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

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強レーザー場中の原子 Atom in an intense laser field

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高強度超短光パルスを発生する方法



Scientific Background on the Nobel Prize in Physics 2018 https://www.nobelprize.org/uploads/2018/10/advanced-physicsprize2018.pdf

— Applications

- Strong-field physics and attosecond science 高強度場物理とアト秒科学
- Laser-plasma acceleration
   レーザープラズマ加速
- High-intensity lasers in industry and medicine
   産業・医療用高強度レーザー

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# How intense is an intense laser field? 強レーザー場とは

# Intensity 強度 10<sup>13</sup>~10<sup>15</sup> W/cm<sup>2</sup>

- Intensity at which the interaction with an atom becomes non-perturbative 原子との相互作用が非摂動論的になり 始める強度。
- \* Effect of laser on the electron ~ Effect of the nucleus
   on the electron
   レーザー場が電子におよぼす影響 ~ 原子核が電子にお
   よぼす影響

# High-field phenomena 高強度場現象

- \* Above-threshold ionization (ATI) 超閾電離
  - \* Ionization upon which an atom absorbs more photons than minimum necessary. 必要以上の光子を吸収してイ オン化する過程
- \* Tunneling ionization トンネル電離
  - \* Ionization by the tunneling effect rather than absorption of photons トンネル効果によるイオン化
- \* High-harmonic generation (HHG) 高次高調波発生
  - \* Generation of harmonics of very high orders 波長変換に よって高次の倍波が発生する現象

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Key concepts キーとなる概念

\* Ponderomotive energy ポンデロモー ティブエネルギー (this week)

■ Quantum paths (trajectories) 量子経
 路 (next week)



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## Single-photon ionization 1光子電離



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### Intensity-dependence of single-photon ionization

### 1光子電離の強度依存性





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## A free electron cannot absorb photons 自由電子は光子を吸えない

inergy conservation  
ニネルギー保存 
$$\frac{p_i^2}{2} + n\hbar\omega = \frac{p_f^2}{2}$$

**Momentum conservation** 

運動量保存

$$\mathbf{p}_i + n\hbar\mathbf{k} = \mathbf{p}_f \qquad \omega = c|\mathbf{k}|$$

\* Solutions exist only for n = 0 → A free electron can neither absorb nor emit photons, because the momentum cannot be conserved 解があるのは、n=0の場合だけ→運動量保存が満たされないた め、自由電子は光子を吸収も放出もできない。

\* Free-free transition possible only near the ion which absorbs the momentum difference 運動量の差を吸収してくれるイオンの近傍

でのみ、free-free遷移が可能

\* Does a rapidly-escaping electron have time to absorb a photon? イオンから逃げていく電子が、光子を吸う暇があるのか?



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wavelength

波長 1064 nm

Xe gas

# Experiments with higher intensity より高強度の実験



FIG. 3. Electron spectra from multiphoton ionization of xenon at 1064-nm. The vertical scales are normalized. The pulse energy F and pressure at which each spectrum is taken is given in the figure. In the spectrum at 0.004 Pa, the background has been subtracted. The estimated intensity is  $F(mJ) \times 2.10^{12}$ W/cm<sup>2</sup>.

Kruit et al., Phys. Rev. A 28, 248 (1983) Group of FOM (Amsterdam)のグループ

Minimum  $E_{kin} = (N+S)\hbar\omega - I_p$ 最小限必要な光子数 余分の光



FIG. 6. Electron-energy spectrum corresponding to absorption of photons above the ionization threshold. Zero energy corresponds to a free electron at rest for either of the two ion-core configurations. The negative energy states represent the ionization energies for the two core configurations and they are thus inverted relative to the normal ion spectrum.

MacIlrath et al., Phys. Rev. A 35, 4611 (1987) Group of AT&T Bell Lab.のグループ

余分の光子数 Extra photons

\* Now certain that ATI is due to free-free transition ATIは、free-free遷移による光子吸収であることが確実に

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FIG. 3. Electron spectra from multiphoton ionization of xenon at 1064-mm. The vertical scales are normalized. The pulse energy F and pressure at which each spectrum is taken is given in the figure. In the spectrum at 0.004 Pa, the background has been subtracted. The estimated intensity is  $F(mJ) \times 2.10^{12}$ W/cm<sup>2</sup>.

Kruit et al., Phys. Rev. A 28, 248 (1983) FOM (アムステルダム)のグループ

At high intensity 高強度では



FIG. 7. Xenon photoelectron spectra for 1064-nm light. The polarization is linear, oriented along the detection axis. The rescale factor at right may be used to obtain the relative rates per unit xenon density.

MacIIrath et al., Phys. Rev. A 35, 4611 (1987) AT&Tベル研のグループ

- \* Comparable peak heights → non-perturbative 吸収光子数によらず、ピークの高さが同程度→非摂動論的
- \* 低次の吸収ピークが消える (peak suppression at low orders)

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# High-order perturbation theory 高次の摂動論

$$i\hbar\frac{\partial\psi}{\partial t} = (H_0 + H_I)\psi$$

$$H_{I} = -\left(e\sum_{i=1}^{n}\mathbf{r}_{i}\right) \cdot \mathbf{E}(t) \quad \text{or $\mathbf{\sharp}$tick} \quad H_{I} = -\frac{e}{m}\left(\sum_{i=1}^{n}\mathbf{p}_{i}\right) \cdot \mathbf{A}(t) + \frac{ne^{2}}{2m}\mathbf{A}^{2}(t)$$
LENGTH FORM
VELOCITY FORM

cross section  
断面積 
$$\sigma_N = \frac{2\pi}{\hbar} \left(\frac{2e^2\hbar\omega}{\epsilon_0 c}\right)^N \sum_f \left|M_{i\to f}^{(N)}\right|^2$$
 unit  
単位 cm<sup>2N</sup>s<sup>N-1</sup>

$$M_{i \to f}^{(N)} = \sum_{j', j'', \cdots, j'''} \frac{\langle i|x|j'\rangle \langle j'|x|j''\rangle \cdots \langle j'''|x|f\rangle}{(E_i + \hbar\omega - E_{j'})(E_i + 2\hbar\omega - E_{j''}) \cdots (E_i + (N-1)\hbar\omega - E_{j'''})}$$

# (N+S)-photon ionization cross section of a hydrogen atom 水素原子の(N+S)光子電離の断面積 (cm<sup>2(N+S)</sup>/W<sup>N+S</sup>/S)

Gontier and Trahin, J. Phys. B 13, 4383 (1980)

	最小限必要な光子数 N				
余分の光子数 S	6 (530 nm)	8 (650 nm)	10 (910 nm)	12 (1082 nm)	
0	1.39×10-69	1.49×10-97	4.51×10-123	3.46×10-149	
1	2.84×10 <sup>-83</sup>	9.85×10-111	7.78×10 <sup>-136</sup>	9.81×10-162	
2	2.92×10-97	2.53×10-124	5.35×10 <sup>-149</sup>	1.10×10-174	
3	2.80×10-111	5.84×10-138	2.61×10-162	1.08×10-187	
4	2.66×10-125	1.35×10-151	1.89×10-175	9.87×10-201	
5	2.32×10-139	2.75×10-165	1.04×10 <sup>-188</sup>	8.91×10-214	
S=0と1が同じに					

S=0と1が向した なる強度 (W/cm<sup>2</sup>)  $\rightarrow$  4.89×10<sup>13</sup>

1.51×10<sup>13</sup>

3.53×10<sup>12</sup>

Equal cross section for S=0 and 2

Intensity at which the interaction becomes non-perturbative 非摂動論的になる強度の目安

 $5.80 \times 10^{12}$ 

longer wavelength → lower intensity 長波長ほど低強度



**Consistent with experiments** 





### **Charged particle in an electromagnetic wave** 雷磁波中の荷電粒子 $\mathbf{E}(\mathbf{r},t) = \frac{1}{2} [\mathbf{E}_0(\mathbf{r},t)e^{-i\omega t} + \text{c.c.}] = |\mathbf{E}_0|\cos(\omega t + \varphi)$ $$\begin{split} \mathbf{B}(\mathbf{r},t) &= \frac{1}{2} [\mathbf{B}_0(\mathbf{r},t)e^{-i\omega t} + \mathrm{c.c.}] = |\mathbf{B}_0|\cos(\omega t + \varphi) \\ \mathbf{r}(t) &= \mathbf{R}(t) + \delta \mathbf{r}(t) \end{split}$$ Microscopic oscillation $\delta \mathbf{r}(t) = \delta \mathbf{r}_0 e^{-i\omega t} + \text{c.c.}$ ミクロな振動運動(振動数ω) $|\delta \mathbf{r}_0 \cdot abla \mathbf{E}_0| \ll |\mathbf{E}_0|$ Macroscopic drift motion マクロなドリフト運動 R(t) $\delta r(t)$ N



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 $|\delta \mathbf{r}_0 \cdot 
abla \mathbf{E}_0| \ll |\mathbf{E}_0|$ 

 $|\delta \mathbf{r}_0 \cdot 
abla \mathbf{B}_0| \ll |\mathbf{B}_0|$ 

$$\mathbf{r}(t) = \mathbf{R}(t) + \delta \mathbf{r}(t)$$
  $\delta \mathbf{r}(t) = \delta \mathbf{r}_0 e^{-i\omega t} + c.c$ 

 $E_0$ ,  $B_0$  rarely change in the scale of  $\delta r_0$  $\delta r_0$ のスケールでは、 $E_0$ ,  $B_0$ はほとんど変わらない。

> $\mathbf{v}(t) = \mathbf{V}(t) + \delta \mathbf{v}(t) \qquad \delta \mathbf{v}(t) = \delta \mathbf{v}_0 e^{-i\omega t} + \text{c.c.}$ or is small but  $\delta \mathbf{v}$  is NOT necessarily small.

Non-relativistic electron velocity 電子の速度は非相対論的

 $\mathbf{V} \times \mathbf{B}_0 \ll \mathbf{E}_0$ 



ンシャル (エネルギー)

# Force acting on the charged particle 荷電粒子に作用する力

$$\begin{split} \mathbf{F} &= q[\mathbf{E}(\mathbf{r}(t),t) + \mathbf{v}(t) \times \mathbf{B}(\mathbf{r}(t),t)] \\ &= q[\mathbf{E}(\mathbf{R} + \delta \mathbf{r},t) + (\mathbf{V} + \delta \mathbf{v}) \times \mathbf{B}(\mathbf{R} + \delta \mathbf{r},t)] \\ &\approx q[\mathbf{E}(\mathbf{R},t) + \delta \mathbf{r} \cdot \nabla \mathbf{E}(\mathbf{R},t) + \mathbf{V} \times \mathbf{B}(\mathbf{R},t) + \delta \mathbf{v} \times \mathbf{B}(\mathbf{R},t)] \\ \text{ime average over many cycles} \implies \text{terms with } e^{\pm i\omega t}, e^{\pm 2i\omega t} \text{ vanish} \\ \mathbf{F} &\approx \frac{q}{2} \left( \delta \mathbf{r}_0^* \cdot \nabla \mathbf{E}_0 + \delta \mathbf{v}_0^* \times \mathbf{B}_0 + \text{c.c.} \right) \\ &= -\frac{q^2}{4m\omega^2} \left[ \mathbf{E}_0 \cdot \nabla \mathbf{E}_0^* + \mathbf{E}_0 \times (\nabla \times \mathbf{E}_0^*) + \text{c.c.} \right] = -\frac{q^2}{4m\omega^2} \nabla |\mathbf{E}_0|^2 \\ \mathbf{F} &= -\nabla U_p(\mathbf{R},t) \qquad U_p(\mathbf{R},t) = \frac{q^2 |\mathbf{E}_0(\mathbf{R},t)|^2}{4m\omega^2} \\ \end{aligned}$$

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## **Ponderomotive force** ポンデロ

$$\mathbf{F} = -\nabla U_p(\mathbf{R}, t) \qquad \qquad U_p(\mathbf{R}, t) = \frac{q^2 |\mathbf{E}_0(\mathbf{R}, t)|^2}{4m\omega^2}$$

(動重力) NDEROMOTIVE POTENT (ENERGY) ポンデロモーティブポテ ンシャル(エネルギー)

- \* Potential force ポテンシャルカ
- \* Proportional to the laser intensity 電磁波の強度に比例
- Independent of the sign of charge (from the beam axis to outside) 電荷の正負によらず向きが同じ(ビームの中心から外へ)
- \* Higher energy for lighter particles (larger effect for electrons than for nuclei and ions) 軽い粒子ほど大きなエネルギー
- \* A charged particle in a laser field has an energy of  $U_p$  by default. 荷電粒子は、レーザー場中にただいるだけでUpのエ ネルギーを持っている。



### ミクロな視点からみた Ponderomotive energy from a microscopic view point

Motion of a charge particle (mass m, charge q) in an oscillating electric field 振動電界中の質量*m*, 電荷qの荷電粒子の運動

 $E(t) = E_0 \sin \omega t$ 

 $m\dot{v} = qE_0\sin\omega t$ 

 $v = -\frac{qE_0}{m\omega}\cos\omega t + \text{drift}$ 並進運動

Energy of quiver motion (jitter motion)のエネルギー



$$\frac{1}{2}mv^{2} = \frac{q^{2}E_{0}^{2}}{2m\omega^{2}}\cos^{2}\omega t$$
 Time average  $\left\langle \frac{1}{2}mv^{2} \right\rangle = \frac{q^{2}E_{0}^{2}}{4m\omega^{2}} = U_{p}$   
For an electron 電子の場合  
 $U_{p}(\text{eV}) = \frac{e^{2}E_{0}^{2}}{4m\omega^{2}} = 9.337 \times 10^{-14}I(\text{W/cm}^{2})\lambda^{2}(\mu\text{m})$ 

\* A charged particle in a laser field has an energy of  $U_p$  by default.

\* 電子(荷電粒子)は、レーザー場中にただいるだけで*U*pのエネルギーを 持っている。

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### **Effective ionization potential** = $I_p + U_p$

実効的なイオン化ポテンシャルがIp+Upになる。





FIG. 7. Xenon photoelectron spectra for 1064-nm light. The polarization is linear, oriented along the detection axis. The rescale factor at right may be used to obtain the relative rates per unit xenon density.

Number of photons necessary for ionization イオン化に必要な光子数

 $n\hbar\omega \ge I_p + U_p$ 

Observed electron energy 観測される電子のエネルギー

$$E_{\rm kin} = [n\hbar\omega - (I_p + U_p)] + U_p = n\hbar\omega - I_p$$





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Bound electrons 束縛電子の場合 Quantum mechanically, AC-stark effect 量子力学的には:ACシュタルクシフトに対応

2nd-order perturbation  
theory 摂動論から  

$$\Delta E = \frac{e^2 E_0^2}{4} \sum_n \frac{2\omega_{ni} |\mu_{in}|^2}{\omega^2 - \omega_{ni}^2} = -\frac{1}{4} \alpha(\omega) E_0^2$$
  
Electric dipole polarizability  
Lorentz oscillator model  
 $m\ddot{x} = -eE_0 \cos \omega t - m\omega_0^2 x$  電気双極子分極率  
 $\alpha = -\frac{e^2}{m(\omega^2 - \omega_0^2)}$   
 $\Delta E = \frac{e^2 E_0^2}{4m(\omega^2 - \omega_0^2)}$   
\* Negative for the ground state  
基底状態では負→dipole trap  
 $\Delta E_g \approx -\frac{e^2 E_0^2}{4m\omega_0^2} \propto -I$   
 $\omega_0 \gg \omega$   
\* Positive for Rydberg atoms and free electrons  $\Im_{2} - F$   
 $\ll U_p = \frac{e^2 E_0^2}{4m\omega^2} \propto I$   
 $U_p \gg |\Delta E_g|$ 

1 m t. 3

# A measure of non-perturbativeness 非摂動論的であることのめやす

### PEAK SUPPRESSION 低次のピークが消える

$U_p \sim \hbar \omega$ $E_0^2 \sim \frac{4m\omega}{e^2}$					
	530 nm	650 nm	910 nm	1082 nm	
Gontier and Trahin	4.89×10 <sup>13</sup>	1.51×10 <sup>13</sup>	5.80×10 <sup>12</sup>	3.53×10 <sup>12</sup>	
$U_p \sim \hbar \omega$	8.9×10 <sup>13</sup>	4.8×10 <sup>13</sup>	1.8×10 <sup>13</sup>	1.0×10 <sup>13</sup>	

\* Order of magnitude and trend consistent オーダーと波長依存性がよく合っている。



wavelength

1064 nm

Xe gas

# **Above-threshold ionization (ATI)**

### roughly at 10<sup>13</sup>~10<sup>14</sup> W/cm<sup>2</sup> intensity in the near-infrared (NIR)



FIG. 3. Electron spectra from multiphoton ionization of xenon at 1064-nm. The vertical scales are normalized. The pulse energy F and pressure at which each spectrum is taken is given in the figure. In the spectrum at 0.004 Pa, the background has been subtracted. The estimated intensity is  $F(mJ) \times 2.10^{12}$ W/cm<sup>2</sup>.

Kruit et al., Phys. Rev. A 28, 248 (1983) Group of FOM (Amsterdam)



FIG. 6. Electron-energy spectrum corresponding to absorption of photons above the ionization threshold. Zero energy corresponds to a free electron at rest for either of the two ion-core configurations. The negative energy states represent the ionization energies for the two core configurations and they are thus inverted relative to the normal ion spectrum.

MacIIrath et al., Phys. Rev. A 35, 4611 (1987) Group of AT&T Bell Lab.



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### Tunneling ionization トンネル電離(トンネルイオン化)



### Conditions of tunneling ionization トンネル電離の条件

**Tunneling rate W is high enough** トンネル確率が十分大きい

$$W \sim I_p \exp\left(-\frac{4\sqrt{2}}{3}\frac{I_p^{3/2}}{E}\right)$$

Field should be sufficiently strong



 $\gamma = 1$ 

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# Keldysh parameter

### **Keldysh parameter**

$$\gamma = \sqrt{\frac{I_p}{2U_p}}$$

 $\gamma>1$ :Multi-photon regime 多光子領域  $\gamma\lesssim 1$ :Tunneling regime トンネル領域



Xe (l<sub>p</sub>=12.13 eV), wavelength1064nm, about  $5.7 \times 10^{13}$  W/cm<sup>2</sup>

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### トンネルイオン化の条件 Conditions of tunneling ionization

**Tunneling rate W is high enough** トンネル確率が十分大きい



Don't forget this!

 $W \sim I_p \exp\left(-\frac{4\sqrt{2}}{3} \frac{I_p^{3/2}}{E}\right)$  Field should be sufficiently strong

**Field oscillation is slow enough** レーザー電場の振動が十分遅い

 $\gamma = \sqrt{rac{I_p}{2U_p}}$   $\gamma \lesssim 1$ : Tunneling regime  $\gamma > 1$ : Multi-photon regime 多光子領域

### **Typical misunderstanding**

terahertz radiation with I THz frequency and 2 MV/cm field strength

 $U_p = 44 \text{ eV}$  tunneling ionization? **NO**!

only 5×109 W/cm<sup>2</sup> too weak for tunneling

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# Change of ionization mechanism with laser intensity レーザー強度によるイオン化の変化



### Chat your student ID number and full name.

# Change of ionization mechanism with laser intensity レーザー強度によるイオン化の変化



# Appendix

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 $\gamma = 1$ 

Xe (Ip=12.13 eV), 波長1064nmで、5.7×1013 W/cm2程度

# なぜ、トンネル電 離でも光電子スペ クトルは離散的な のか?



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電子経路の量子力学的干渉 Volkov波動関数 個々の経路 i の持つ位相  $\exp\left(\frac{iS(t_r^{(i)})}{\hbar}\right)$  $\Psi_V(z,t) = \exp\left[i(k+eA(t))z - \frac{iS(t)}{\hbar}\right]$  $S(t) = \int dt L = \int_{-\infty}^{t} dt' \left[ \frac{(k + eA(t'))^2}{2m} + I_p \right]$ 作用(action)  $= 2U_p \left[ \left( 1 + \frac{\cos(2\omega t)}{2} \right) t - \frac{3}{4\omega} \sin(2\omega t) \right] + I_p t$  $P(k) \propto \left| \sum_{r} \exp\left(\frac{iS(t_r^{(i)})}{\hbar}\right) \right|$ 光電子の運動量分布 Electric field — Vector potential Unit cell  $\propto \cos^2\left(\frac{\Delta S}{2}\right) \left\|\sum_{r} \exp\left(\frac{iS(t_r^{(1j)})}{\hbar}\right)\right\|$ i=2i = 3i=1Vector potential A(t) Electric field F(t) サイクル内干渉 サイクル間干渉  $\Delta S = S(t_r^{(2,1)}) - S(t_r^{(1,1)})$  $0 t_r^{(1,1)} t_r^{(2,1)}$ 1 2 3 Time (optical cycle) Intracycle interference

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$$E_{\rm kin} = n\hbar\omega - I_p - U_p$$



\* 光電子スペクトルの離散的な ピーク \*電子経路のサイクル間干渉 による \*トンネル電離が、レーザー 場の周期で起こるため \* ポンデロモーティブシフト が自然に出てくる

まとめ

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# 強度 10<sup>13</sup>~10<sup>15</sup> W/cm<sup>2</sup>のレーザー場中のイオン化

- \* 超閾電離(Above-threshold ionization, ATI)
  - \* 必要以上の光子を吸収してイオン化する過程(光 子の観点)
- \*トンネル電離
  - \*トンネル効果によるイオン化(電磁波の観点)
- \* 光電子スペクトルは離散的なピークからなる
   \* free-free遷移による光子の吸収(原子物理の観点)
  - \*トンネル電離で周期的に出てくる電子の干渉

