高強度赤外光源によるアト秒科学と将来展望

東京大学　物性研究所
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マイクロ固体フォトニクス研究会
第7回レーザー学会「小型集積レーザー」専門委員会
第7回科学技術交流財団「ジャイアント・マイクロフォトニクス研究会」
2022年12月22日

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(Kennesaw State Univ.)

KENNESAW STATE UNIVERSITY

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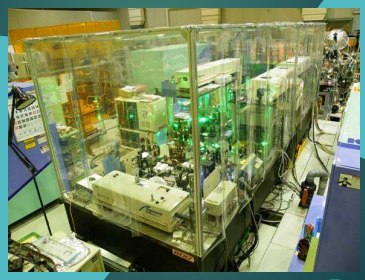
高強度極短パルスレーザーと高強度場物理学



Ti:sapphire CPA laser

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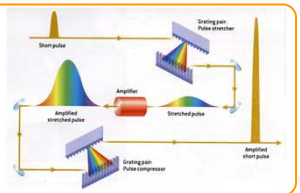
高強度超短パルスレーザーの誕生 (1985-1994)

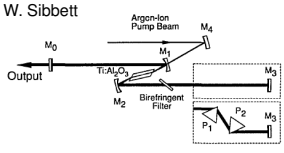
チャープパルス増幅法 CPA (1985)
D. Strickland and G. A. Mourou

2018 Physics



チタンサファイアレーザー (1986)
P. Moulton

カーレンズモード同期法 (1991)
W. Sibbett



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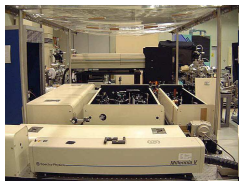
テーブルトップTWレーザー (1985-1994)

Strong field physics has become available at university-scale labs.

Nova @ LLNL (1980's)

Pulse energy	4kJ
Duration	4ns
Peak power	1 TW
Repetition	1 /hours
Wavelength	1054 nm



10-Hz TiS @ ISSP (1997)

Pulse energy	0.48 J
Duration	22 fs
Peak power	22 TW
Repetition	10 Hz
Wavelength	800 nm

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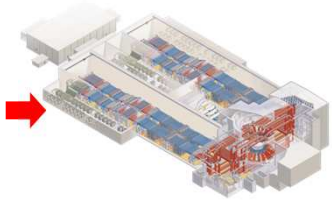
レーザー核融合を目指したペタワットレーザー (2014)

Innertial confinement fusion (ICF, レーザー核融合) still continues.



Nova @ LLNL (1980's)		National Ignition Facility @ LLNL (2014)	
Pulse energy	4kJ	Pulse energy	4.1 MJ
Duration	4ns	Duration	20 ns
Peak power	1 TW	Peak power	200 TW
Repetition	1 /hours	Repetition	1 /hours
Wavelength	1054 nm	Wavelength	350 nm

ブレークイーブン達成 (2022)

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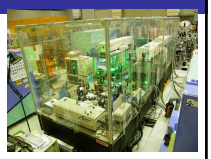
高強度場物理学 – エネルギの時間･空間的集中

$$\text{Peak power [W]} = \frac{\text{Pulse energy [J]}}{\text{Duration [sec]}}$$

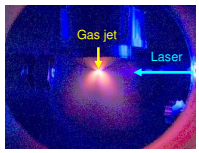
1 TW = 10^{12} W, 20 mJ, 20 fs



Cf. 全世界で連続的に作られている電力 ~ 2 TW.

$$\text{Intensity [W/cm}^2\text{]} = \frac{\text{Peak power [W]}}{\text{Spot area [cm}^2\text{]}}$$

10^{15} W/cm^2, 1TW (=10^{12} W), ϕ0.3mm



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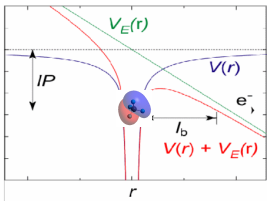
高強度場物理学を特徴付ける物理量

Distorted Coulomb potential by optical E field



振動電場中の自由電子
ポンデロモーティブポテンシャル Up

$$U_p = \frac{e^2E^2}{4m\omega^2} \propto I\lambda^2$$

~ 60 eV @ 10^{15} W/cm^2, 800 nm

束縛電子
イオン化ポテンシャル I_p

IP ~ 10..20 eV (neutral atoms/molecules)

- レーザーの光電場がクーロンポテンシャルを変形させる。
- Up と Ip が同程度になると、トンネルイオン化が起こる。
- 放出された光電子は、振動エネルギー Up(=10-100 eV)をもつ。

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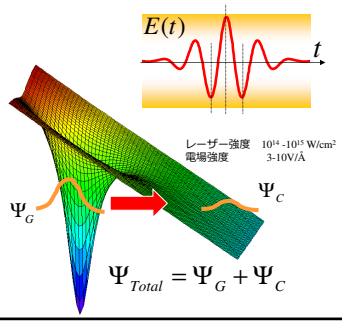
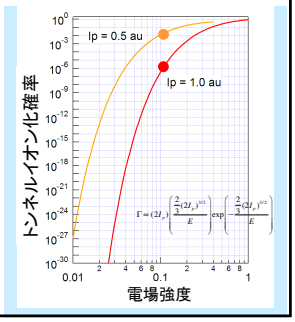
トンネルイオン化における極端な非線形性

- トンネルイオン化は電場の瞬間強度のピークで起こる(サブサイクル過程)
- 最も束縛の弱い外殻電子が放出される
- 電子の波動関数をコヒーレントに分割する(束縛状態＋非束縛状態)





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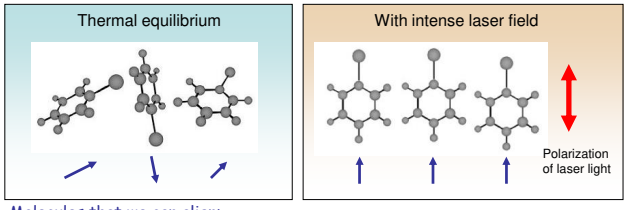
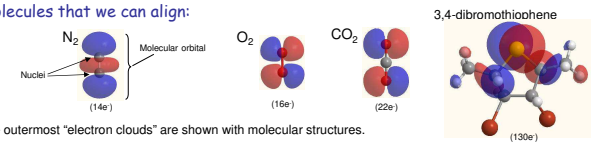
高強度光電場下の現象(1)
分子配列・分子配向

At 10^{13} W/cm^2, we can align some molecules.



Molecules that we can align:



The outermost "electron clouds" are shown with molecular structures.

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高強度光電場下の現象 (2)
多光子イオン化とトンネルイオン化

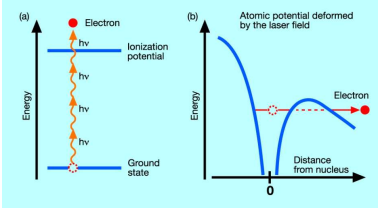
同時に相互作用しうる光子数

$$\rho = \frac{I\lambda^3}{(\hbar\omega)c} \approx 2\times10^{10}\ \text{ph / cubic wavelength}$$

[@ 10^{14} W/cm^2, 1.06 μm]

Keldysh parameter:

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



多光子イオン化
光子性
$\gamma \gg 1$

トンネルイオン化
波動性
$\gamma < 1$

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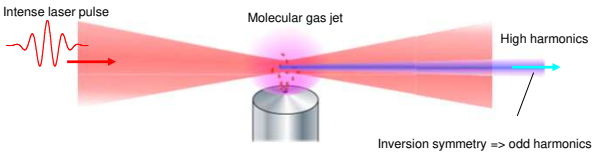
高強度光電場下の現象 (3)
高次高調波発生

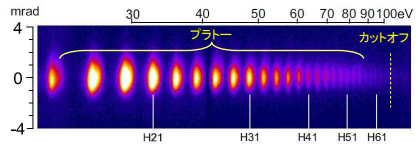
At $10^{14\text{-}15}$ W/cm^2, we can produce attosecond soft X-ray pulses.



Inversion symmetry => odd harmonics

$$\hbar\omega_X = (2m+1)\hbar\omega_L$$

Spatially-resolved spectrum of high harmonics (false color)



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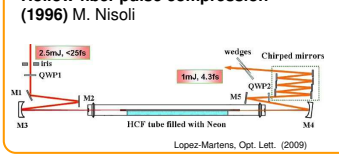
高強度極短パルスレーザー (1996-2000)
波形制御された光パルス

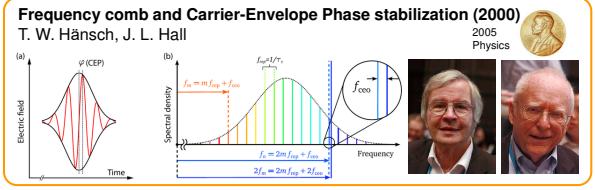
Hollow-fiber pulse compression
(1996) M. Nisoli



Lopez-Martens, Opt. Lett. (2009)

Frequency comb and Carrier-Envelope Phase stabilization (2000)
T. W. Hänsch, J. L. Hall

2005 Physics



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高強度極短パルスレーザーのエネルギー効率

Average power
Pav=100 W

LD励起固体レーザー
Nd:YLF(1 kHz)
Nd:YAG(10 kHz)
Nd:YVO_4(100 kHz)
100 ns

Pump 527/532 nm

Pav=20 W
Thermal limit

チャープパルス増幅
チタンサファイアレーザー
30 fs

- 10-20 mJ, 30 fs @ 1 kHz
- Wavelength 800 nm
- Carrier-envelope phase stable

Pav=1 W
Waveguide
(Unscalable in enegy)

非線形
- Gas-filled hollow-core fiber
- Dispression compensation mirrors

5 fs

Waveform-controlled few-cycle pulses

Near IR
800 nm

- 0.5-1 mJ, 5 fs @ 1 kHz
- Wavelength 700-900 nm
- Carrer-envelope phase stable (active feedback)

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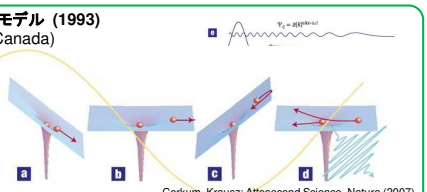
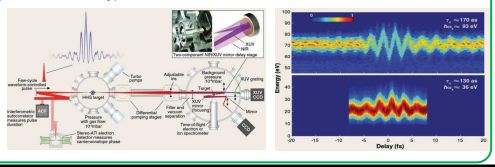
アト秒科学の誕生

高次高調波発生の3段階モデル (1993)
Paul B. Corkum (NRC, Canada)



Corkum, Krausz: Attosecond Science, Nature (2007)

EUVアト秒パルス発生 (2001)
Ferenc Krausz (MPQ, Germany)



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Summary

1990 • **Key technologies of intense USP lasers were invented in 1980's.**

2000 • **Ti:sapphire laser technology was matured.**
• **Attosecond Science emerged.**

2010 Next topic

Energy upshift from EUV to Soft X rays
- Attosecond science was established by Ti:sapphire lasers.
- New light sources in IR and MIR need to be developed.

2020

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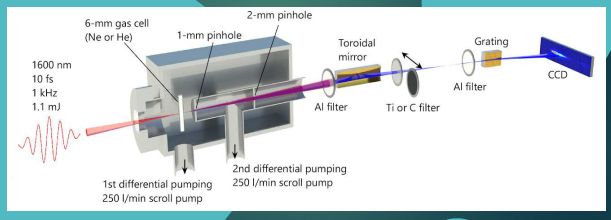
赤外領域の高強度極短パルス光源と軟X線アト秒分光



Generation of attosecond soft X-ray pulses

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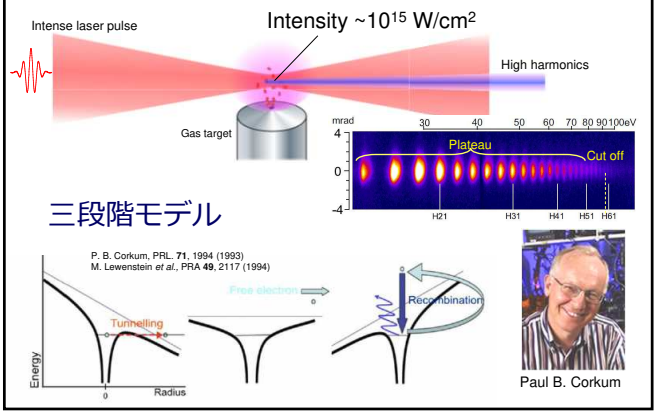
高次高調波発生 (HHG)



三段階モデル

Paul B. Corkum

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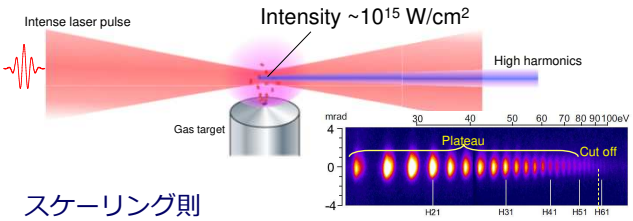
高次高調波発生 (HHG)



スケーリング則

- 最大光子エネルギー $\hbar\omega_{max} \propto U_P \propto I\lambda^2$
- 変換効率 $\eta \propto \lambda^{-5.5}$

軟X線アト秒科学のためには、長波長かつ高出力の光源が必要。

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keV HHG by Colorado-Wien group (2012)

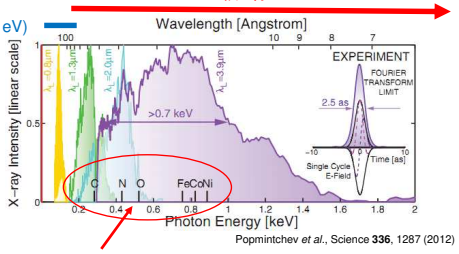
Can we produce isolated attosecond pulses?

Can we construct attosecond spectroscopy?

3.9 μm (MIR)
Multi-cycle, CEP unstable pulses
Low rep rate (10Hz)

極端紫外線
(EUV, <200 eV)

軟X線



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

元素吸収端 => 元素選択性のある超高速分光 (NEXAFS, EXAFS)
軽元素 (生命科学･化学): 200-500 eV
遷移金属 (物質科学): 500-800 eV

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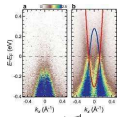
軟X線を用いた分光･イメージング
電子状態を探る「最良の手段」

光電子分光
band structures, electron in k-space

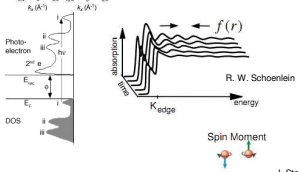


Okazaki, Shin

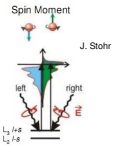
吸収分光

Near edge (XANES)
local electronic structure, bonding geometry, oxidation state

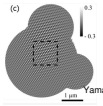
Extend fine structure (EXAFS)
local atomic structure and coordination



磁気二色性の利用
spin, magnetization



回折･イメージング
long-range orbital/magnetic order



The University of Tokyo
The Institute for Solid State Physics

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Outline

- **赤外領域の高強度極短パルス光源**
- レーザー軟X線過渡吸収分光

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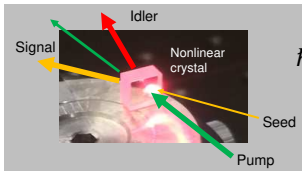
光パラメトリック増幅(OPA)による超広帯域増幅

Optical Parametric Amplifier (OPA)



$$\hbar\omega_{pump} = \hbar\omega_{signal} + \hbar\omega_{idler}$$
$$\boldsymbol{k}_{pump} = \boldsymbol{k}_{signal} + \boldsymbol{k}_{idler}$$

Ultra-broadband OPA in the *degenerate* condition

Phase mismatch $\boldsymbol{\Delta k} = \boldsymbol{k}_{signal} + \boldsymbol{k}_{idler} - \boldsymbol{k}_{pump}$

Degenerate OPA => Odd terms = 0

$$\Delta k = \Delta k_0 + \left(\frac{\partial k_s}{\partial \omega_s} - \frac{\partial k_i}{\partial \omega_i}\right)\Delta\omega + \frac{1}{2!}\left(\frac{\partial^2 k_s}{\partial \omega_s^2} + \frac{\partial^2 k_i}{\partial \omega_i^2}\right)(\Delta\omega)^2 + \frac{1}{3!}\left(\frac{\partial^3 k_s}{\partial \omega_s^3} - \frac{\partial^3 k_i}{\partial \omega_i^3}\right)(\Delta\omega)^3 + \frac{1}{4!}\left(\frac{\partial^4 k_s}{\partial \omega_s^4} + \frac{\partial^4 k_i}{\partial \omega_i^4}\right)(\Delta\omega)^4 + \ldots$$
$$\simeq 2\frac{1}{4!}\frac{\partial^4 k_s}{\partial \omega_s^4}(\Delta\omega)^4 + 2\frac{1}{6!}\frac{\partial^6 k_s}{\partial \omega_s^6}(\Delta\omega)^6 + \ldots$$

Zero GDD => The 2nd order term = 0
ex. BBO (zero GDD at 1450 nm, pump 725 nm)
BIBO (zero GDD at 1580 nm, pump 790 nm)

BBO: β-BaB_2O_4
BIBO: BiB_3O_6

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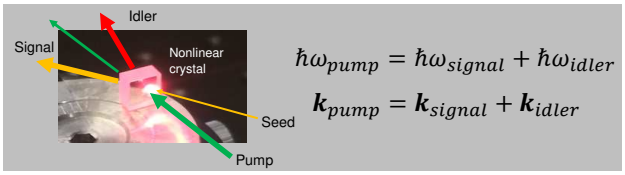
光パラメトリック増幅(OPA)による超広帯域増幅

Optical Parametric Amplifier (OPA)



$$\hbar\omega_{pump} = \hbar\omega_{signal} + \hbar\omega_{idler}$$

$$\boldsymbol{k}_{pump} = \boldsymbol{k}_{signal} + \boldsymbol{k}_{idler}$$

Ultra-broadband OPA in the *degenerate* condition

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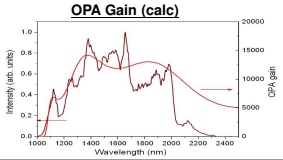
Zero GDD => The 2nd order term = 0

ex. BBO (zero GDD at 1450 nm, pump 725 nm)
BIBO (zero GDD at 1580 nm, pump 790 nm)

OPA Gain (calc)



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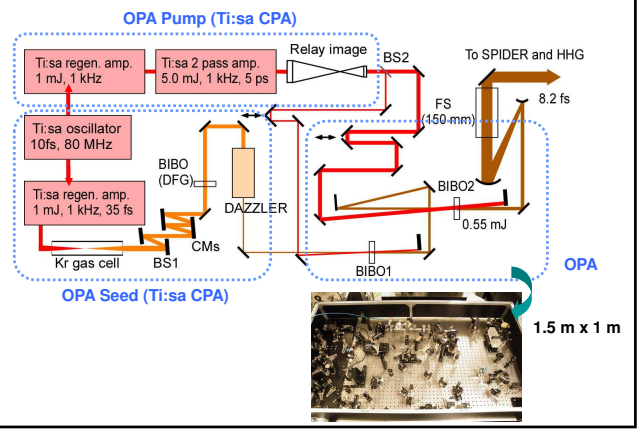
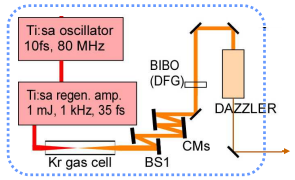
位相安定な高強度赤外OPCPA光源



26

位相安定な赤外シードパルス発生

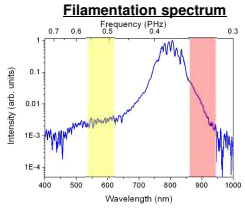


TYPE 2 in BIBO

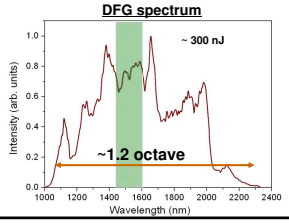
570 nm (e) – 900 nm (e) = 1555 nm (o)

CEP passively stabilized

Filamentation spectrum



DFG spectrum



27

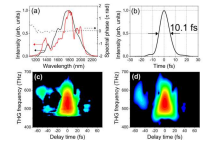
高強度赤外OPCPA光源の出力

Output

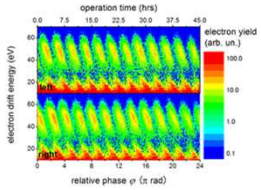
Wavelength	1.6 μm
Pulse energy	1.5 mJ
Repetition rate	1 kHz
Pulse duration	10 fs
CEP	Passively stable Controlled by AOPDF

Ishii *et al*, Opt. Lett. **37**, 4182 (2012)
Ishii *et al*, J. Opt. **20**, 014003 (2018)

FROG measurement of 10.1-fs pulse

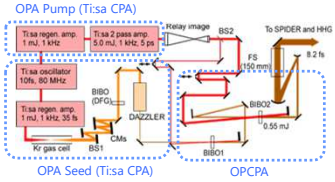


CEP-scanned stereo ATI for 45 hours



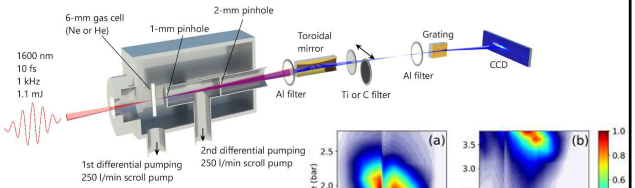
H. Geiseler *et al*, Appl. Phys. B 117, 941 (2014)

Setup of BIBO-OPCPA (original setup)

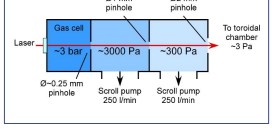


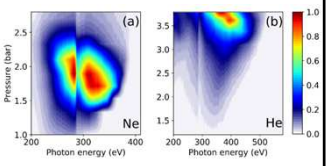
28

軟X線アト秒パルス発生 (1)



Differential pumping scheme





	Optimum pressure	Cutoff
Ne	1.9 bar	380 eV
He	3.6 bar	520 eV

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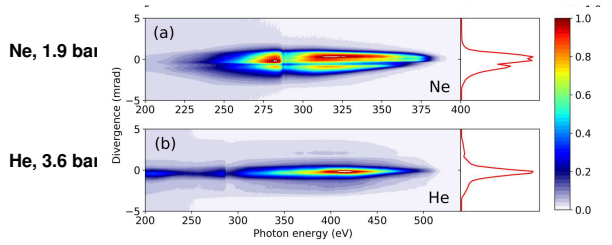
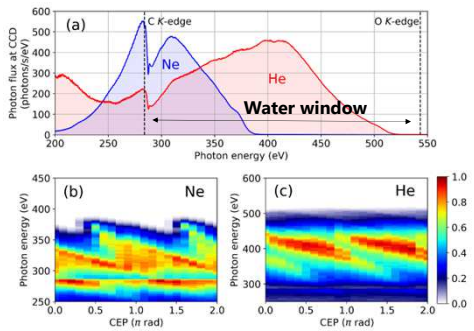
軟X線スペクトルとビーム品質



	Optimum pressure	Cutoff	Divergence (FWHM)
Ne	1.9 bar	380 eV	2.0 mrad
He	3.6 bar	520 eV	0.9 mrad

30

「水の窓」領域での軟X線アト秒パルス発生



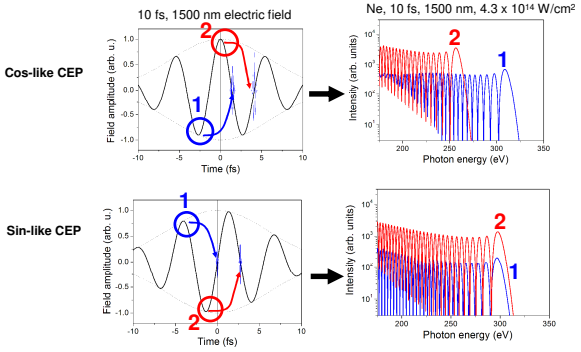
✓ Clear CEP dependence
→ Indication of isolated attosecond pulse generation

31

位相敏感なカットオフ構造と孤立アト秒パルス発生

Haworth *et al.*, Nature Phys. **3**, 52 (2007)

SFA-based HHG simulation
Ne, 10 fs, 1500 nm, 4.3 x 10^{14} W/cm^2



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Outline

- 赤外領域の高強度極短パルス光源
- レーザー軟X線過渡吸収分光

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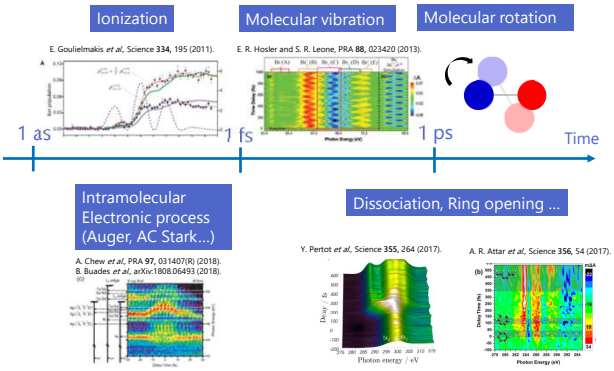
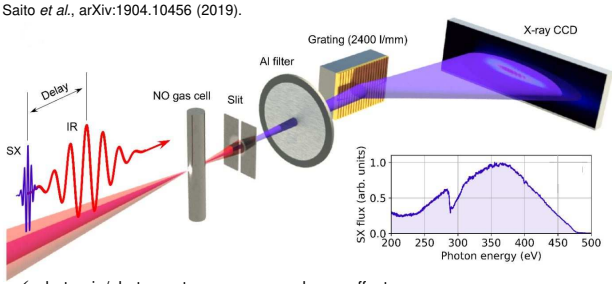
原子・分子における超高速過渡吸収分光



34

軟X線領域でのアト秒過渡吸収分光

Saito *et al.*, arXiv:1904.10456 (2019).



✓ photon-in/photon-out => no space charge effect
✓ Temporal resolution <1 fs (path length actively stabilized <100 as)
✓ Large delay range (up to ~100 ps)

- Target gas: NO
- HHs from He
- Total acquisition time: ~4 hours

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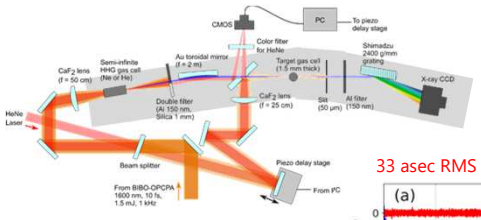
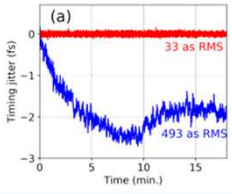
軟X線アト秒分光の実験配置

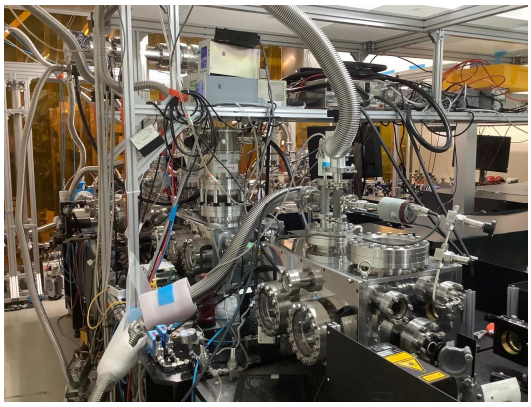


- HHG from He (2.4 bar)
- Sample gas: NO (0.1 bar)
- Pulse duration: SX <1 fs, IR 10 fs
- Attosecond delay stabilization (optional)

33 asec RMS for ~20 min



36



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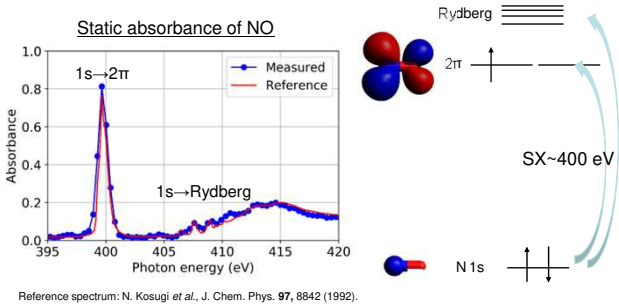
窒素K吸収端 (400 eV)でのNO分子のアト秒分光



Reference spectrum: N. Kosugi *et al.*, J. Chem. Phys. **97,** 8842 (1992).

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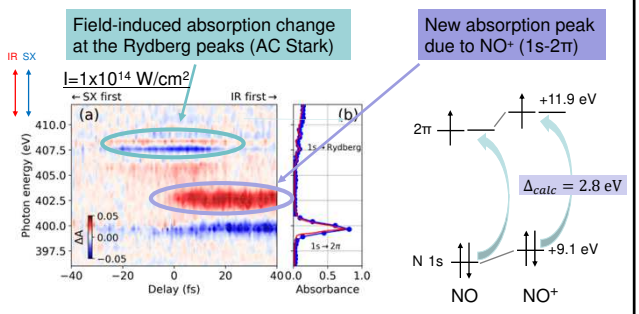
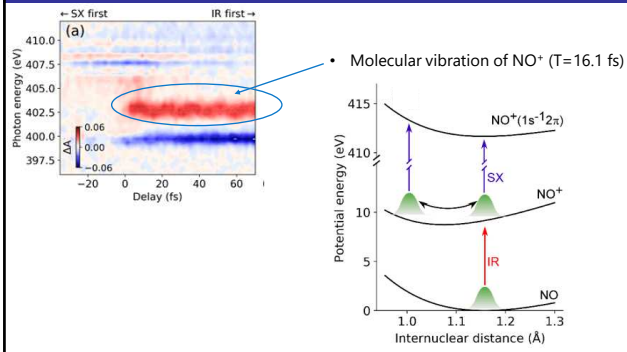
電子ダイナミクス



✓ Peak position of the 1s-2π is shifted after ionization.
(experiment: ~3.0 eV, calculation: 2.8 eV)

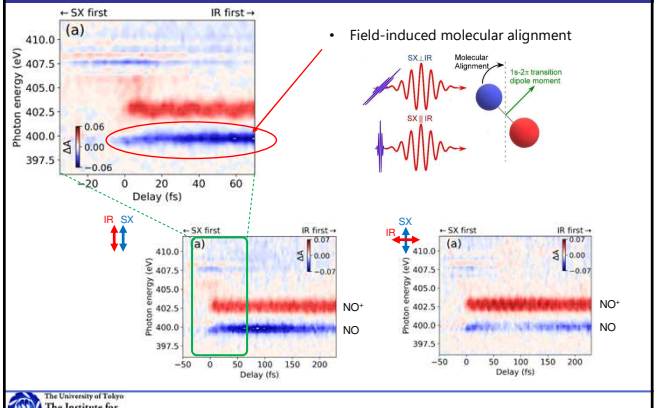
39

振動ダイナミクス



40

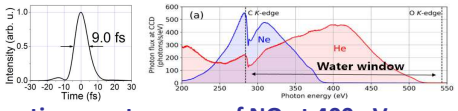
回転ダイナミクス



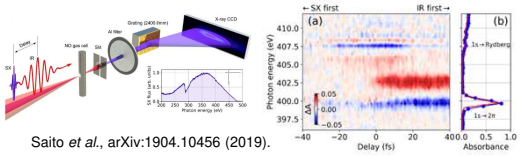
41

Part I. Summary

✓ **BIBO-based OPCPA for intense IR pulses (1.6 μm)**
Output: 1.5 mJ, 10 fs (sub-2-cycles), CEP-stable @ 1kHz
「水の窓」領域での孤立アト秒パルス発生が実現した。



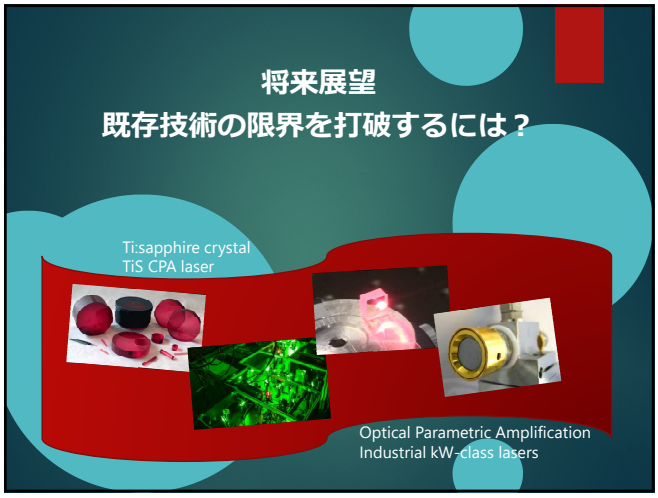
✓ **Transient absorption spectroscopy of NO at 400 eV**
多様な階層の量子ダイナミクスの観測が全光学的に可能となった。



Saito *et al.*, arXiv:1904.10456 (2019).

42

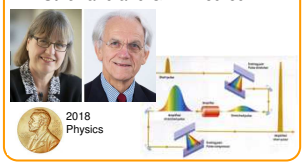
将来展望
既存技術の限界を打破するには？



43

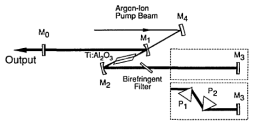
高強度超短パルスレーザーの要素技術 (1985-)

Chirped Pulse Amplification (1985)
D. Strickland and G. A. Mourou

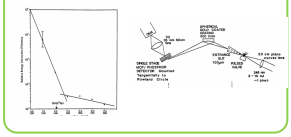


Ti:sapphire laser(1986)
P. Moulton

Kerr-lens Mode lock laser (1991)
W. Sibbett



High Harmonic Generation (1987)
McPherson, C. K. Rhodes



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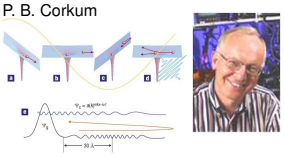
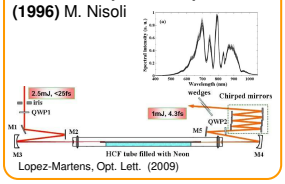
高強度超短パルスレーザーの要素技術 (1990-)

The three step model (1993)
P. B. Corkum



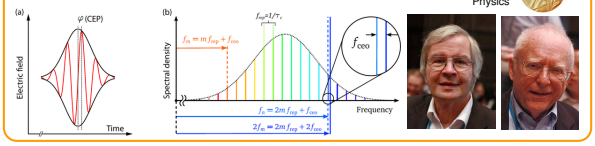
Corkum, Krausz: Attosecond Science, Nature (2007)

Hollow-fiber pulse compression (1996) M. Nisoli



Lopez-Martens, Opt. Lett. (2009)

Frequency comb and Carrier-Envelope Phase stabilization (2000)
T. W. Hänsch, J. L. Hall
2005
Physics



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この30年間で体験したこと

- 要素技術がブレークスルーにつながる。
- 要素技術 => システム化には、時間がかかる。
- システム技術の成熟により、科学が劇的に進む。
- 科学の進展により、新しい技術要請が生まれる。

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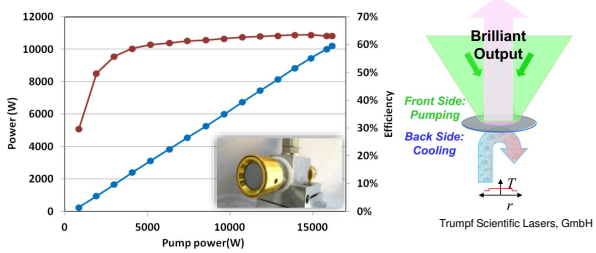
薄ディスクレーザー
「kW scalable」なレーザーの登場

- "Thin disk" gain media for efficient cooling



Trumpf Scientific Lasers, GmbH

- 600-W picosecond lasers are commercially available. Power scalable to >1 kW.

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高強度極短パルスレーザーのエネルギー効率
（チタンサファイアレーザー）

Average power
Pav=100 W

LD-pumped solid-state lasers
Nd:YLF(1 kHz)
Nd:YAG(10 kHz)
Nd:YVO_4(100 kHz)

100 ns

Coherent REVOLUTION

Pump 527/532 nm

Pav=20 W
Thermal limit

Chirped pulse amplification (CPA) Ti:sapphire laser

30 fs

- 10-20 mJ, 30 fs @ 1 kHz
- Wavelength 800 nm
- Carrier-envelope phase stable

Ti:sapphire CPA (ISSP, UTokyo)

Pav=1 W

Nonlinear pulse compression
- Gas-filled hollow-core fiber
- Dispersion compensation mirrors

5 fs

Waveguide
(Unscalable in enegy)

Hollow fiber @ KSU

Waveform-controlled few-cycle pulses

Near IR
800 nm

- 0.5-1 mJ, 5 fs @ 1 kHz
- Wavelength 700-900 nm
- Carrier-envelope phase stable（active feedback）

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次世代OPCPA光源のエネルギー効率

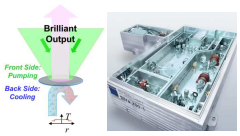
Average power
Pav=600 W

Chirped pulse amplification (CPA) thin-disk Yb:YAG laser

1 ps

Scalable in power up to multi-kW

- 60 mJ, 1 ps @ 10 kHz
- Wavelength 1030 nm



Trumpf Scientific Lasers, GmbH

Pav=60 W

Optical parametric chirped-pulse amplification (OPCPA)

15 fs

No thermal load

Waveform-controlled few-cycle pulses

MIR
2200 nm

- 6 mJ, 15 fs @ 10 kHz
- Wavelength 2200 nm
- Carrer-envelope phase stable（no feedback）

BIBO-OPA（ISSP, UTokyo）

49

常温接合 Distributed Face Cooling

平等グループ
Opt. Mat. Express **7**, 3214 (2017).

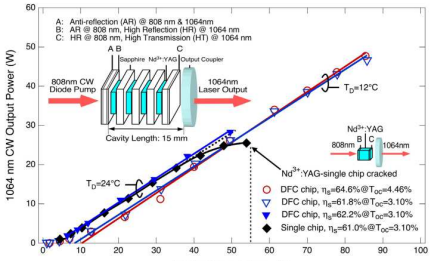


Fig. 3. Round-trip cavity loss and the output power from 9-disk Sapphire/Nd^{3+}:YAG DFC chip under continuous wave (CW) laser pump. T_D stands for chiller temperature for diode laser.

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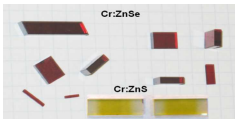
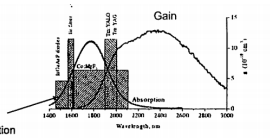
中赤外域の新固体レーザー

IPG Photonics（多結晶）

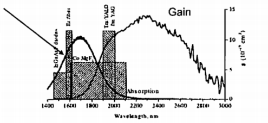


吸収スペクトルと蛍光スペクトル



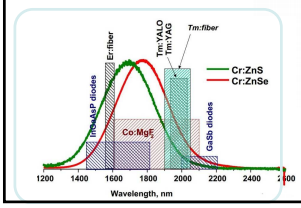
Cr^{2+}:ZnSe

absorption



Cr^{2+}:ZnS

吸収スペクトルと励起光源



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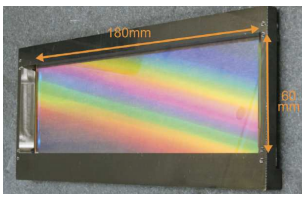
高効率な透過型回折格子

レーザー学会産業賞「奨励賞」受賞



特徴
- 優れた回折効率（≥95%、1250本/mm、中心波長：800nm/1040nm）
- 高精度つなぎ露光による大型素子の実現（≥180mm、幅：60mm）
- 薄板合成石英による高耐久性 および 優れた環境安定性
- 半導体露光装置の能力を活用した発展性
- 微細化：>6,000本/mm　大型化：≧300mm

Canon

T. Seki
T. Kitamura
Y. Kuramoto
T. Sukegawa

東京理科大学 TOKYO UNIVERSITY OF SCIENCE

Watanabe group
Chun Zhou
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C. Zhou *et al.* APEX **4** (2011) 072701
C. Zhou *et al.* Opt. Exp. **22** (2014) 5995

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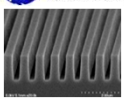
Kobayashi Group
Y. Kobayashi



Itatani Group
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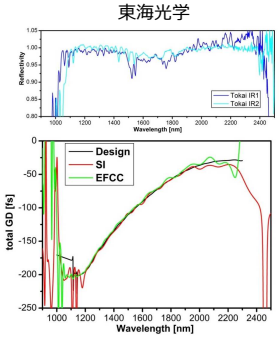

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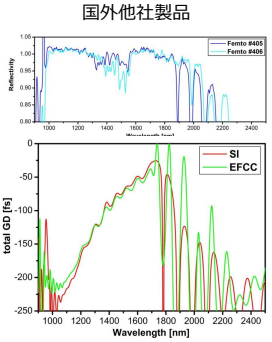

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（光学素子開発）

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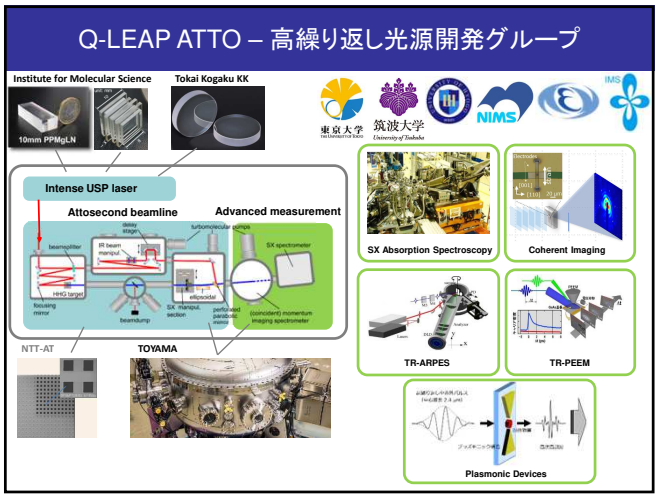
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Attosecond beamline
Advanced measurement
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HHG target
ellipsoidal
beamdump
(coincident) momentum imaging spectrometer
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