (2003)



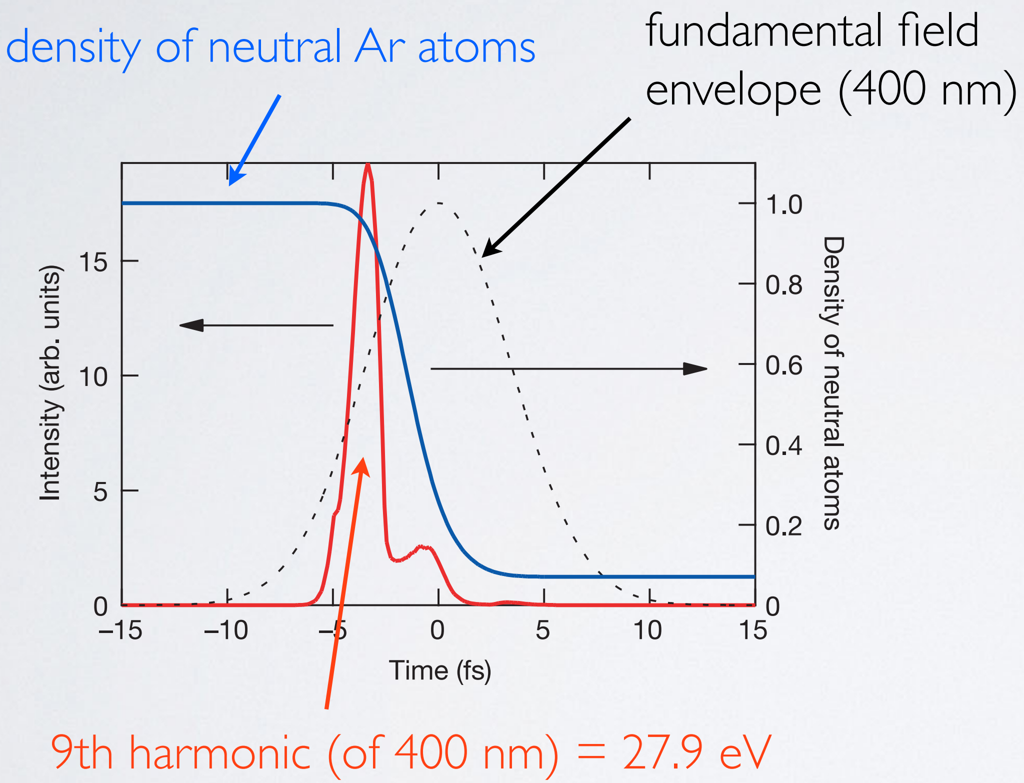


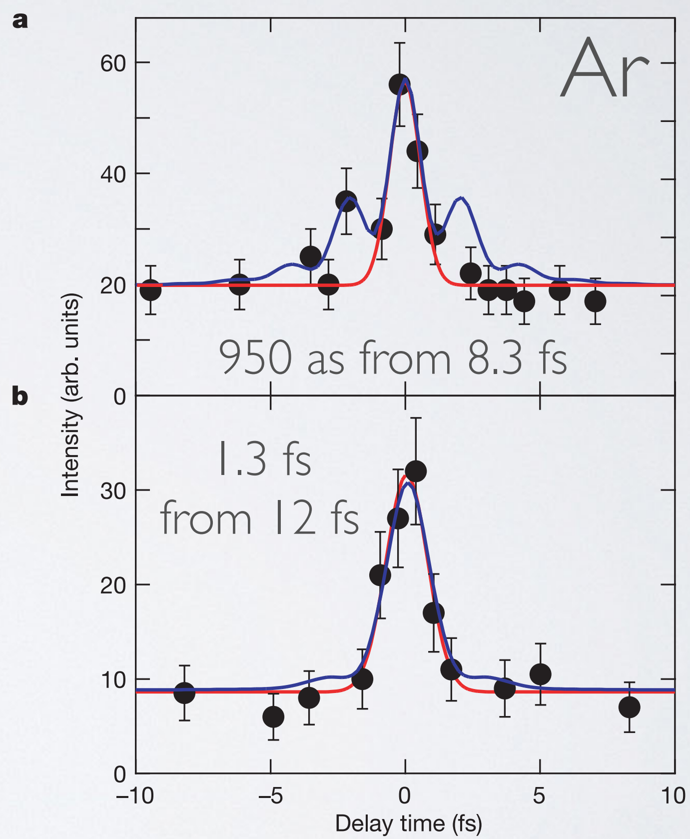
Goulielmakis et al. Science 320, 1614 (2008)

Light emission takes place only once. → **Attosecond (10^{-18} sec) pulse**

IONIZATION SHUTTER

HHG is suppressed when neutral atoms are depleted





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

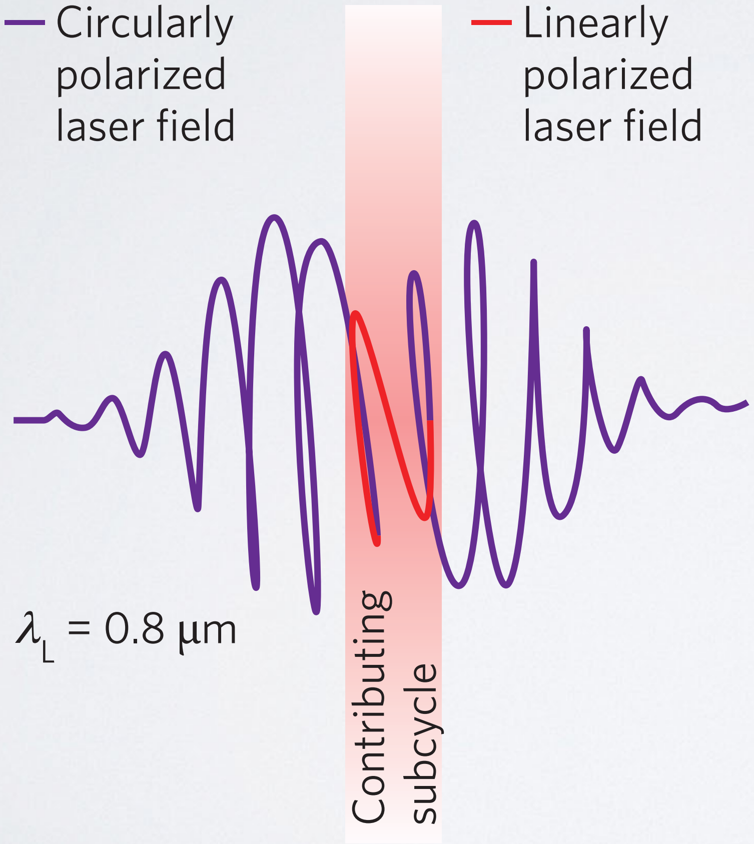
Sekikawa *et al.*, Nature 432, 605 (2004)

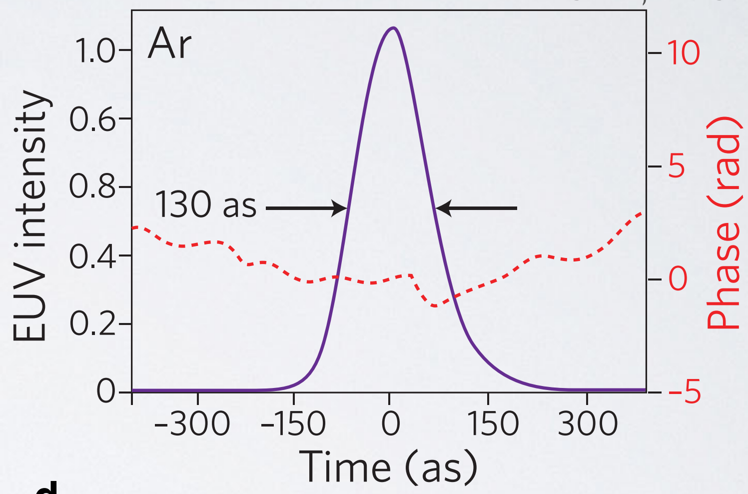
POLARIZATION GATING (PG)

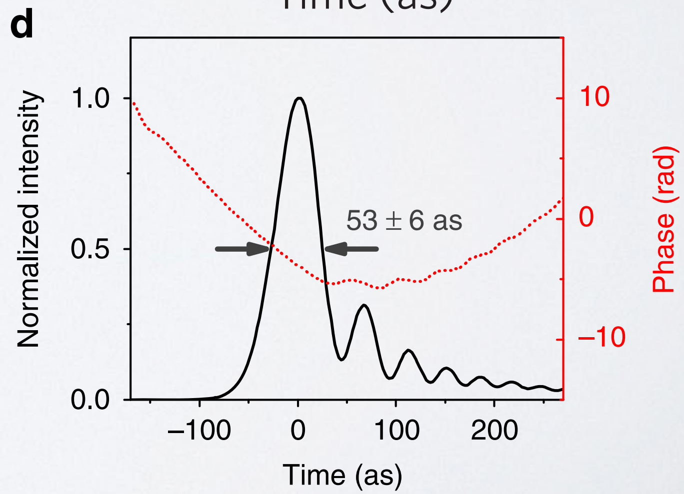
HHG is suppressed when circular polarization is used

counter-rotating circularly polarized pulses with a delay

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







Lin et al. (2017)

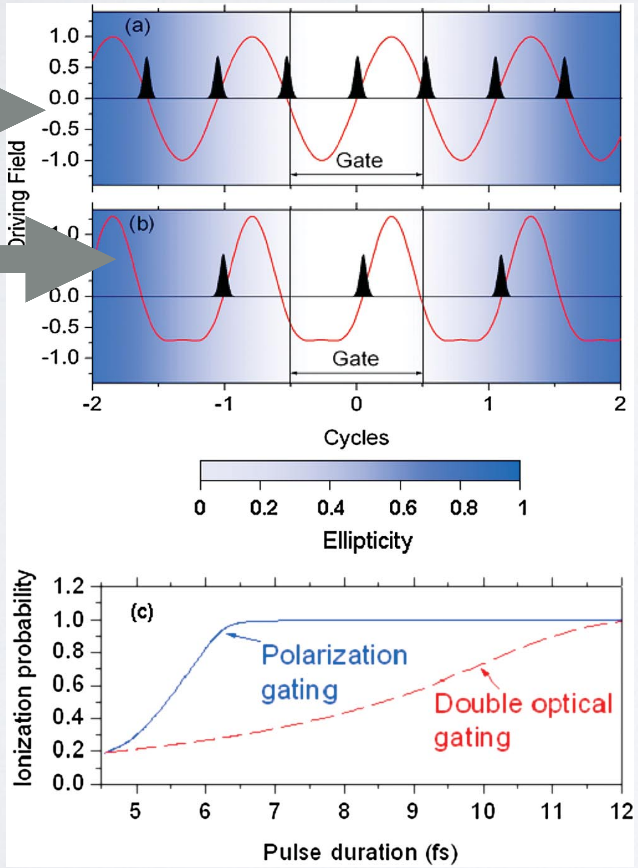
DOUBLE OPTICAL GATING (DOG)

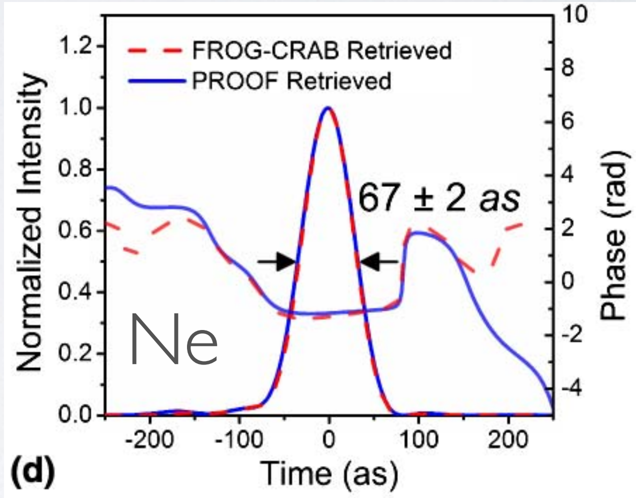
Polarization gating + two-color gating

ω

$\omega + 2\omega$

with second-harmonic field





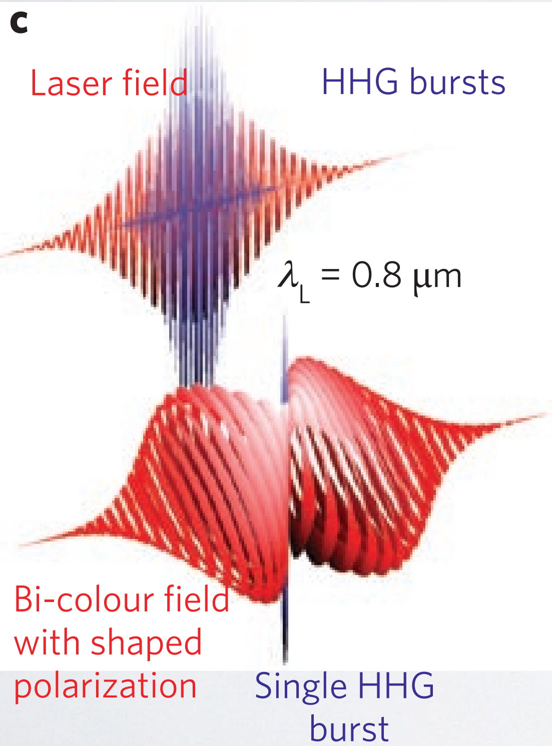
IAP generation from a ~10 fs pulse

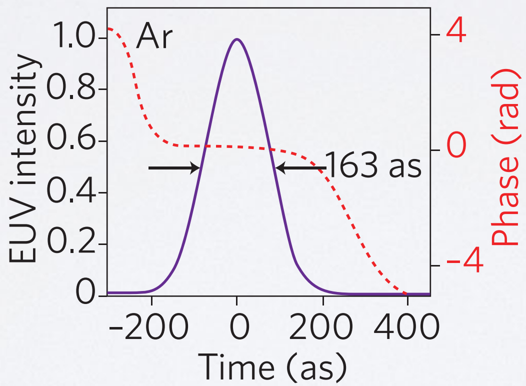
Mashiko et al., PRL 2008, 103906 (2008)

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

GENERALIZED DOUBLE OPTICAL GATING (GDOG)

Elliptical instead of circular polarization





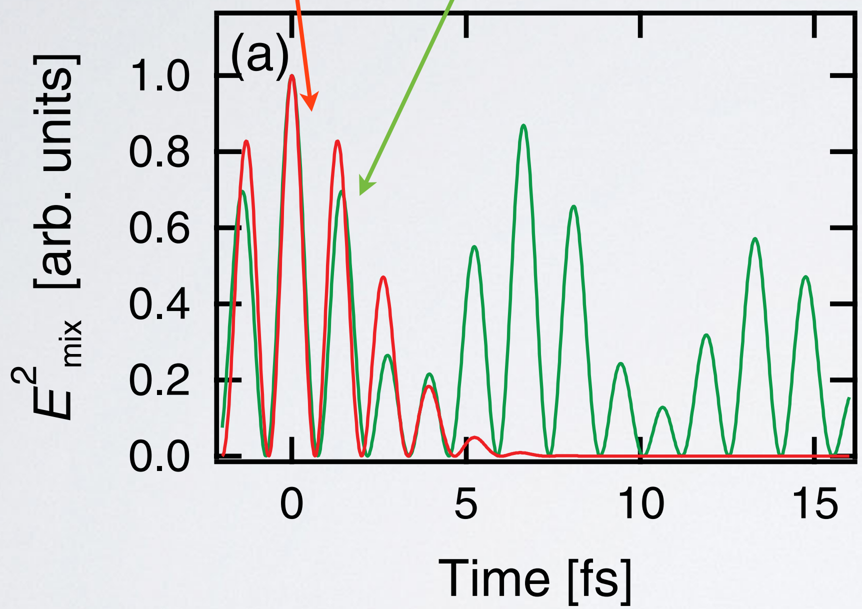
IAP generation from a > 20 fs pulse without need of carrier-envelope stabilization

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

INFRARED TWO-COLOR SYNTHESIS

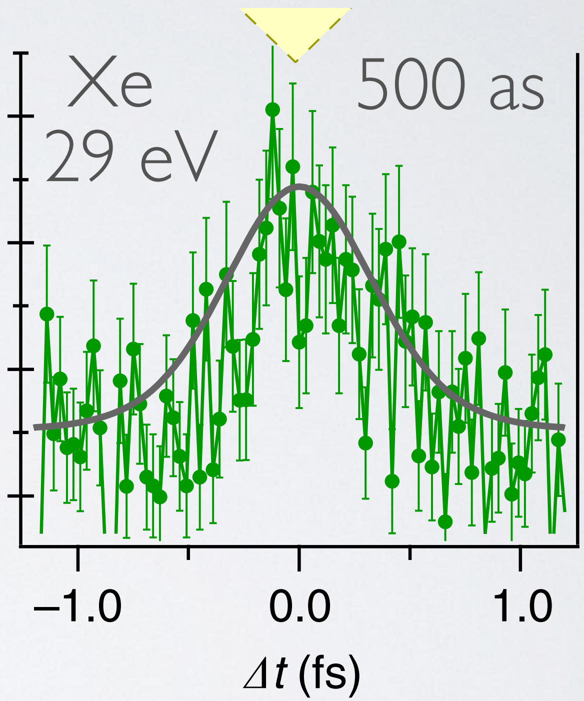
800 nm + 1300 nm two-color driving field

800 nm   800 nm + 1300 nm



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

autocorrelation trace

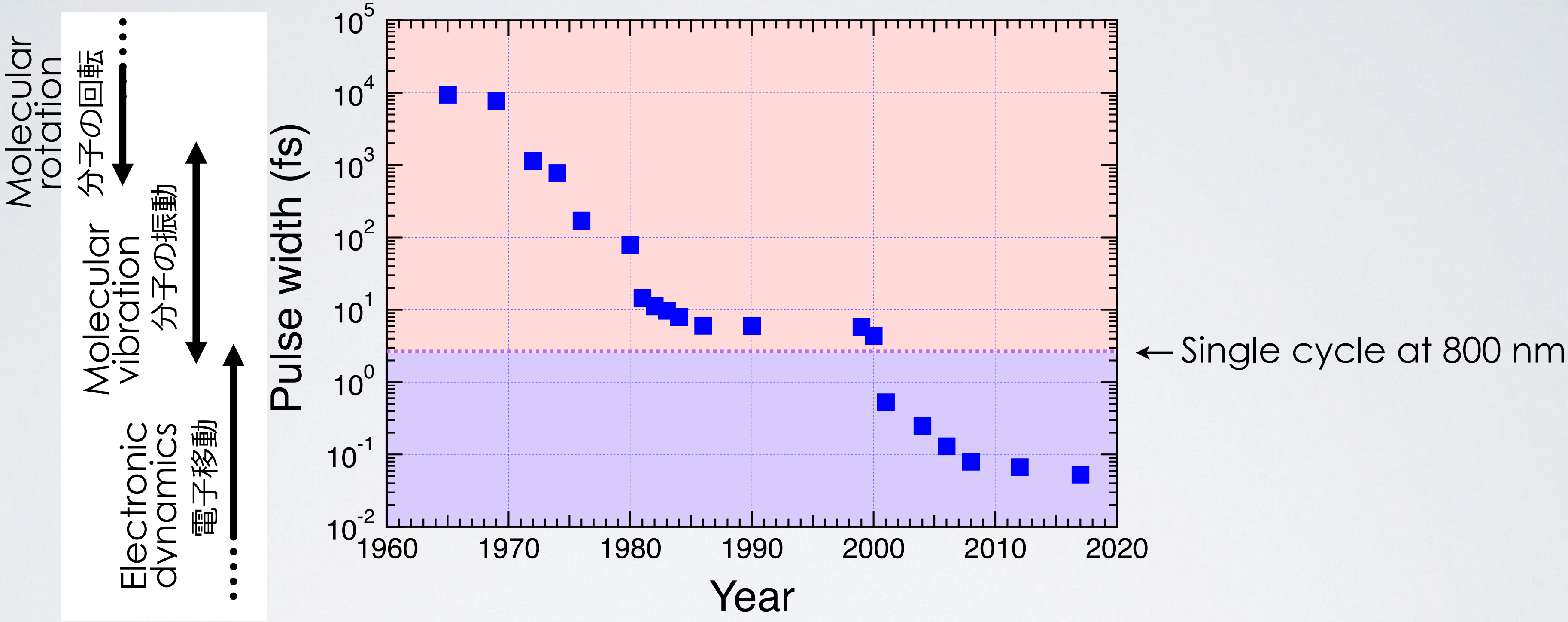


Takahashi et al., Nat. Commun. 4, 2691 (2013)

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

more than 100 times more energetic than previously reported

FROM FEMTOSECOND TO ATTOSECOND



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

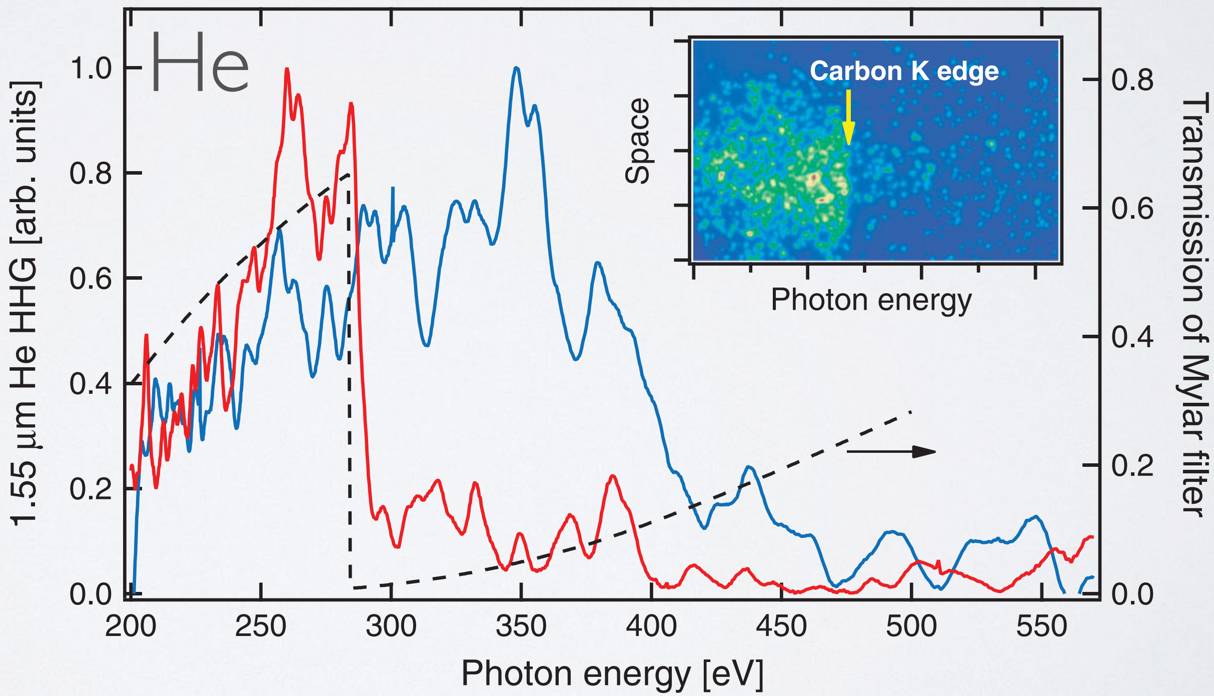
spectral range between the *K*-absorption edges of C (284 eV) and O (543 eV)

→ absorbed by biological samples but not by water

→ attractive for high-contrast biological imaging

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

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

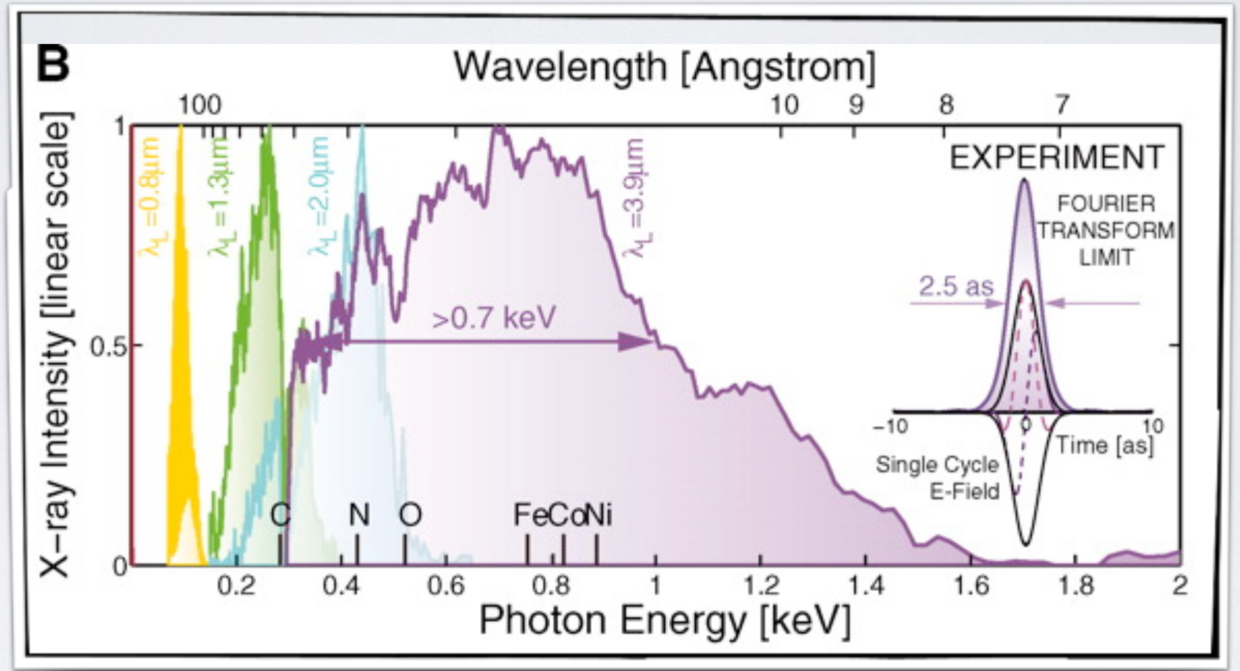


Takahashi et al., PRL 101, 253901 (2008)

keV HHG

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

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



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

a new type of laser-based radiation source

supplementary materials

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}

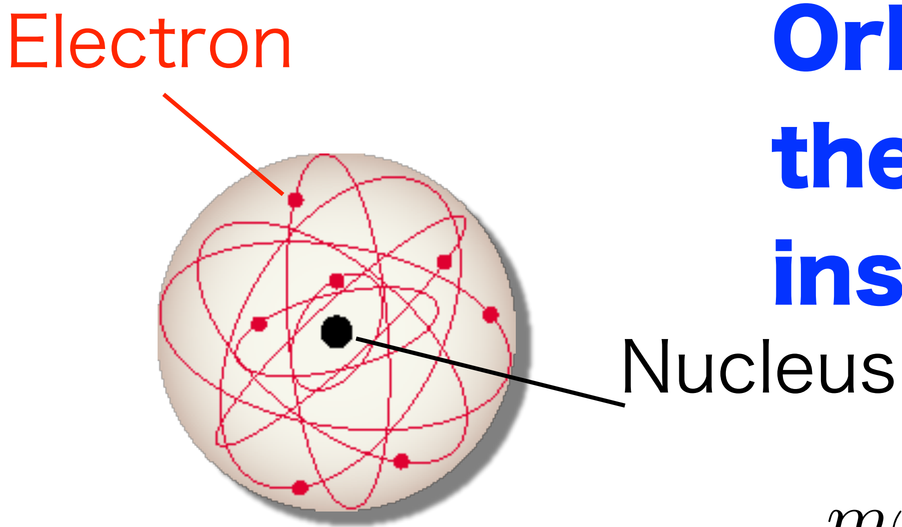
Light propagates during 30 fs …

$$3 \times 10^8 (\mathrm{m/s}) \times 30 \times 10^{-15} (\mathrm{s}) = 9 \times 10^{-6} (\mathrm{m}) = 9\ \mu\mathrm{m}$$

Why so short pulses?

necessary ‘shutter speed’ for snapping ultrafast motion

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

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

A method to analyze ultrafast processes with a laser field.

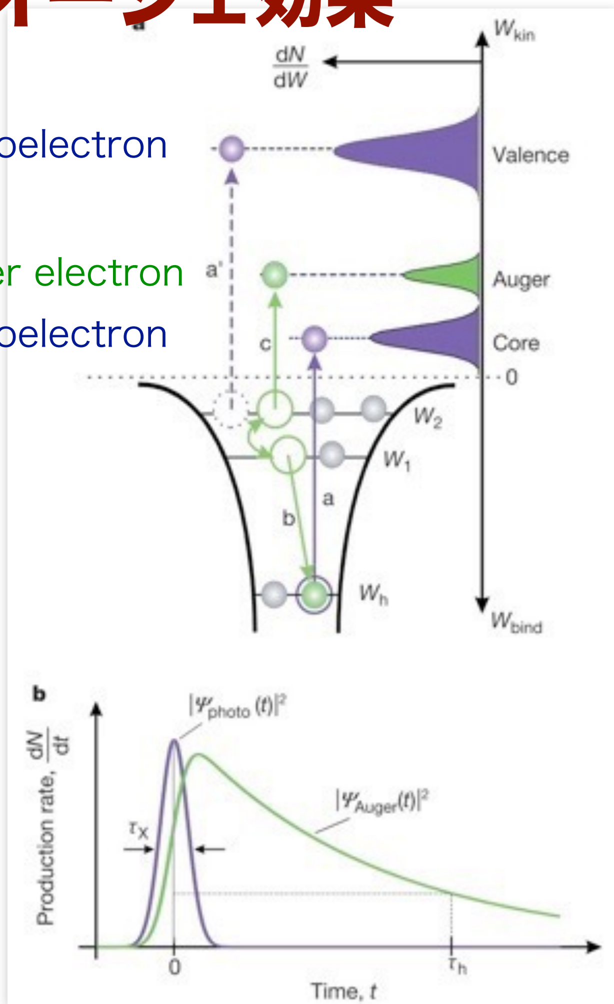
Auger effect

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

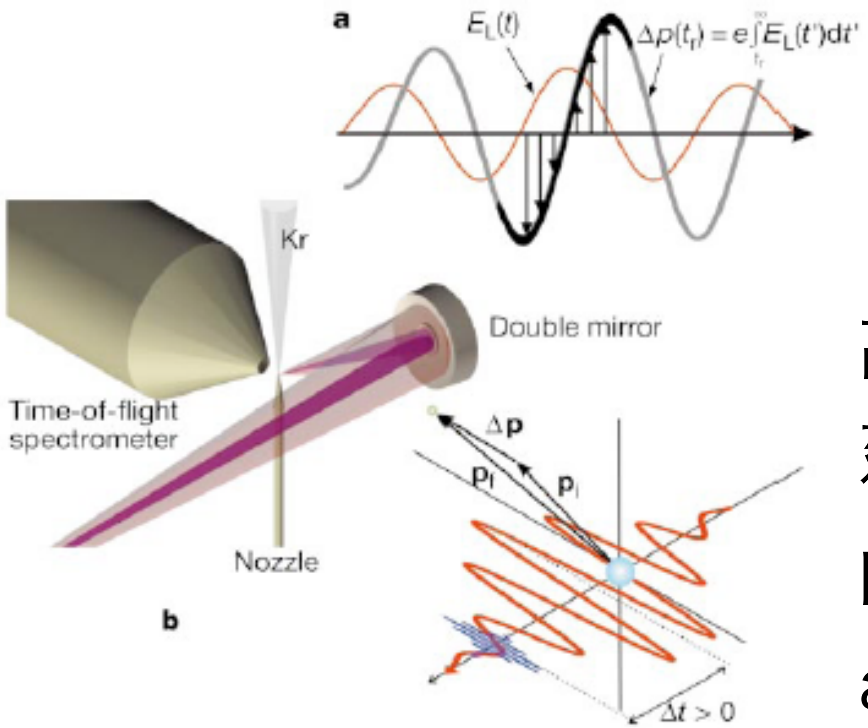
Observation of the ejection of Auger electrons

→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）

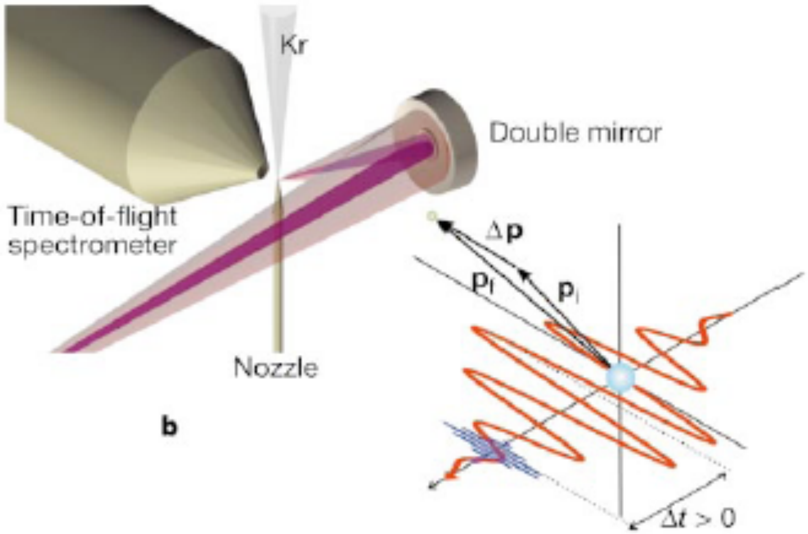
How to measure the electron ejection time?



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）

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

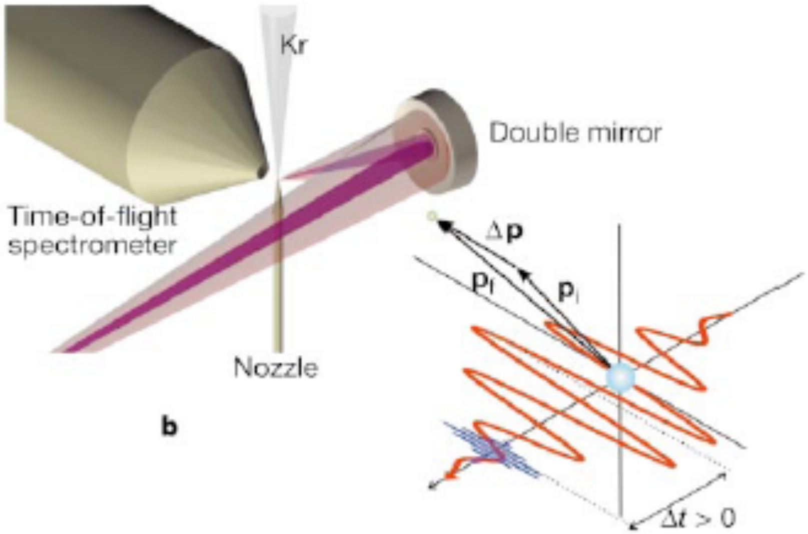
検出器での運動量 Momentum at the detector $p = p_0 + \Delta p$

$$\Delta p = -e\int_{t_r}^{\infty} E(t)dt = -eA(t_r) \approx \frac{eE_0(t)}{\omega}\sin(\omega t_r + \phi) = \sqrt{4mU_p(t_r)}\sin(\omega t_r + \phi)$$

検出器での運動エネルギー Kinetic energy at the detector

$$W \approx W_0 + \frac{p_0 \Delta p}{m} = W_0 + \sqrt{8W_0U_p(t_r)}\sin(\omega t_r + \phi)$$

How to measure the electron ejection time?

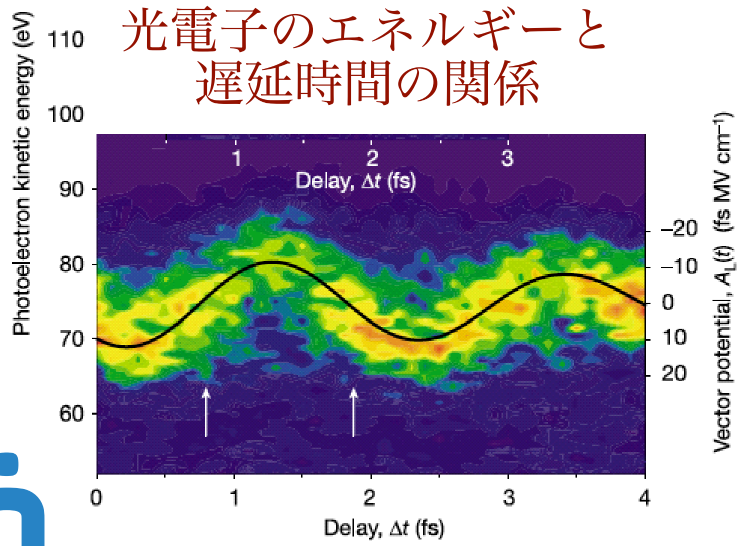


検出器での運動エネルギー

$$W \approx W_0 + \sqrt{8W_0U_p(t_r)}\sin(\omega t_r + \phi)$$

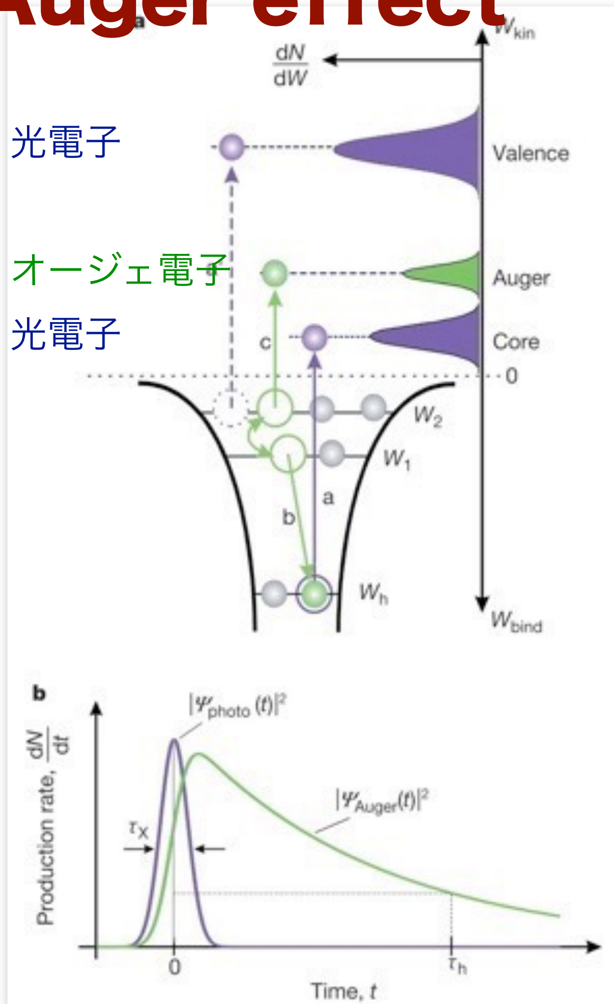
Electron kinetic energy

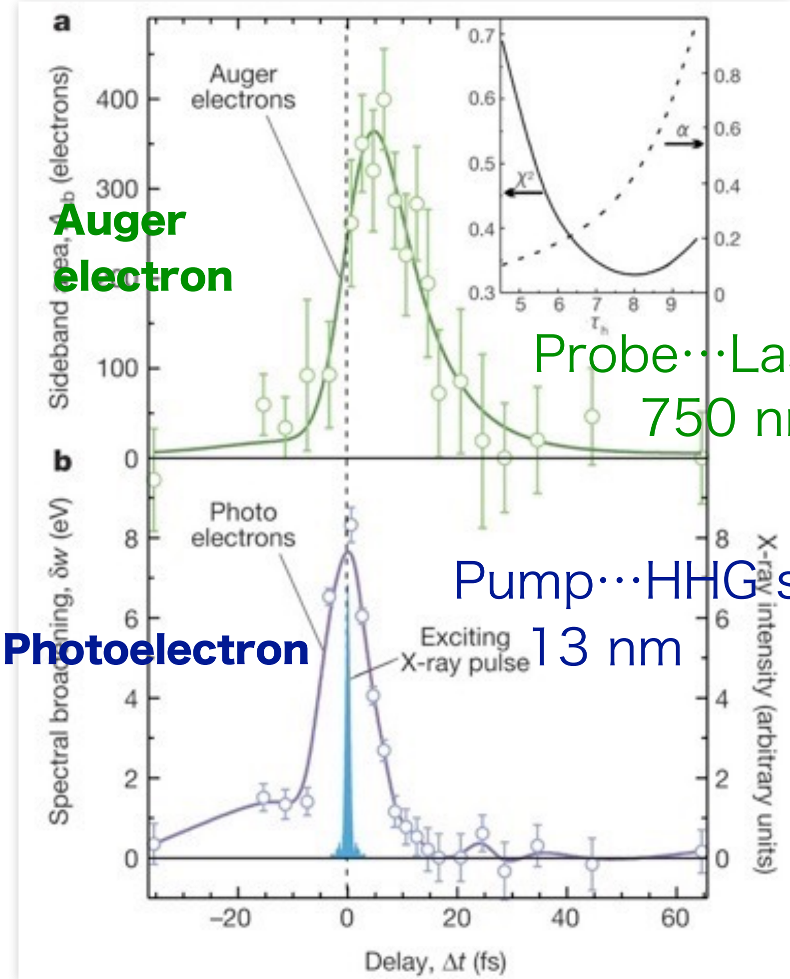
Ejection time



Life time of the Auger decay～8 fs

Auger effect





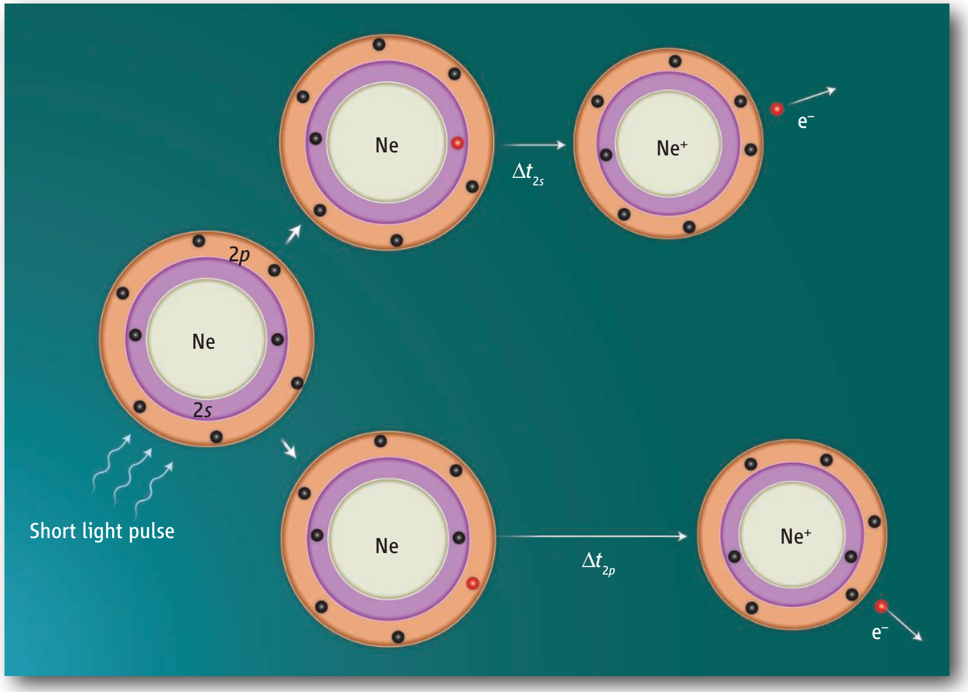
10フェムト秒程度の超高速過程が見える！

Ultrafast process ~ 10 fs

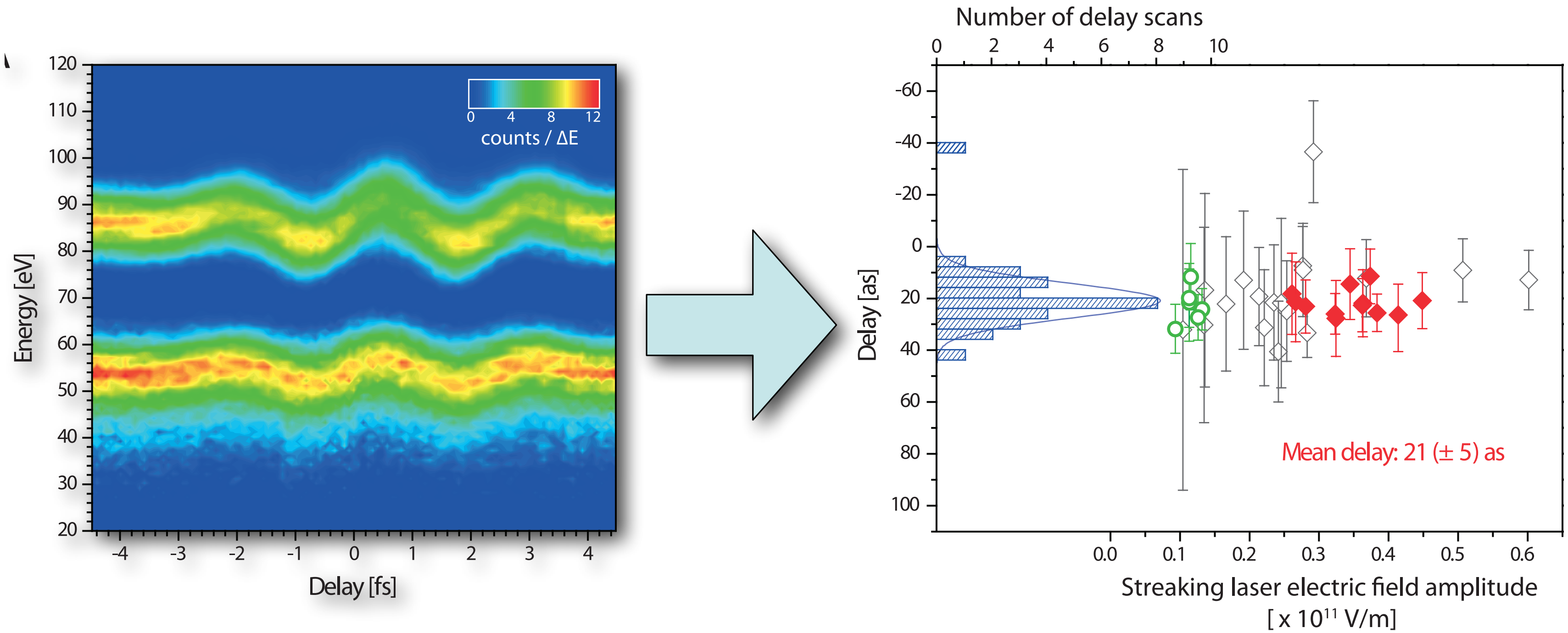
Delay in photoemission
光電効果には何アト秒かかるか？

When Does Photoemission Begin?

The photoelectric effect is usually considered instantaneous.



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



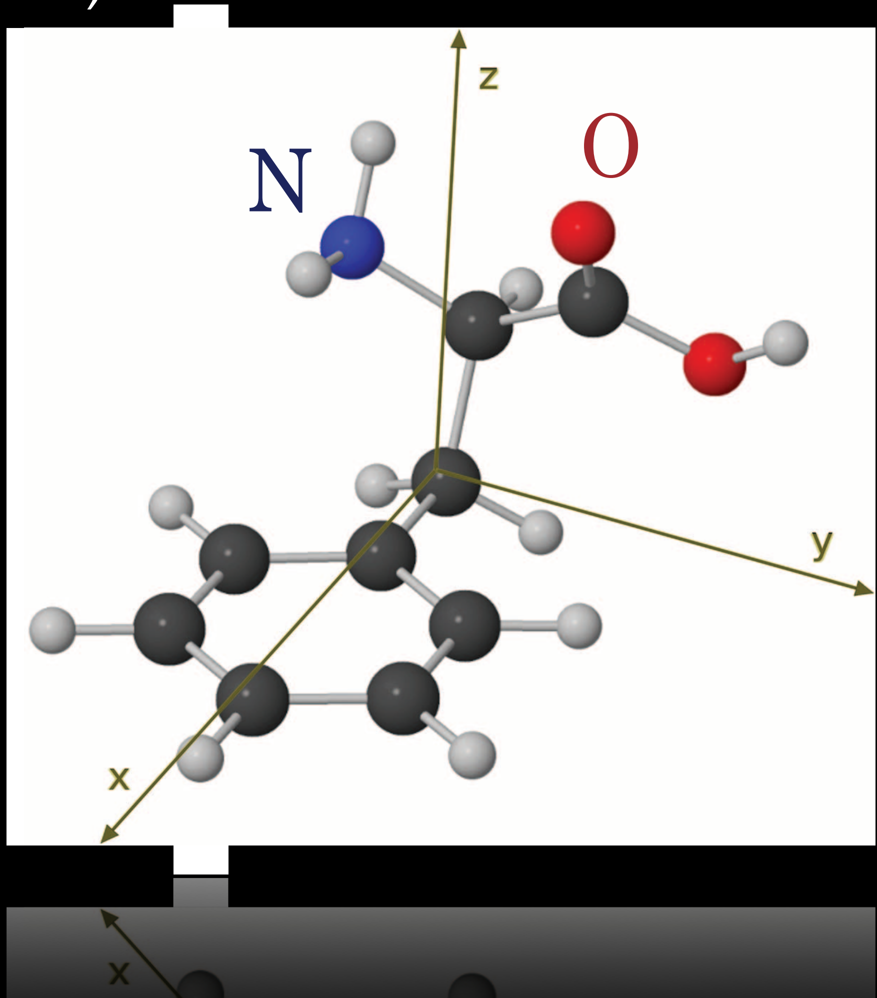
Schultze *et al.*, Science 328, 1658 (2010)

フェニルアラニン

Ultrafast electron dynamics in phenylalanine initiated through ionization by attosecond pulses

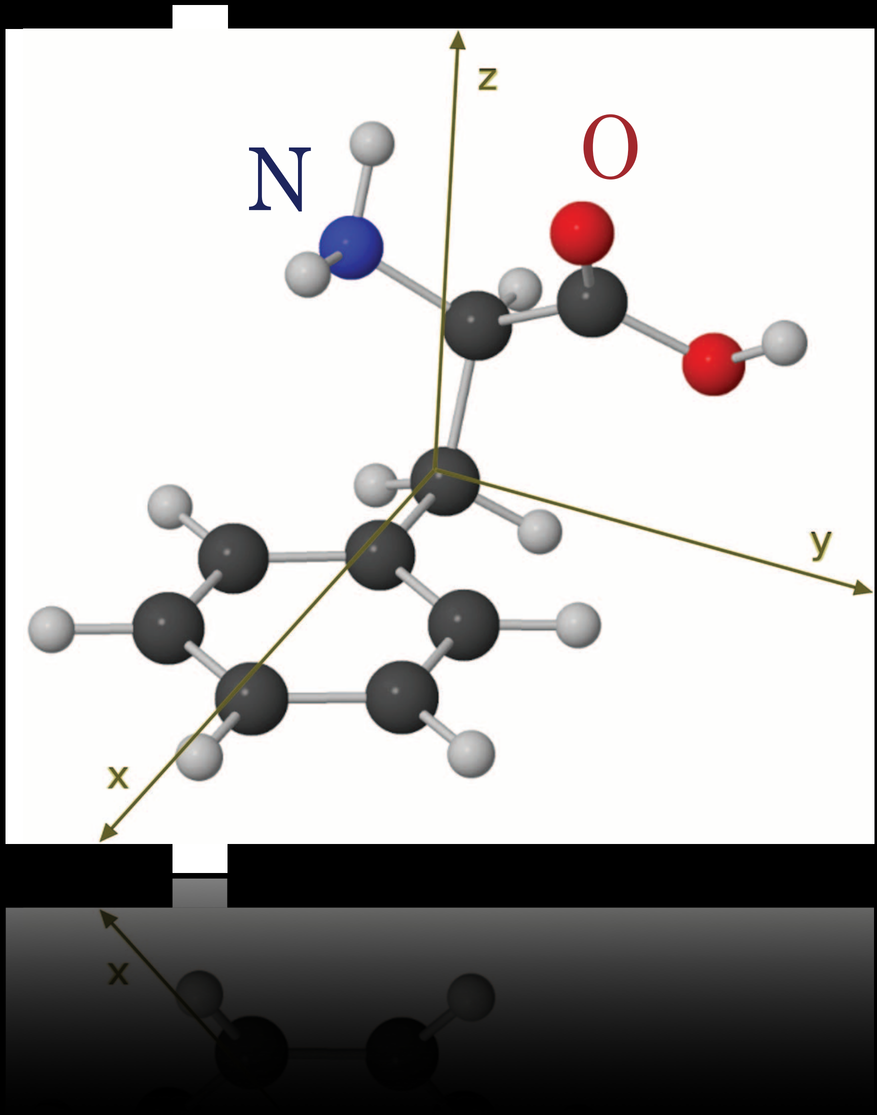
amino acid
アミノ酸

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



biological effect of ionizing radiation

放射線の生物効果



pump

sub-300 as XUV

15-35 eV

probe

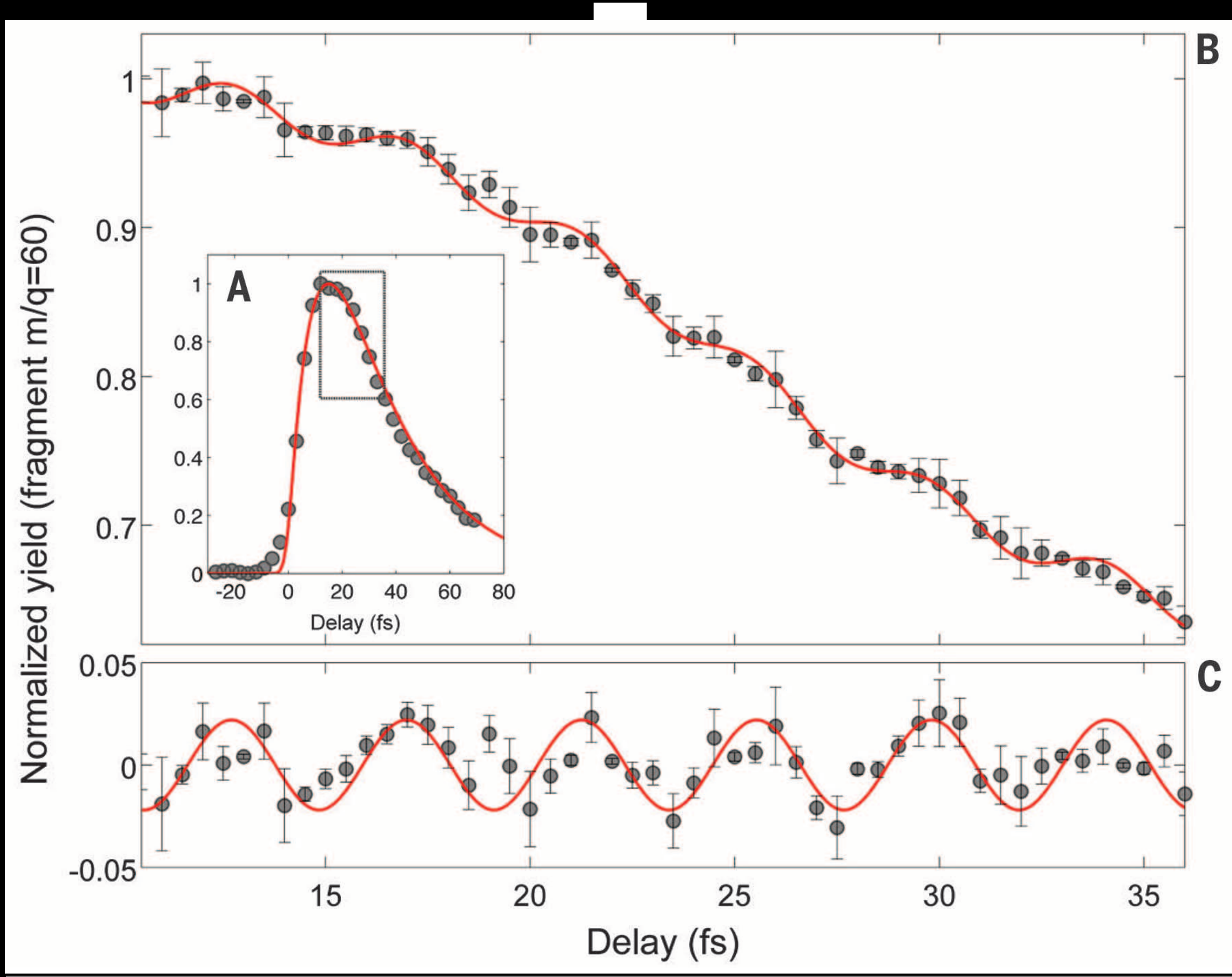
4 fs VIS/NIR

1.77 eV / 700 nm

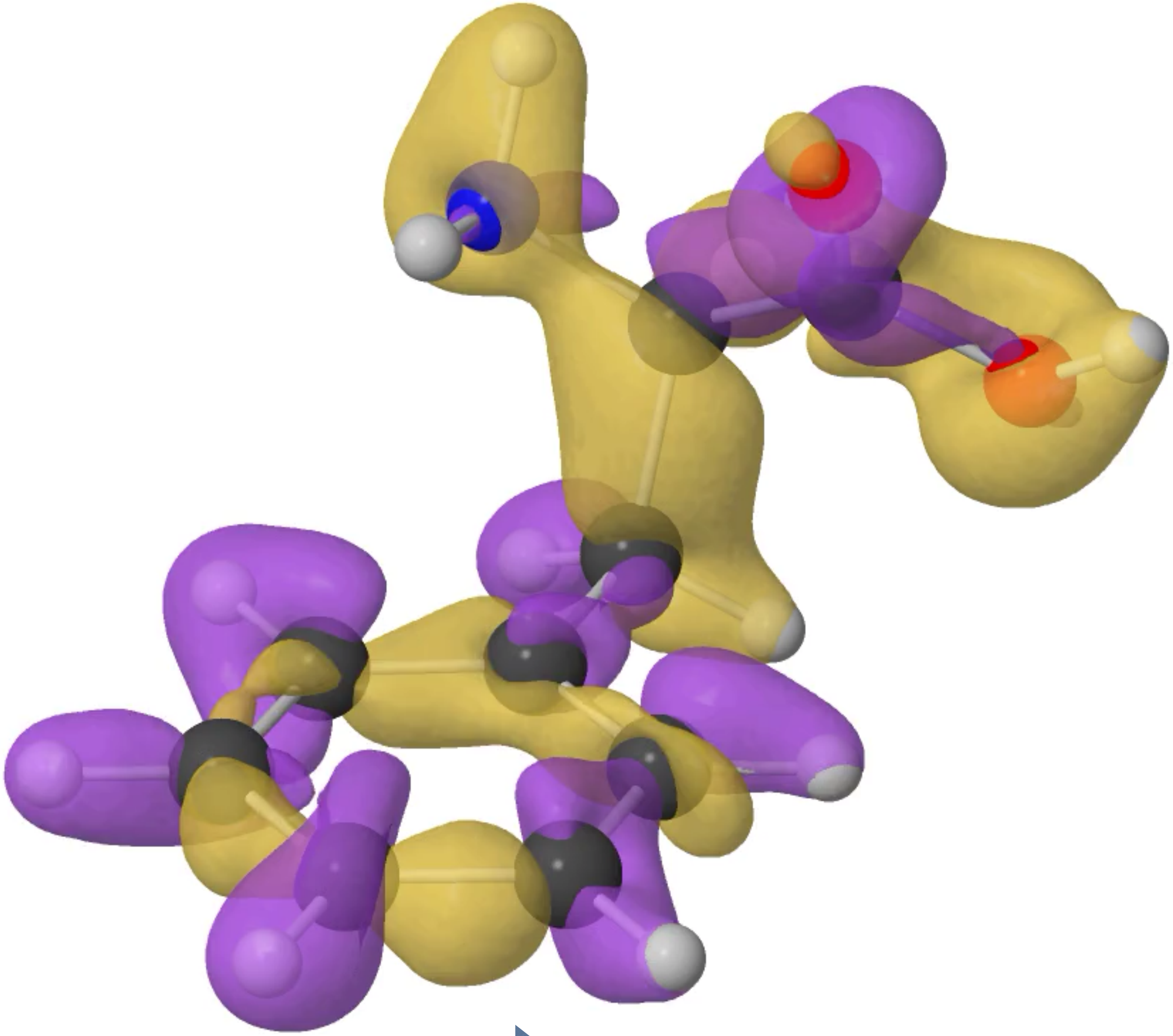
detect

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

dication yield oscillates with period ~ 4.3 fs



assigned to electron dynamics in the molecule



0.00 fs

report assignment