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Advanced Laser and Photon Science
レーザー・光量子科学特論E

4. High-harmonic generation
高次高調波発生

HARMONIC GENERATION
高調波発生

Process whereby integer multiples of the original light's frequency (fundamental frequency) are generated.

crystal, gas

Linear optical effect
線形光学効果

ω → 結晶等 → ω

Material response is linear in light intensity

Nonlinear optical effect
非線形光学効果

ω → 結晶等 → $\omega, 2\omega, 3\omega, 4\omega, 5\omega, \cdots$

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

frequency conversion
波長変換

$$D = \varepsilon_0 E + P$$

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

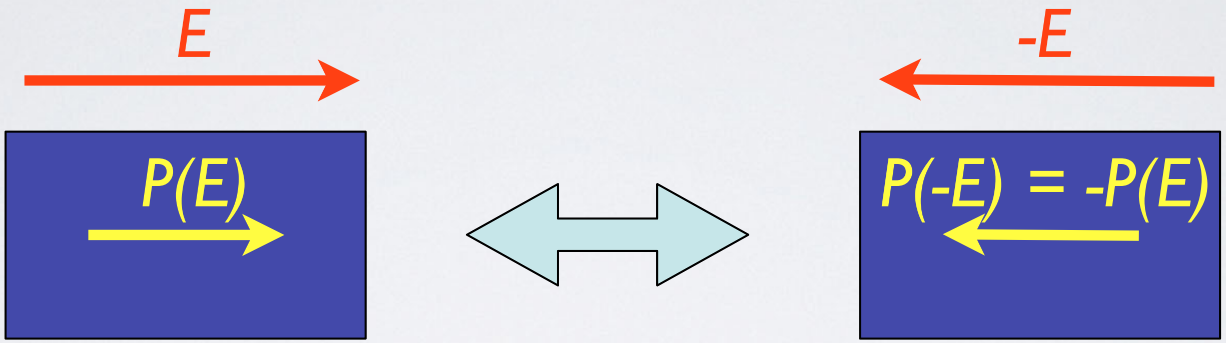
非線形分極　nonlinear

線形分極　linear polarization

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

2ω：2次高調波(2nd harmonic)
3ω：3次高調波(3rd harmonic)
.....

反転対称な物質では、偶数次はゼロ
Even-order components vanish for a medium with inversion symmetry



$$P(E) = \epsilon_0 \left[\chi^{(1)} E + \chi^{(2)} E^2 + \chi^{(3)} E^3 + \chi^{(4)} E^4 + \cdots\right]$$

$$P(-E) = \epsilon_0 \left[-\chi^{(1)} E + \chi^{(2)} E^2 - \chi^{(3)} E^3 + \chi^{(4)} E^4 - \cdots\right]$$

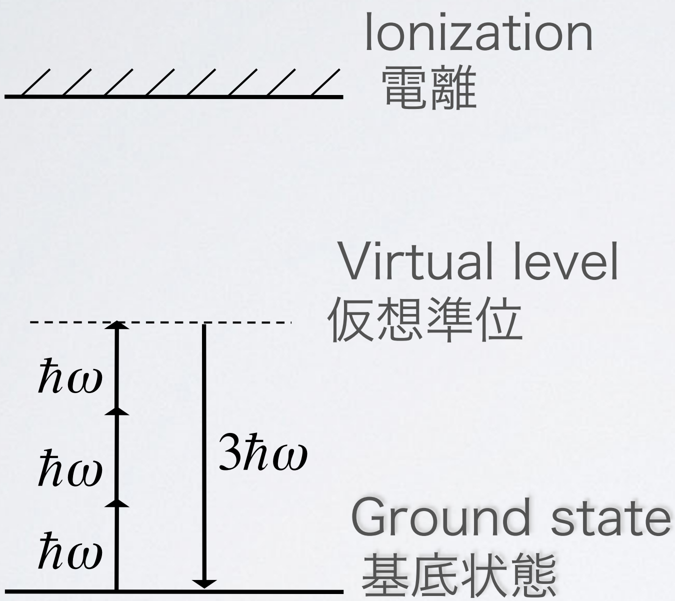
$$-P(E) = \epsilon_0 \left[-\chi^{(1)} E - \chi^{(2)} E^2 - \chi^{(3)} E^3 - \chi^{(4)} E^4 - \cdots\right]$$

$$\Rightarrow \quad \chi^{(2)} = 0, \chi^{(4)} = 0, \cdots$$

摂動論的高調波発生

PERTURBATIVE HARMONIC GENERATION

3rd harmonic
３次高調波発生の概念図



Transition matrix element　遷移行列要素

$$
\begin{aligned}
M_{\mathrm{THG}} = \sum_{h,i,f} \Bigg\{ & \frac{\boldsymbol{\epsilon}_3 \cdot \mathbf{D}_{1h}\boldsymbol{\epsilon}_1 \cdot \mathbf{D}_{hi}\boldsymbol{\epsilon}_1 \cdot \mathbf{D}_{ij}\boldsymbol{\epsilon}_1 \cdot \mathbf{D}_{j1}}{(3\omega_1 - \omega_h)(2\omega_1 - \omega_i)(\omega_1 - \omega_j)} \\
& + \frac{\boldsymbol{\epsilon}_1 \cdot \mathbf{D}_{1h}\boldsymbol{\epsilon}_3 \cdot \mathbf{D}_{hi}\boldsymbol{\epsilon}_1 \cdot \mathbf{D}_{ij}\boldsymbol{\epsilon}_1 \cdot \mathbf{D}_{j1}}{(-\omega_1 - \omega_h)(2\omega_1 - \omega_i)(\omega_1 - \omega_j)} \\
& + \frac{\boldsymbol{\epsilon}_1 \cdot \mathbf{D}_{1h}\boldsymbol{\epsilon}_1 \cdot \mathbf{D}_{hi}\boldsymbol{\epsilon}_3 \cdot \mathbf{D}_{ij}\boldsymbol{\epsilon}_1 \cdot \mathbf{D}_{j1}}{(-\omega_1 - \omega_h)(-2\omega_1 - \omega_i)(-\omega_1 - \omega_j)} \\
& + \frac{\boldsymbol{\epsilon}_1 \cdot \mathbf{D}_{1h}\boldsymbol{\epsilon}_1 \cdot \mathbf{D}_{hi}\boldsymbol{\epsilon}_1 \cdot \mathbf{D}_{ij}\boldsymbol{\epsilon}_3 \cdot \mathbf{D}_{j1}}{(-\omega_1 - \omega_h)(-2\omega_1 - \omega_i)(-3\omega_1 - \omega_j)}
\end{aligned}
$$

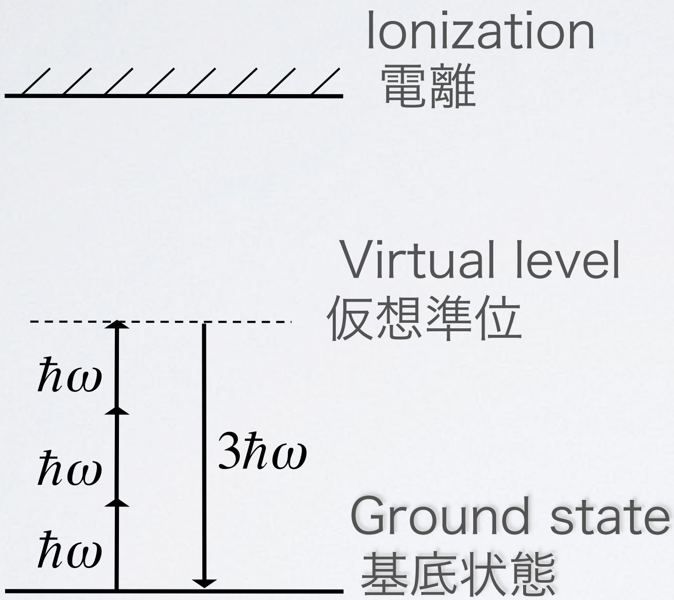
次数が高くなるほど、発生効率は減少。

order ↑ ➡ efficiency ↓

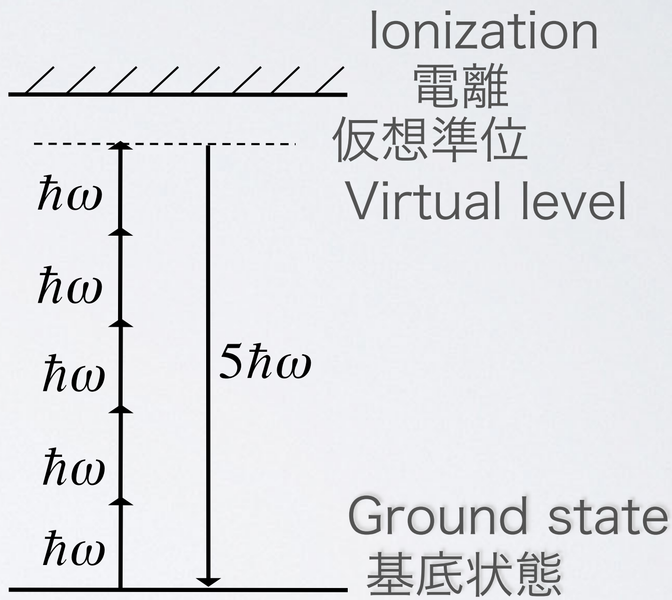
摂動論的高調波発生

(PERTURBATIVE HARMONIC GENERATION)

3rd harmonic
３次高調波



5th harmonic
５次高調波



次数が高くなるほど、発生効率は減少。

order ↑ ➡ efficiency ↓

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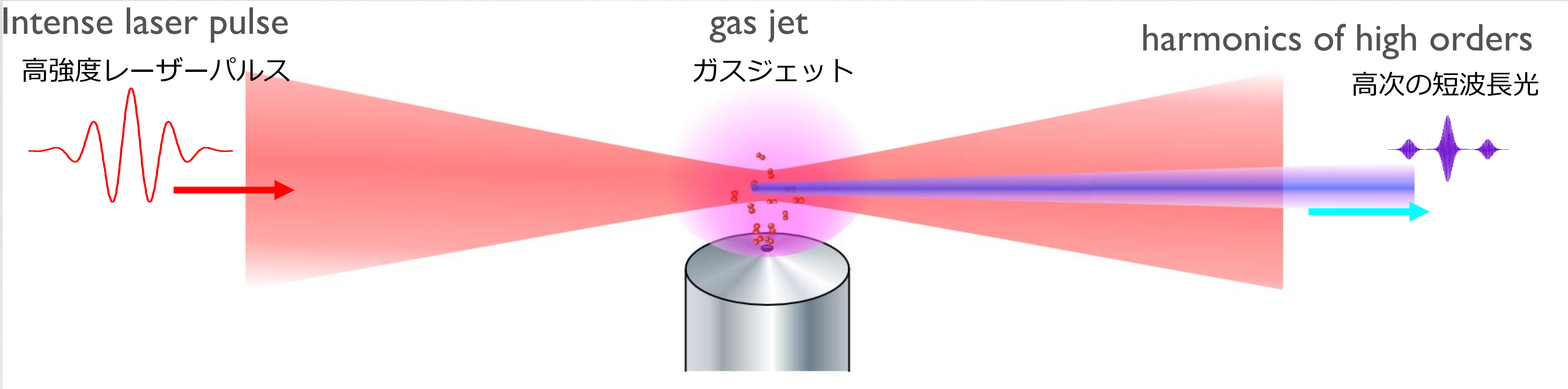
高次高調波発生
HIGH-HARMONIC GENERATION (HHG)

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.

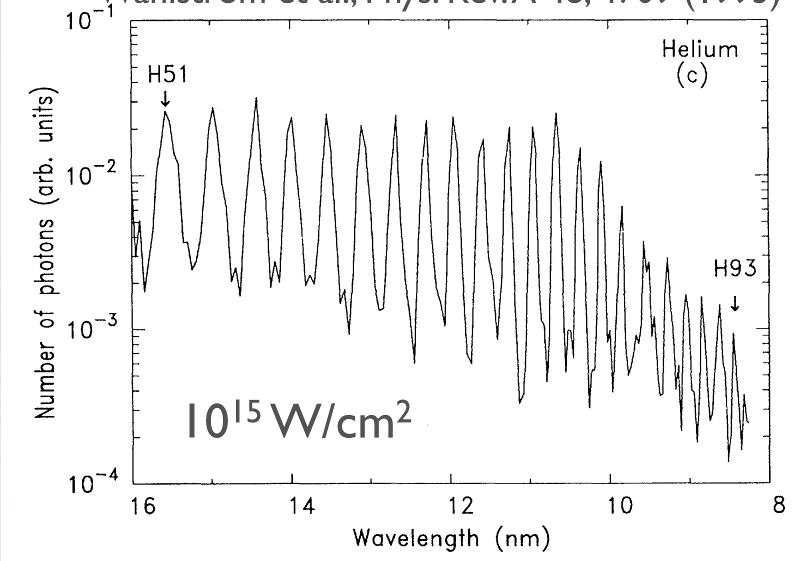
新しい極端紫外・軟エックス線光源として注目される。

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

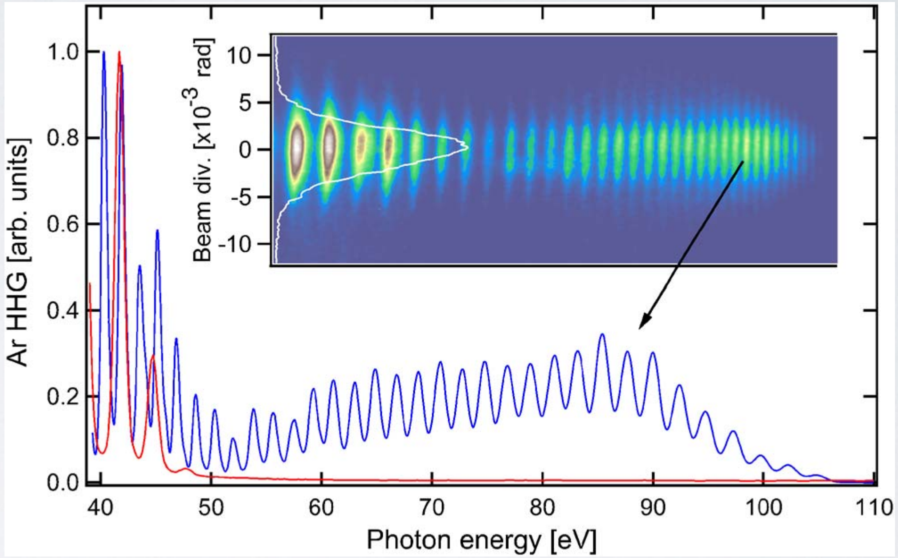
How high orders?

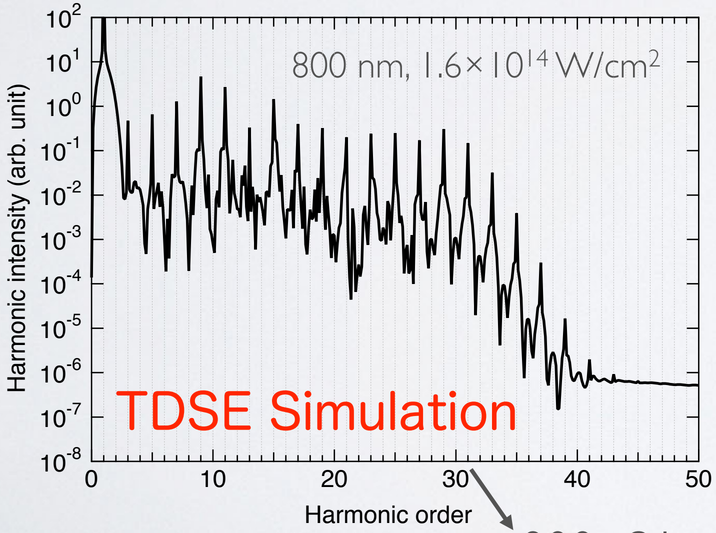
HARMONIC SPECTRUM 高調波スペクトル

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



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





800÷31= 26 nm

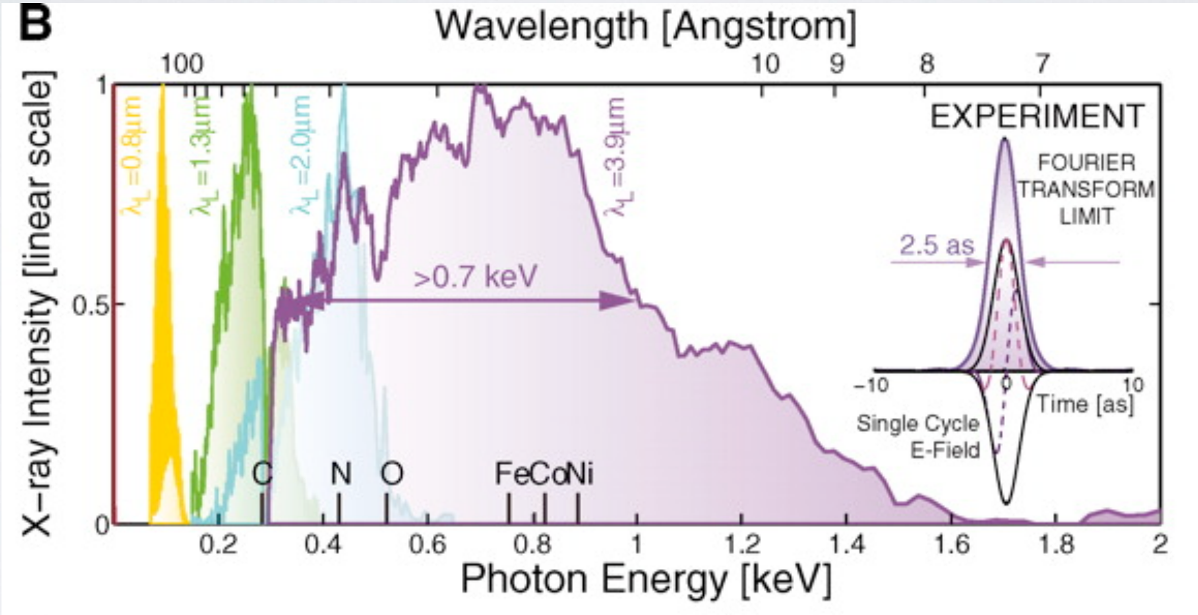
Only odd orders 奇数次のみ

gas is a medium of inversion symmetry

期待媒質は反転対称

Even up to 1.6 keV, > 5000 orders

almost x-ray!



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

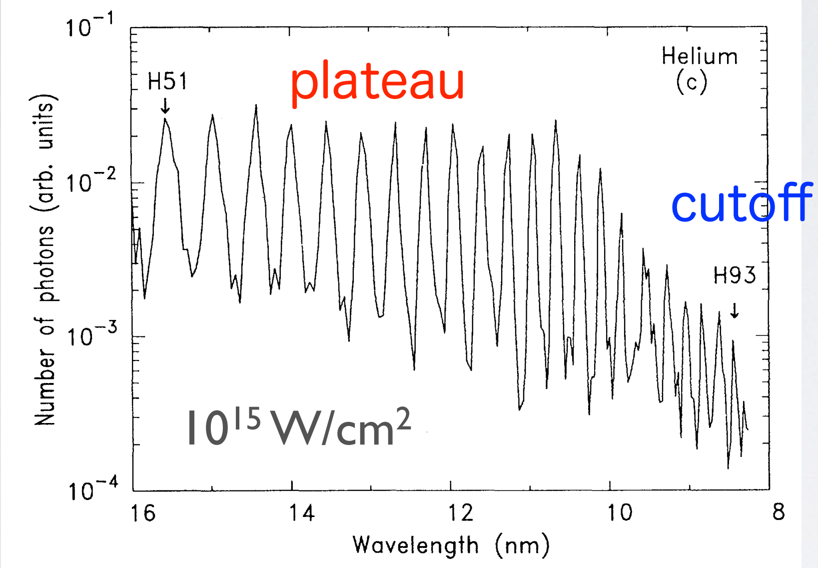
a new type of laser-based radiation source

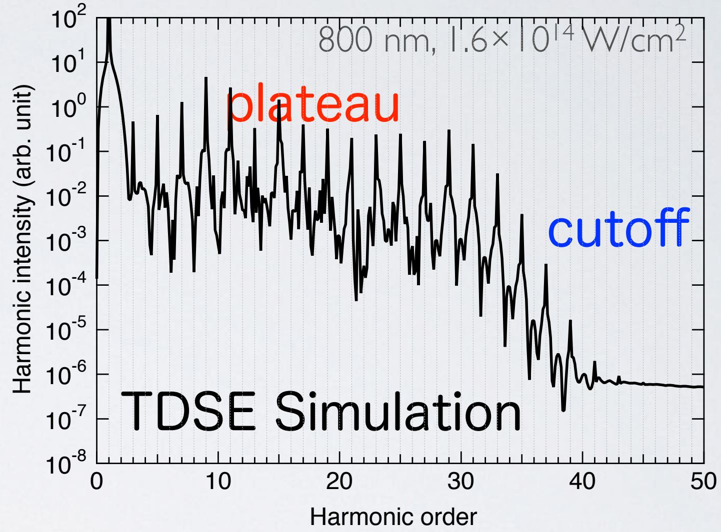
レーザーをベースにした新しいタイプの放射線源

Plateau and cutoff プラトーとカットオフ

- 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

$$E_c \approx I_p + 3.17 U_p \qquad U_p(\mathrm{eV}) = \frac{e^2 E_0^2}{4m\omega^2} = 9.3 \times 10^{-14} I(\mathrm{W/cm^2}) \lambda^2 (\mu\mathrm{m})$$

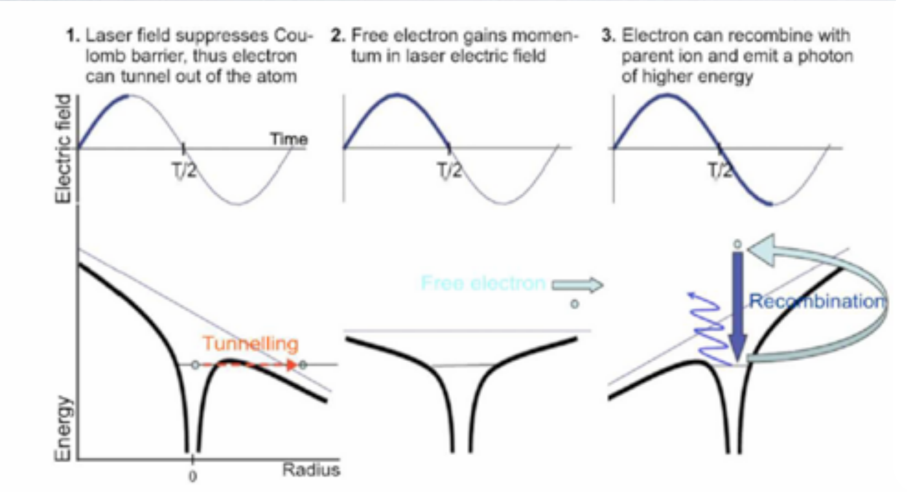
ポンデロモーティブエネルギー ponderomotive energy

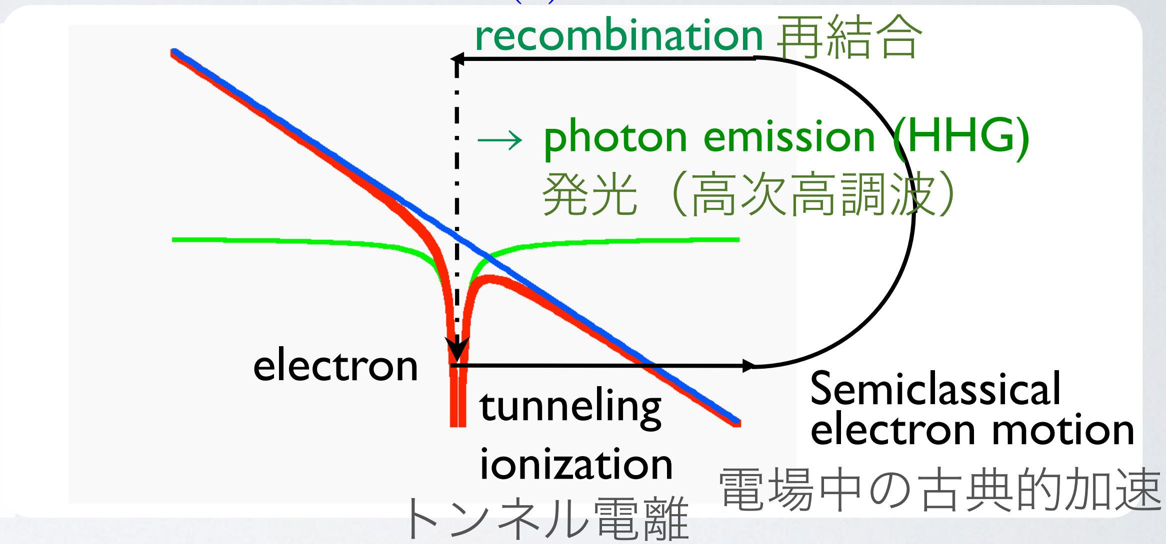
These features cannot be understood as perturbative harmonic generation.

摂動論的には解釈できない

高次高調波発生のメカニズム
3 ステップモデル 3-STEP MODEL

Laser field レーザー電場 $E(t) = E_0 \cos \omega t$






Paul B. Corkum

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)

3-STEP MODEL OF HHG
高次高調波発生の３ステップモデル

Ionization at $t = t_0$ with vanishing initial velocity at origin
時刻 t_0 でイオン化。原点に初速ゼロで出現

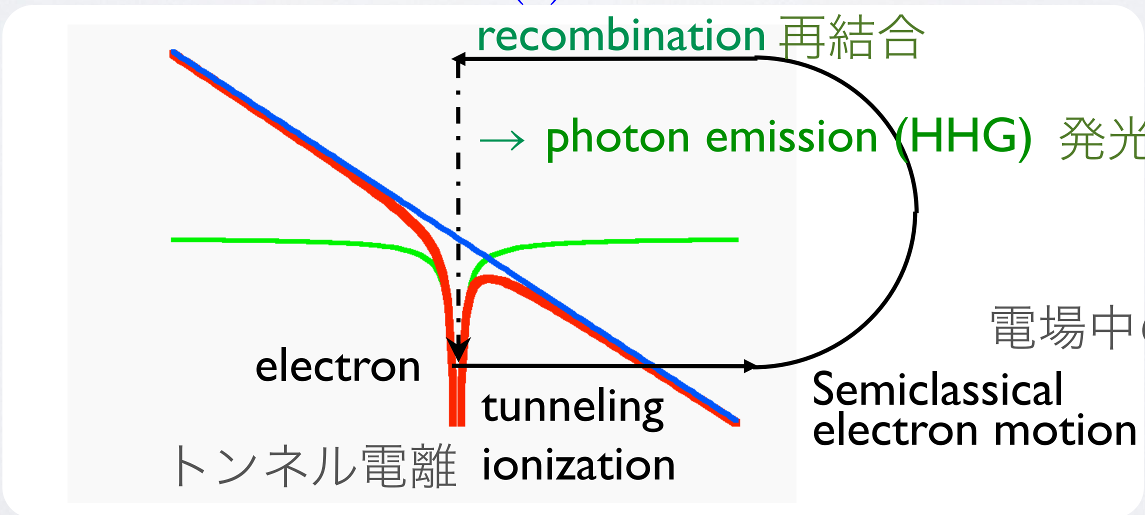
$$m\ddot{z} = -eE_0 \cos \omega t \qquad \dot{z}(t_0) = 0 \qquad z(t_0) = 0$$

Normalization 規格化 $\phi = \omega t \quad \phi_0 = \omega t_0$

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

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

Laser field レーザー電場 $E(t) = E_0 \cos \omega t$



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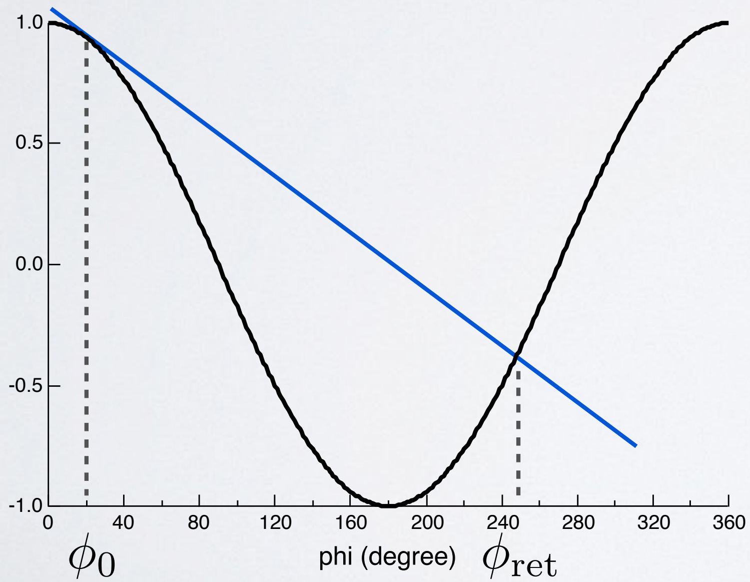
TIME (PHASE) OF RECOMBINATION
再衝突時刻

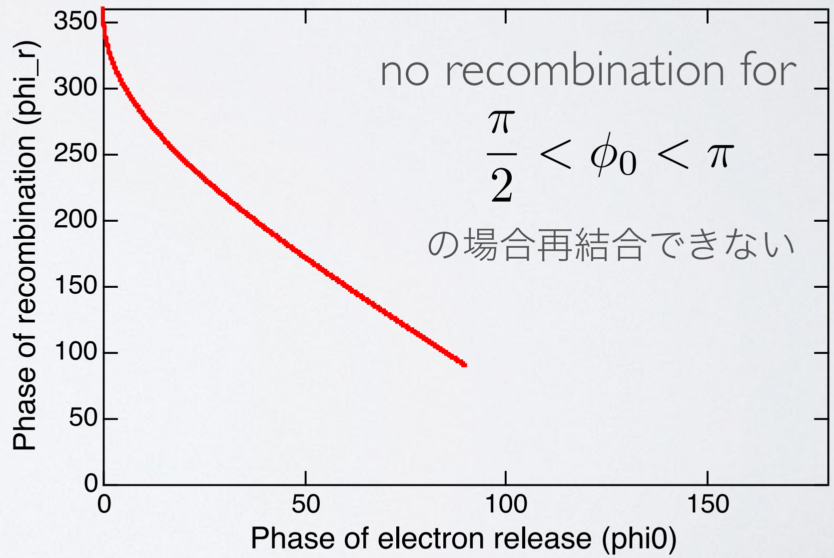
$$z = 0 \longrightarrow (\cos\phi_{\rm ret} - \cos\phi_0) + (\phi_{\rm ret} - \phi_0)\sin\phi_0 = 0$$

$$(\cos\phi)'|_{\phi_0} = \frac{\cos\phi_{\rm ret} - \cos\phi_0}{\phi_{\rm ret} - \phi_0}$$

phase of ionization vs phase of recombination

イオン化時刻と再衝突時刻の関係





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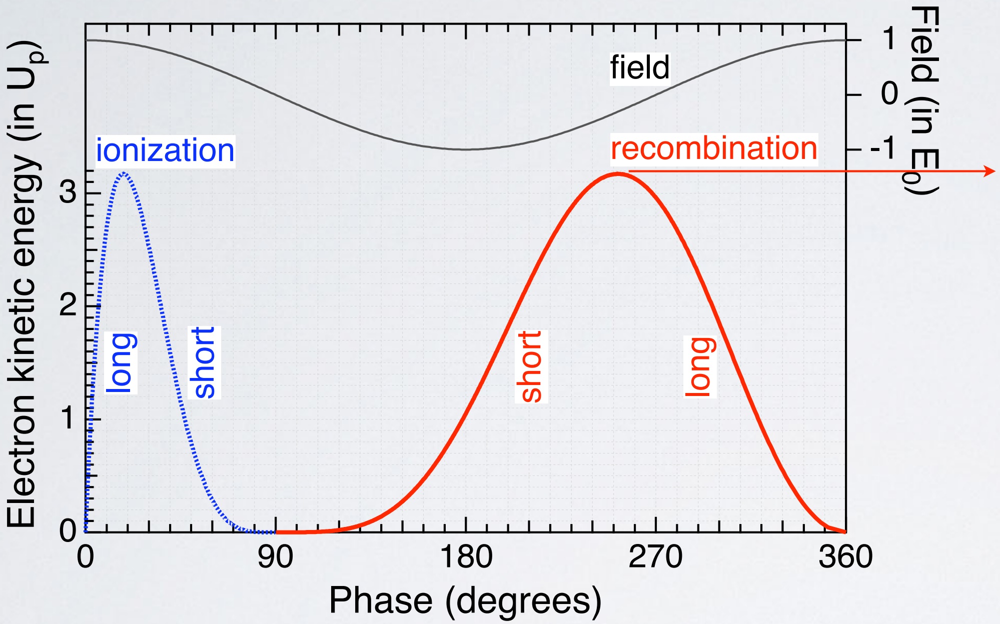
Simple explanation of the cut-off law

カットオフ則のシンプルな説明



There is the maximum kinetic energy which is classically allowed.

再結合時の運動エネルギーの最大値

$$3.17U_p$$

cut-off カットオフ

$$E_c = I_p + 3.17U_p$$

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

同じ高調波次数（光子エネルギー）に対応するイオン化時刻と再結合時刻のペアは２つある。

short trajectory
long trajectory

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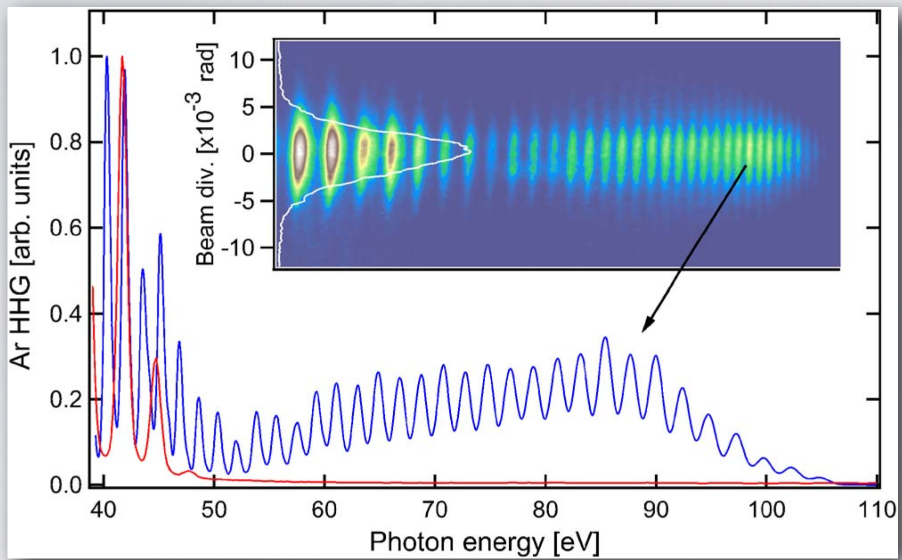
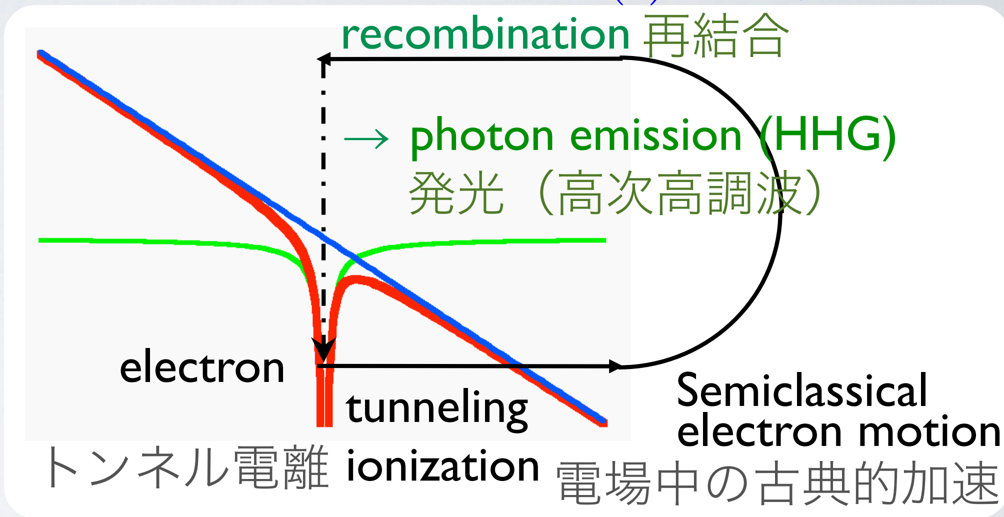
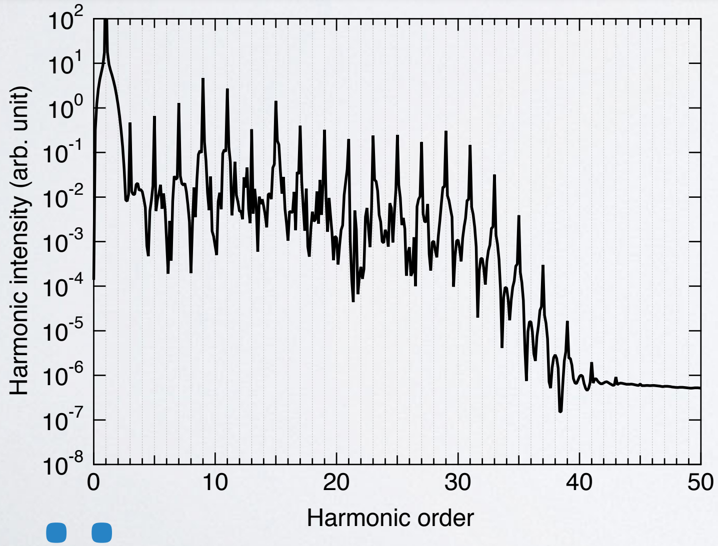
WHY DO HARMONIC SPECTRA CONSIST OF DISCRETE PEAKS? なぜ、高次高調波スペクトルは離散的なのか？

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



Laser field レーザー電場 $E(t) = E_0 \cos \omega t$

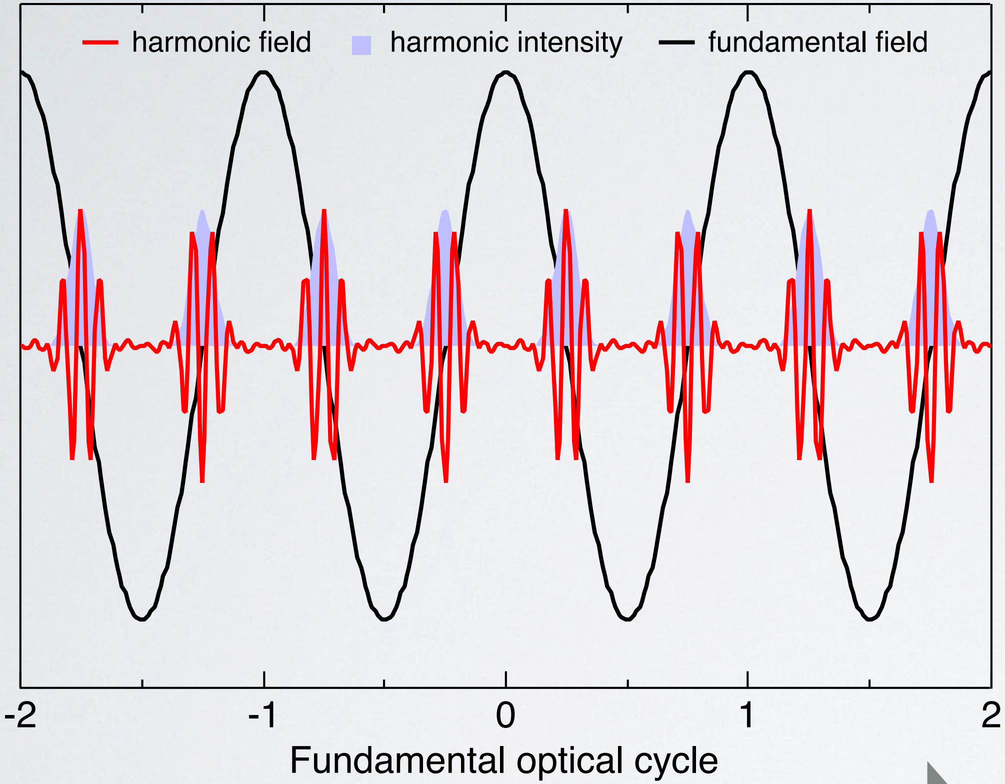




This is repeated every half cycle with an alternating phase

トンネル電離と高調波の発生は、レーザーの半周期ごとに起こる。

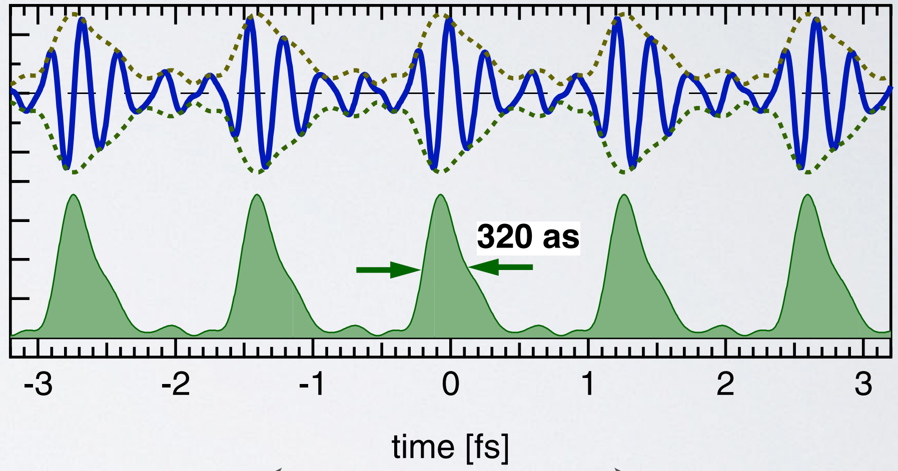
Harmonic electric field
高調波の電場波形の概念図



$$f(t) = -f(t - \pi/\omega_0)$$

Experimentally measured
実験的にも観測されている

Nabekawa et al., Phys. Rev. Lett. 97, 153904 (2006)



laser cycle レーザーの１周期(2.7 fs)

Spectrum only consists of odd harmonics.

基本波の奇数倍の周波数成分のみを含む。

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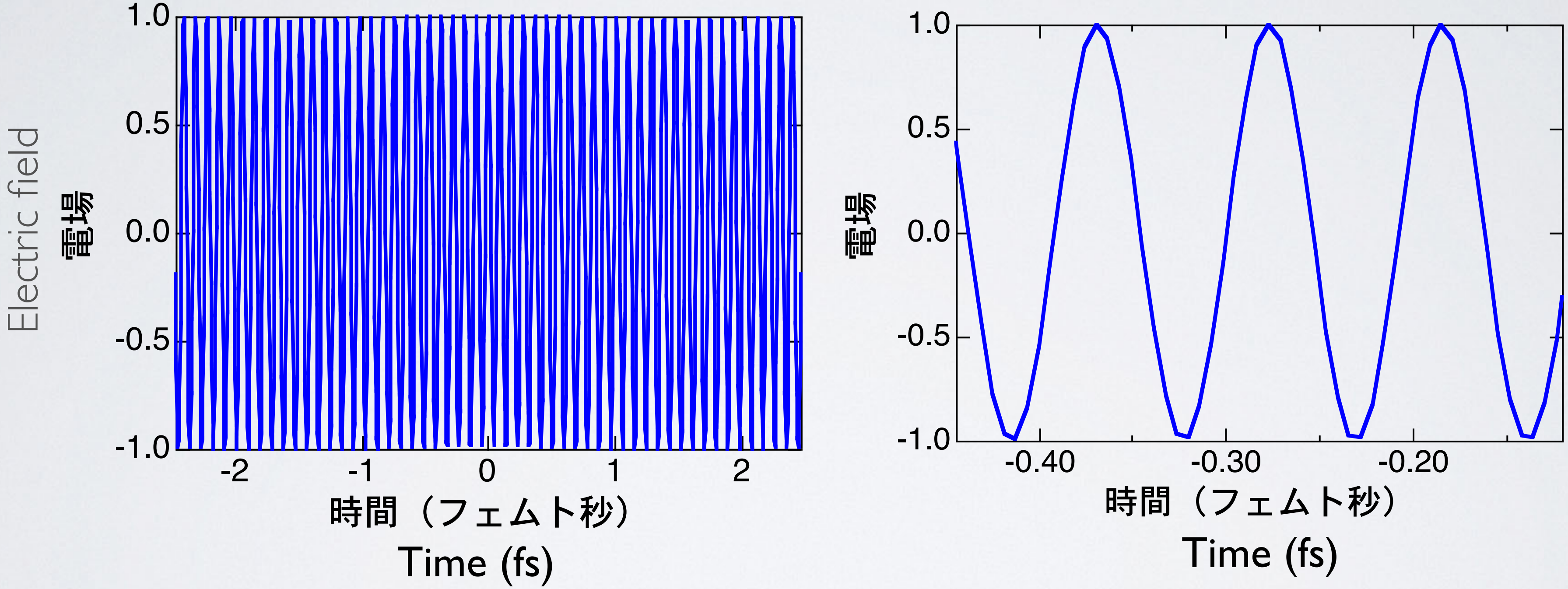
SINGLE FREQUENCY COMPONENT
１つの次数のみが存在するときの光電界

$$E_h(t) = E_q \cos(q\omega + \phi_q) = E_{2n+1} \cos[(2n+1)\omega + \phi_{2n+1}]$$

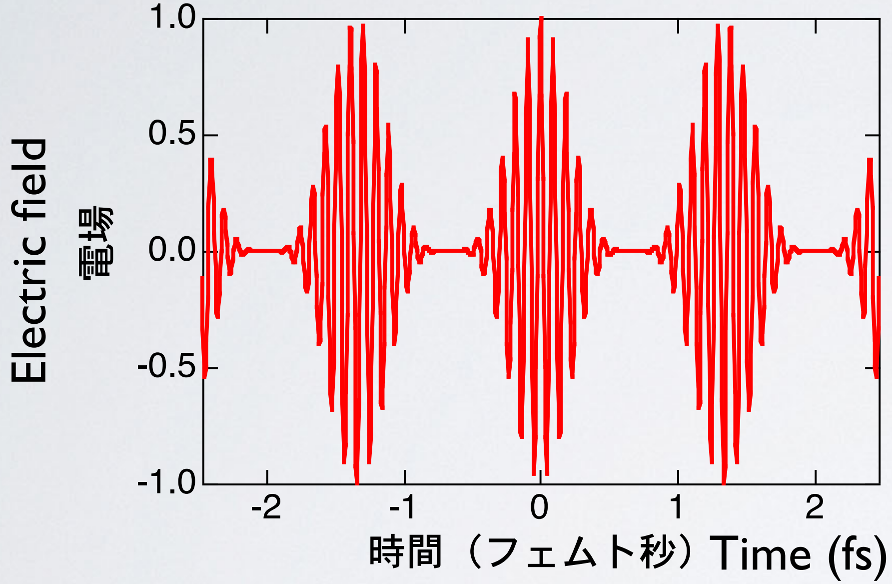


Continuous wave (no pulse)

連続波（パルスではない）

MULTIPLE (ODD) HARMONIC COMPONENTS
複数の次数（奇数次）が混在するときの光電界

$$E_h(t) = \sum_q E_q \cos(q\omega + \phi_q) = \sum_q E_{2n+1} \cos[(2n+1)\omega + \phi_{2n+1}]$$



- attosecond pulse train (APT)
  アト秒パルス列になっている
- bursts repeated every half cycle of the fundamental laser
  パルスの間隔は、基本波の半周期
- adjacent pulses have an opposite phase
  隣り合うパルスは位相が反転

equispaced frequency components
等間隔の周波数成分

⟷

train of repeated pulses
等間隔のパルス列

We don’t need “photons” to understand harmonic generation

DISCRETE PEAKS OF ODD HARMONICS CAN BE INTERPRETED IN TWO WAYS.

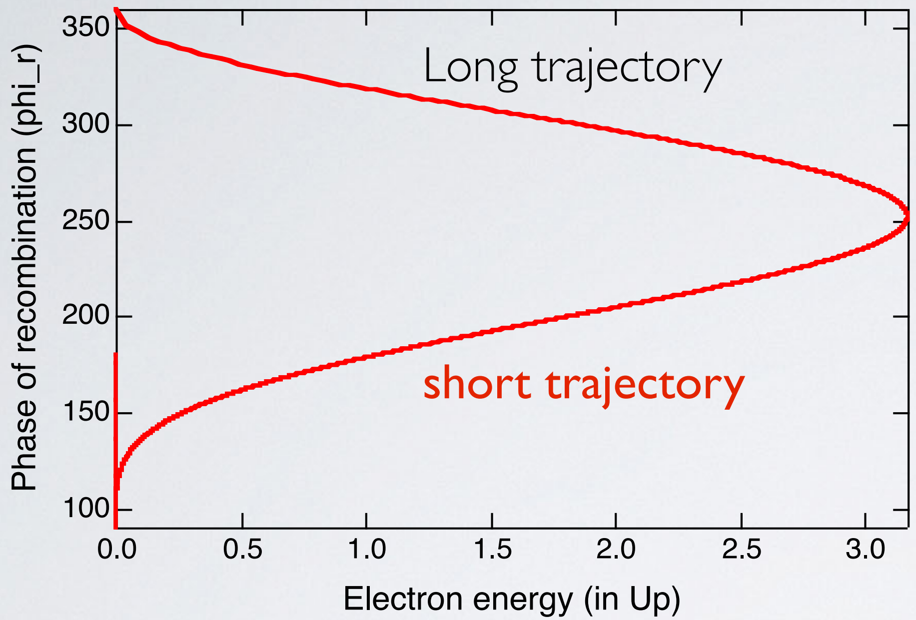
高次高調波の奇数次のみを含む離散的なピークは、二通りに解釈できる。

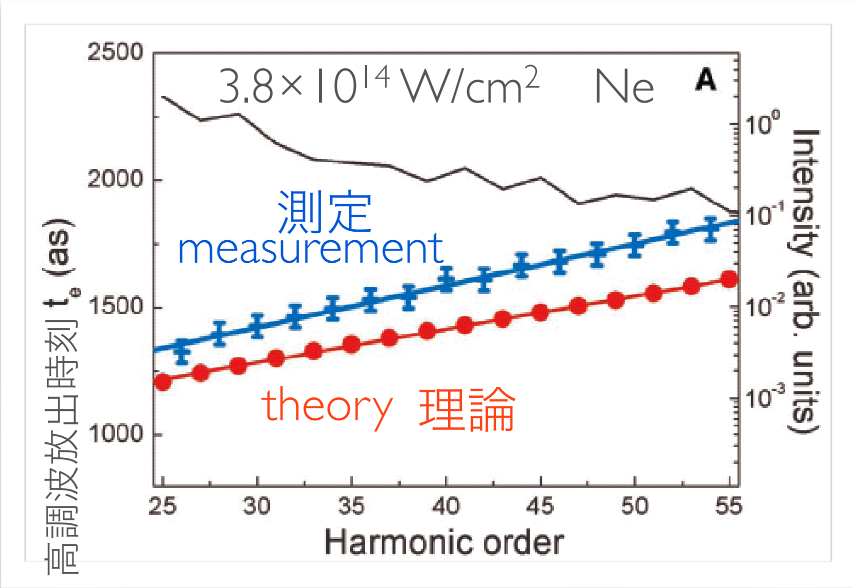
- Integer number of photon energy + inversion symmetry
  光子エネルギーの整数倍＋反転対称性

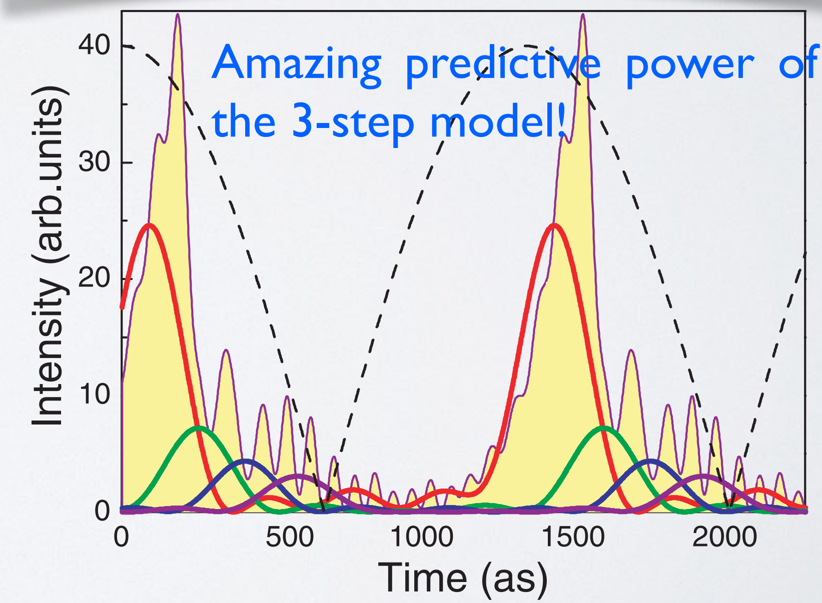
- **Light emission repeated every half cycle** (with alternating phase)
  基本波の半サイクルごとに、反対の位相で光放出

Time of emission depends on harmonic order

次数によって高調波の発生時刻が異なる







Mairesse et al., Science 302, 1540 (2003)
Varju et al., J. Mod. Opt. 52, 379 (2005)

ショートトラジェクトリーの場合
低次が先に高次が後で発生する。

For the case of short trajectory
Higher-order components emitted later

→ **Positive chirp**
ポジティブチャープ

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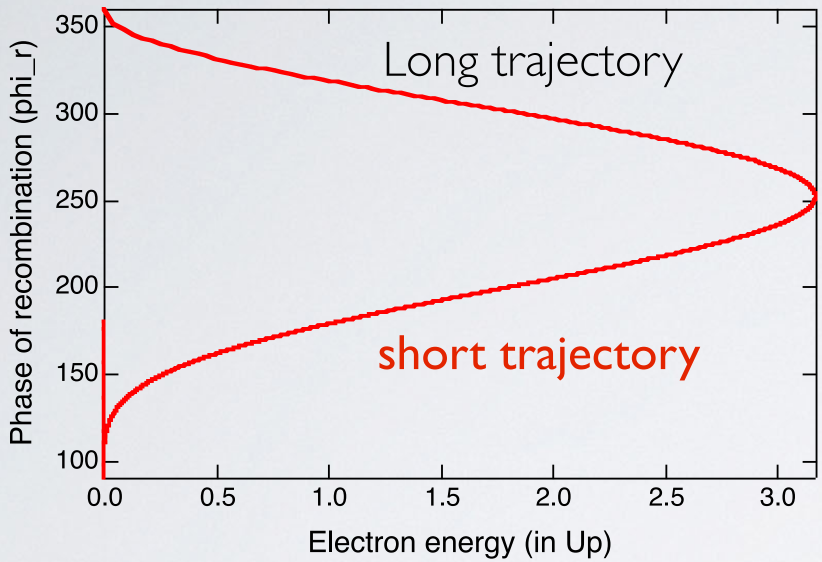
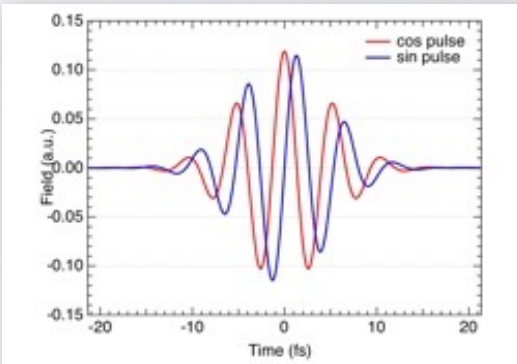
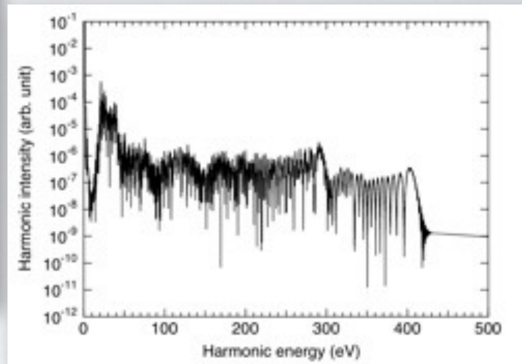
Time of emission depends on harmonic order

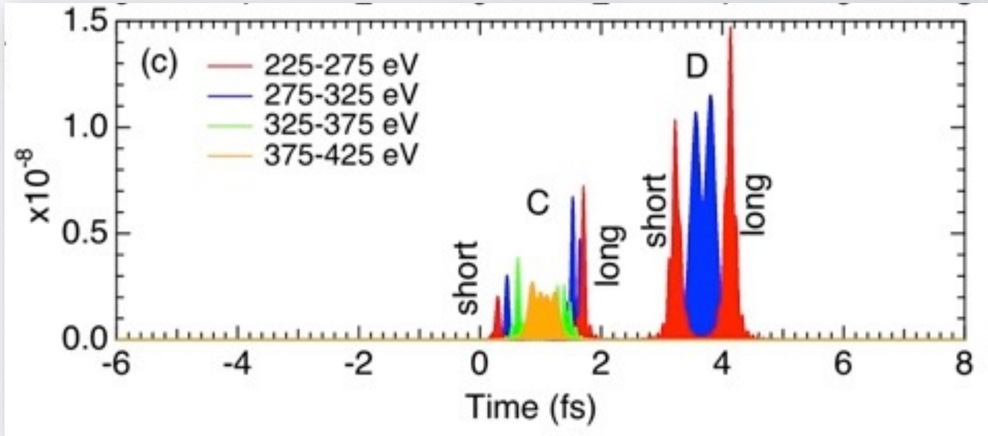
次数によって高調波の発生時刻が異なる



TDSE simulation







for the short trajectory → **Positive chirp**

for the long trajectory → **Negative chirp**

K. L. Ishikawa, "High-harmonic generation" in Advances in Solid-State Lasers, ed. by M. Grishin (INTECH, 2010) 439-464

高次高調波発生の量子論

Quantum theory of high-harmonic generation

Lewenstein model

Lewenstein et al., Phys. Rev. A 49, 2117 (1994)

高強度場近似
Strong-field approximation (SFA)

- 励起状態の寄与を無視　The contribution of all the excited bound states can be neglected.
- 連続状態に対する原子のポテンシャルの効果を無視（連続状態を平面波で近似）The effect of the atomic potential on the motion of the continuum electron can be neglected.
- 基底状態の減少を無視　The depletion of the ground state can be neglected.

$$i\frac{\partial\psi(\mathbf{r},t)}{\partial t} = \left[-\frac{1}{2}\nabla^2 + V(\mathbf{r}) + zE(t)\right]\psi(\mathbf{r},t)$$

双極子モーメント

Time-dependent dipole moment

$$x(t) \equiv \langle \psi(\mathbf{r},t) \mid z \mid \psi(\mathbf{r},t)\rangle$$

$$= i\int_{-\infty}^{t} dt' \int d^3\mathbf{p}\, \langle\varphi(\mathbf{r})e^{iI_pt}|z|\mathbf{p}+\mathbf{A}(t)\rangle \exp\left[-i\int_{t'}^{t} dt''\left(\frac{[\mathbf{p}+\mathbf{A}(t'')]^2}{2}\right)\right] \langle\mathbf{p}+\mathbf{A}(t')|zE(t')|\varphi(\mathbf{r})e^{iI_pt'}\rangle + \text{c.c.}$$

3-step model

recombination 再結合

motion in the laser field レーザー場中での運動

ionization イオン化

$$= i\int_{-\infty}^{t} dt' \int d^3\mathbf{p}\, \langle\varphi(\mathbf{r})e^{iI_pt}|z|\mathbf{p}+\mathbf{A}(t)\rangle \exp\left[-i\int_{t'}^{t} dt''\left(\frac{[\mathbf{p}+\mathbf{A}(t'')]^2}{2}\right)\right] \langle\mathbf{p}+\mathbf{A}(t')|zE(t')|\varphi(\mathbf{r})e^{iI_pt'}\rangle + \text{c.c.}$$

Dipole moment between the recolliding wave packet and the ground state

再衝突電子波束と基底状態との間の双極子モーメント

recolliding electron wave packet 再衝突電子波束

ionization イオン化

$$x(t) = i\int_{-\infty}^{t} dt' \int d^3\mathbf{p}\, d^*(\mathbf{p}+\mathbf{A}(t)) \cdot \exp[-iS(\mathbf{p},t,t')] \cdot E(t')d(\mathbf{p}+\mathbf{A}(t')) + \text{c.c.}$$

transition dipole

transition dipole 遷移双極子行列要素

半古典的作用積分 semiclassical action

$$S(\mathbf{p},t,t') = \int_{t'}^{t} dt''\left(\frac{[\mathbf{p}+\mathbf{A}(t'')]^2}{2} + I_p\right)$$

HARMONIC SPECTRUM
= FOURIER TRANSFORM OF DIPOLE MOMENT

高調波スペクトル＝双極子モーメントのフーリエ変換

$$\hat{x}(\omega_h) = i \int_{-\infty}^{\infty} dt \int_{-\infty}^{t} dt' \int d^3\mathbf{p}\, d^*(\mathbf{p}+\mathbf{A}(t)) \cdot \exp[i\omega_h t - iS(\mathbf{p},t,t')] \cdot E(t') d(\mathbf{p}+\mathbf{A}(t')) + \mathrm{c.c.}$$

5 重積分　five-dimensional integral

↓

鞍点解析　saddle-point analysis

cf. path integral 経路積分

saddle-point analysis (SPA)

$\boxed{\text{Saddle-point equations}}$ solutions 解 ➞ trajectories トラジェクトリー

$$\frac{[p+A(t')]^2}{2} = -I_p$$

tunneling ionization
トンネル電離

t' time of ionization
t time of recombination

$$\int_{t'}^{t} [p+A(t'')]dt'' = 0$$

recombines at the location of ionization
イオン化と再結合の位置が同じ

$$\frac{[p+A(t)]^2}{2} + I_p = \omega_h$$

harmonic photon energy
= kinetic energy at recombination + ionization potential
高調波の光子エネルギー ＝ 再結合時の運動エネルギー ＋ イオン化ポテンシャル

$$\hat{x}(\omega_h) = \sum_s \left(\frac{\pi}{\epsilon + \frac{i}{2}(t_s - t'_s)}\right)^{3/2} \frac{i2\pi}{\sqrt{\det S''(t,t')|_s}} d^*(p_s + A(t_s))$$
$$\times \exp[i\omega_h t_s - iS(p_s, t_s, t'_s)]E(t'_s)d(p_s + A(t'_s)),$$

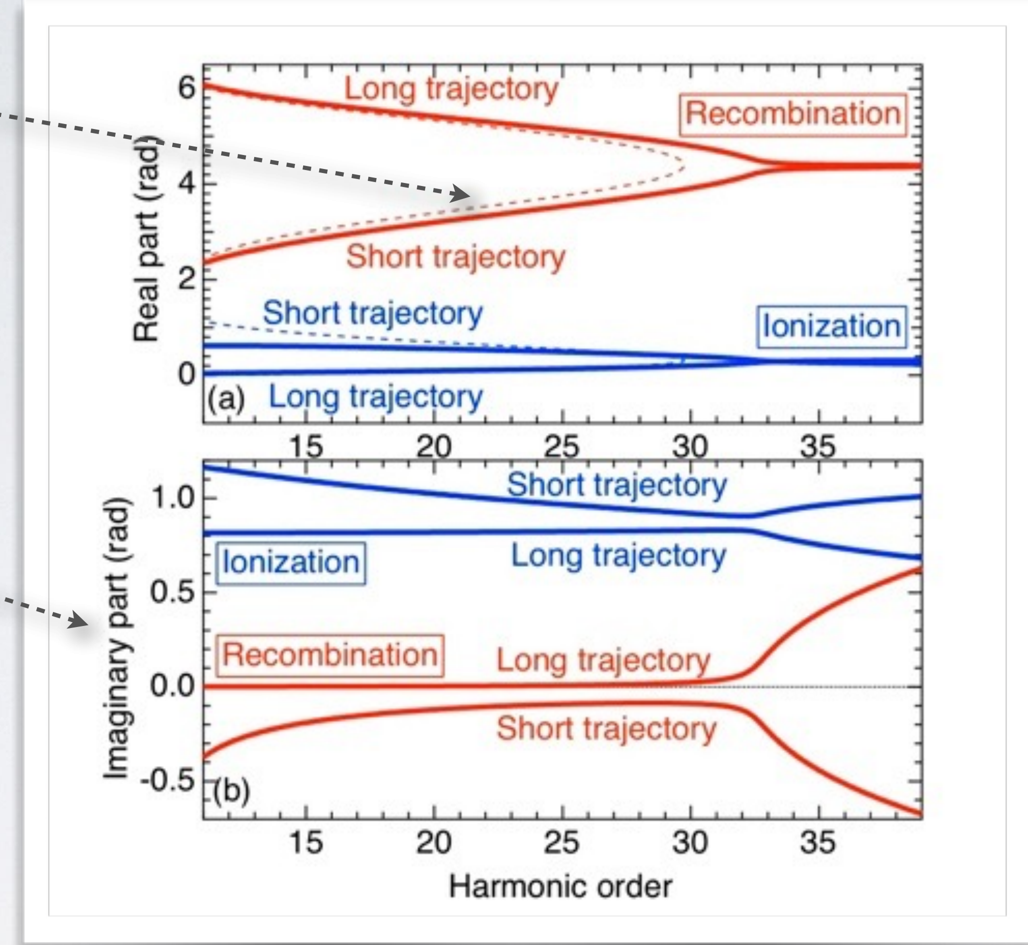
➡ physically corresponds to the 3-step model
3ステップモデルに物理的に対応

Example of saddle-point solutions　鞍点解の例

$E(t) = E_0 \cos \omega t$　　Ar (I_p = 15.7596 eV)　　$1.6 \times 10^{14}\ \mathrm{W/cm^2}$

Real part (top) and imaginary part (bottom) of $\phi' = \omega t'$　$\phi = \omega t$　の実部（上）と虚部（下）

dashed lines:
3-step model
破線は３ステップモデルの解



cutoff カットオフ

$E_c = 3.17U_p + gI_p$

$(g \approx 1.3)$

interpretted as
"tunnling time"

「トンネル時間」
に対応すると解釈
されている

- The 3-step model is a good approximation to the quantum-mechanical Lewenstein model → Success of the 3-step model
  - ３ステップモデルは、量子力学的なLewensteinモデルのよい近似になっている。
  → ３ステップモデルの成功の理由

attosecond pulse train (APT)

アト秒パルス列

と

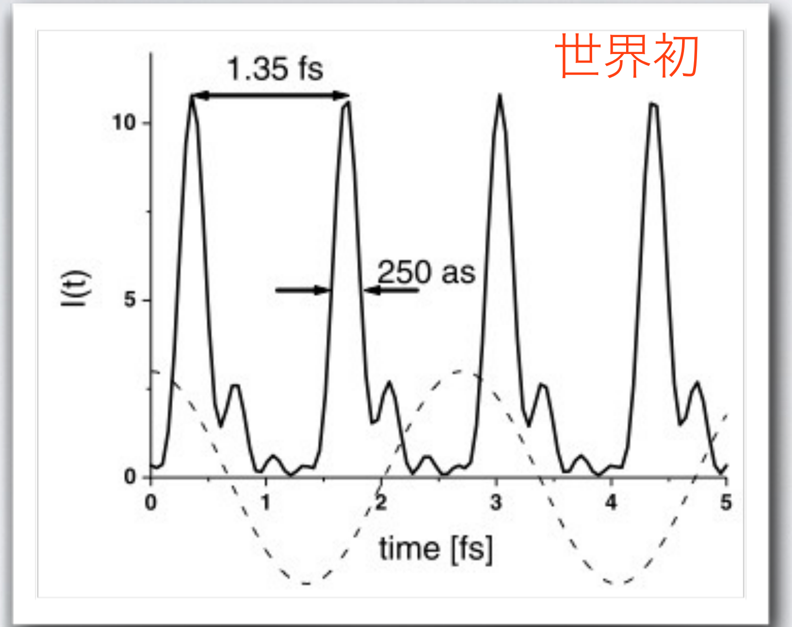
単独アト秒パルス

isolated attosecond pulse (IAP)

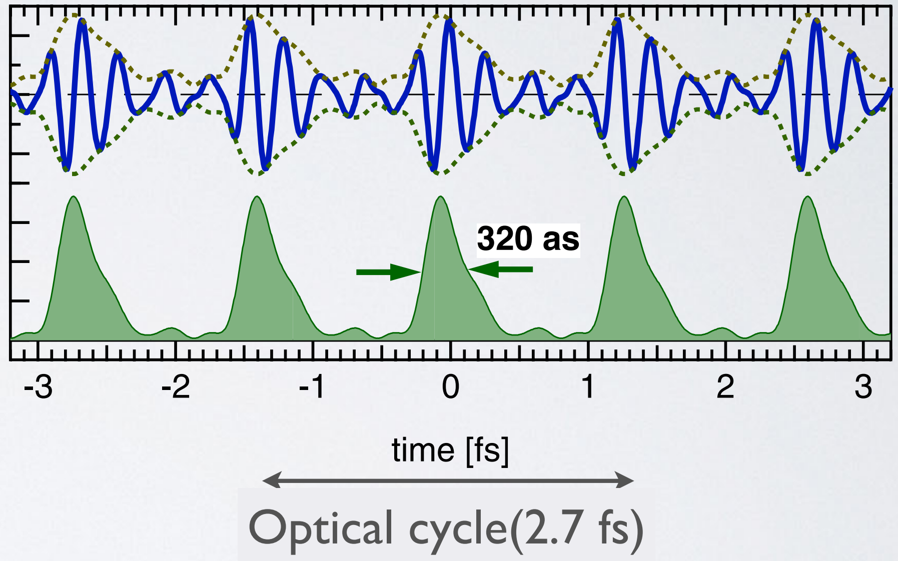
High-order harmonics are generated as attosecond bursts repeated each half cycle of the fundamental laser (attosecond pulse train)

高次高調波は、基本波レーザーの半周期ごとにアト秒のバーストとして発生する（アト秒パルス列）

Paul et al., Science 292, 1689 (2001)



Nabekawa et al., Phys. Rev. Lett. 97, 153904 (2006)



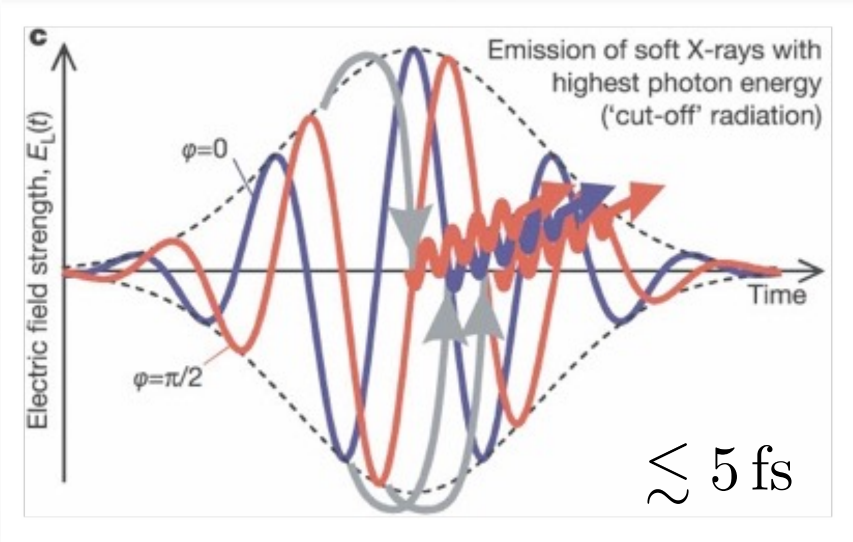
Only one burst → Isolated attosecond pulse (IAP)

１回だけバーストが発生するようにすれば、単独パルスになる。

Isolated attosecond pulse generation by a few-cycle laser pulse

単独アト秒パルス

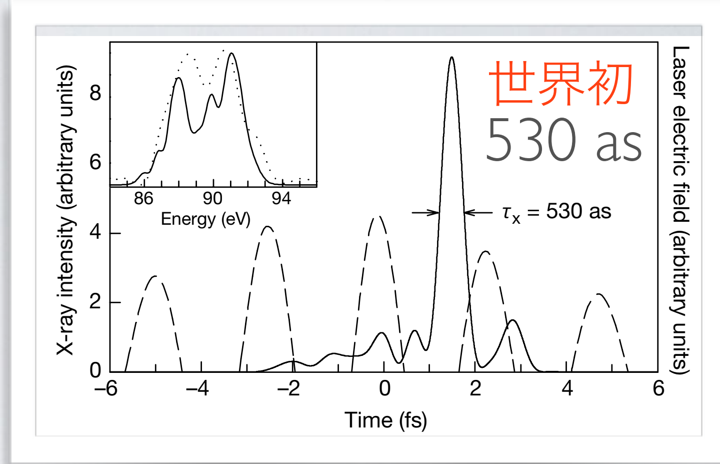
Baltuska et al. Nature 421, 611 (2003)

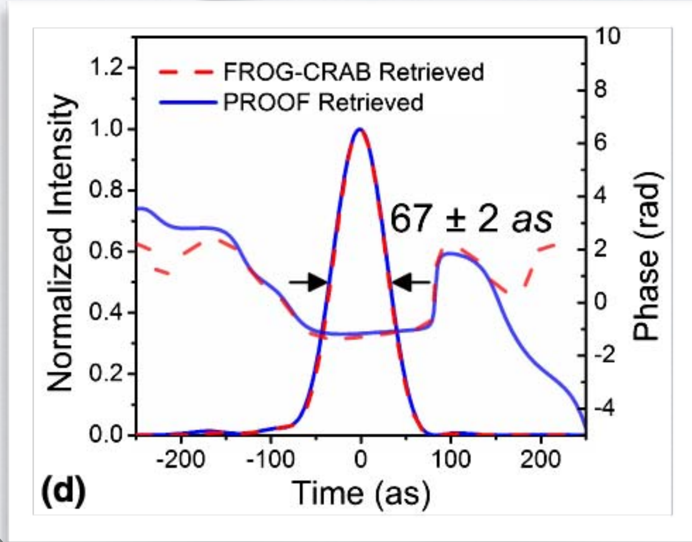


Light emission takes place only once.

光の放出は１回だけ

Hentschel et al. Nature 414, 509 (2001)





Zhao et al. (2012)

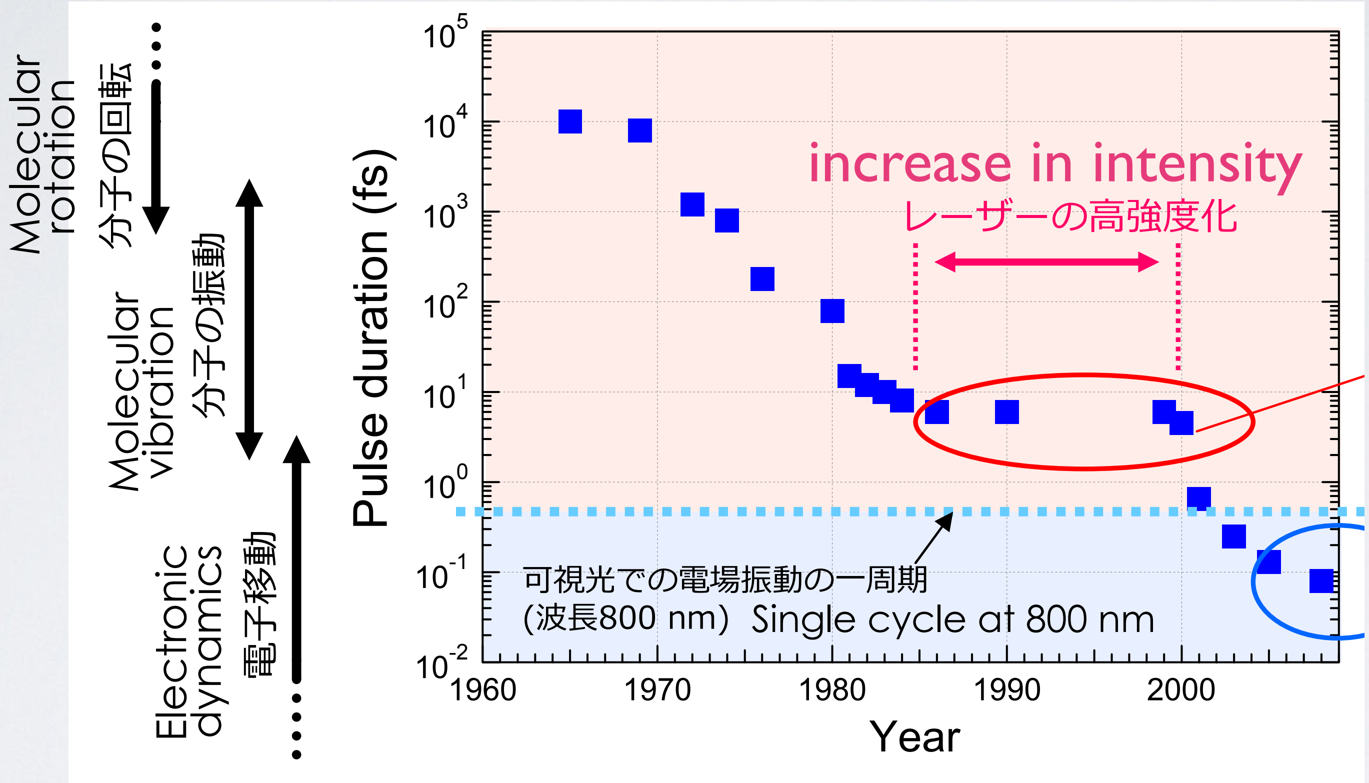
Attosecond (10^{-18} sec) pulse

アト秒パルス

more details in *Quantum Beam Engineering*

From femtosecond to attosecond

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



(courtesy of Prof. J. Itatani)

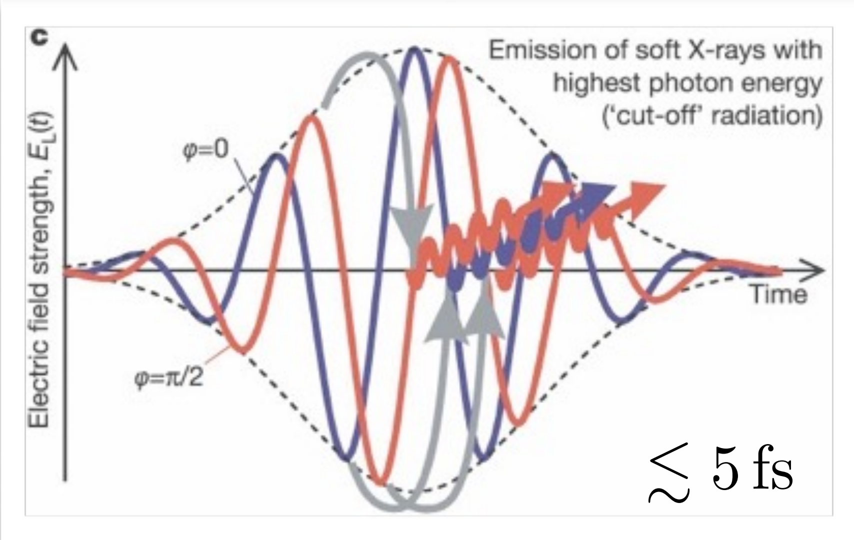
0.1attosecond！　0.1アト秒！

supplementary materials

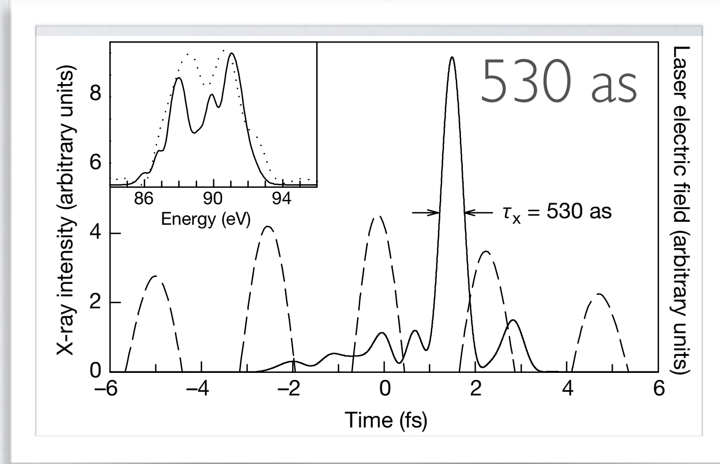
How to generate IAP

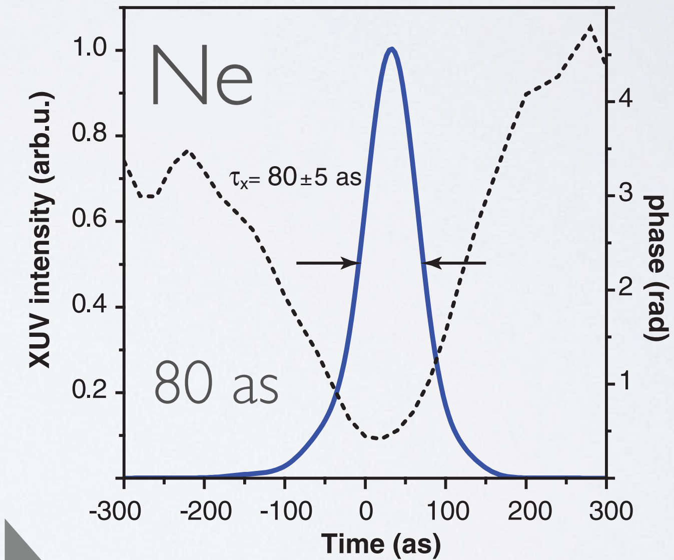
Isolated attosecond pulse generation by a few-cycle laser pulse

Baltuska et al. Nature 421, 611 (2003)



Hentschel et al. Nature 414, 509 (2001)





Goulielmakis et al. Science 320, 1614 (2008)

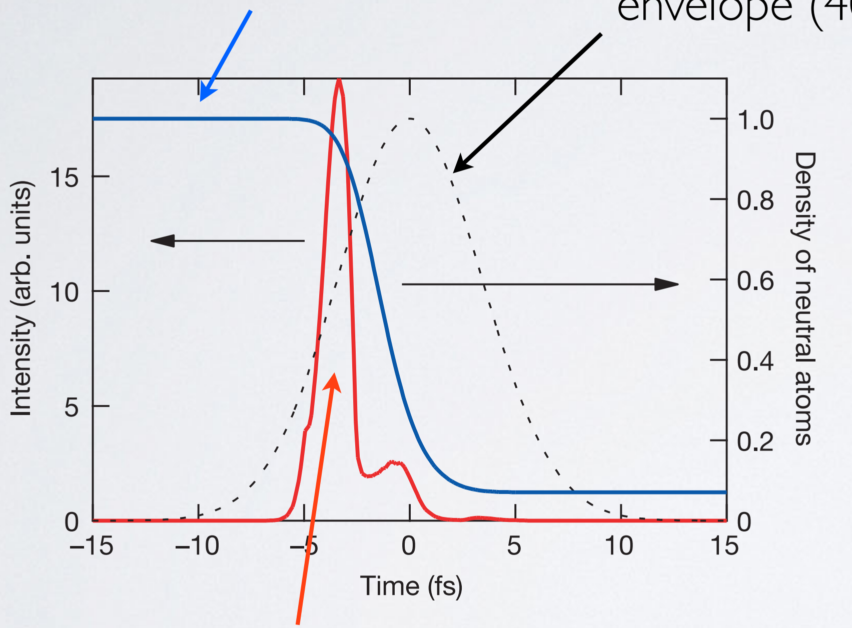
Light emission takes place only once. → **Attosecond (10^{-18} sec) pulse**

IONIZATION SHUTTER

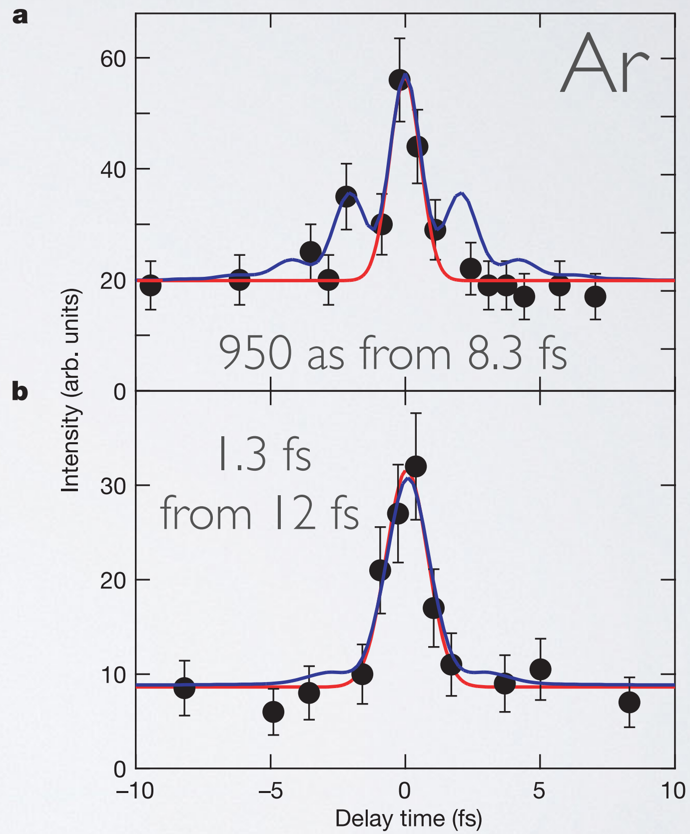
HHG is suppressed when neutral atoms are depleted

density of neutral Ar atoms

fundamental field envelope (400 nm)



9th harmonic (of 400 nm) = 27.9 eV



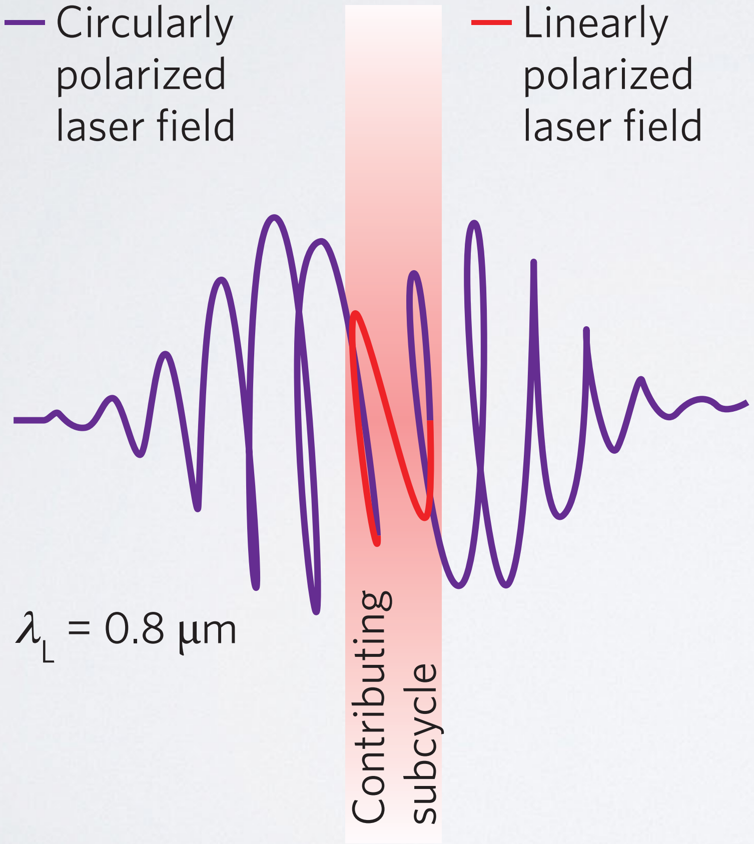
Isolated sub-fs pulse generation from a ~10 fs pulse

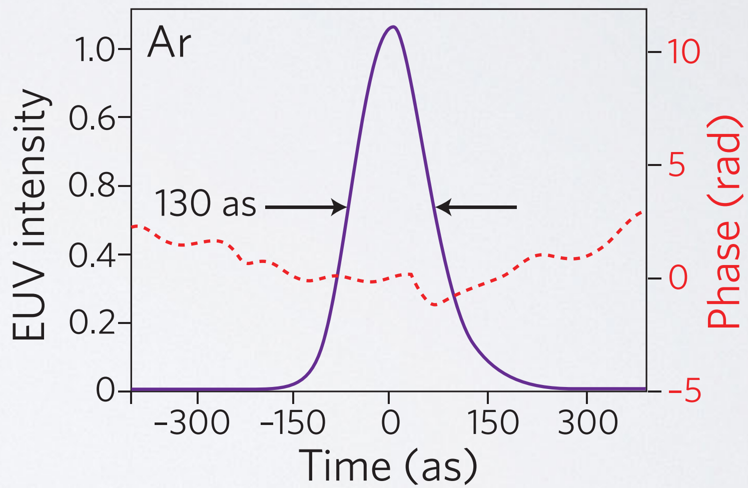
Sekikawa *et al.*, Nature 432, 605 (2004)

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)

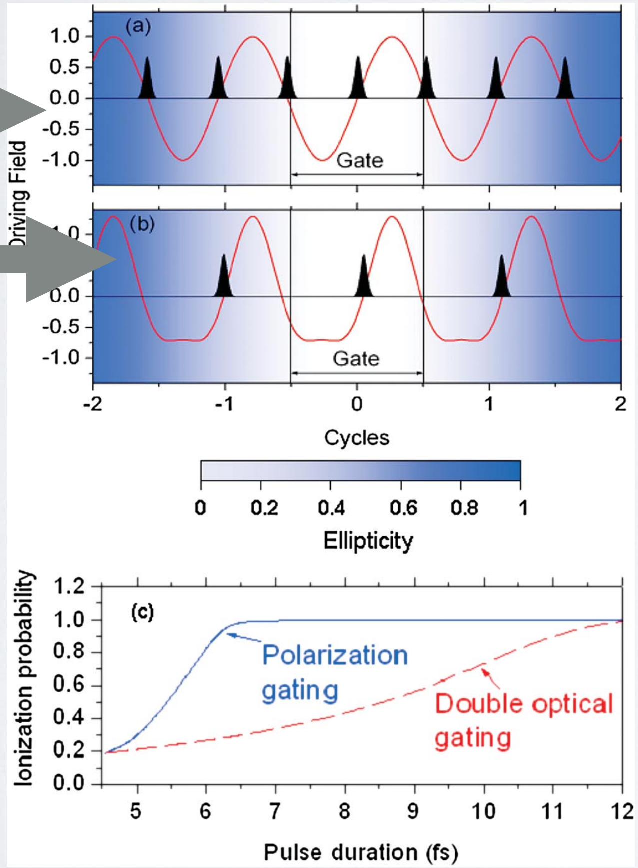
DOUBLE OPTICAL GATING (DOG)

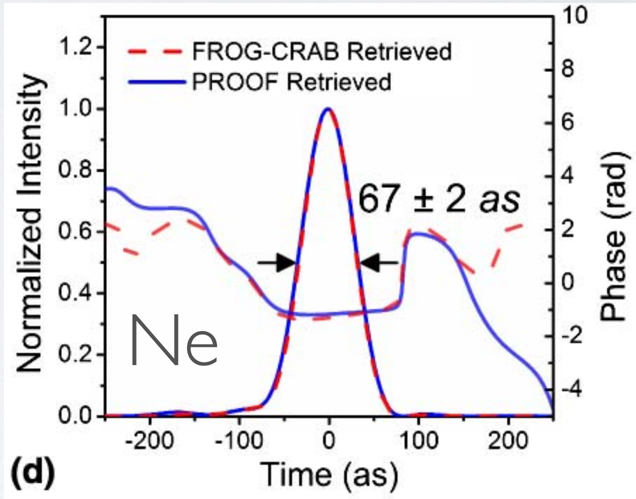
Polarization gating + two-color gating

ω

$\omega + 2\omega$

with second-harmonic field





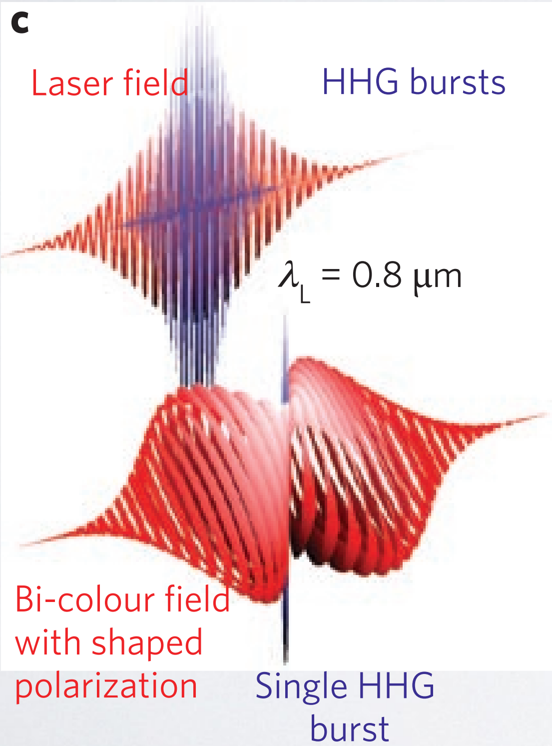
IAP generation from a ~10 fs pulse

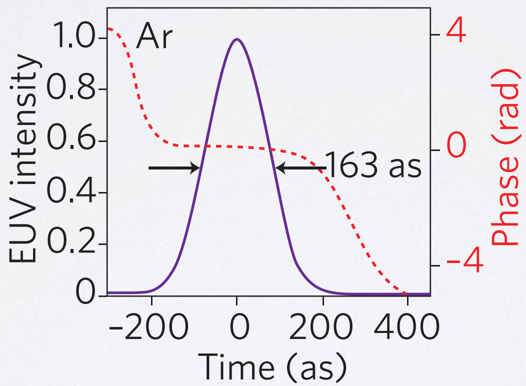
Mashiko et al., PRL 2008, 103906 (2008)

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

GENERALIZED DOUBLE OPTICAL GATING (GDOG)

Elliptical instead of circular polarization





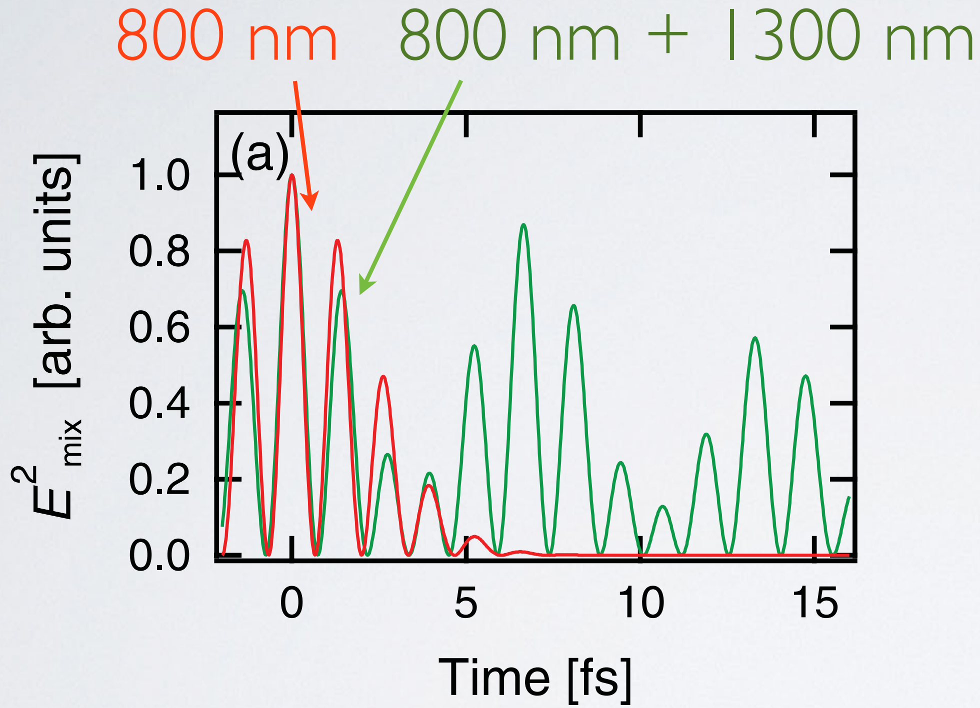
IAP generation from a > 20 fs pulse without need of carrier-envelope stabilization

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

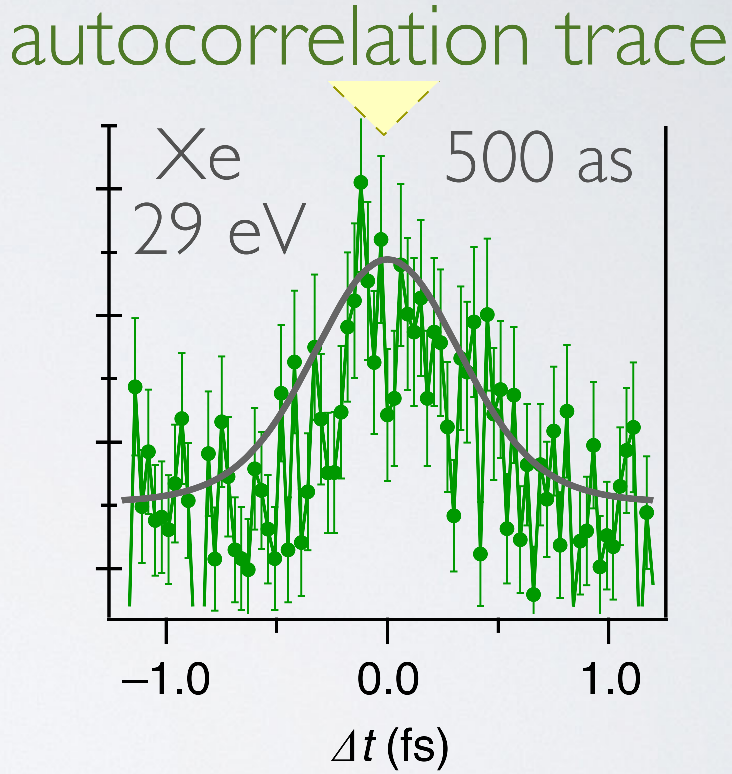
K. L. Ishikawa

INFRARED TWO-COLOR SYNTHESIS

800 nm + 1300 nm two-color driving field



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

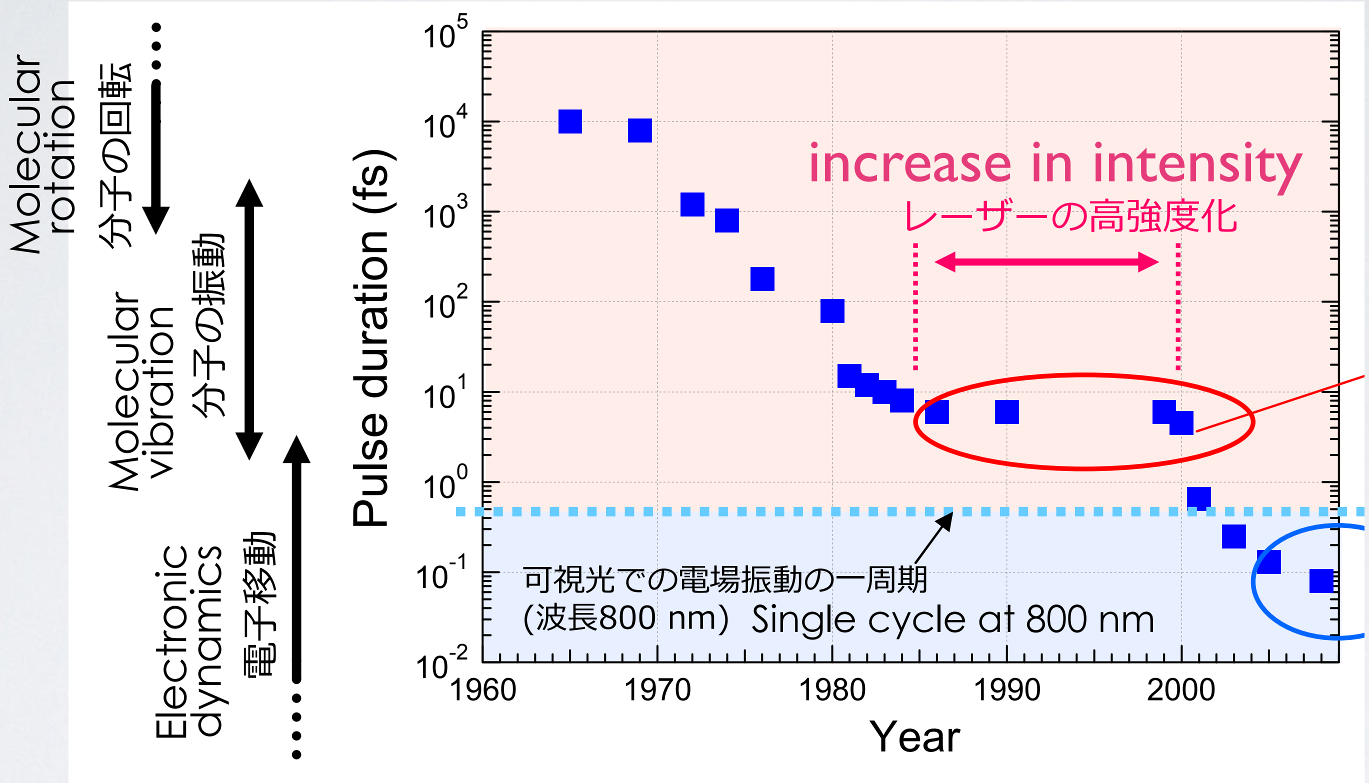


Takahashi et al., Nat. Commun. 4, 2691 (2013)

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



(courtesy of Prof. J. Itatani)

Quest for higher photon energy (shorter wavelength)

cutoff $$E_c = I_p + 3.17U_p$$

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

Longer fundamental wavelength is advantageous

Optical parametric chirped-pulse amplification (OPCPA)

WATER-WINDOW HHG

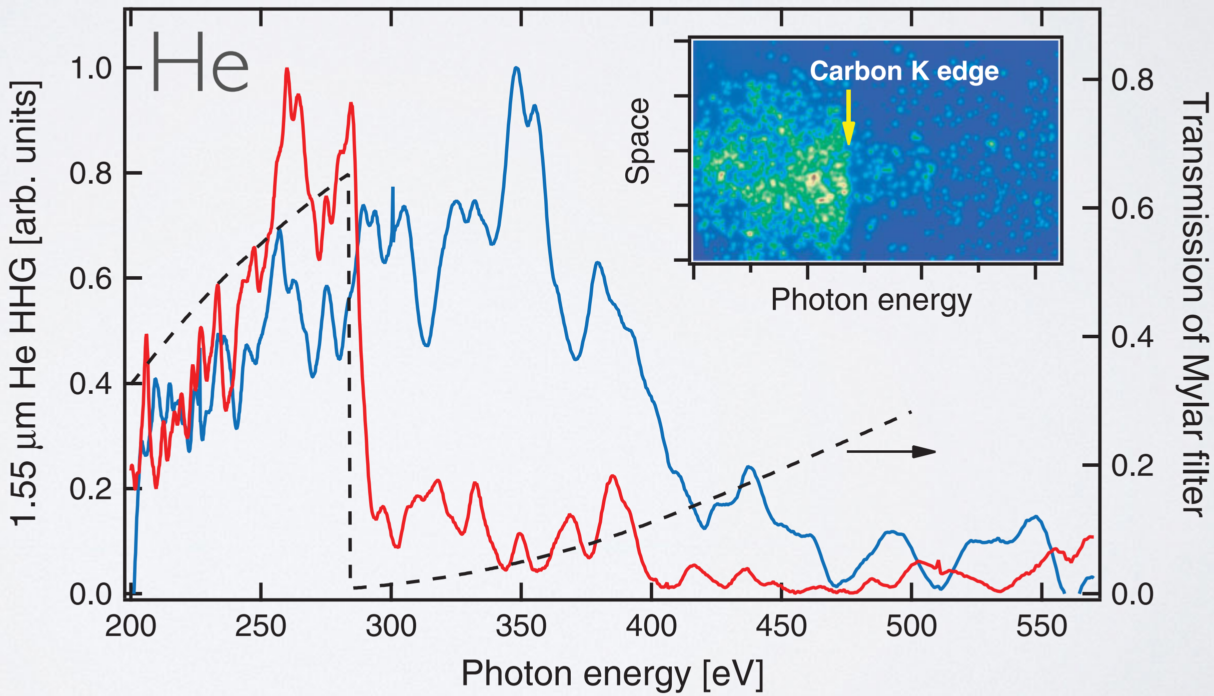
spectral range between the *K*-absorption edges of C (284 eV) and O (543 eV)

➡ absorbed by biological samples but not by water

➡ attractive for high-contrast biological imaging

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

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



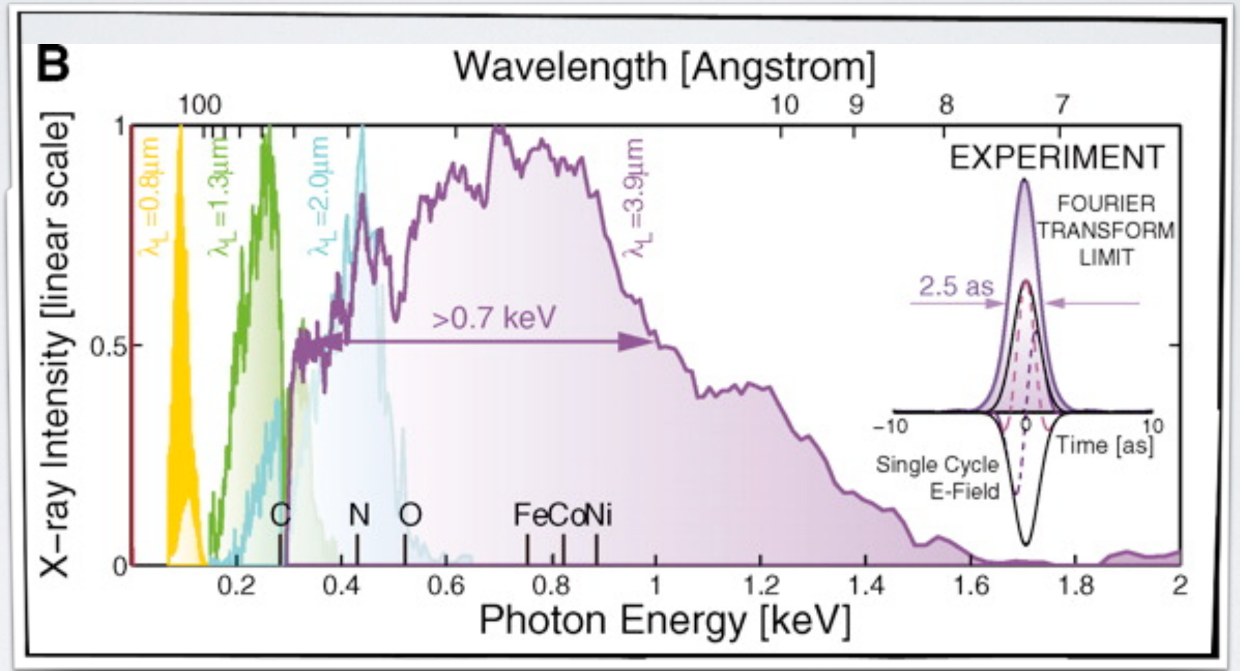
Takahashi et al., PRL 101, 253901 (2008)

keV HHG

Even up to 1.6 keV, > 5000 orders

almost x-ray!

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

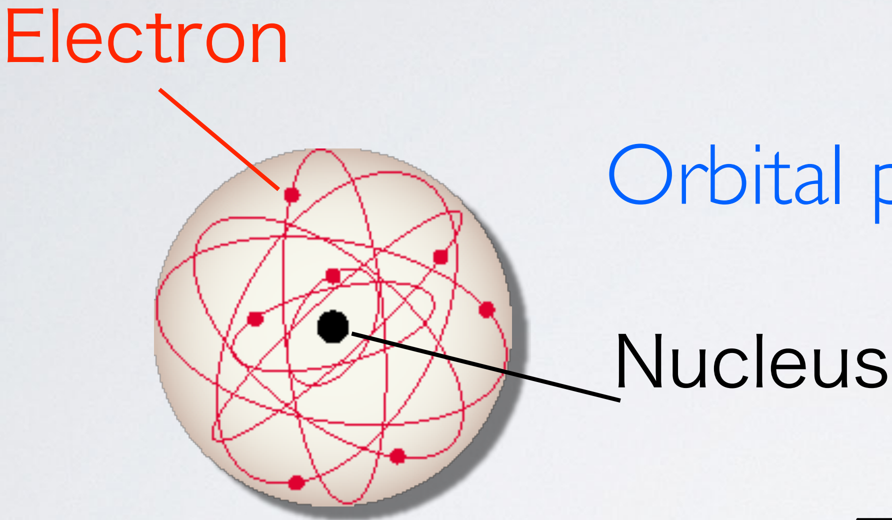


Popmintchev et al., Science 336, 1287 (2012)

a new type of laser-based radiation source

ATTOSECOND SCIENCE

atomic unit of time = 24 attoseconds



Orbital period of the Bohr electron

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

$$\Rightarrow \quad T = \frac{2\pi}{\omega} = 2\pi\sqrt{\frac{4\pi\epsilon_0 m r^3}{e^2}} = 152 \text{ as} = 2\pi \text{ a.u.}$$

real-time observation and time-domain control of **atomic-scale electron dynamics**