Advanced Radiation Application

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-order harmonic generation & attosecond science

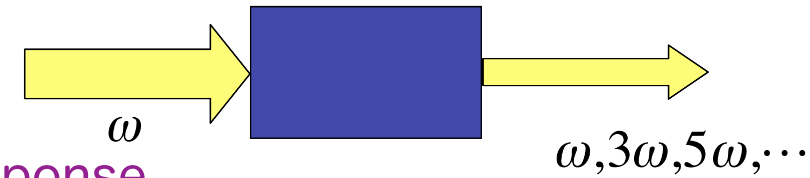
High-harmonic generation
高次高調波発生

高調波発生 (Harmonic generation)

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、 $\chi^{(2)} = 0$

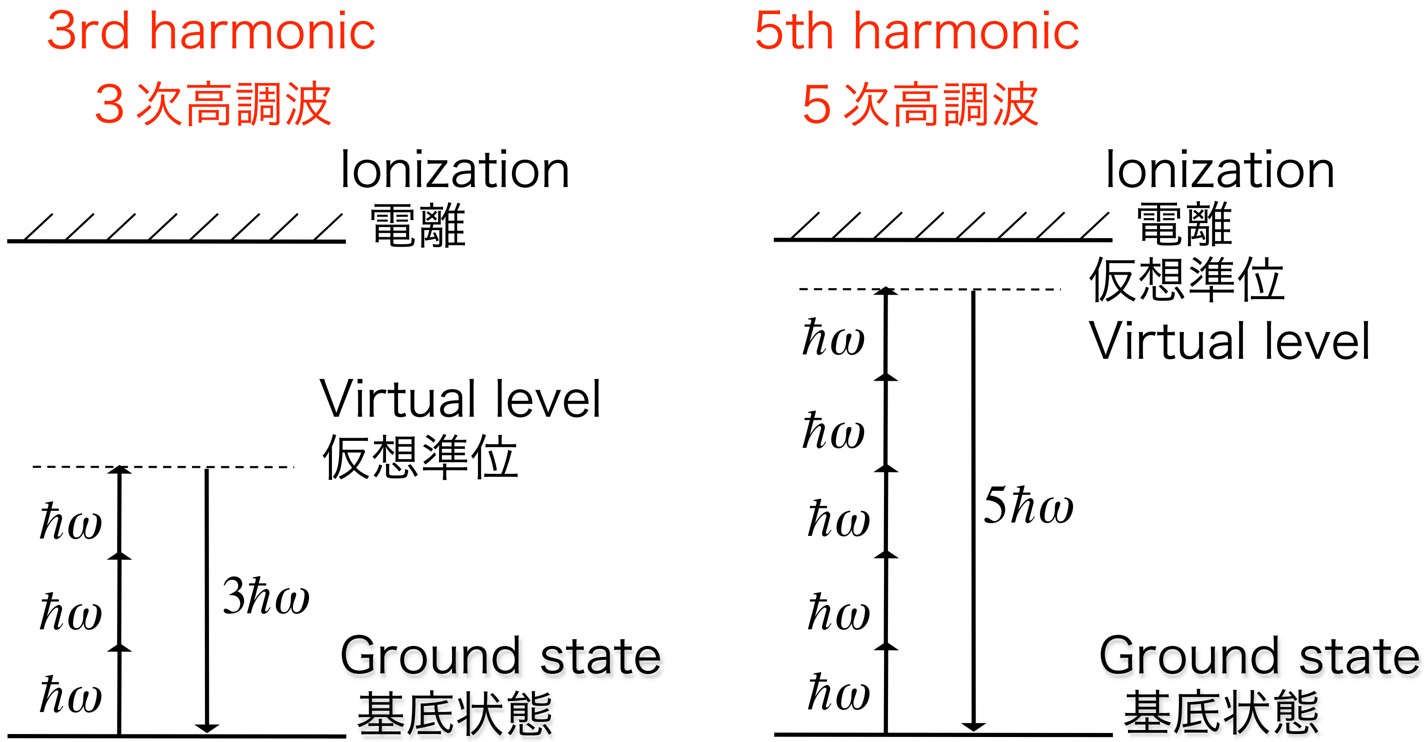
for a medium with inversion symmetry

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

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)

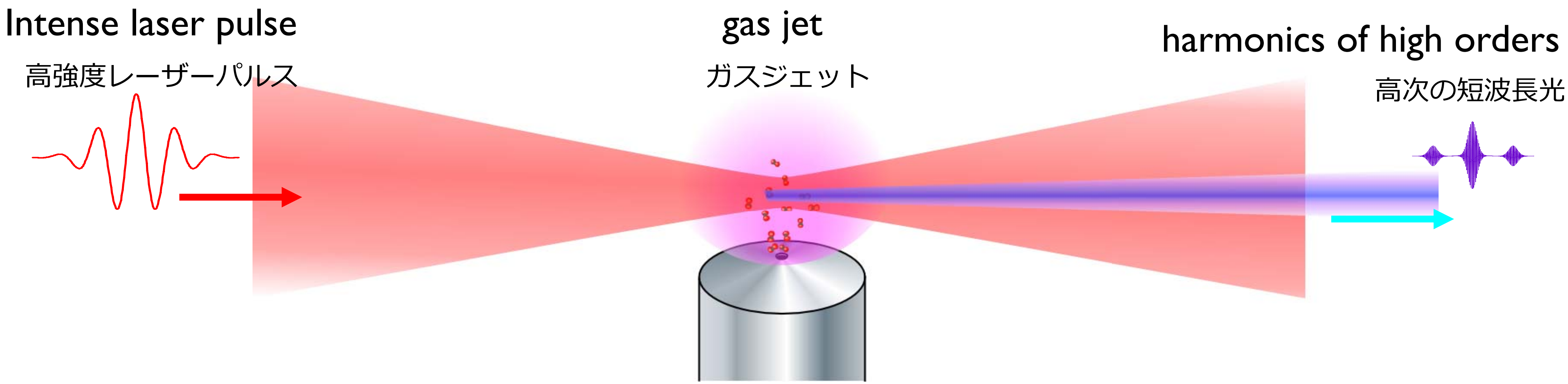


次数が高くなるほど、発生効率は減少。

Harmonic order ↑ ➡ Efficiency ↓

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

discovered in 1987



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

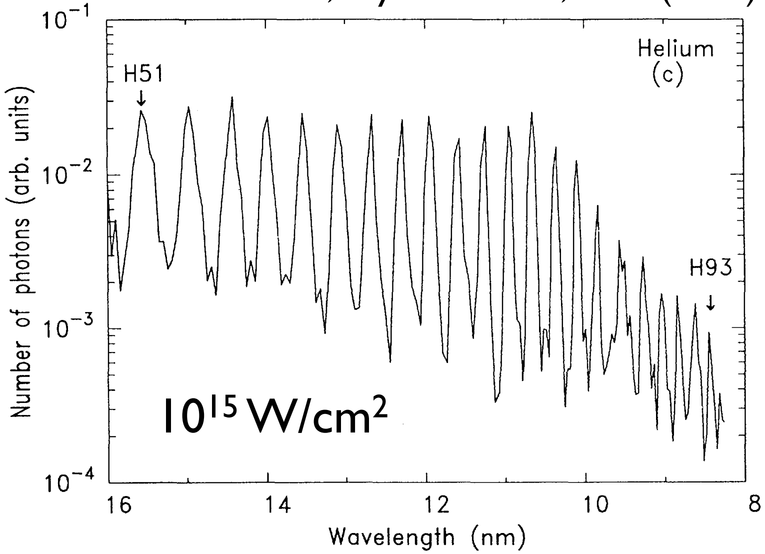
新しい極端紫外・軟エックス線光源として注目される。

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

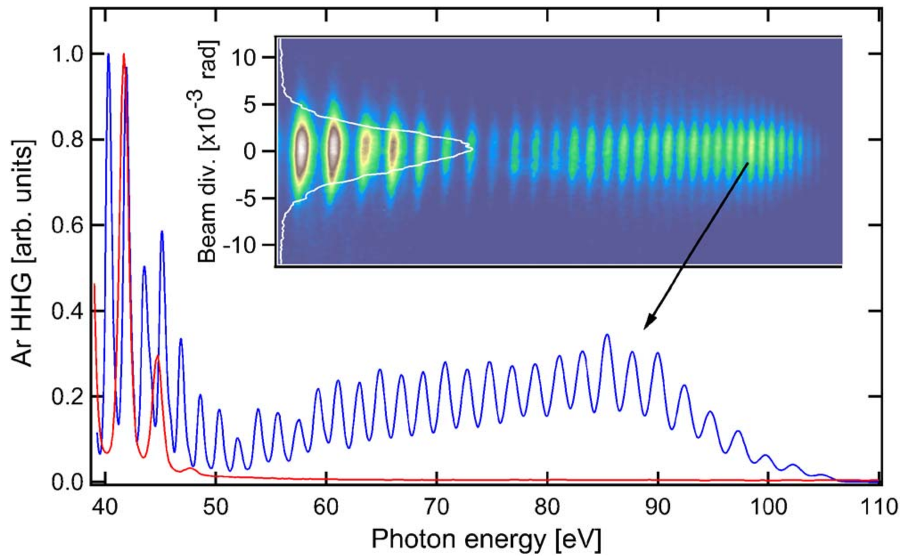
How high orders?

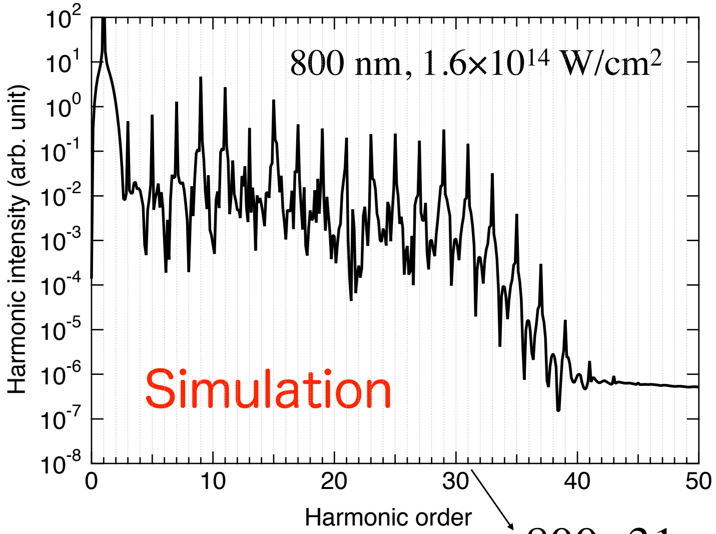
Harmonic spectrum 高調波スペクトル

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



Takahashi et al., Appl. Phys. Lett. 93, 041111 (2008)





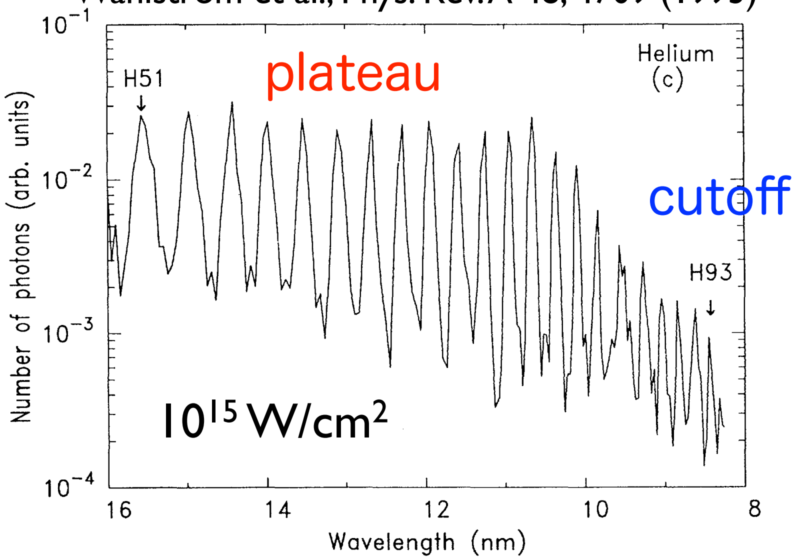
800÷31= 26 nm

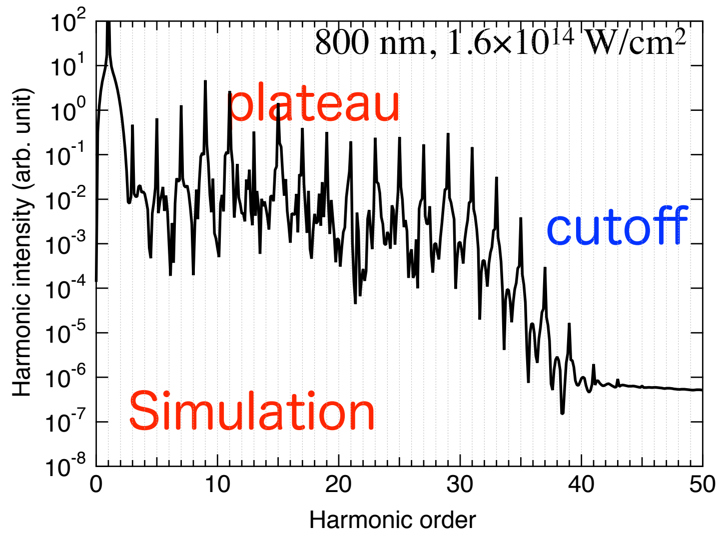
Only odd orders

奇数次のみ

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)

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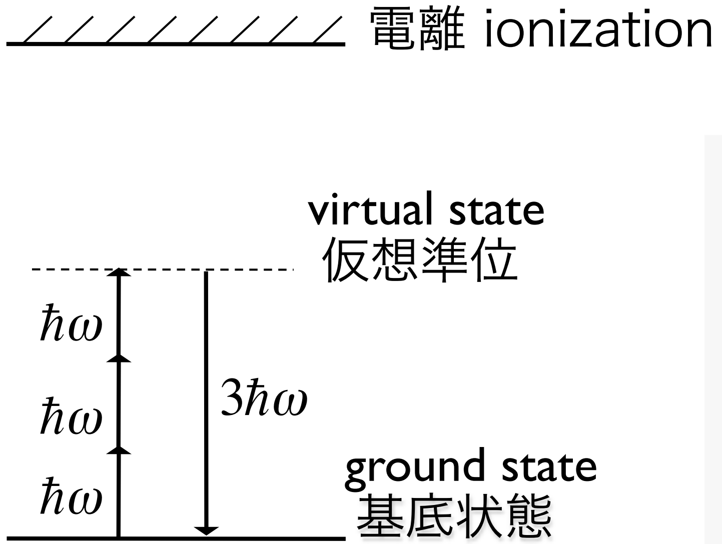
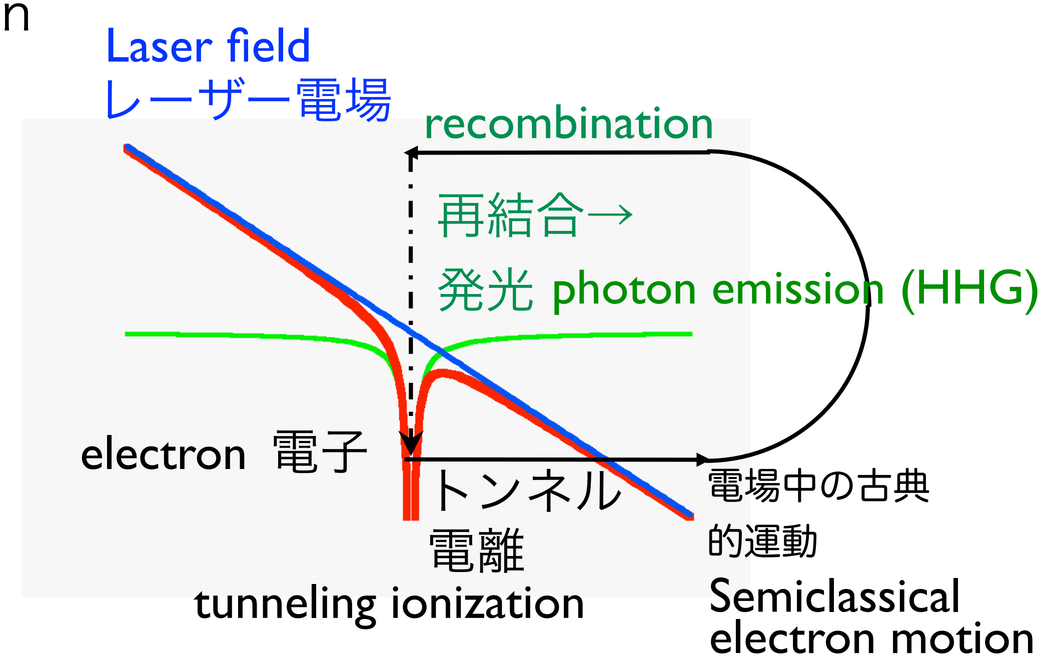
高次高調波発生のメカニズム
Mechanism of HHG

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

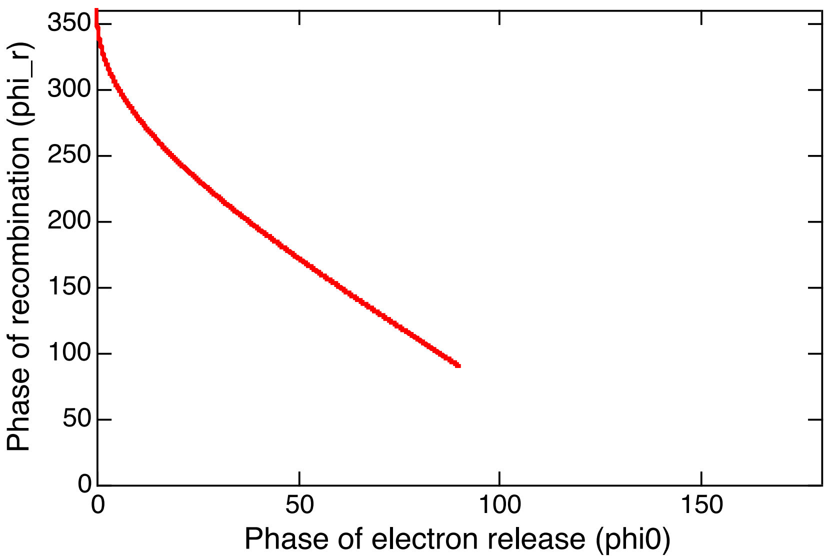
Paul B. Corkum, Phys. Rev. Lett. 71, 1994 (1993)

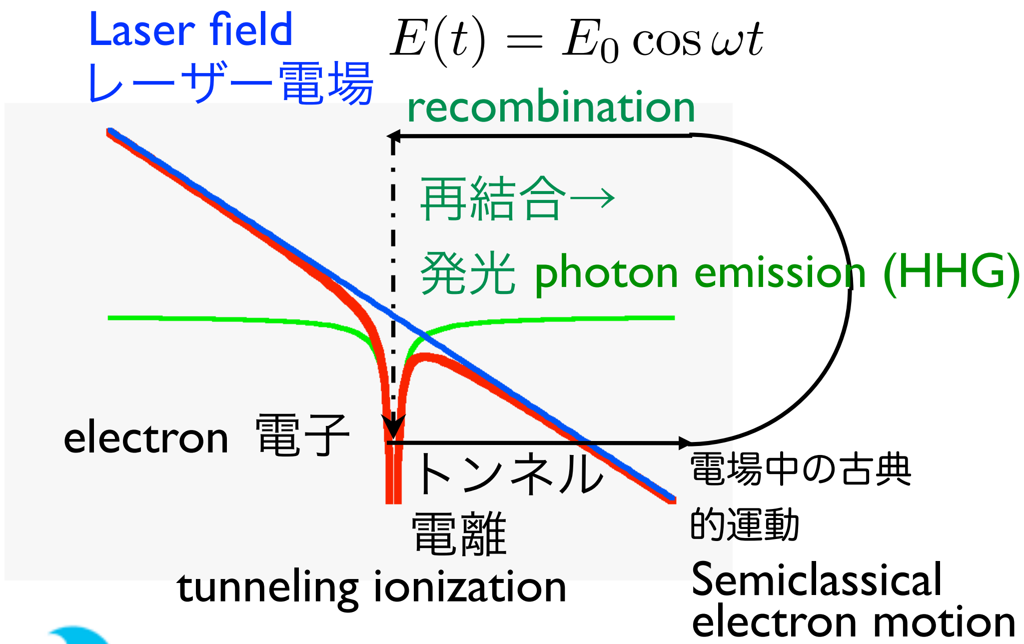
Ionization at $\omega t_0 = \phi_0$

$$z = \frac{E_0}{\omega_2}\left[(\cos\phi - \cos\phi_0) + (\phi - \phi_0)\sin\phi_0\right]$$

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

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





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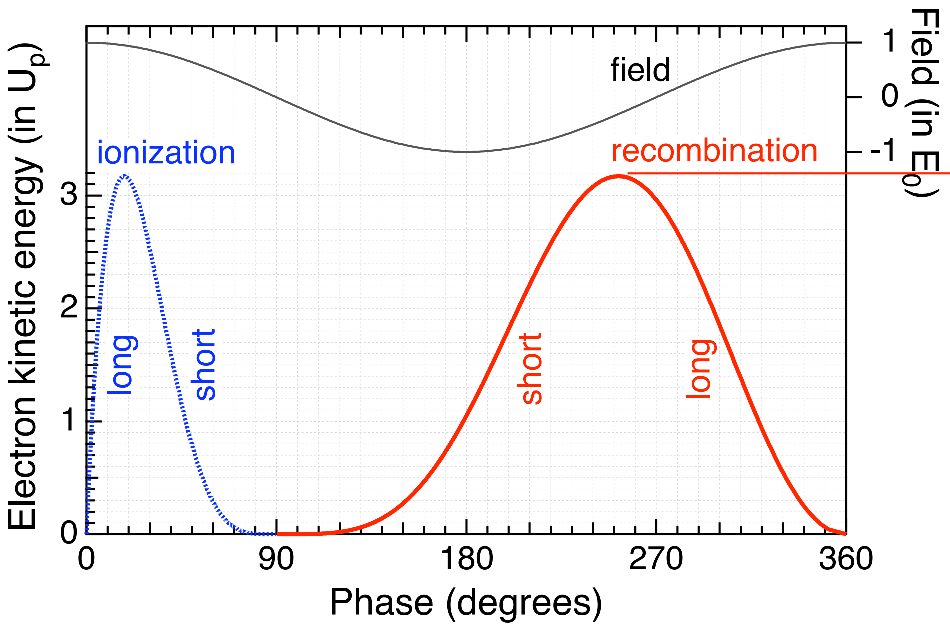
高次高調波発生の３ステップモデル
3-step model of HHG

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.

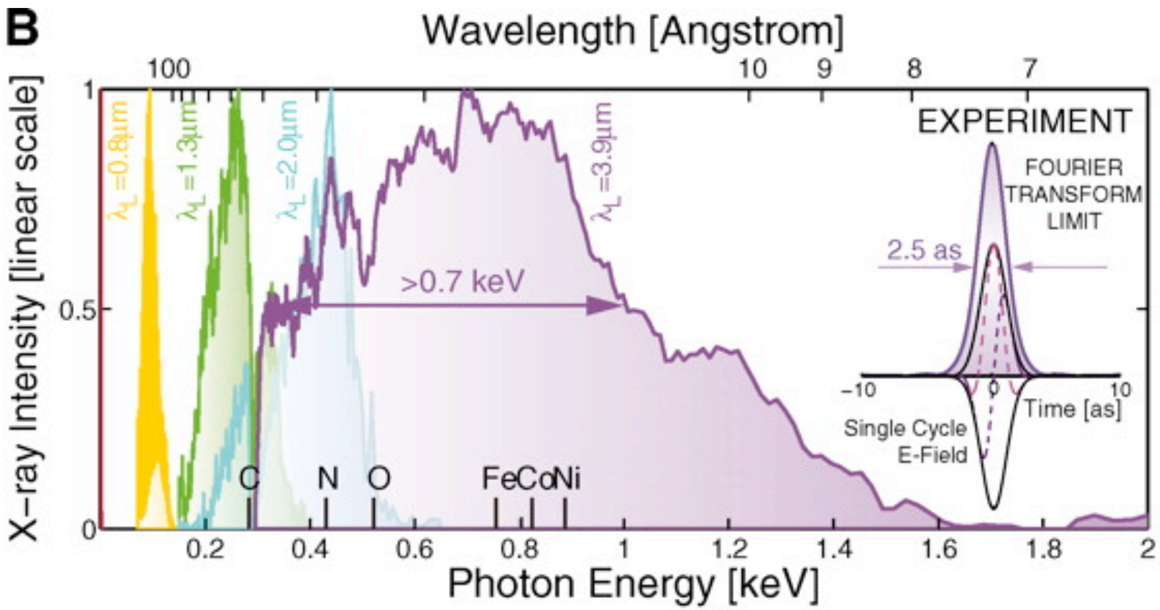
$$E_c = I_p + 3.17U_p$$

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

Short and long trajectories

Even up to 1.6 keV, > 5000 orders

almost x-ray!

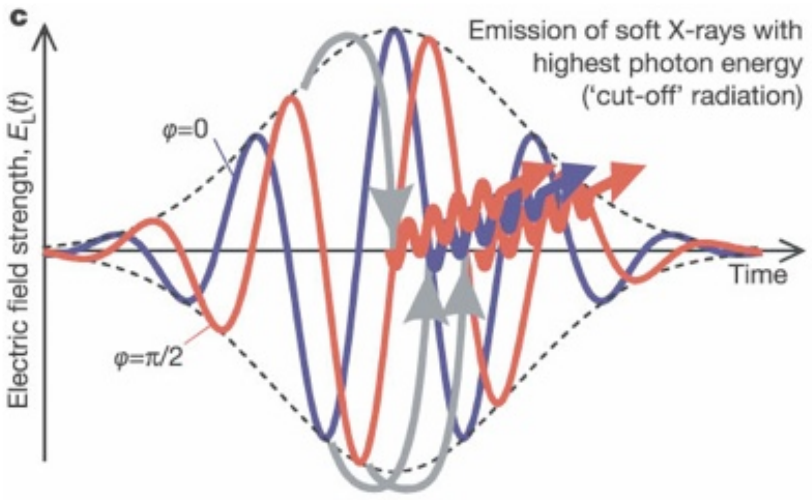


Popmintchev et al., Science 336, 1287 (2012)

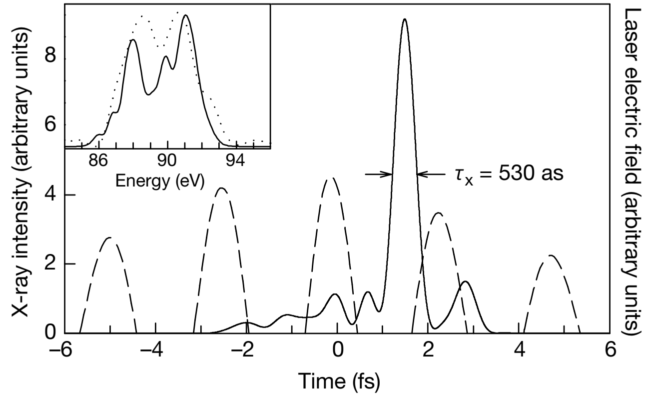
a new type of laser-based radiation source

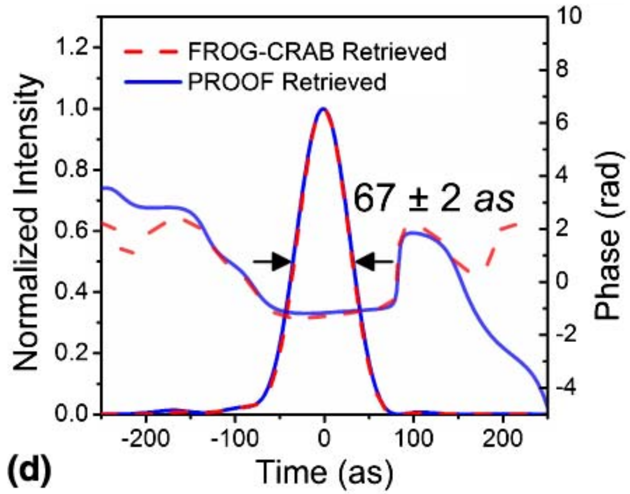
レーザーをベースにした新しいタイプの放射線源

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)

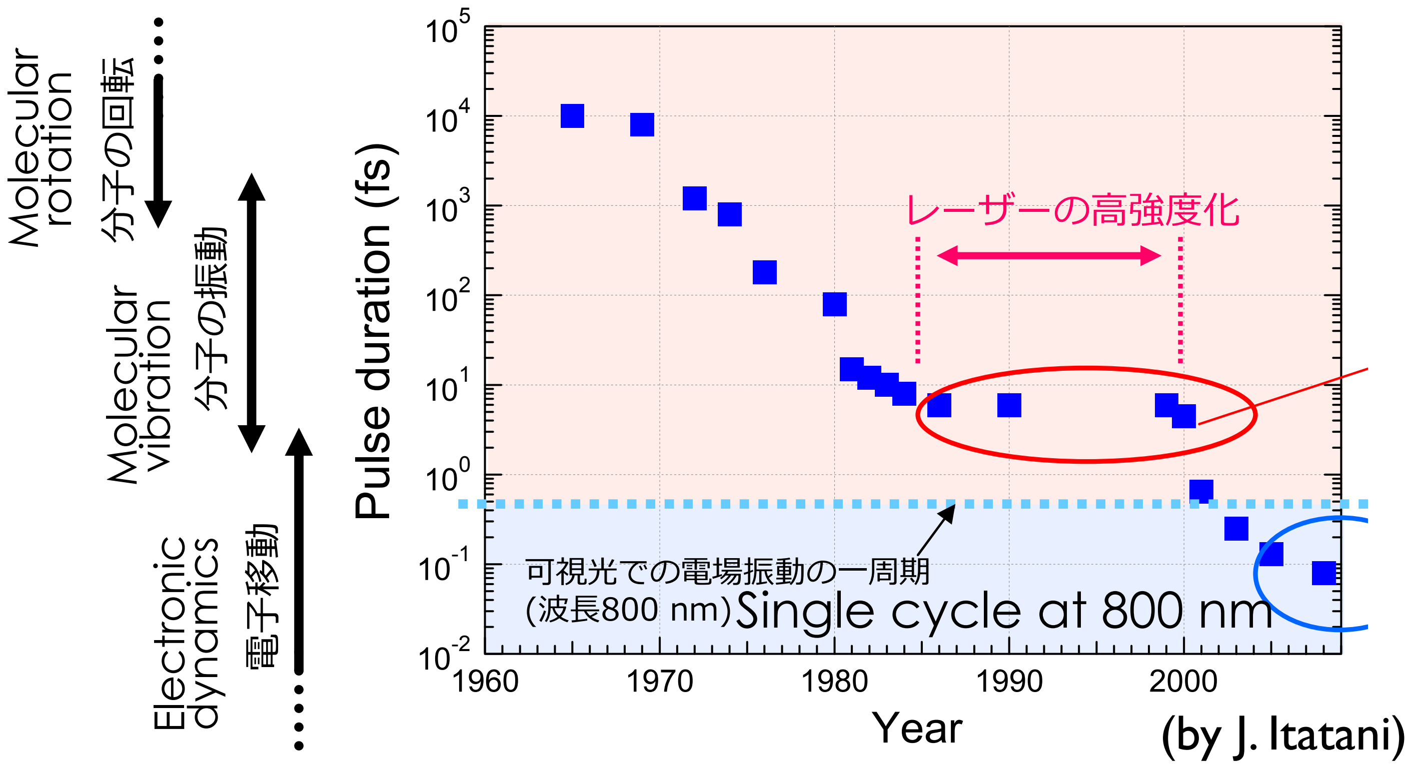
Light emission takes place only once.

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



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}

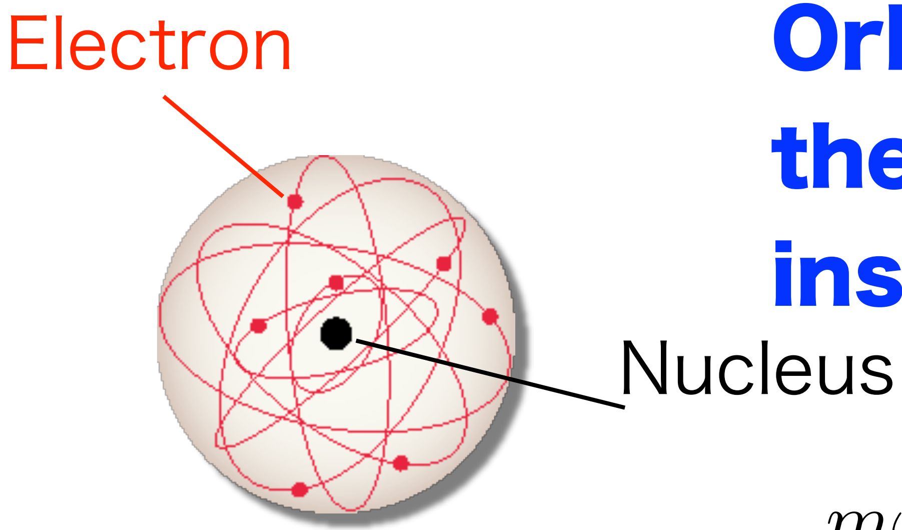
Light propagates during 30 fs ⋯

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

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.

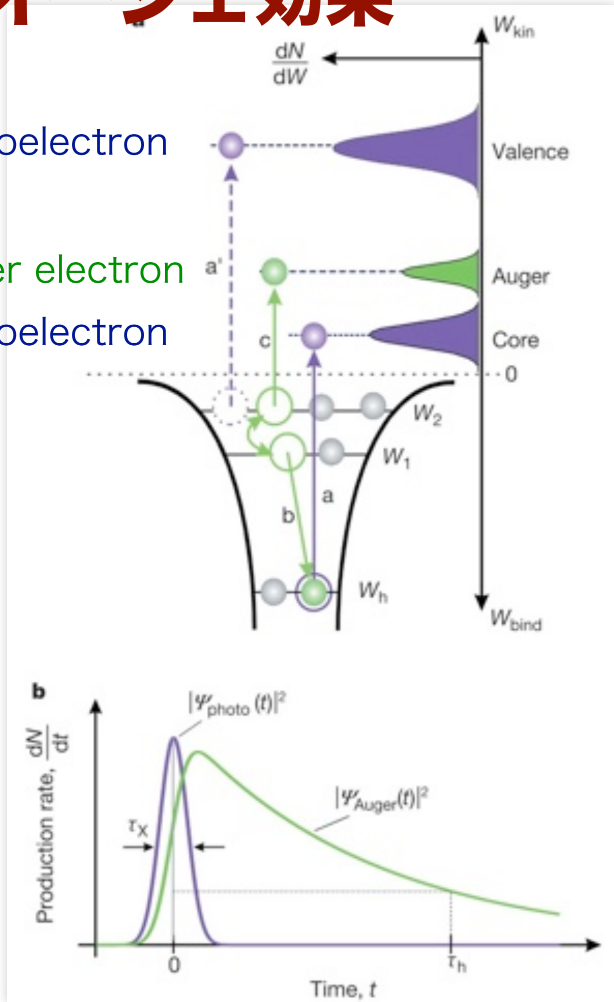
Auger effect

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

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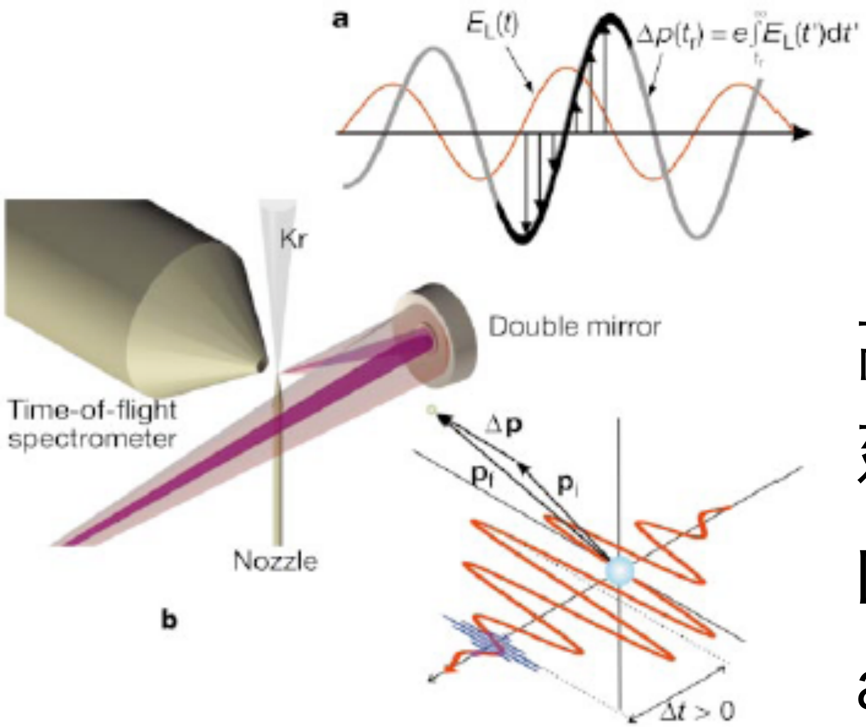
UT-PSC Photon Science Center of the University of Tokyo

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）

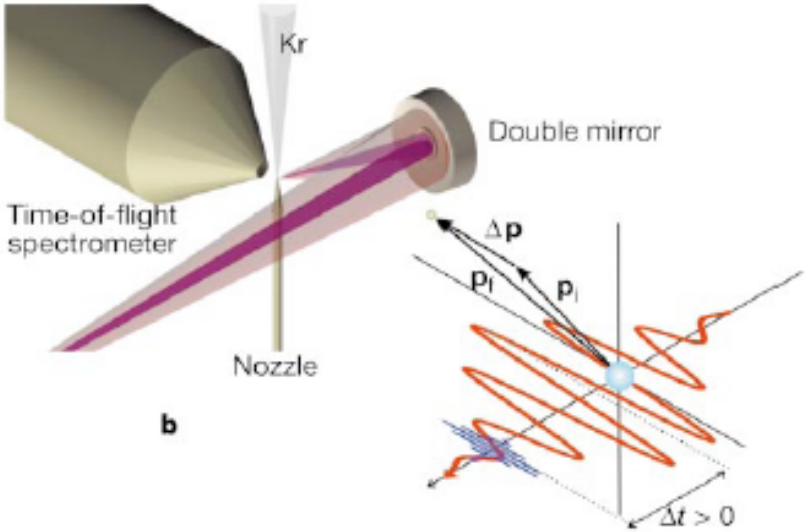
How to measure the electron ejection time?



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）

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

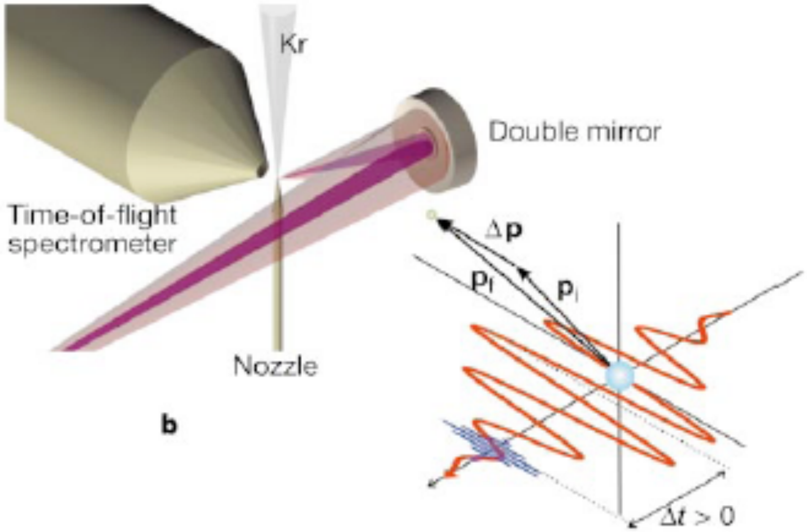
検出器での運動量 Momentum at the detector $p = p_0 + \Delta p$

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

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?



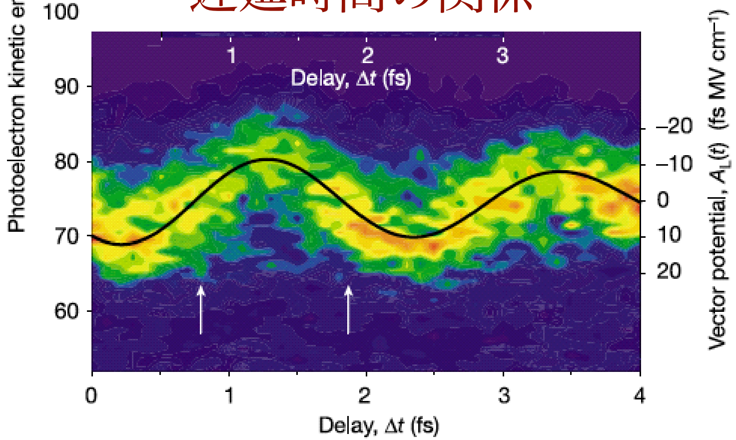
検出器での運動エネルギー

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

Electron kinetic energy

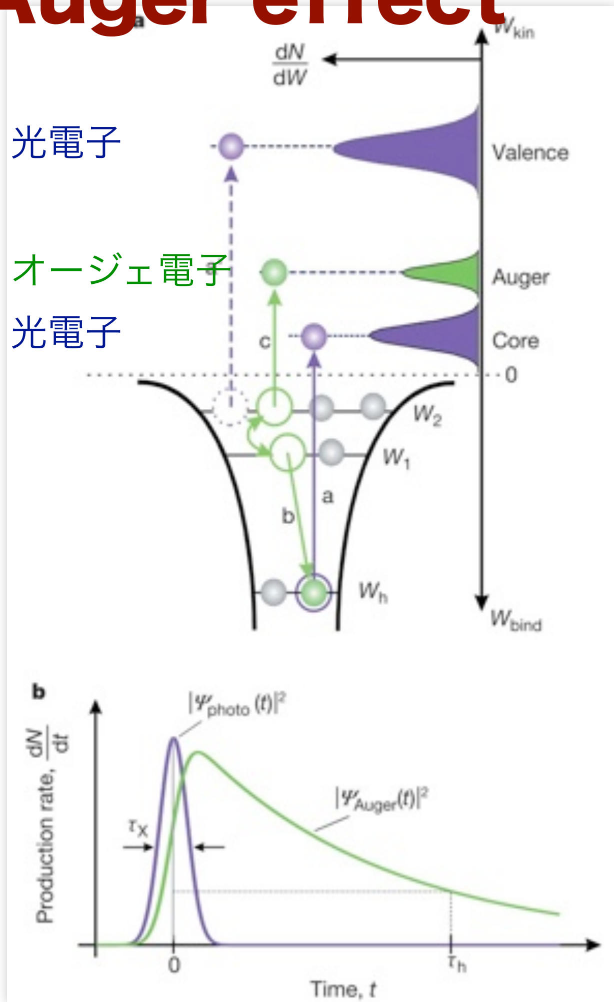
Ejection time

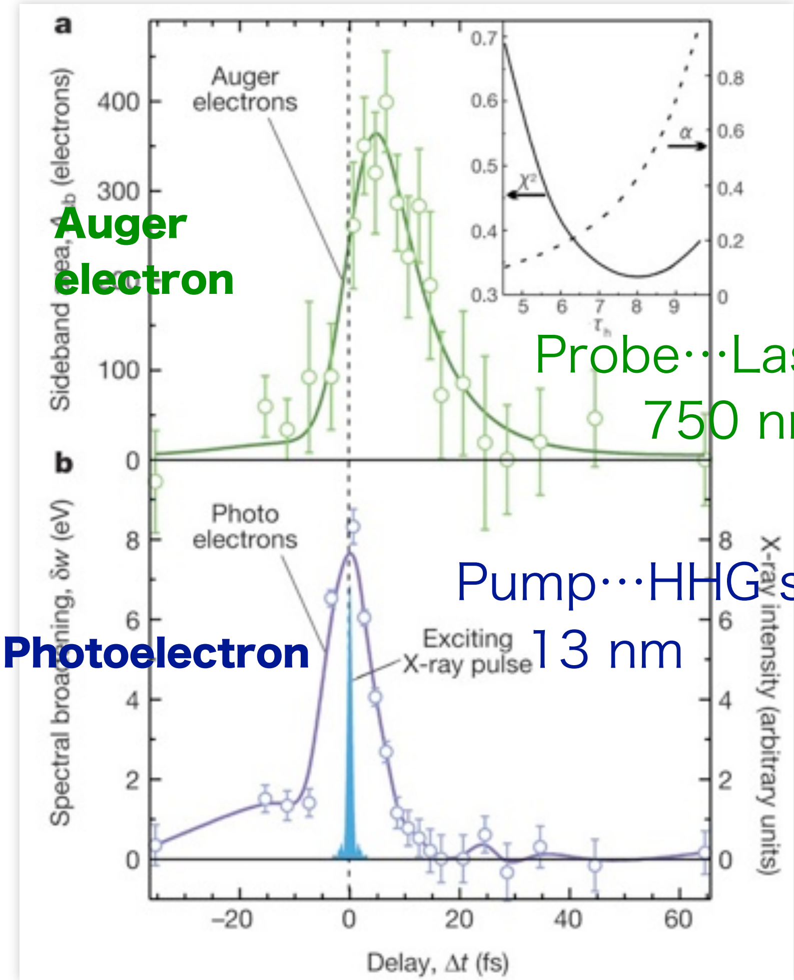
光電子のエネルギーと
遅延時間の関係



Life time of the Auger decay～8 fs

Auger effect





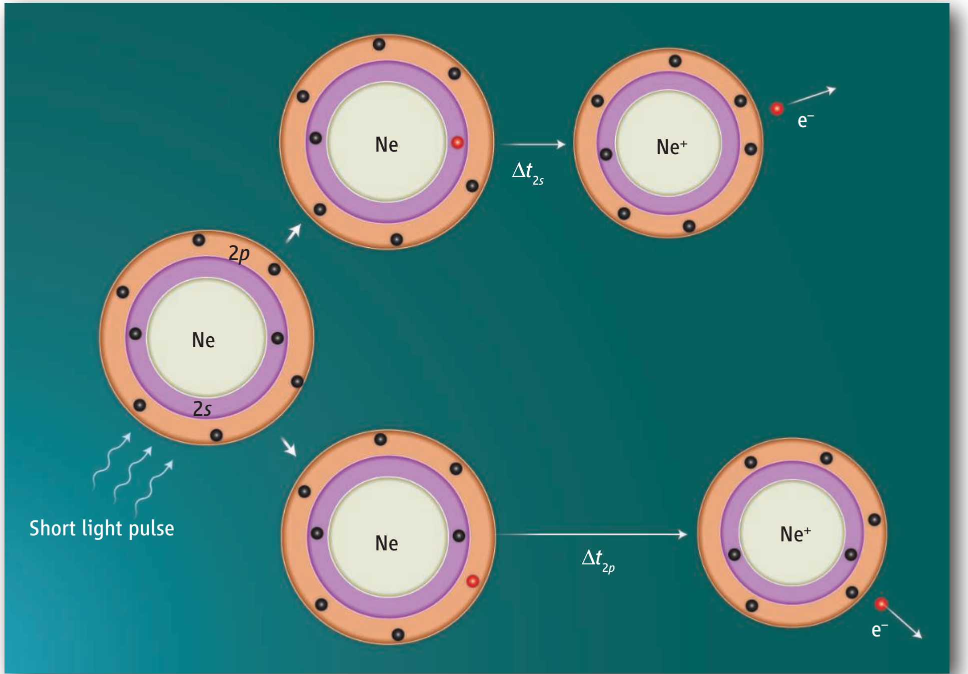
10フェムト秒程度の超高速過程が見える！

Ultrafast process ~ 10 fs

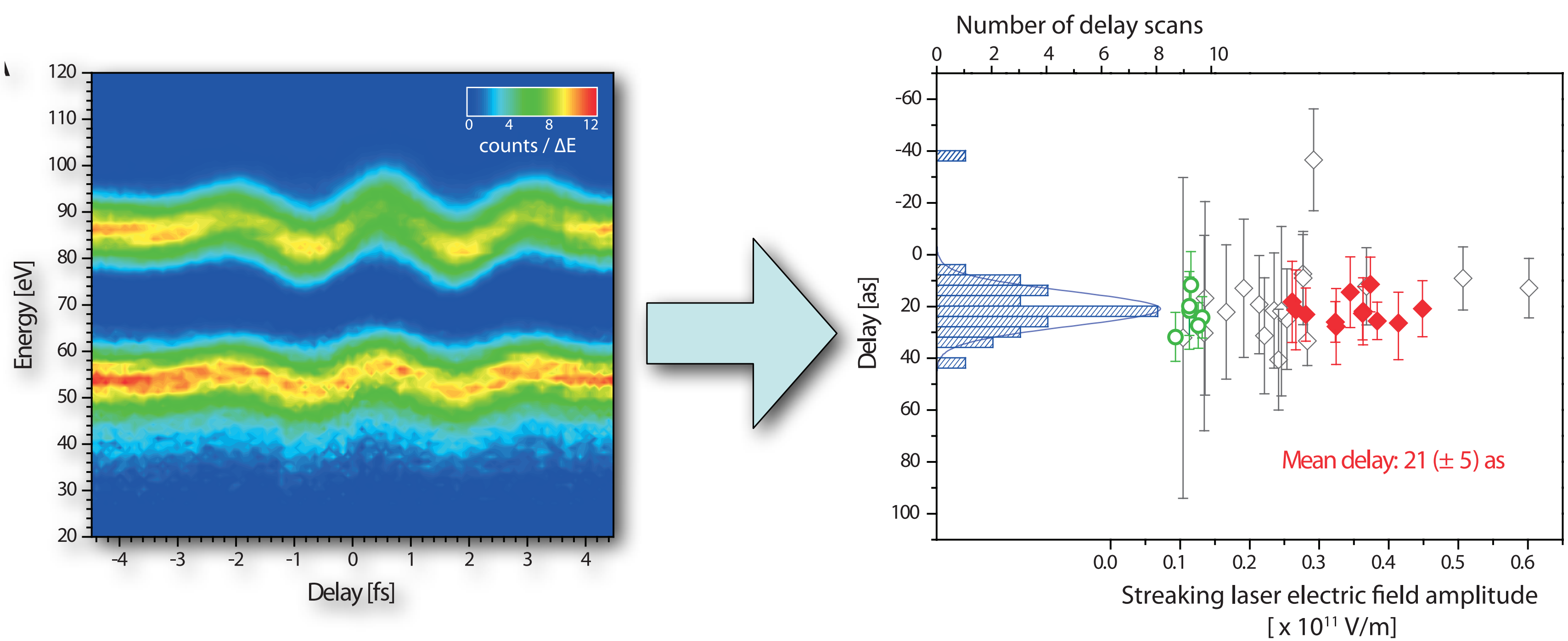
Delay in photoemission
光電効果には何アト秒かかるか？

When Does Photoemission Begin?

The photoelectric effect is usually considered instantaneous.

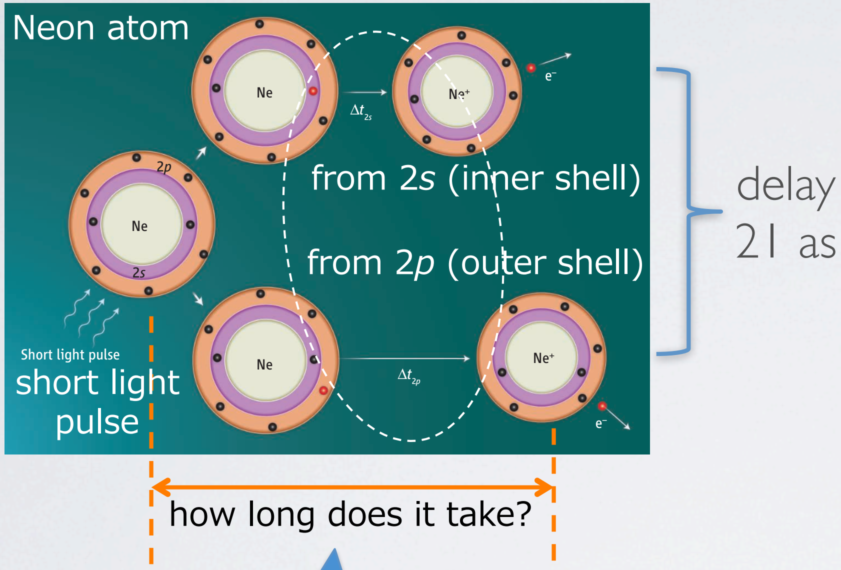


The 2s electron appears to come out 21 attoseconds earlier than the 2p electron!



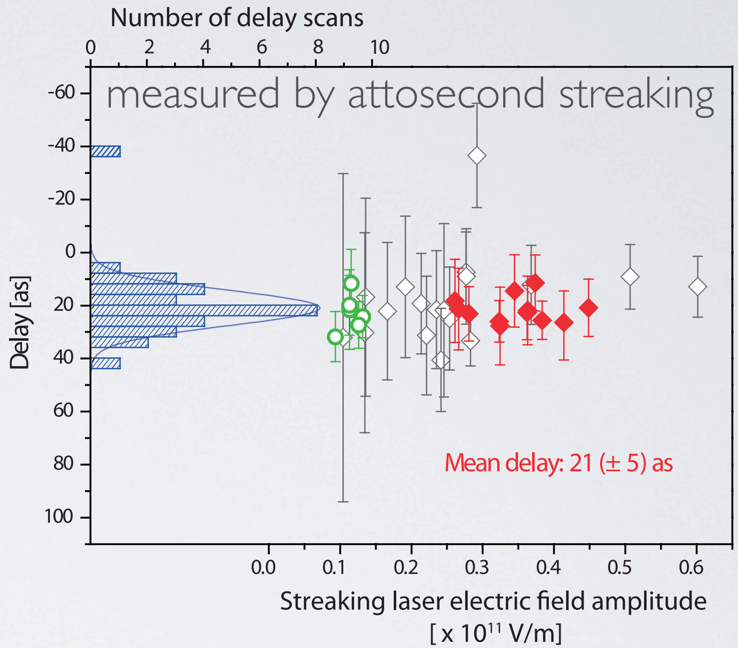
Schultze *et al.*, Science 328, 1658 (2010)

Delay in photoemission



What is happening?

Dynamic multi-electron correlation?

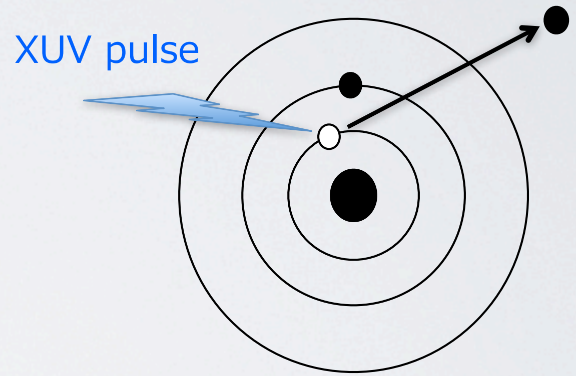


Schultze *et al.*, Science 328, 1658 (2010)

Mechanism

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

Time-dependent ab-initio simulation of inner-shell photoionization of an excited He atom (e.g., 1*s*2*p*)



Method: Time-dependent Schrödinger equation (TDSE)

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

$$H_{\text{atom}}=-\frac{1}{2}\nabla_{r_1}^2-\frac{1}{2}\nabla_{r_2}^2-\frac{2}{r_1}-\frac{2}{r_2}+\frac{1}{r_{12}}$$

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

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

$$\Lambda_{l_1l_2}^L(\hat{\mathbf{r}}_1,\hat{\mathbf{r}}_2)=\sum_m\langle l_1ml_2-m|L0\rangle Y_{l_1m}(\hat{\mathbf{r}}_1)Y_{l_2,-m}(\hat{\mathbf{r}}_2)$$

Coupled spherical harmonics

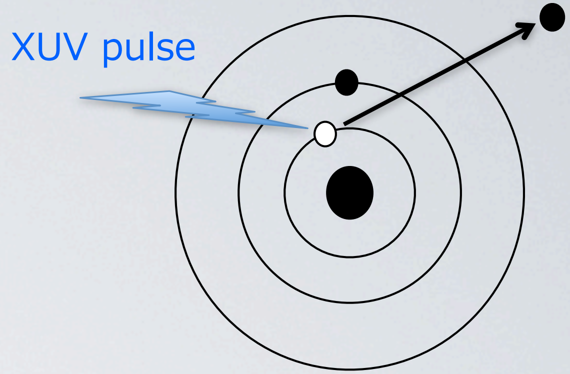
Discretization of $P_{l_1l_2}^L(r_1,r_2,t)$ on (r_1,r_2) grid

$$r_1\rightarrow\left(j_1-\frac{1}{2}\right)\Delta r \qquad r_2\rightarrow\left(j_2-\frac{1}{2}\right)\Delta r$$

$$\Rightarrow \quad P_{l_1l_2}^L(r_1,r_2,t)\Rightarrow P_{l_1l_2j_1j_2}^L(t)$$

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

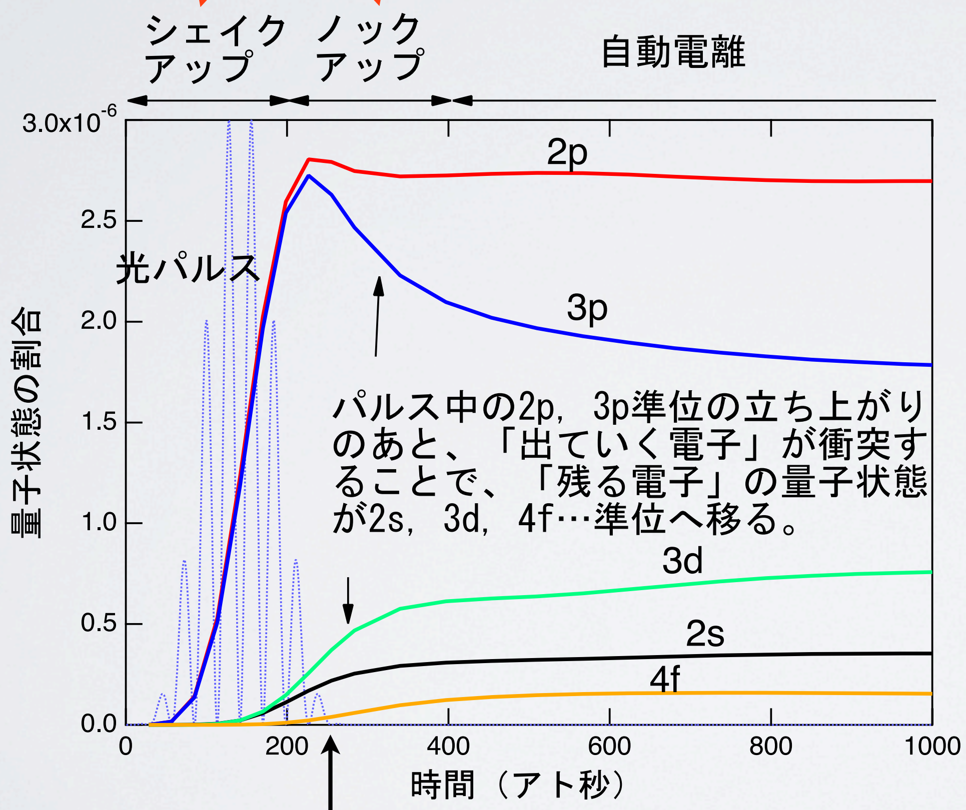
3D simulation

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

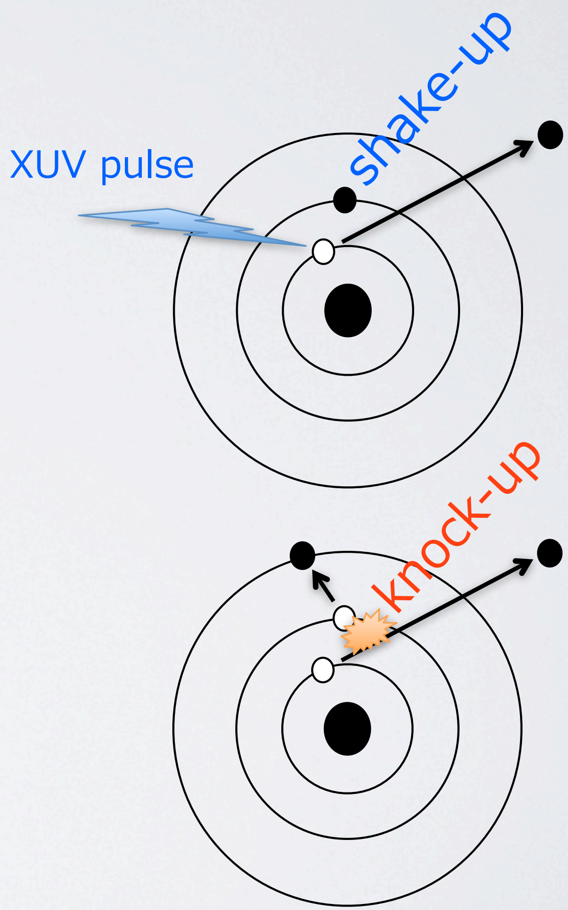
temporal evolution of the ionic state

two distinct time scales

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



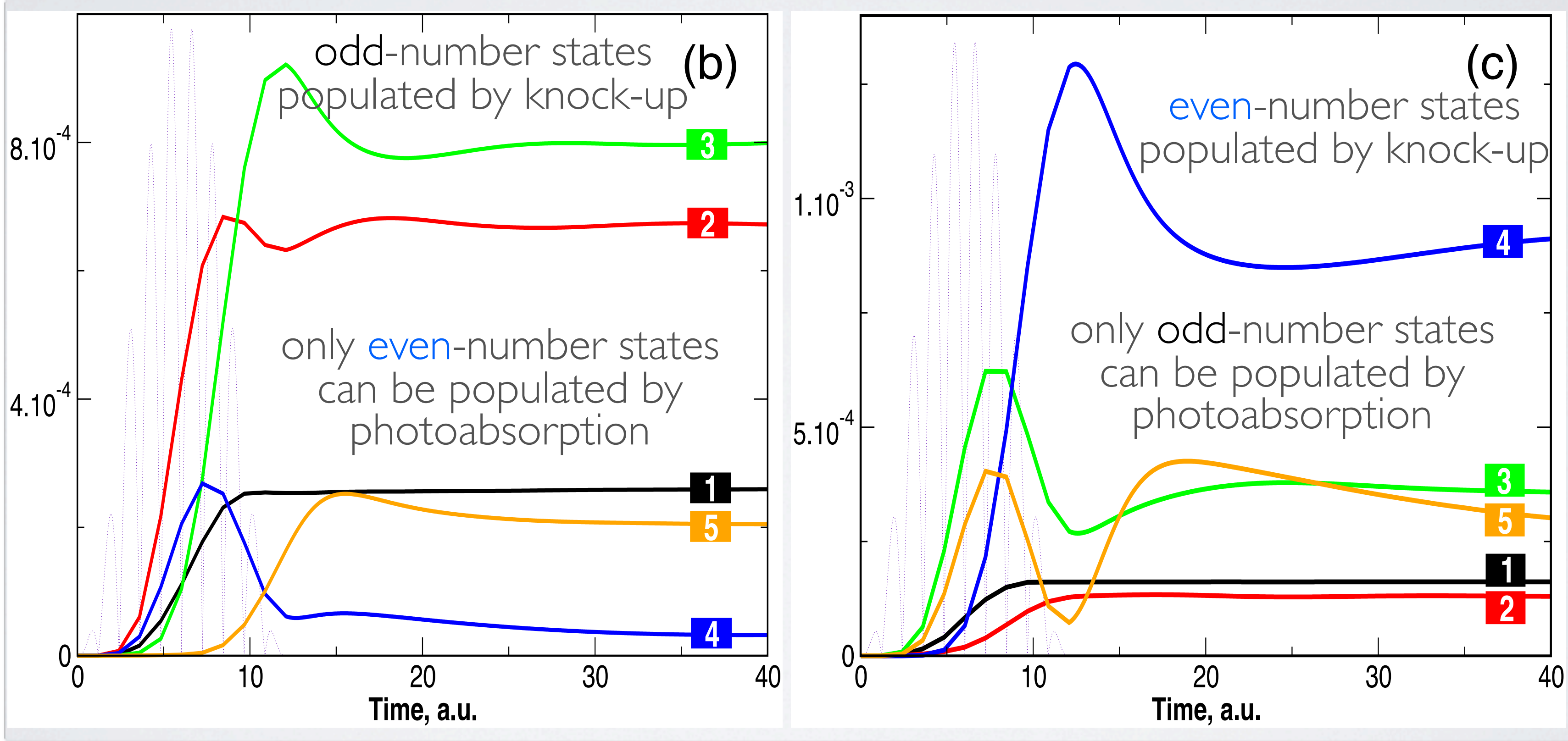
the pulse ends



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

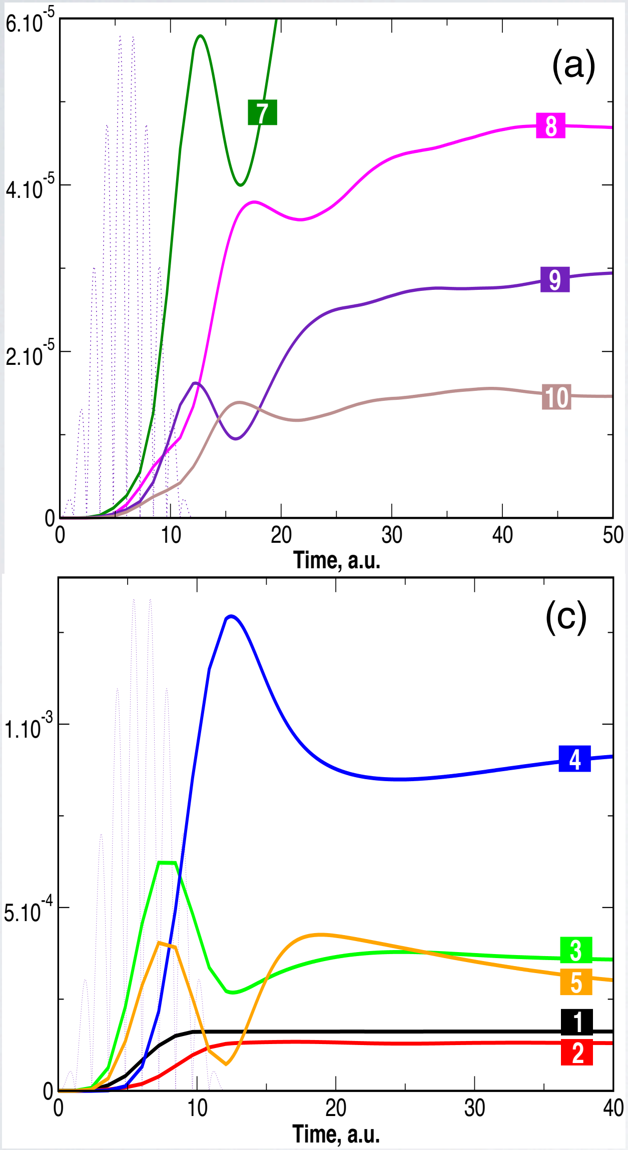


Sukiasyan, Ishikawa, Ivanov, Phys. Rev. A 86, 033423 (2012)

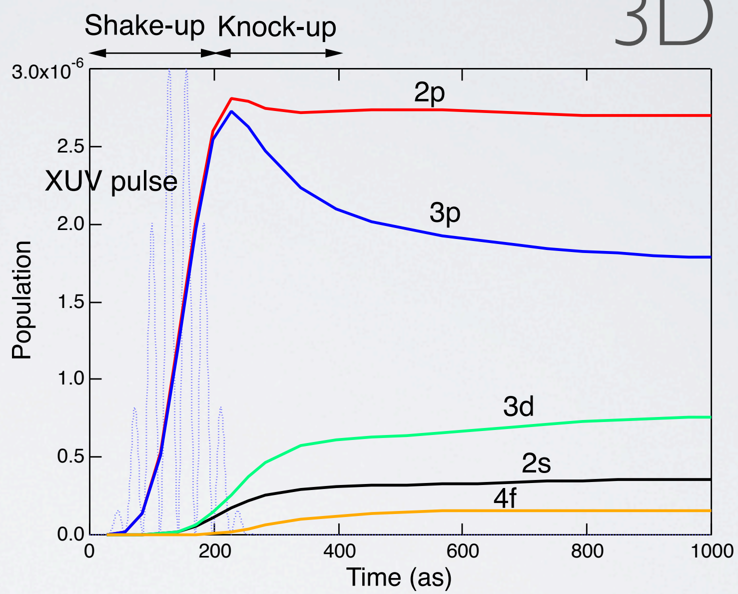
knock-up lasts longer for higher ionic channels.

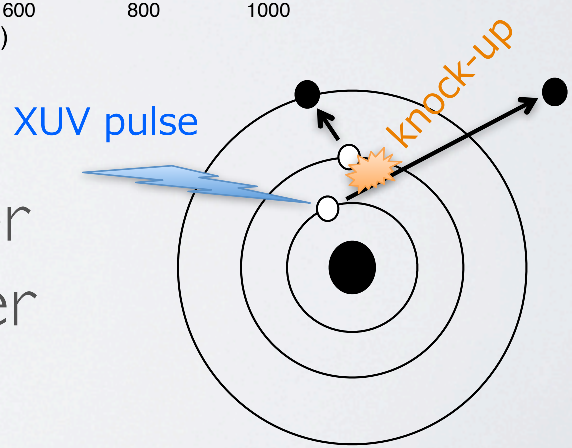
1D

from the 2nd excited atom



3D



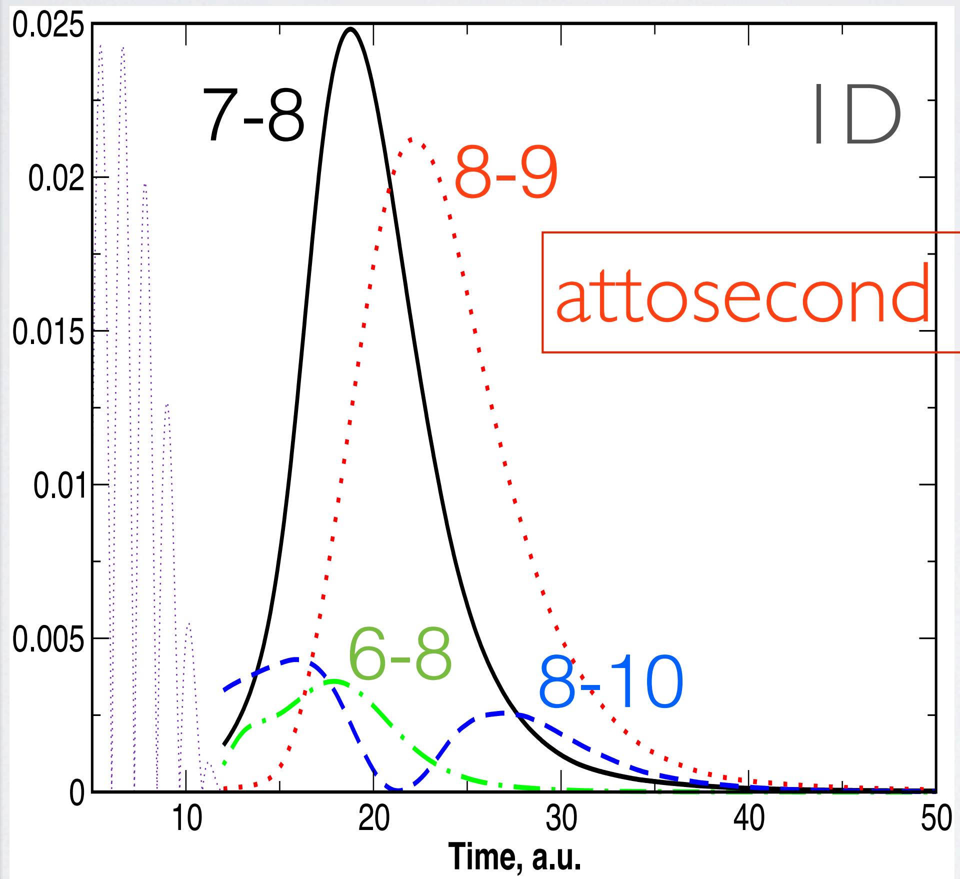


reflects the larger radii of the higher excited states

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

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

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



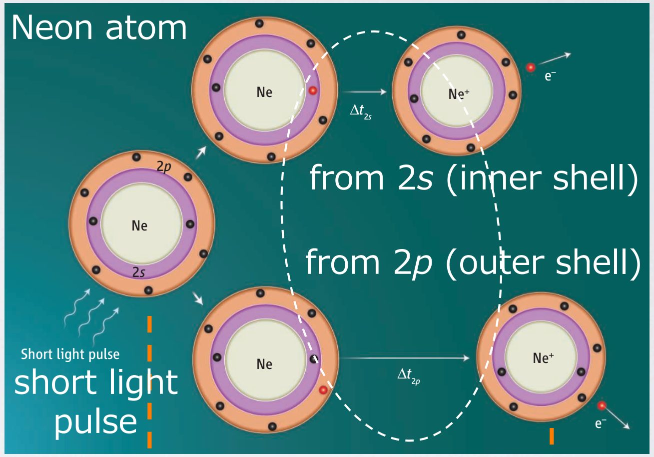
attosecond cascades

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



how long does it take?

photon absorption

electron emission

What is happening?

Dynamic multi-electron correlation

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