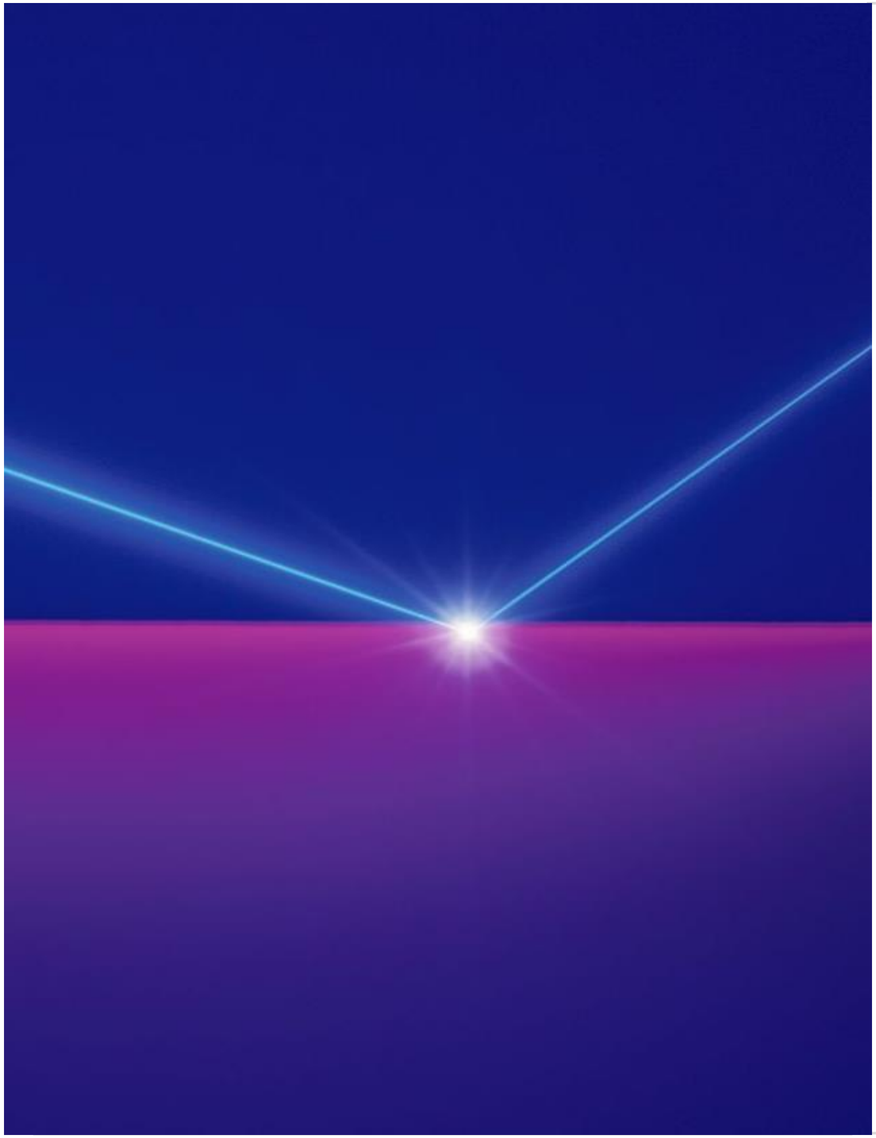

# 阿秒科学和凝聚态物理

Sheng Meng（孟胜)
Institute of Physics,
Chinese Academy of Sciences
2024.5.13

# OUTLINE

I. What is atto?

II. Brief history of “time” (ultrafast studies)

III. The rise of attosecond science

IV. Attoscience in condensed matter
  - development of attosecond technology
  - attosecond dynamics & applications

V. Advanced Attosecond Laser Facility

VI. Outlook

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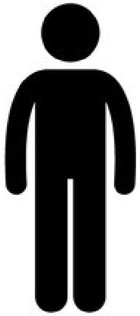
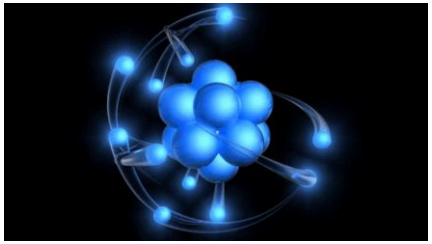
# What is ATTO?

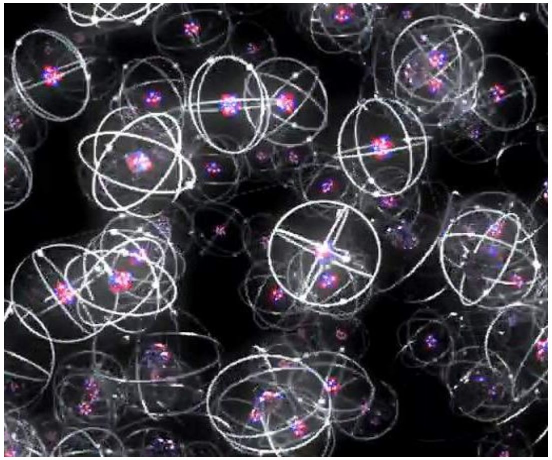
**1 attosecond=0.000 000 000 000 000 001 s**

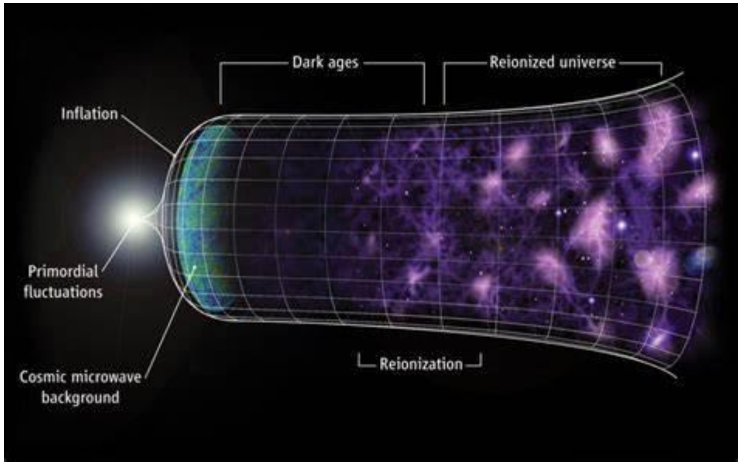




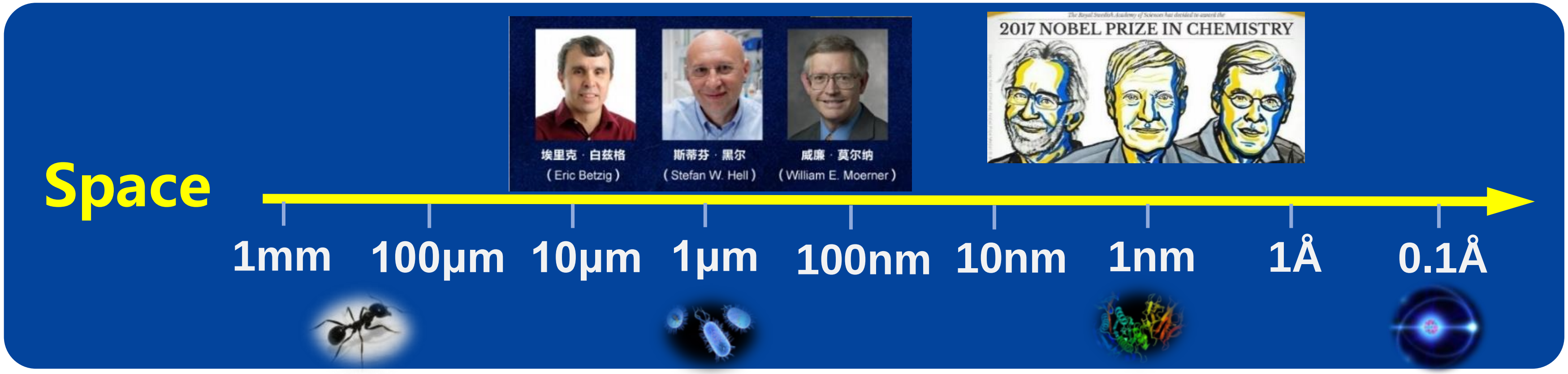
| $10^{27}$ m | 1 m | $10^{-10}$ m |
|---|---|---|
| $10^{18}$ s | 1 s | $10^{-18}$ s |

# The “Forth” Dimension





# Ultimate space-time scales for the study of matter



**Space**

1mm 100μm 10μm 1μm 100nm 10nm 1nm 1Å 0.1Å

埃里克 · 白兹格 (Eric Betzig)

斯蒂芬 · 黑尔 (Stefan W. Hell)

威廉 · 莫尔纳 (William E. Moerner)

2017 NOBEL PRIZE IN CHEMISTRY

**Time**

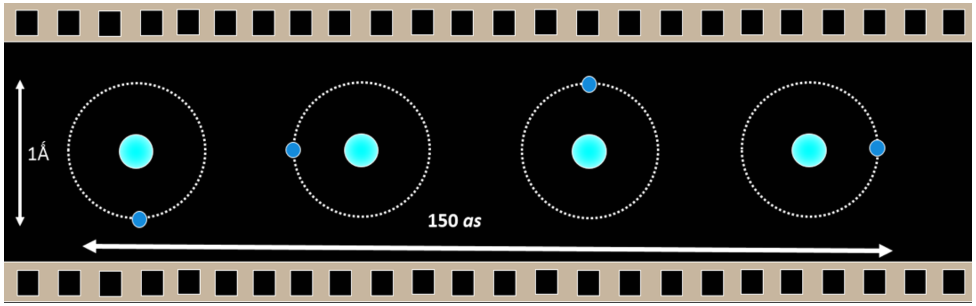
1ns 100ps 10ps 1ps 100fs 10fs 1fs 100as 10as

Zewail Nobel prize 1999

THE NOBEL PRIZE IN PHYSICS 2018

THE NOBEL PRIZE IN PHYSICS 2023

Unknown physics



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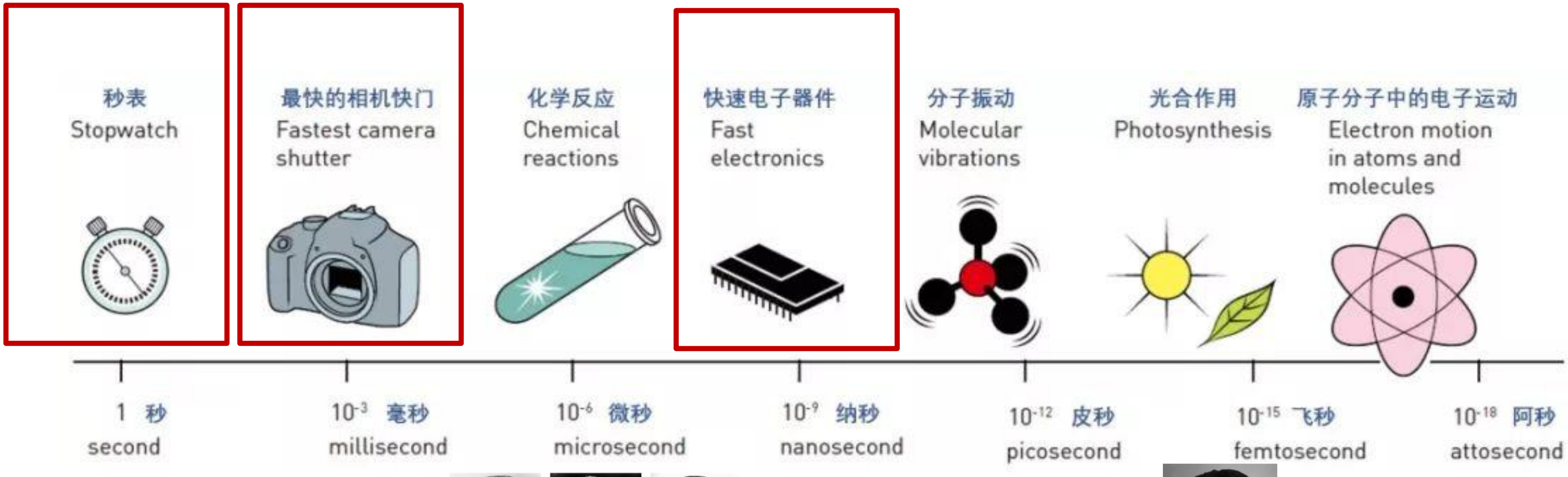
# “时间”简史



秒表
Stopwatch

最快的相机快门
Fastest camera shutter

化学反应
Chemical reactions

快速电子器件
Fast electronics

分子振动
Molecular vibrations

光合作用
Photosynthesis

原子分子中的电子运动
Electron motion in atoms and molecules

1 秒
second

$10^{-3}$ 毫秒
millisecond

$10^{-6}$ 微秒
microsecond

$10^{-9}$ 纳秒
nanosecond

$10^{-12}$ 皮秒
picosecond

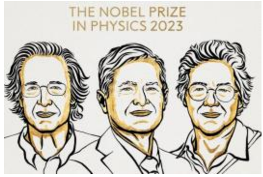
$10^{-15}$ 飞秒
femtosecond

$10^{-18}$ 阿秒
attosecond

Photo from the Nobel Foundation archive.
Manfred Eigen

Photo from the Nobel Foundation archive.
Ronald George Wreyford Norrish

Photo from the Nobel Foundation archive.
George Porter



# 迈布里奇-斯坦福1878年拍摄赛马奔跑的照片

# Stroboscopic photography (频闪照相)

Harold Edgerton (MIT, 1930s-1950s)

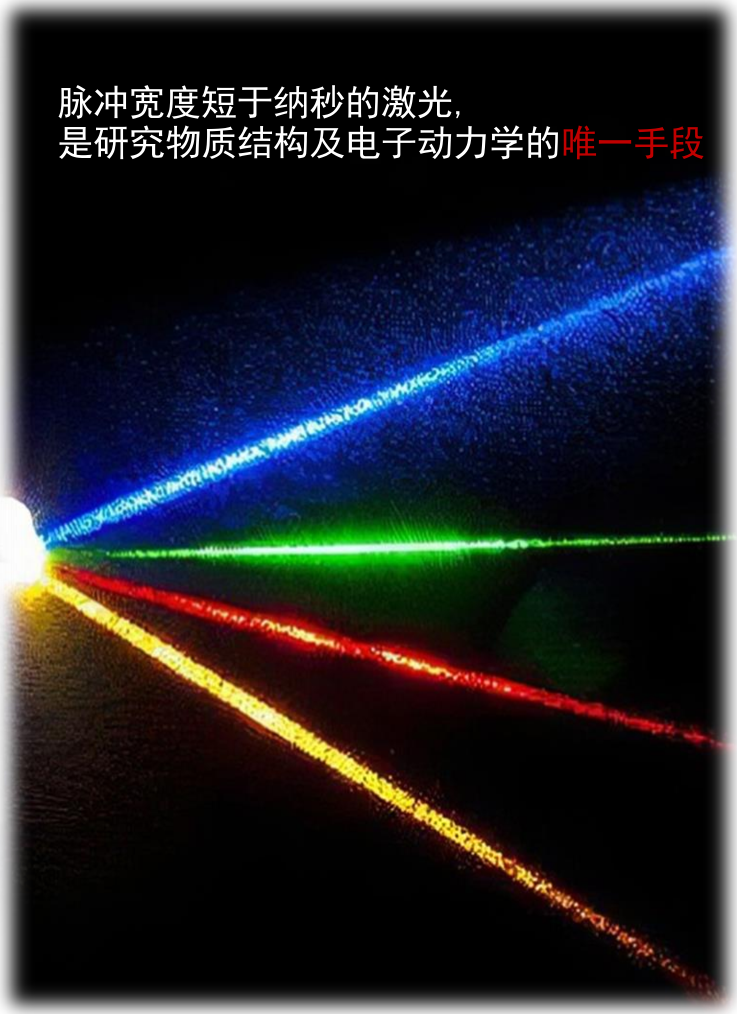
# 激光的出现，带来新机会

A. Einstein T. H. Maiman

光电效应 (1905)
受激辐射 (1917)

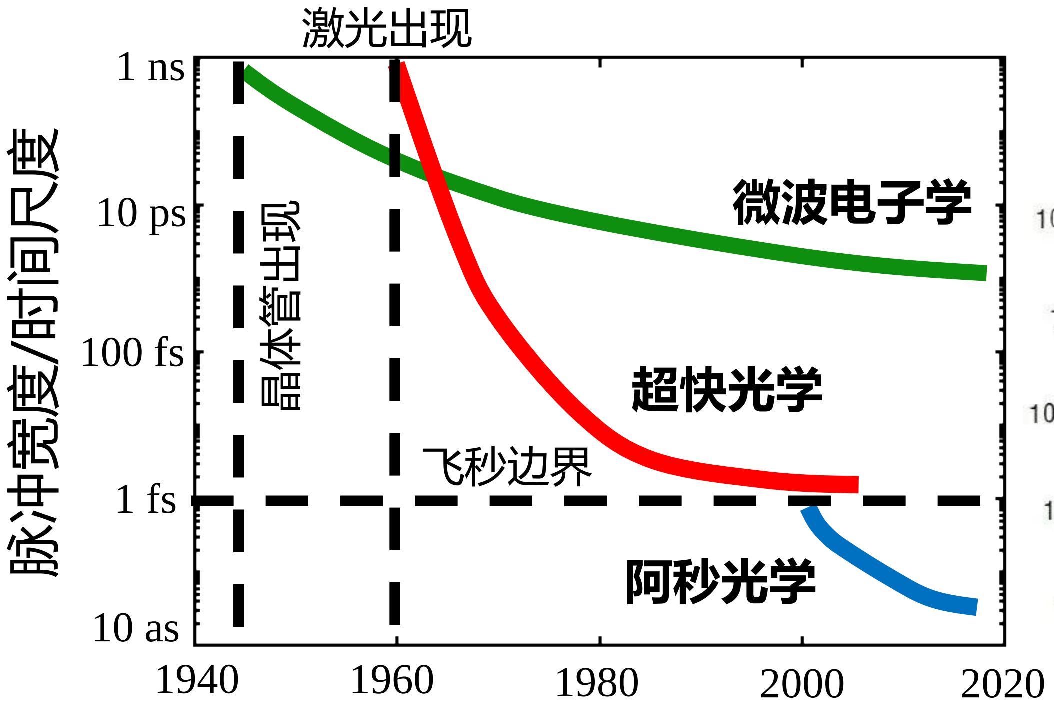
首台激光器
(1960)

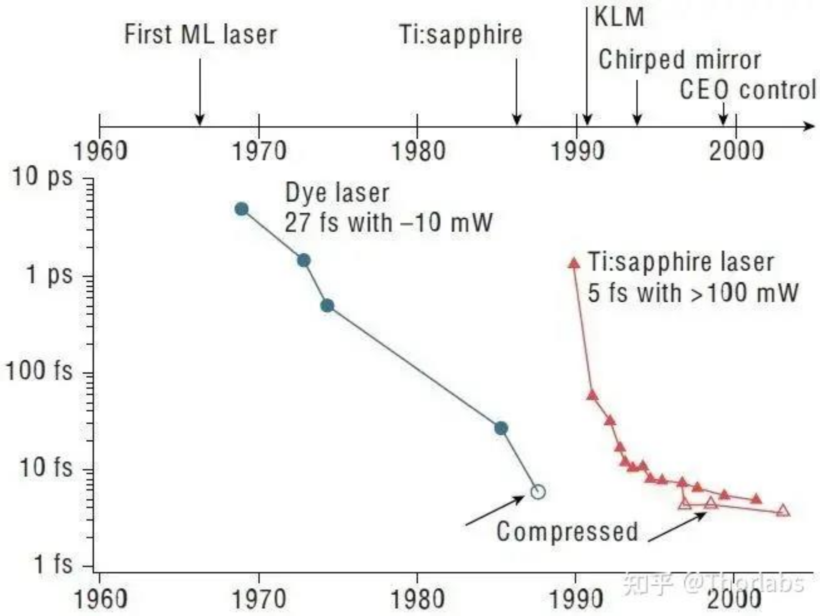
“要有光，激光”



脉冲宽度短于纳秒的激光，
是研究物质结构及电子动力学的唯一手段

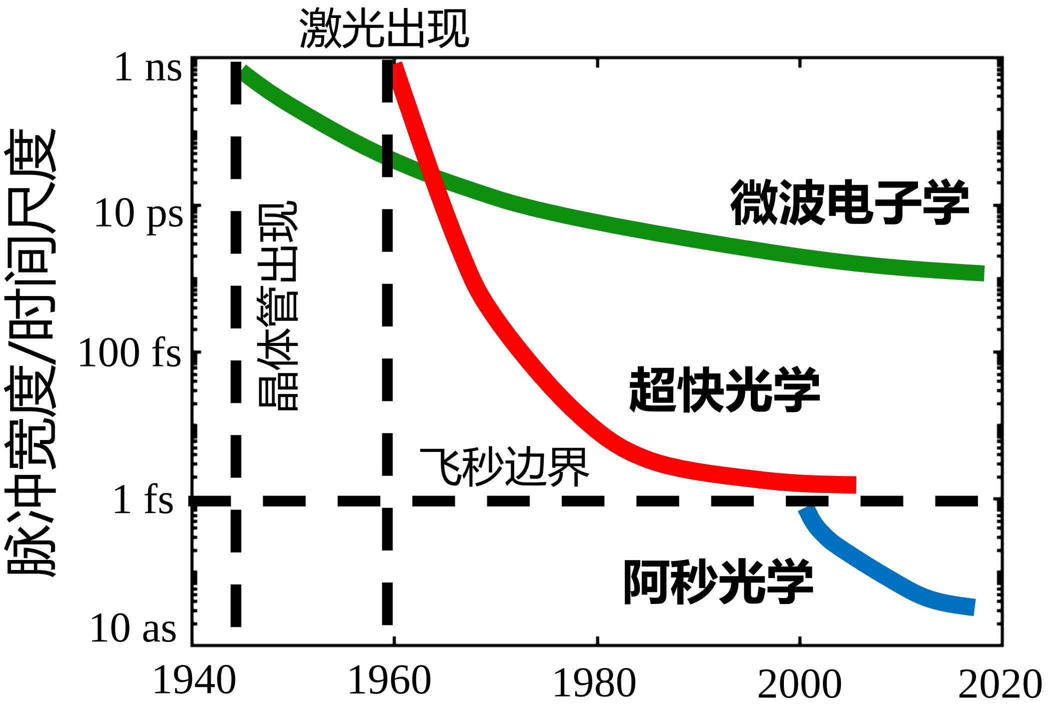
# 超快激光和超快过程





- 1960，红宝石激光器 ~1 ms
- 1961，调Q技术 ~100 ns
- 1966，主动锁模 <1 ns
- 1976，可饱和染料吸收体 <1 ps
- 1980s，对撞脉冲锁模 ~30 fs
- 1980s-1990s，啁啾脉冲压缩、克尔透镜锁模 ~5 fs
- 1980s-1990s，高次谐波和阿秒脉冲串产生 <1 fs

# 超快激光进入阿秒时代



NEWS AND ANALYSIS

## Attosecond lasers come of age

New laser facilities will allow physicists to study events that last for just billionths of a billionth of a second, such as the motion of electrons inside atoms. **Alexander Hellemans** reports

Taking a still picture of a moving object requires an exposure time that is short enough to effectively freeze the motion of that object. In the case of atoms moving within molecules, this time is of the order of a femtosecond ($10^{-15}$ s), which therefore requires the use of ultrashort laser pulses.

Although femtosecond pulses are now common in physics and chemistry, even they are too slow to study the motion of electrons, which takes place on the timescale of attoseconds ($10^{-18}$ s). But over the last two years, separate groups of researchers based in France, Austria, Sweden and the Netherlands have managed to generate and observe light pulses that last for just several

accelerators. As for studies of electrons in atoms, they could provide information about the fundamental quantum-mechanical behaviour of the hydrogen atom.

### Proof of the pudding

The technique used to create attosecond pulses was put forward in the early 1990s by several researchers, notably Paul Corkum of the National Research Council in Ottawa, Canada. Known as high-order harmonics generation, this technique involves using the electric field of femtosecond optical laser pulses to ionize rare-gas atoms and then accelerate the electrons away from the parent ions. When the electric field changes direction half a wavelength later, the electrons are driven back to the parent ions. Upon collision with the ions, the electrons

X-ray spectacle – physicists are generating ultrafast X-ray pulses by irradiating rare-gas atoms with

Ferenc Krausz, Vienna University of Technology

emitted ultraviolet beam and half of the original optical beam into a second target of rare-gas atoms and then altering the relative phase of the two beams. Observing changes in the energy spectrum of any electrons ejected from the gas provided the information they needed to work out the relative phase of the different harmonics.

In November 2001 Ferenc Krausz of the Vienna University of Technology and colleagues announced that they had used a similar technique to observe pulses lasting 650 attoseconds. But unlike Agostini, Balcou, Muller and co-workers, the Vienna team was able to single out individual pulses, and it remains the only research group able to do so. Although attosecond pulses have since also been created at AMOLF and at Lund University in Sweden, these pulses –

like those created at the LOA – have been confined to pulse trains.

Krausz's group is able to isolate individual pulses because it uses a laser that generates very short optical pulses – lasting just 5 femtoseconds – whereas other groups use pulses lasting 50 or 60 femtoseconds. The 5 femtosecond pulses contain only about two cycles of the optical laser light and therefore only about four attosecond pulses. It is then possible to filter out the few extraneous pulses. Krausz has also teamed up with Theodor Hänsch of the Max Planck Institute of Quantum Optics near Munich to control the temporal structure of attosecond pulses. To do this they developed so-called phase-

Last November researchers at Saclay reported making trains with pulses just 130 attoseconds long. The aim is to get down to 10 attoseconds, which, says Agostini, is the lower limit of harmonic generation because the harmonics produced by the rare-gas targets are never completely in phase.

Krausz says that about a dozen institutions in Europe, the US and Japan will shortly be equipping themselves with attosecond lasers that use the technology developed at Vienna. These labs will take about a year to get the pulse-generation and diagnostic technology set up, and should then start carrying out experiments. Altogether, it can take two or three years to master the generation of attosecond pulses, according to Vrakking. "Several labs are now trying hard and are quite close," he says.

10 physicsweb.org PHYSICS WORLD FEBRUARY 2004

## 阿秒激光时代来临

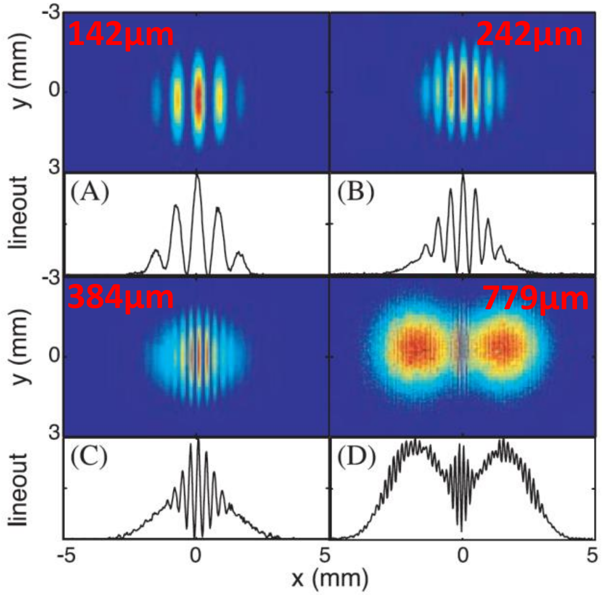
**新的激光装置允许物理学家研究仅发生在100亿亿分之一（$10^{-18}$）秒时间内的事件，如原子中电子的运动**

--Physics World, FEBRUARY 2004

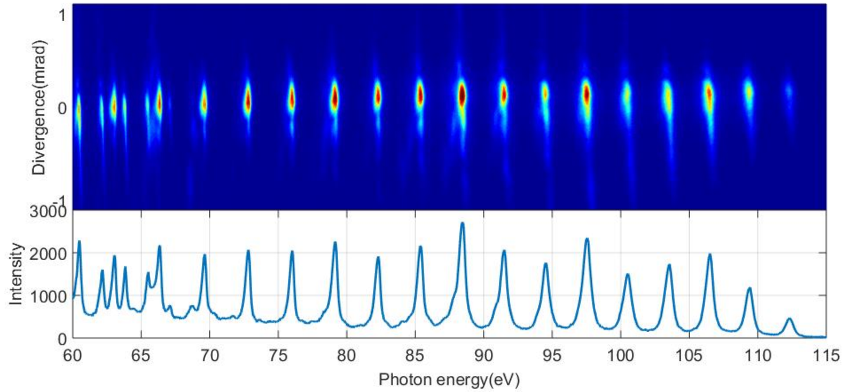
◆ **2001，欧洲首次产生孤立阿秒脉冲 650as**
◆ **2002，阿秒脉冲入选世界十大科学进展**
◆ **2012，阿秒脉冲突破70as**
◆ **2013，国内首次160as，现已突破80as**
◆ **2019，欧盟ELI-ALPS部分建成投入使用**

# 阿秒激光重要特征

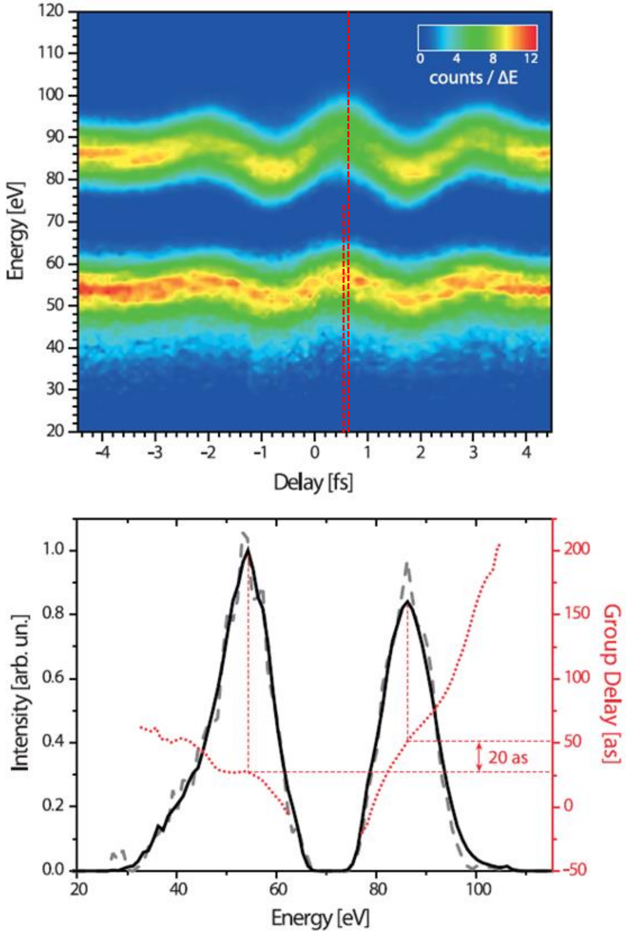
## 阿秒激光同时具有高度时空相干性和超高时间分辨能力



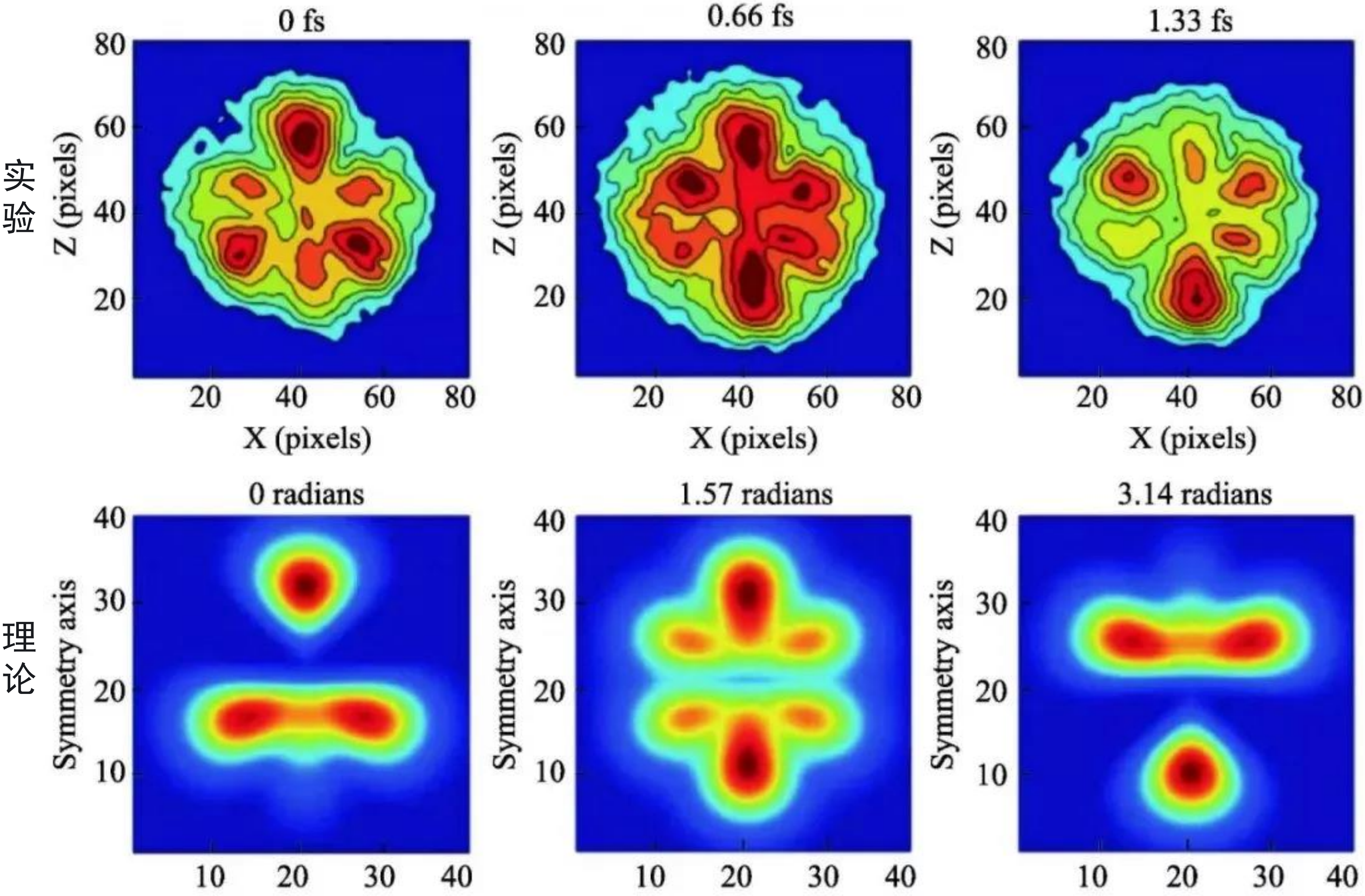
阿秒脉冲空间相干性



阿秒脉冲时间相干性



Ne原子2s、2p电离时差20阿秒



2017年, 加拿大的维伦纽夫研究组采用阿秒脉冲串联合红外激光电场实现了对氖原子的阿秒电子波包的成像

Villeneuve et al., Science 356,1150 (2017)

# 阿秒激光是世界科技进步的强大驱动力

阿秒脉冲:
追踪和控制电子运动

固体物理学

纳米技术

原子物理学

医学

物理化学

X射线/ 粒子源

分子生物学

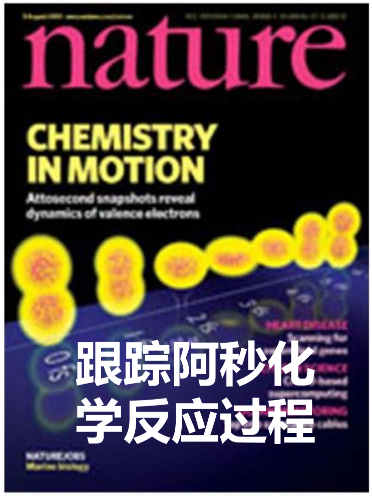
信息技术

生物医学

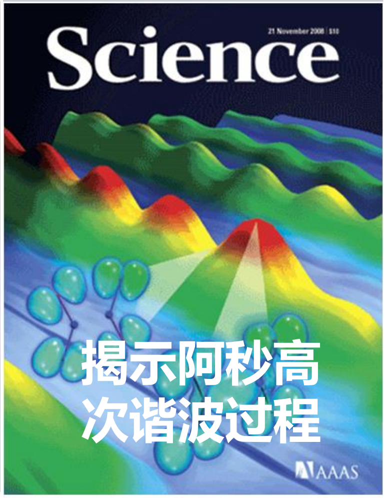
计量学

阿秒物理

阿秒光谱学



跟踪阿秒化学反应过程



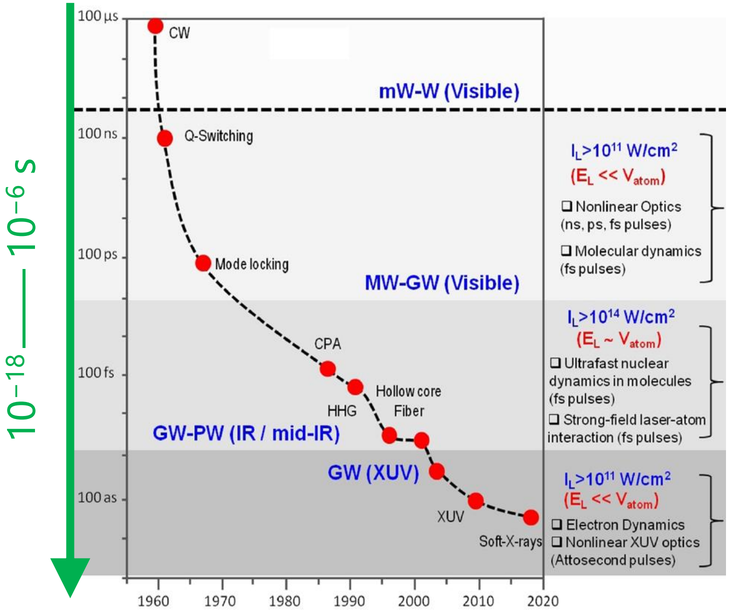
揭示阿秒高次谐波过程

记录阿秒量级瞬态过程

# 超快超强激光

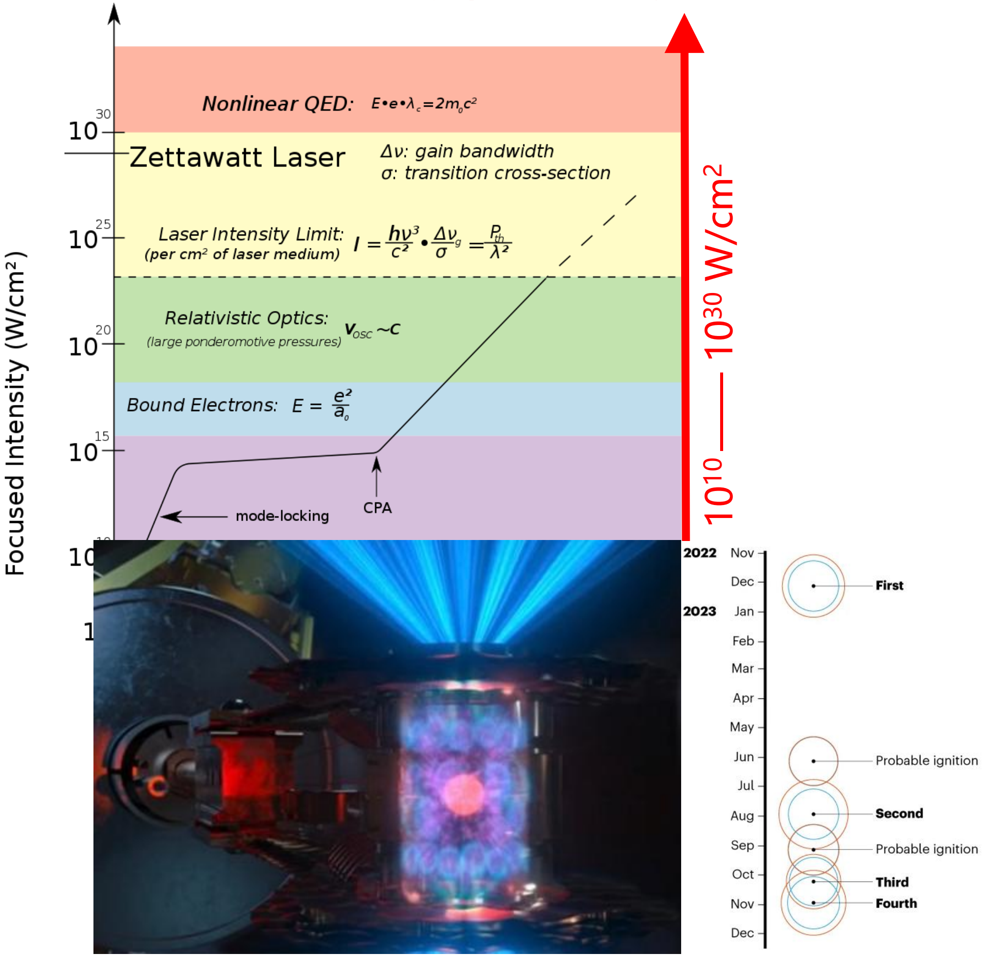
## 脉冲宽度不断压缩

**超高空间、时间分辨率**



## 激光强度不断增加

**强光-物质的相互作用，非平衡态调控**



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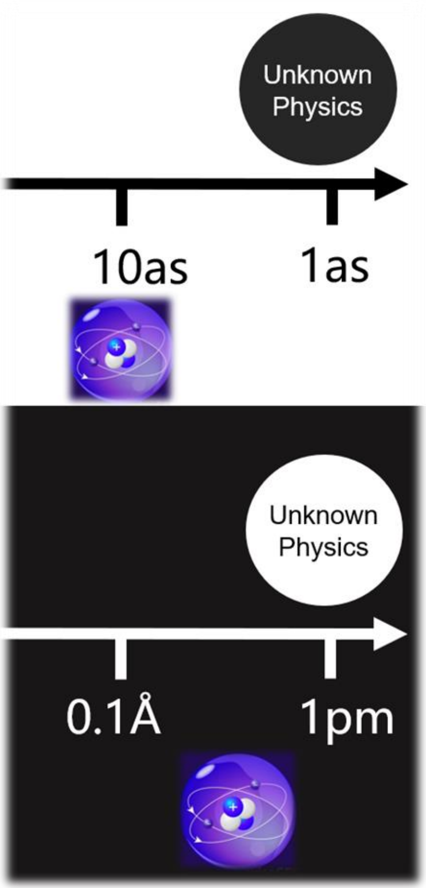
# 阿秒激光的出现

## 挑战?

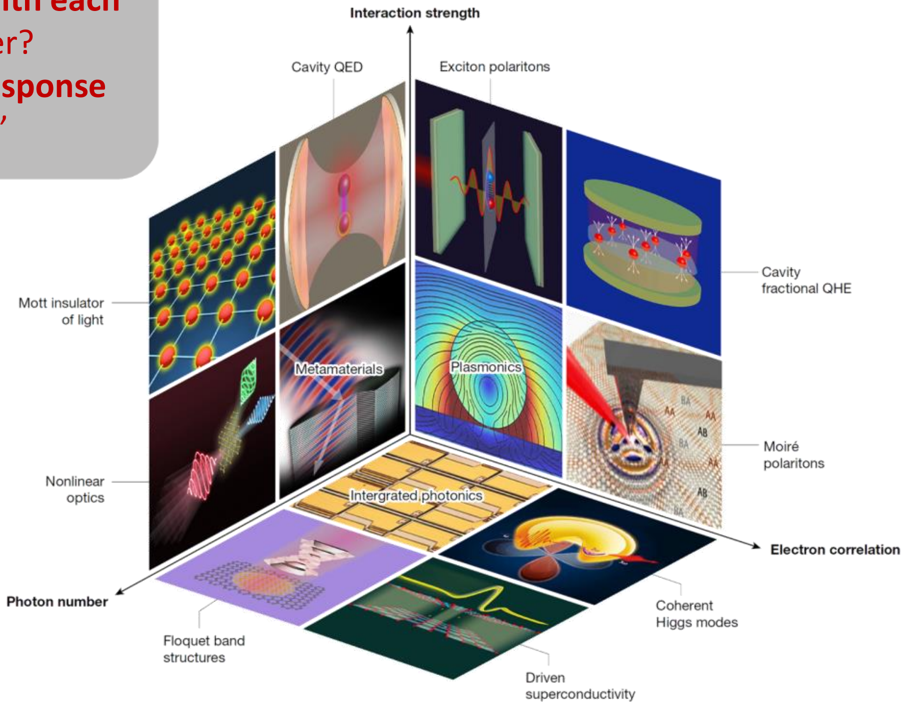


“How do **electrons interact with each** other inside solid matter?
What is the **ultimate time response**
**of electronic** matter?”

Paul Corkum Anne L’ Huillier Ferenc Krausz

2022 Wolf Prize in Physics

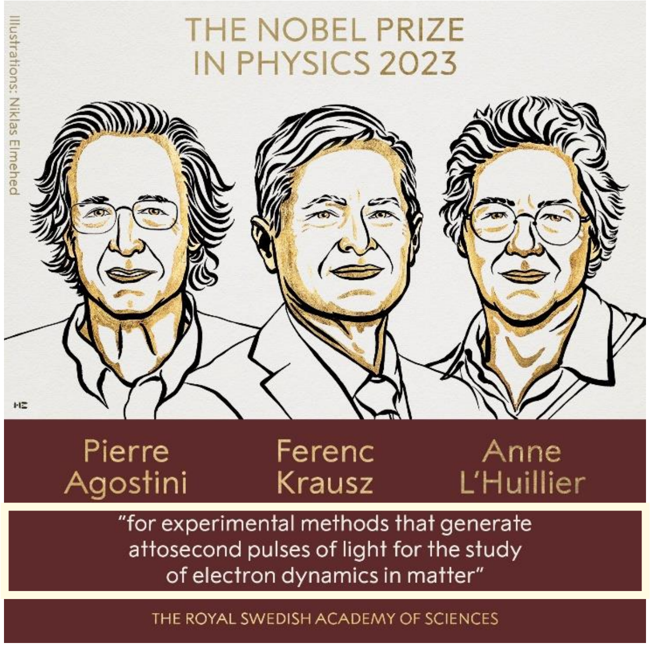
“for their pioneering contributions to the fields of ultrafast laser science and attosecond physics, and for demonstrating time-resolved imaging of electron motion in atoms, molecules, and solids”



J. Bloch et al., Nature 606, 41–48 (2022).

# 阿秒激光的产生

解决挑战，从飞秒到阿秒



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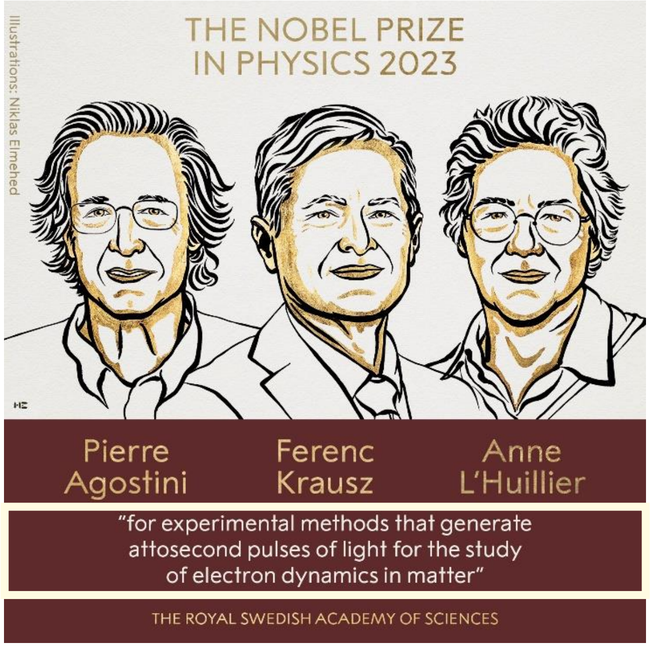
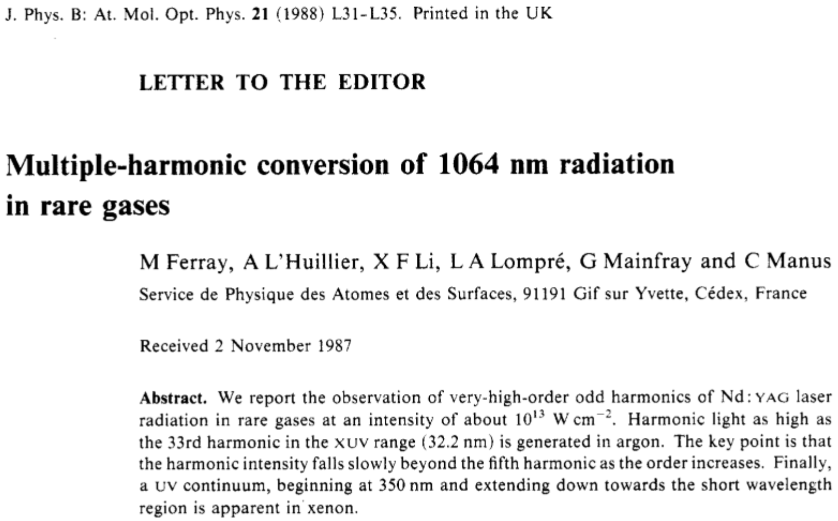
## 解决挑战，从飞秒到阿秒



原子多电离 (1982-1985)
高次谐波的产生 (1987-1992)
阿秒脉冲串理论提出 (1996-2000)

Anne L`Huillier



J. Phys. B: At. Mol. Opt. Phys. 21 (1988) L31-L35. Printed in the UK

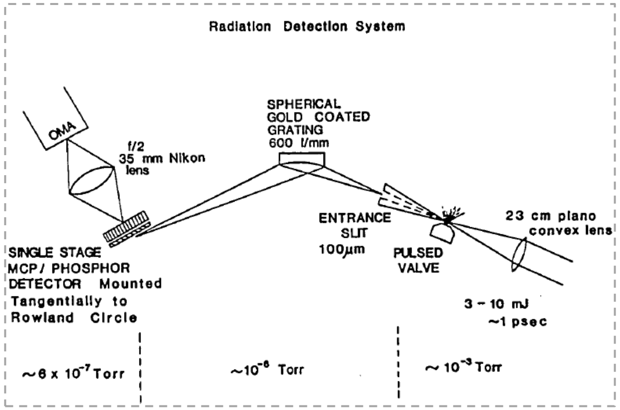
LETTER TO THE EDITOR

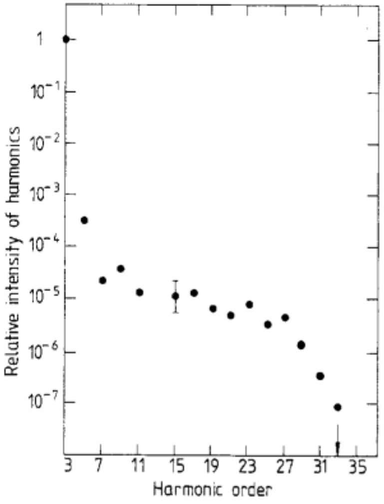
**Multiple-harmonic conversion of 1064 nm radiation in rare gases**

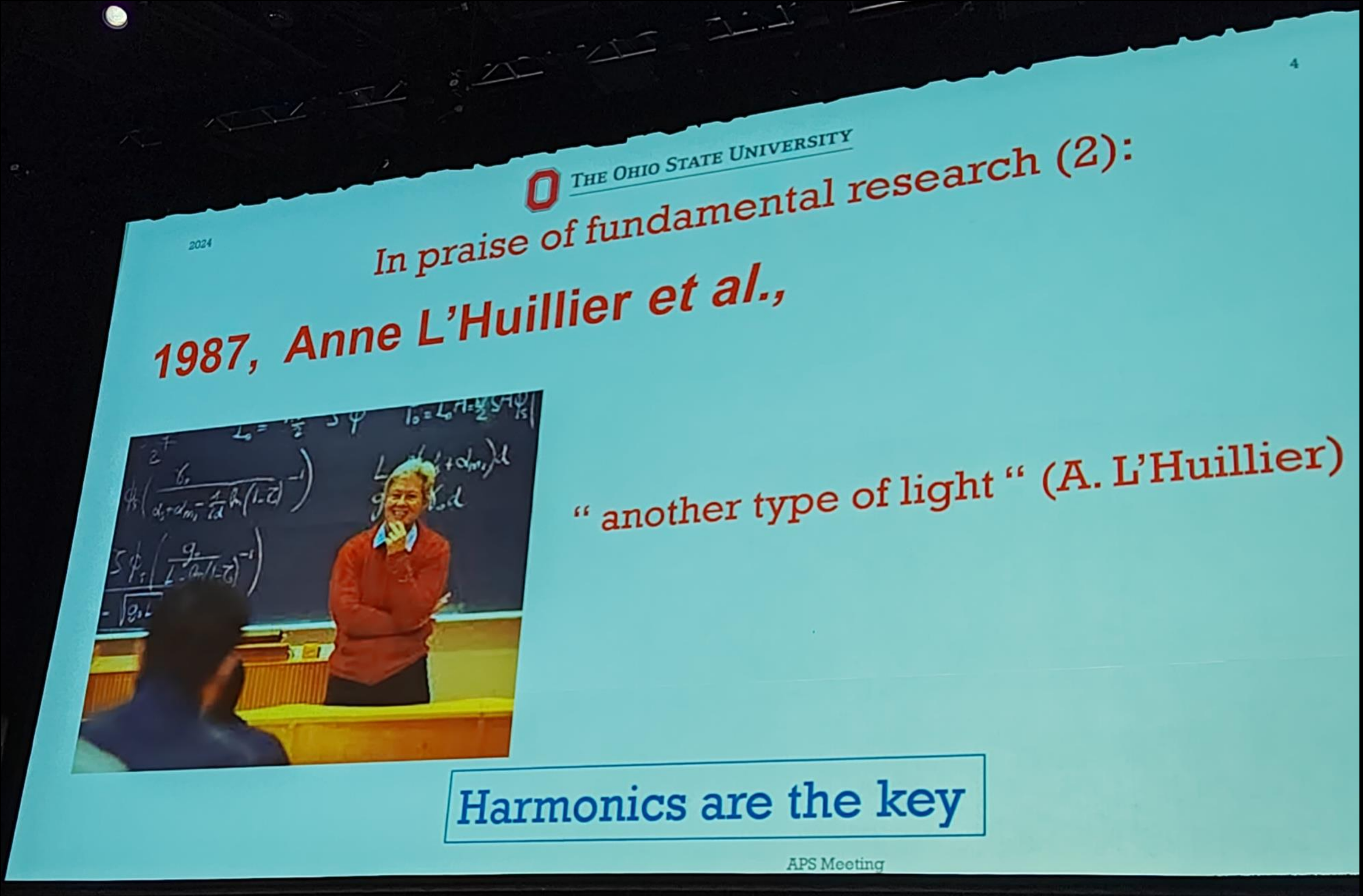
M Ferray, A L'Huillier, X F Li, L A Lompré, G Mainfray and C Manus
Service de Physique des Atomes et des Surfaces, 91191 Gif sur Yvette, Cédex, France

Received 2 November 1987

**Abstract.** We report the observation of very-high-order odd harmonics of Nd:YAG laser radiation in rare gases at an intensity of about $10^{13}$ W cm$^{-2}$. Harmonic light as high as the 33rd harmonic in the XUV range (32.2 nm) is generated in argon. The key point is that the harmonic intensity falls slowly beyond the fifth harmonic as the order increases. Finally, a UV continuum, beginning at 350 nm and extending down towards the short wavelength region is apparent in xenon.







THE OHIO STATE UNIVERSITY

## In praise of fundamental research (2):

**1987, Anne L’Huillier *et al.*,**

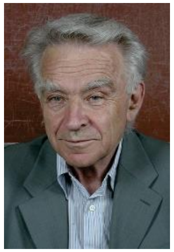
“ another type of light “ (A. L’Huillier)

**Harmonics are the key**

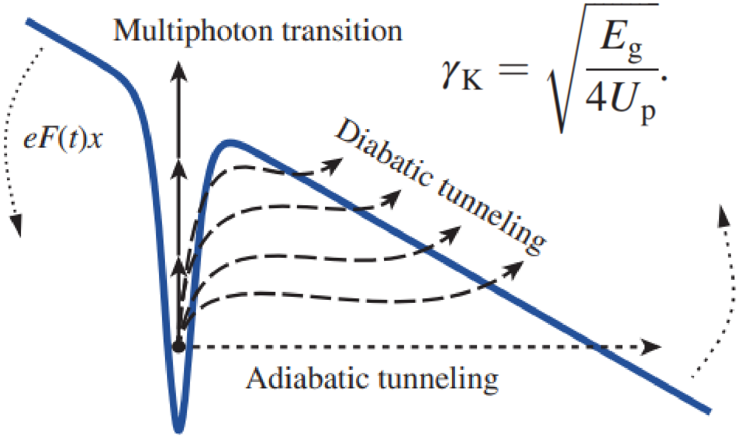
Courtesy: P. Agostini

# 阿秒激光的产生

解决挑战，从飞秒到阿秒



Leonid Keldysh

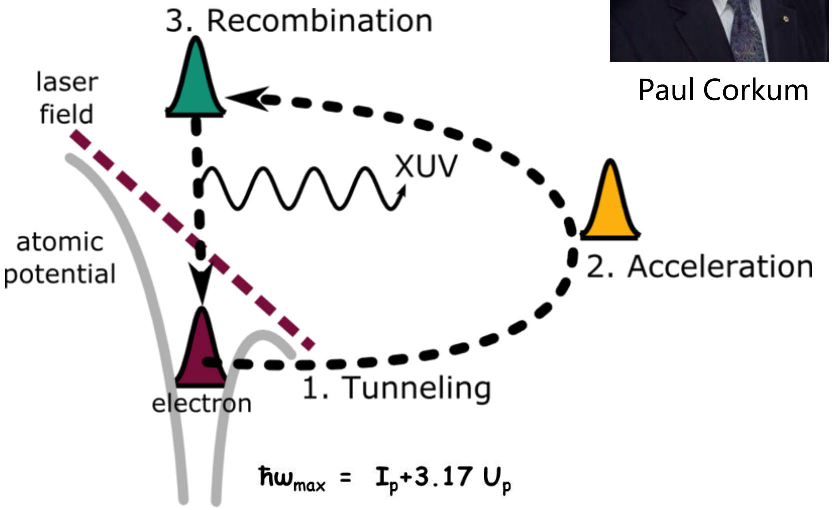


$$\gamma_K = \sqrt{\frac{E_g}{4U_p}}.$$

隧穿电离Keldysh模型 (1965)

L. V. Keldysh, Sov. Phys. JETP 20, 1307 (1965).
S. Yu et al., Rev. Mod. Phys. 90, 021002 (2018).

Paul Corkum



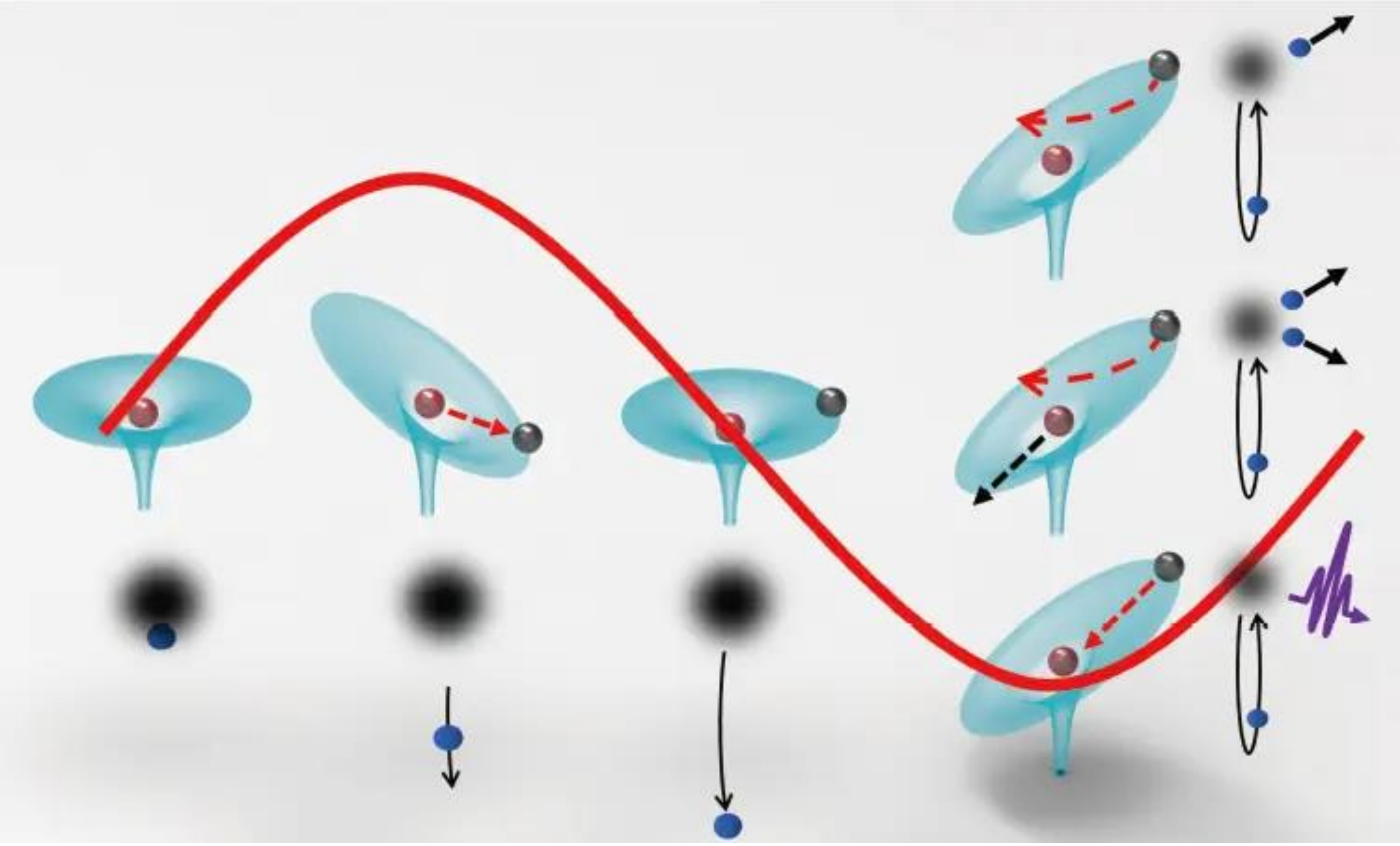
$$\hbar\omega_{max} = I_p + 3.17\, U_p$$

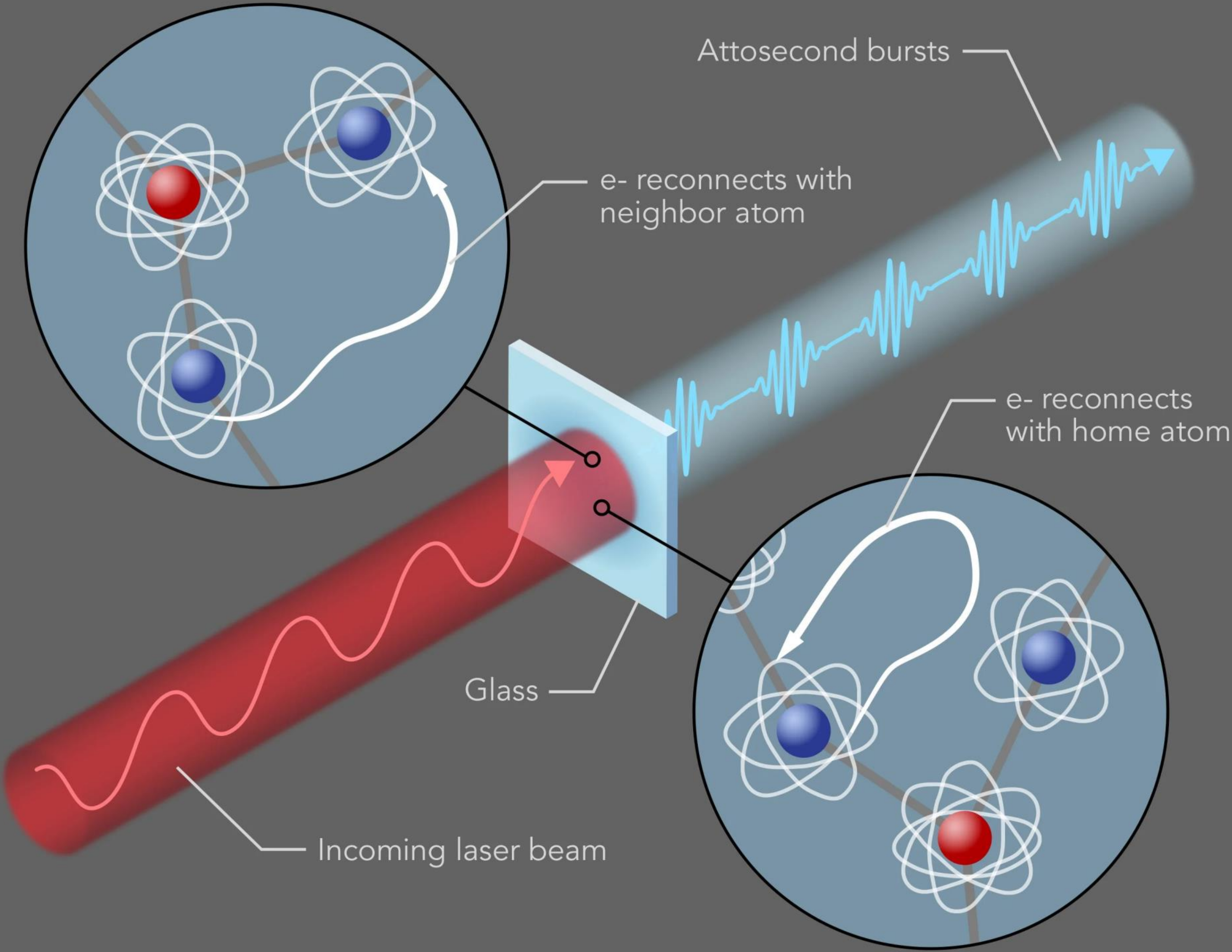
三步模型：

强光场下描述原子中电子的运动 (1993)

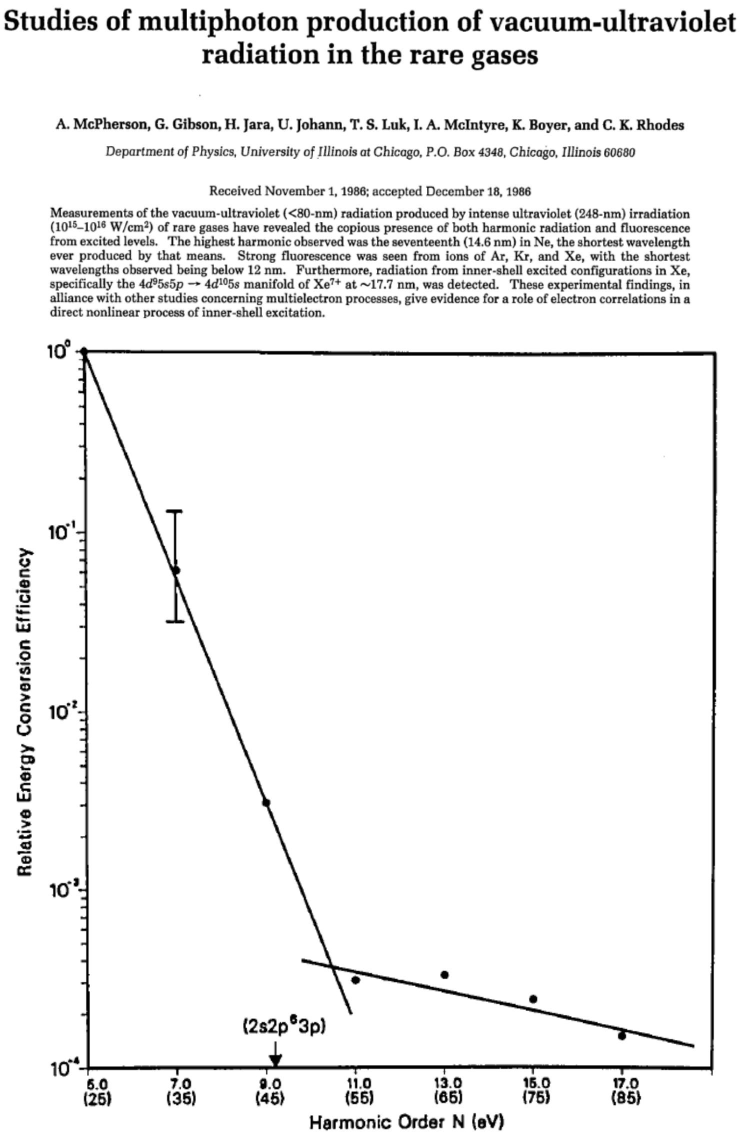
P. B. Corkum, Phys. Rev. Lett. 71, 1994 (1993).

# Rescattering or Three-Step Model




Attosecond bursts
e- reconnects with neighbor atom
e- reconnects with home atom
Glass
Incoming laser beam

# High harmonics reported earlier



## Studies of multiphoton production of vacuum-ultraviolet radiation in the rare gases

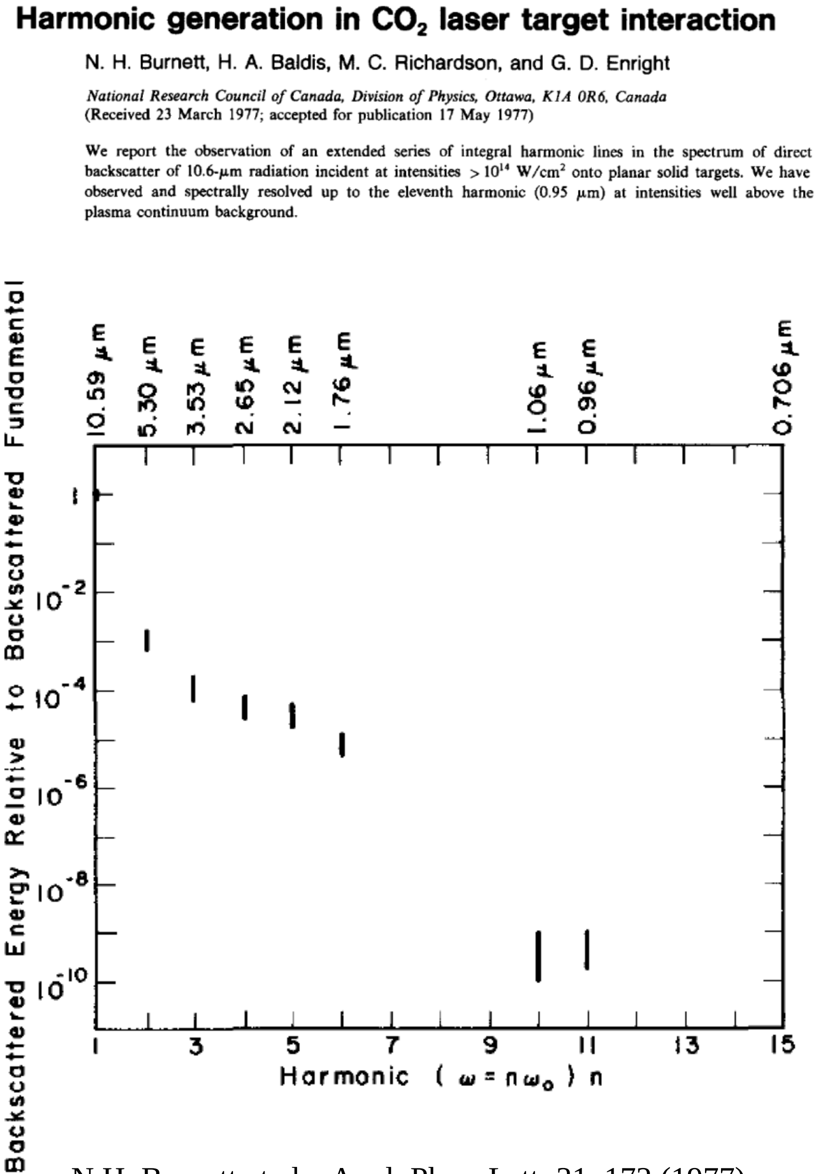
**A. McPherson, G. Gibson, H. Jara, U. Johann, T. S. Luk, I. A. McIntyre, K. Boyer, and C. K. Rhodes**

*Department of Physics, University of Illinois at Chicago, P.O. Box 4348, Chicago, Illinois 60680*

Received November 1, 1986; accepted December 18, 1986

Measurements of the vacuum-ultraviolet (<80-nm) radiation produced by intense ultraviolet (248-nm) irradiation ($10^{15}$–$10^{16}$ W/cm$^2$) of rare gases have revealed the copious presence of both harmonic radiation and fluorescence from excited levels. The highest harmonic observed was the seventeenth (14.6 nm) in Ne, the shortest wavelength ever produced by that means. Strong fluorescence was seen from ions of Ar, Kr, and Xe, with the shortest wavelengths observed being below 12 nm. Furthermore, radiation from inner-shell excited configurations in Xe, specifically the $4d^95s5p \rightarrow 4d^{10}5s$ manifold of Xe$^{7+}$ at ~17.7 nm, was detected. These experimental findings, in alliance with other studies concerning multielectron processes, give evidence for a role of electron correlations in a direct nonlinear process of inner-shell excitation.

A. McPherson et al., J. Opt. Soc. Am. B 4, 595 (1987).



## Harmonic generation in $CO_2$ laser target interaction

N. H. Burnett, H. A. Baldis, M. C. Richardson, and G. D. Enright

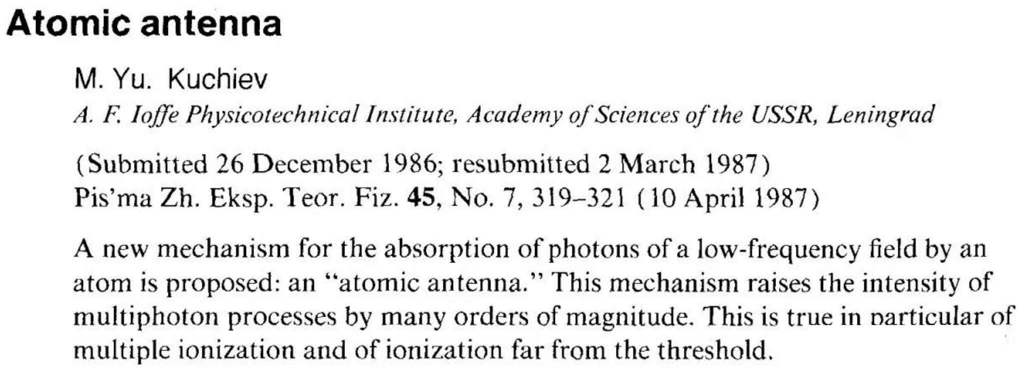
*National Research Council of Canada, Division of Physics, Ottawa, K1A 0R6, Canada*

(Received 23 March 1977; accepted for publication 17 May 1977)

We report the observation of an extended series of integral harmonic lines in the spectrum of direct backscatter of 10.6-μm radiation incident at intensities > $10^{14}$ W/cm$^2$ onto planar solid targets. We have observed and spectrally resolved up to the eleventh harmonic (0.95 μm) at intensities well above the plasma continuum background.

N.H. Burnett et al., Appl. Phys. Lett. 31, 172 (1977).

# Theoretical explanations



## Atomic antenna

M. Yu. Kuchiev
*A. F. Ioffe Physicotechnical Institute, Academy of Sciences of the USSR, Leningrad*

(Submitted 26 December 1986; resubmitted 2 March 1987)
Pis'ma Zh. Eksp. Teor. Fiz. **45**, No. 7, 319–321 (10 April 1987)

A new mechanism for the absorption of photons of a low-frequency field by an atom is proposed: an "atomic antenna." This mechanism raises the intensity of multiphoton processes by many orders of magnitude. This is true in particular of multiple ionization and of ionization far from the threshold.

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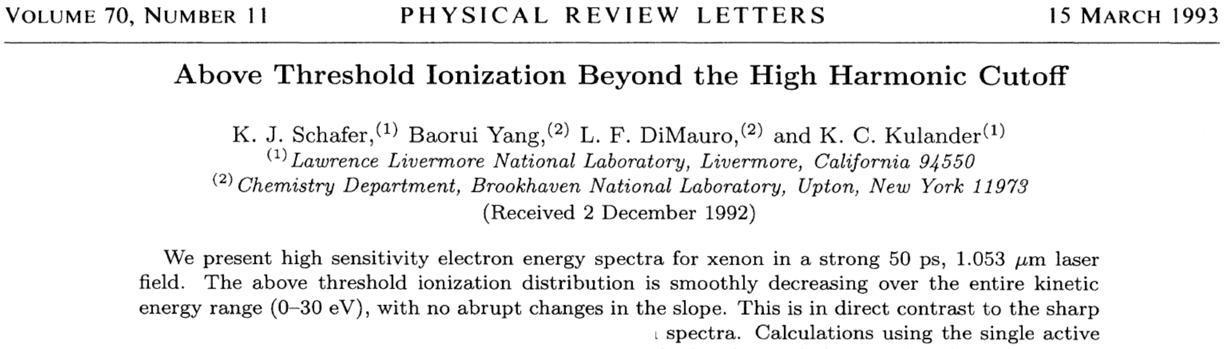

VOLUME 70, NUMBER 11 PHYSICAL REVIEW LETTERS 15 MARCH 1993

## Above Threshold Ionization Beyond the High Harmonic Cutoff

K. J. Schafer,(1) Baorui Yang,(2) L. F. DiMauro,(2) and K. C. Kulander(1)
(1) *Lawrence Livermore National Laboratory, Livermore, California 94550*
(2) *Chemistry Department, Brookhaven National Laboratory, Upton, New York 11973*

(Received 2 December 1992)

We present high sensitivity electron energy spectra for xenon in a strong 50 ps, 1.053 $\mu$m laser field. The above threshold ionization distribution is smoothly decreasing over the entire kinetic energy range (0–30 eV), with no abrupt changes in the slope. This is in direct contrast to the sharp
spectra. Calculations using the single active
th the observed electron distributions. These
etween the electron and photon emission from

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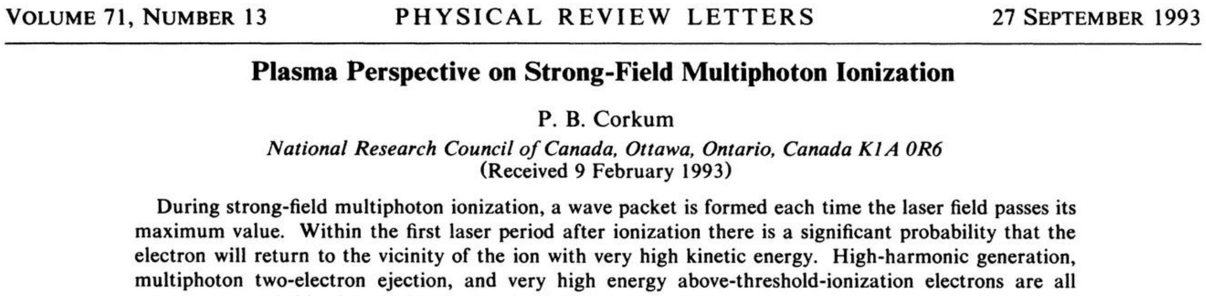

VOLUME 71, NUMBER 13 PHYSICAL REVIEW LETTERS 27 SEPTEMBER 1993

## Plasma Perspective on Strong-Field Multiphoton Ionization

P. B. Corkum
*National Research Council of Canada, Ottawa, Ontario, Canada K1A 0R6*

(Received 9 February 1993)

During strong-field multiphoton ionization, a wave packet is formed each time the laser field passes its maximum value. Within the first laser period after ionization there is a significant probability that the electron will return to the vicinity of the ion with very high kinetic energy. High-harmonic generation, multiphoton two-electron ejection, and very high energy above-threshold-ionization electrons are all consequences of this electron-ion interaction. One important para
of these effects is the rate at which the wave packet spreads in th
electric field; another is the laser polarization. These will be crucial

PACS numbers: 32.80.Rm

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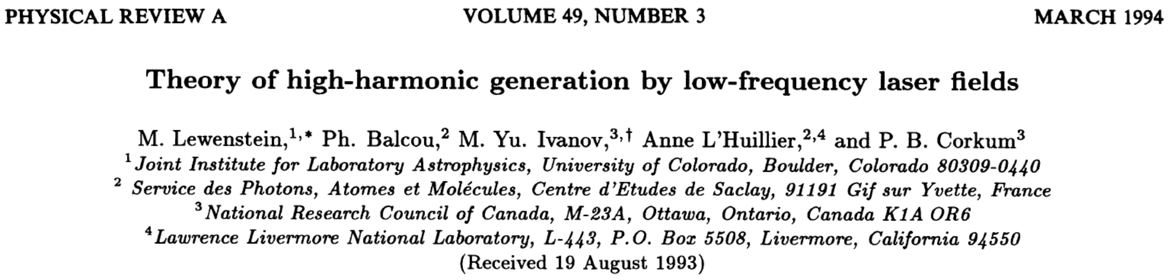


PHYSICAL REVIEW A VOLUME 49, NUMBER 3 MARCH 1994

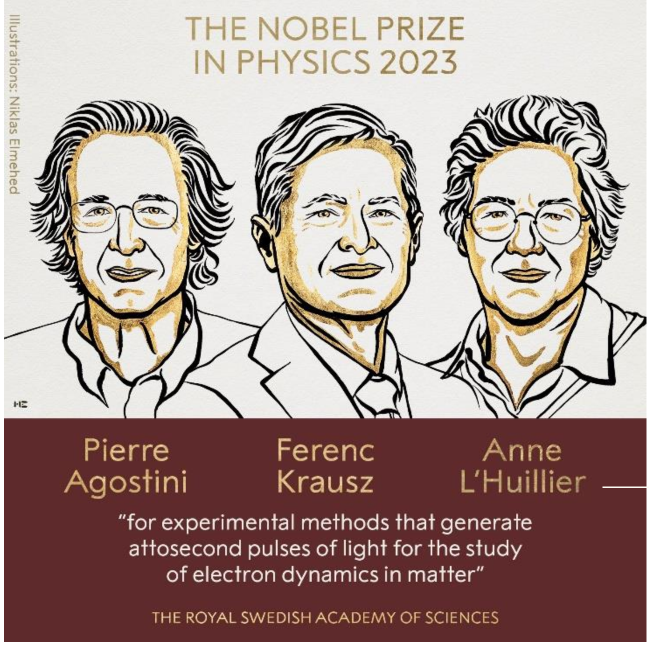
## Theory of high-harmonic generation by low-frequency laser fields

M. Lewenstein,[1,*] Ph. Balcou,[2] M. Yu. Ivanov,[3,†] Anne L'Huillier,[2,4] and P. B. Corkum[3]
[1] *Joint Institute for Laboratory Astrophysics, University of Colorado, Boulder, Colorado 80309-0440*
[2] *Service des Photons, Atomes et Molécules, Centre d'Etudes de Saclay, 91191 Gif sur Yvette, France*
[3] *National Research Council of Canada, M-23A, Ottawa, Ontario, Canada K1A 0R6*
[4] *Lawrence Livermore National Laboratory, L-443, P.O. Box 5508, Livermore, California 94550*

(Received 19 August 1993)

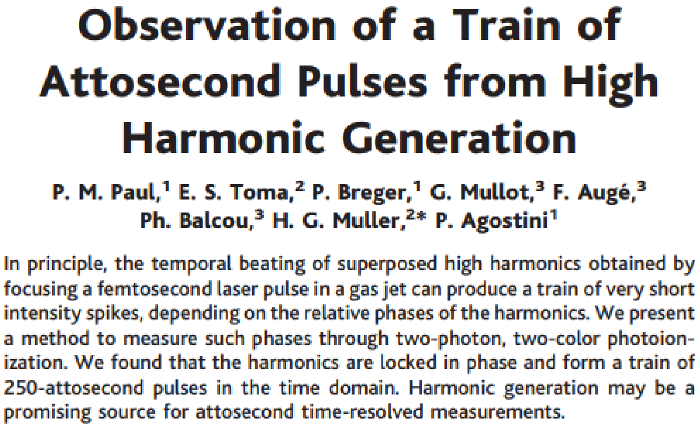
# 阿秒激光的产生

## 解决挑战，从飞秒到阿秒



"for experimental methods that generate attosecond pulses of light for the study of electron dynamics in matter"

Pierre Agostini

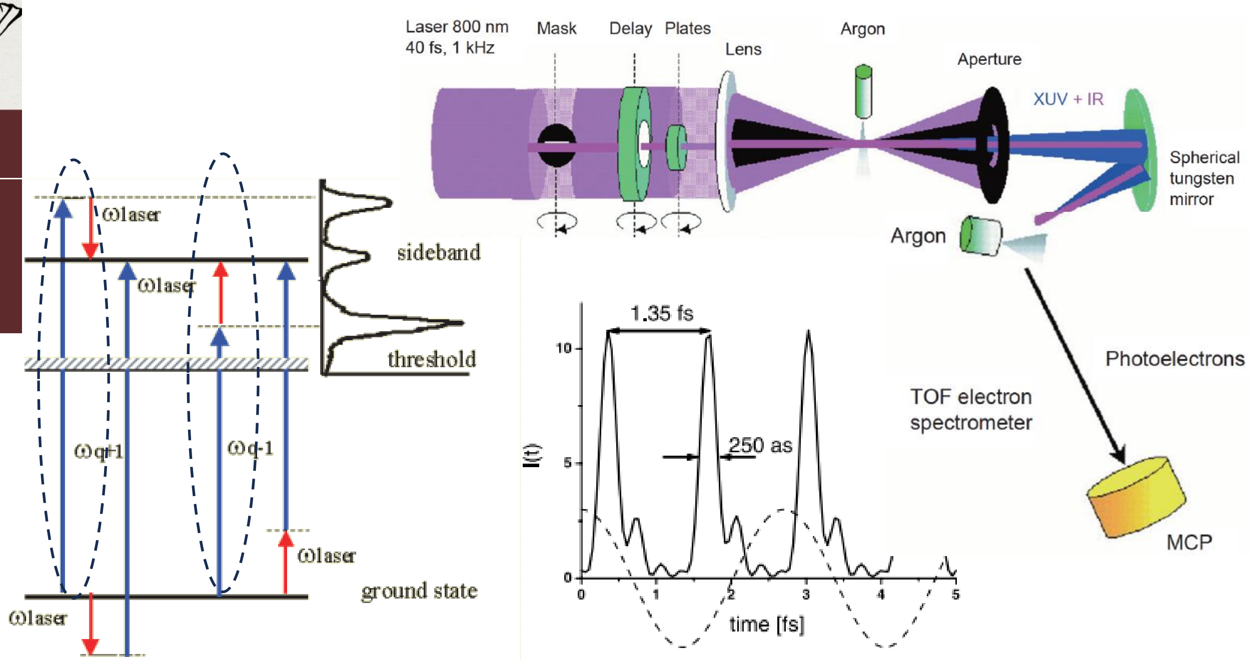


### Observation of a Train of Attosecond Pulses from High Harmonic Generation

P. M. Paul,[1] E. S. Toma,[2] P. Breger,[1] G. Mullot,[3] F. Augé,[3]
Ph. Balcou,[3] H. G. Muller,[2]* P. Agostini[1]

In principle, the temporal beating of superposed high harmonics obtained by focusing a femtosecond laser pulse in a gas jet can produce a train of very short intensity spikes, depending on the relative phases of the harmonics. We present a method to measure such phases through two-photon, two-color photoionization. We found that the harmonics are locked in phase and form a train of 250-attosecond pulses in the time domain. Harmonic generation may be a promising source for attosecond time-resolved measurements.

强场阈上电离 (1982-1985)
首次实现阿秒脉冲串 (2001)



**Reconstruction of attosecond beating by interference of two-photon transitions (RABBITT)**

THE OHIO STATE UNIVERSITY

2024

1

# From Attos to Zeptos

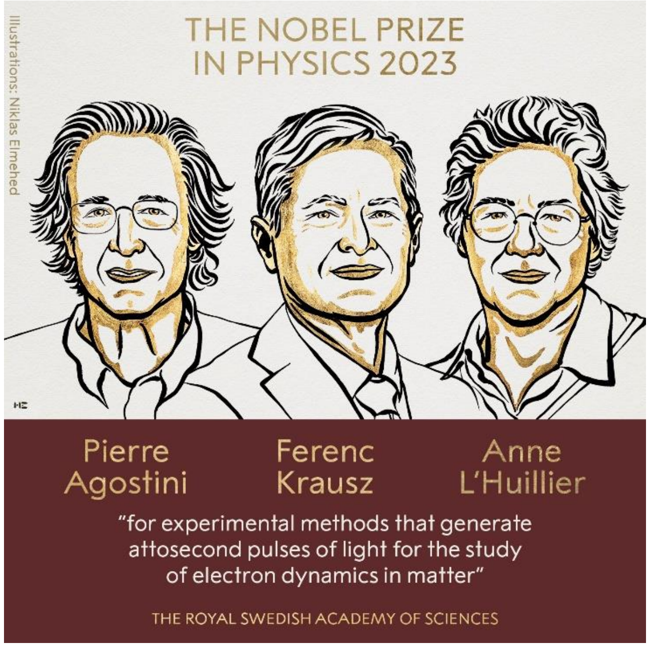
In praise of fundamental research

Agostini-DiMauro
Ultra-fast Atomic Physics Research Group

APS Meeting

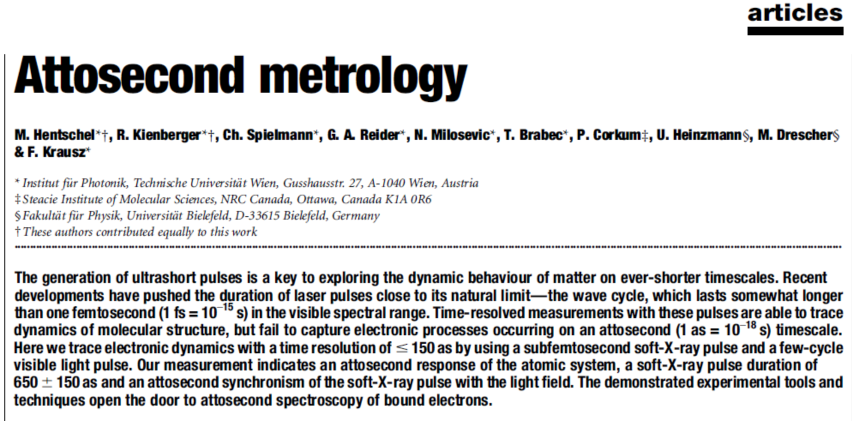
# 阿秒激光的产生

解决挑战，从飞秒到阿秒



飞秒放大激光 (1997)
首次实现孤立阿秒脉冲 (2001)

Ferenc Krausz



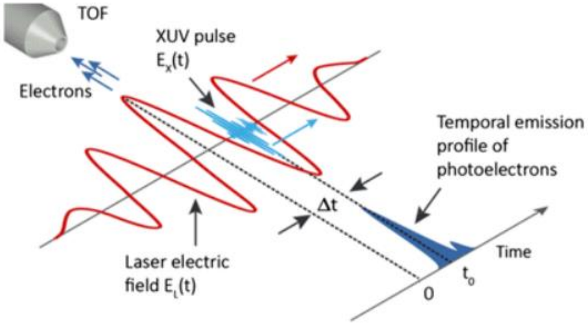
**articles**

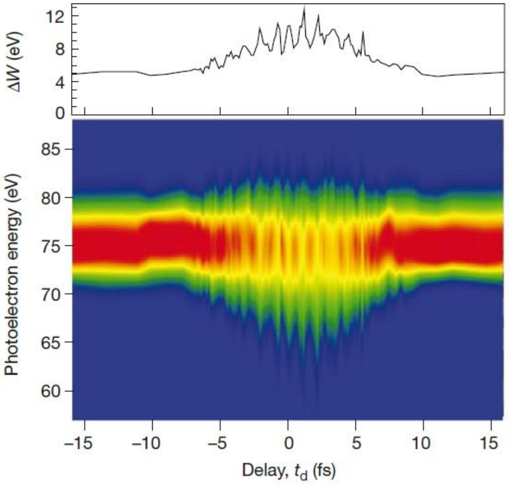
**Attosecond metrology**

**M. Hentschel*†, R. Kienberger*†, Ch. Spielmann*, G. A. Reider*, N. Milosevic*, T. Brabec*, P. Corkum‡, U. Heinzmann§, M. Drescher§ & F. Krausz***

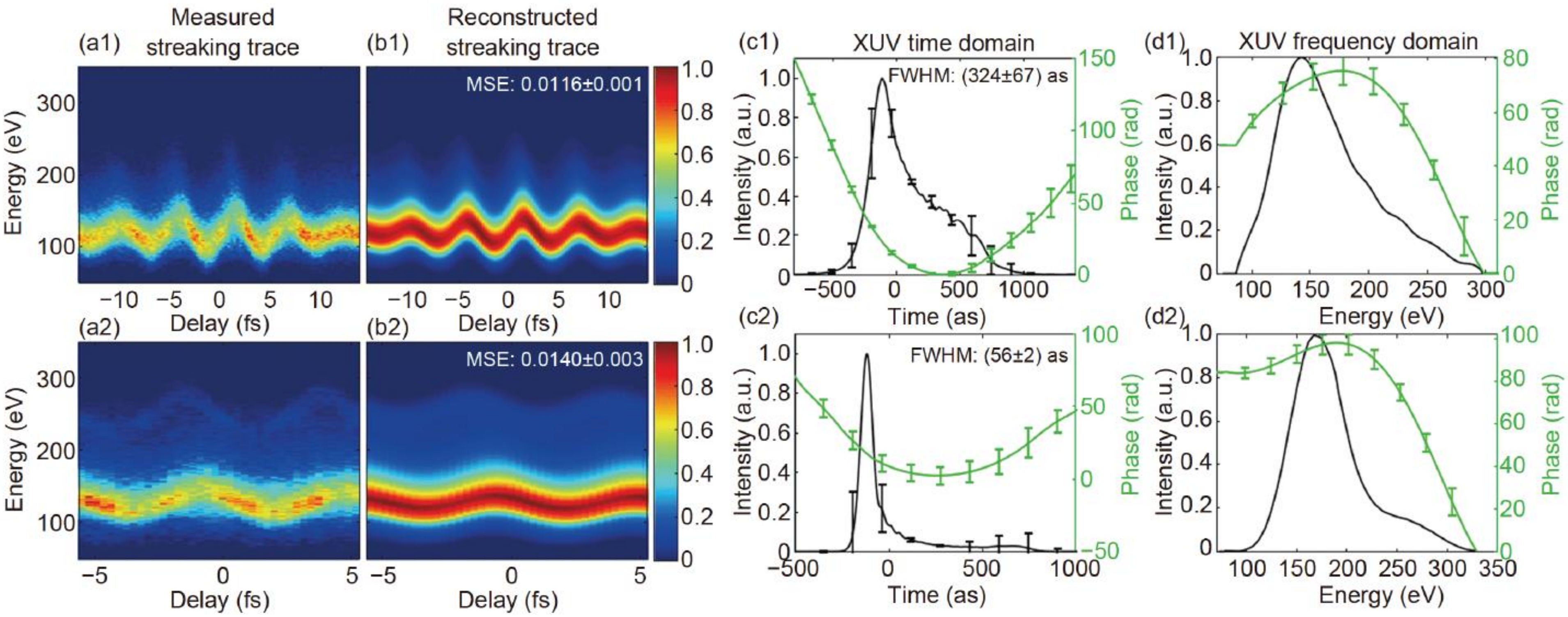
** Institut für Photonik, Technische Universität Wien, Gusshausstr. 27, A-1040 Wien, Austria*
*‡ Steacie Institute of Molecular Sciences, NRC Canada, Ottawa, Canada K1A 0R6*
*§ Fakultät für Physik, Universität Bielefeld, D-33615 Bielefeld, Germany*
*† These authors contributed equally to this work*

**The generation of ultrashort pulses is a key to exploring the dynamic behaviour of matter on ever-shorter timescales. Recent developments have pushed the duration of laser pulses close to its natural limit—the wave cycle, which lasts somewhat longer than one femtosecond (1 fs = $10^{-15}$ s) in the visible spectral range. Time-resolved measurements with these pulses are able to trace dynamics of molecular structure, but fail to capture electronic processes occurring on an attosecond (1 as = $10^{-18}$ s) timescale. Here we trace electronic dynamics with a time resolution of ≤ 150 as by using a subfemtosecond soft-X-ray pulse and a few-cycle visible light pulse. Our measurement indicates an attosecond response of the atomic system, a soft-X-ray pulse duration of 650 ± 150 as and an attosecond synchronism of the soft-X-ray pulse with the light field. The demonstrated experimental tools and techniques open the door to attosecond spectroscopy of bound electrons.**





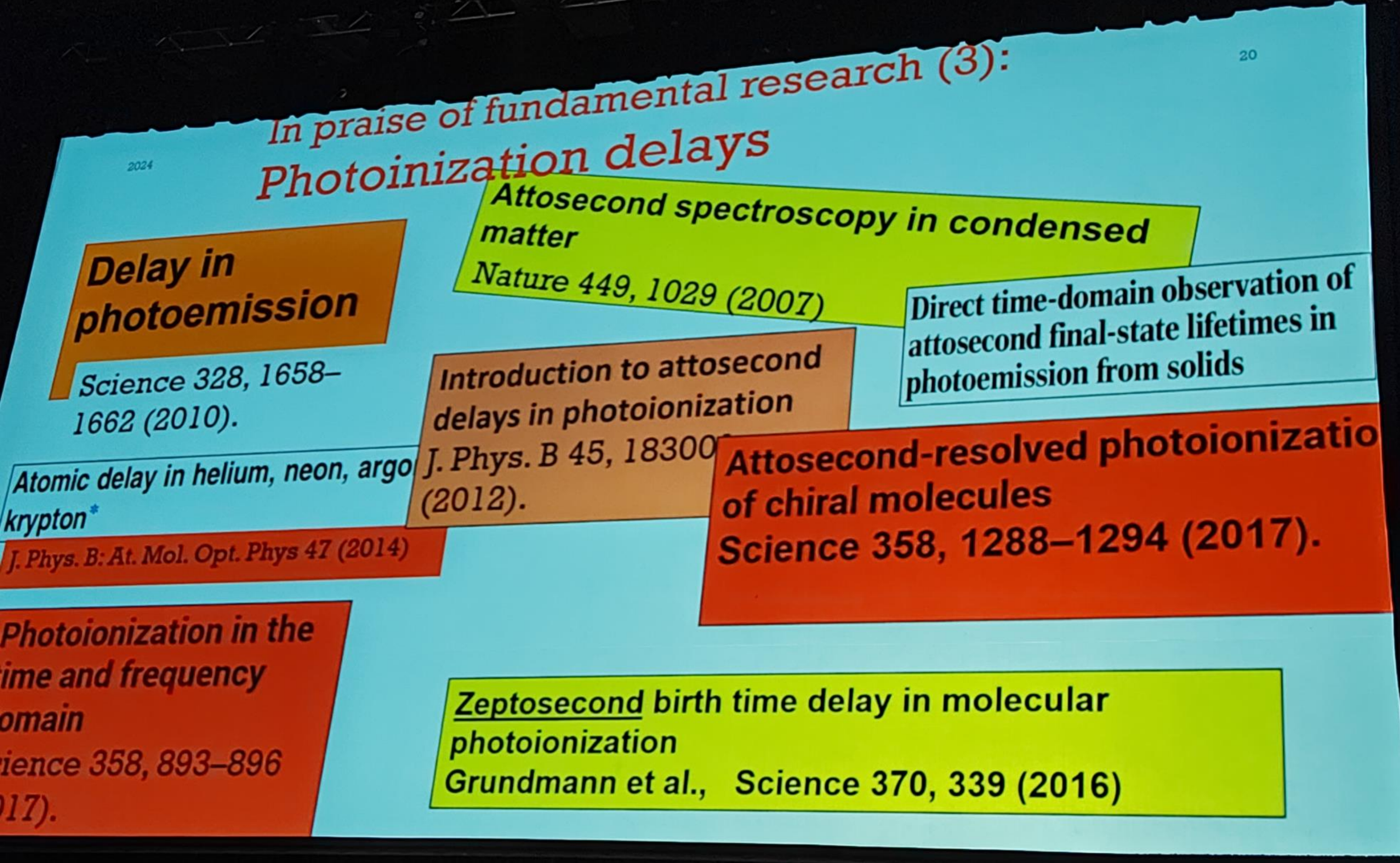
# Measuring single isolated attosecond pulse



Z. Zhu et al. Sci. Rep. 10, 1-7 (2020)


MAX-PLANCK-INSTITUT
FÜR QUANTENOPTIK
BMZ
Hans-Kopfermann Str. 1

2023.6.2



# In praise of fundamental research (3):
# Photoinization delays

**Delay in photoemission**
*Science 328, 1658–1662 (2010).*

**Attosecond spectroscopy in condensed matter**
Nature 449, 1029 (2007)

**Direct time-domain observation of attosecond final-state lifetimes in photoemission from solids**

**Introduction to attosecond delays in photoionization**
J. Phys. B 45, 18300
(2012).

***Atomic delay in helium, neon, argo krypton****
*J. Phys. B: At. Mol. Opt. Phys 47 (2014)*

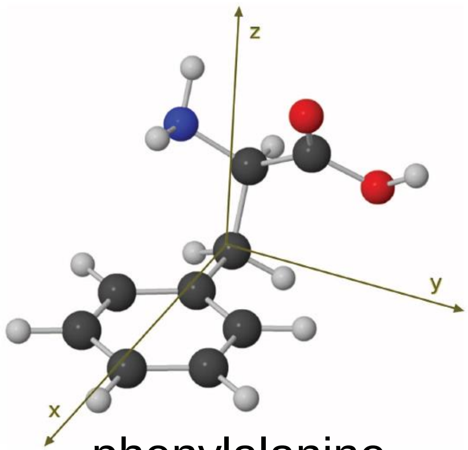
**Attosecond-resolved photoionizatio of chiral molecules**
**Science 358, 1288–1294 (2017).**

***Photoionization in the ime and frequency omain***
*ience 358, 893–896 17).*

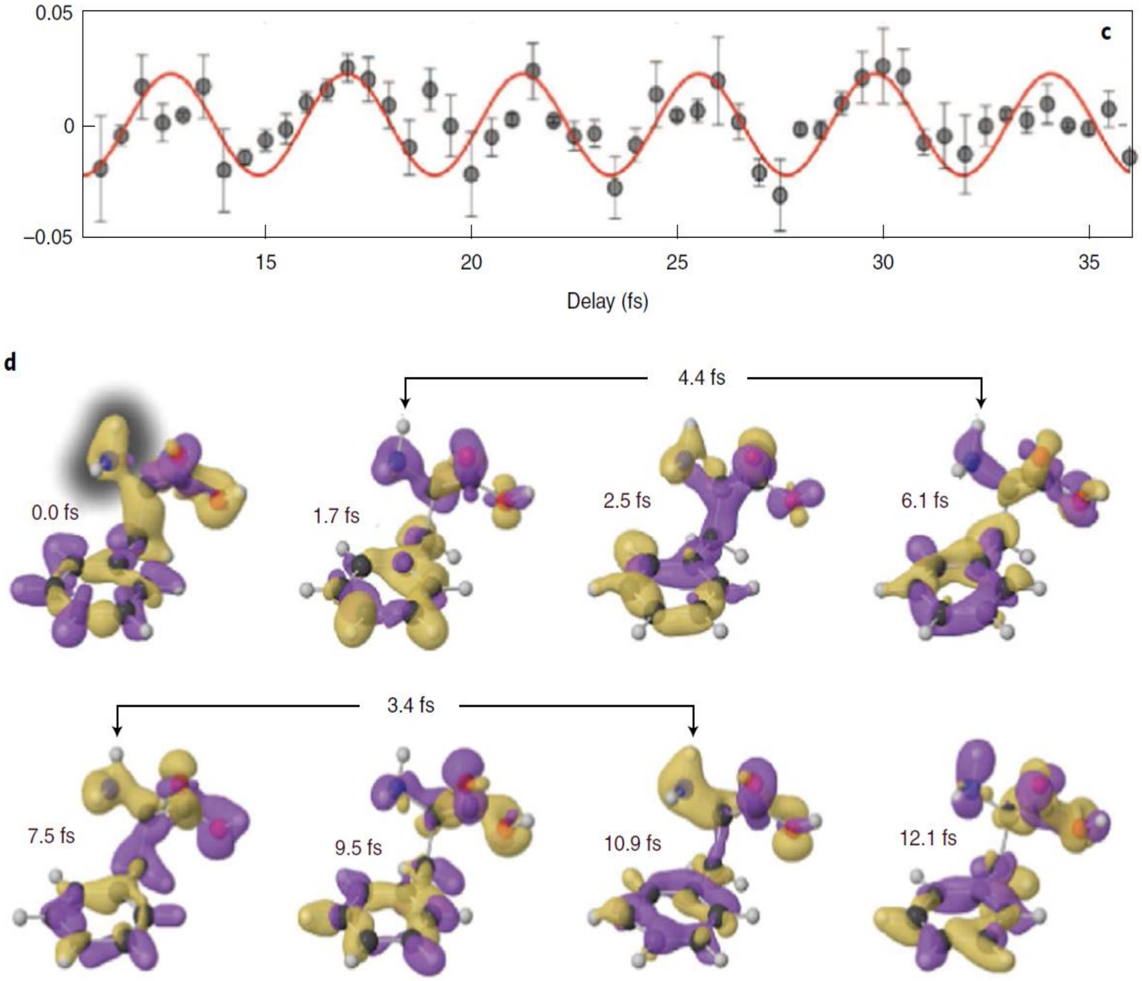
**Zeptosecond birth time delay in molecular photoionization**
**Grundmann et al., Science 370, 339 (2016)**

Courtesy: P. Agostini

# Ultrafast charge migration

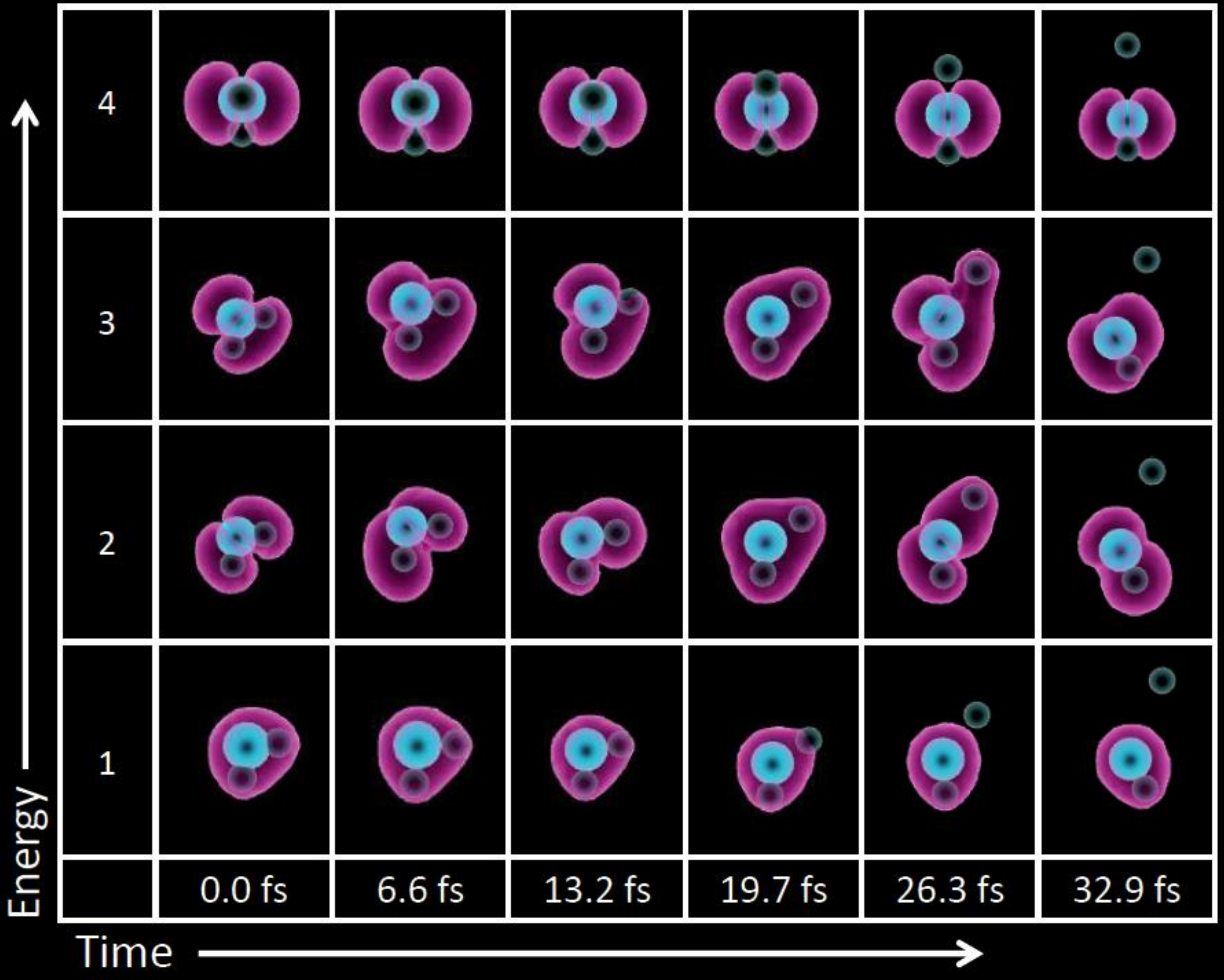


phenylalanine



F. Calegari et al., Science 346, 336 (2014).

# 水分子光分解



L. Yan, S. Meng. Sci China PMA 60,027032(2017).

# OUTLINE

I. What is atto?

II. Brief history of “time” (ultrafast studies)

III. The rise of attosecond science

IV. Attoscience in condensed matter
  - development of attosecond technology
  - attosecond dynamics & applications

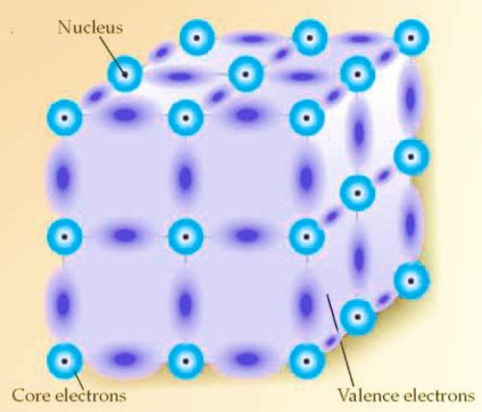
V. Advanced Attosecond Laser Facility

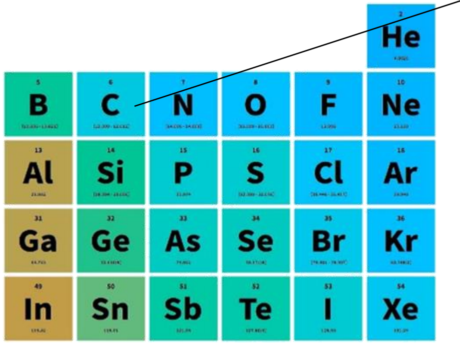
VI. Outlook

# 身边的物质世界：凝聚态物质

基本概念

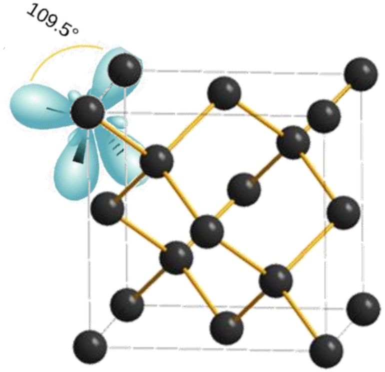
原子结构和周围的电子决定了材料的性质





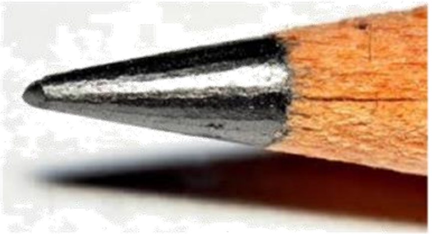
|  |  |  |  |  |  |
|---|---|---|---|---|---|
|  |  |  |  |  | He |
| B | C | N | O | F | Ne |
| Al | Si | P | S | Cl | Ar |
| Ga | Ge | As | Se | Br | Kr |
| In | Sn | Sb | Te | I | Xe |

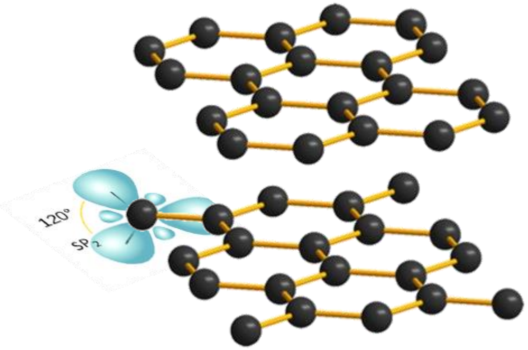
¥50000/g



Cubic

¥0.01/g

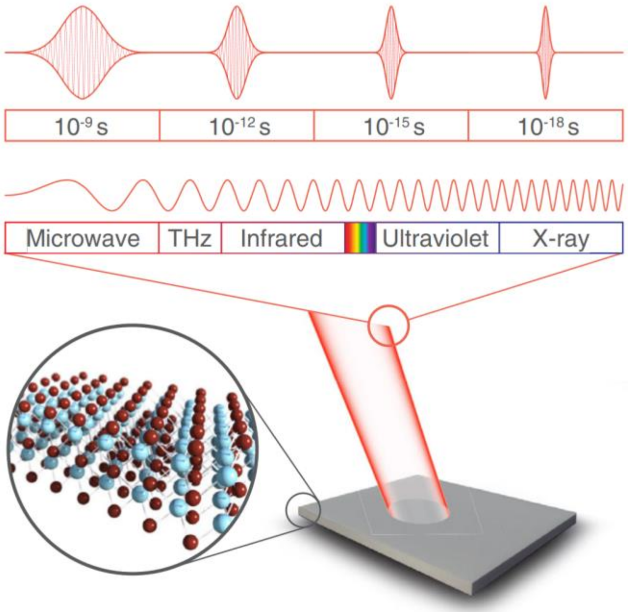


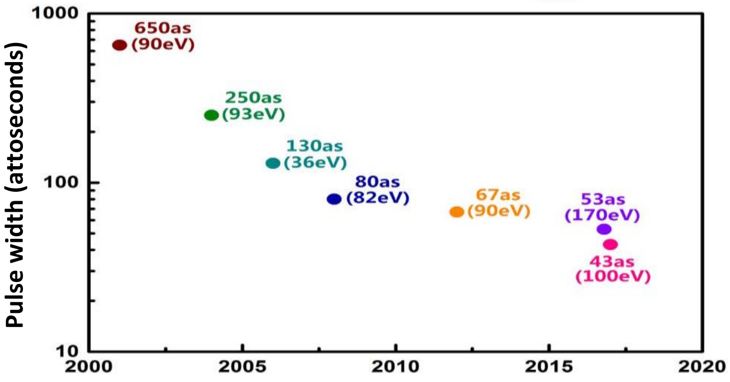


Hexagonal

# Developments in ultrafast techniques

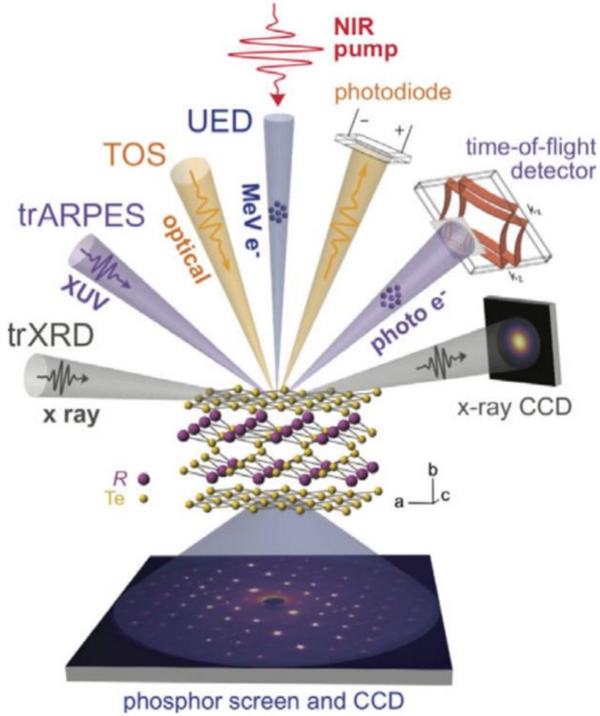
## Laser





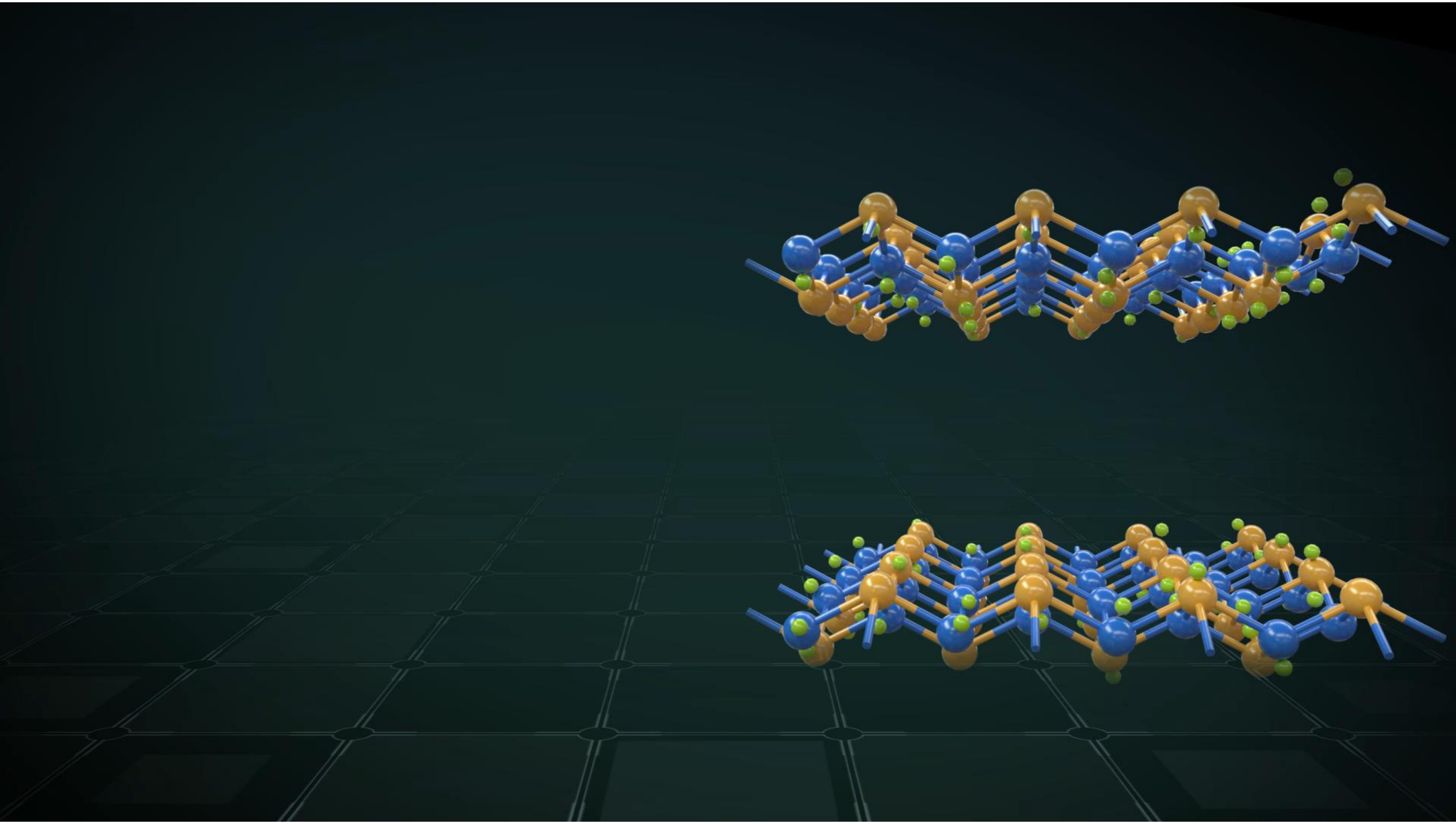
Rev. Mod. Phys. 93, 041002 (2021)

## Probes



Phys. Rev. Lett. 123, 097601 (2019)

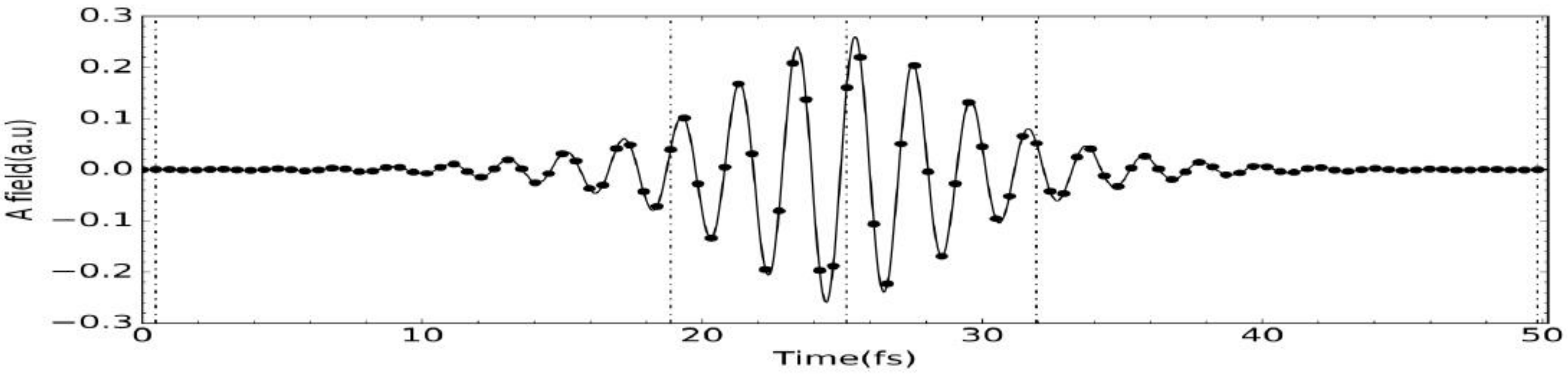
# Ultrafast light-matter Interaction

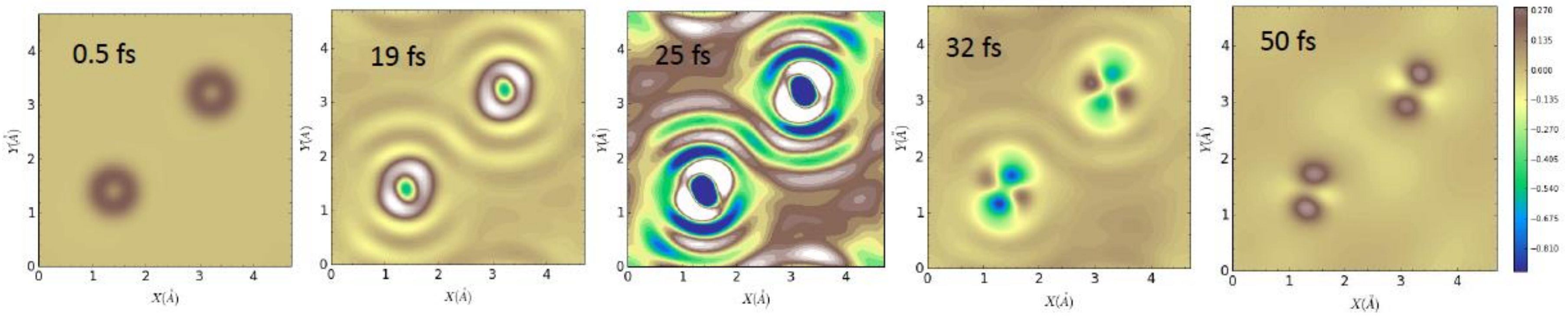


Nature Phys 13, 1056–1068 (2017).

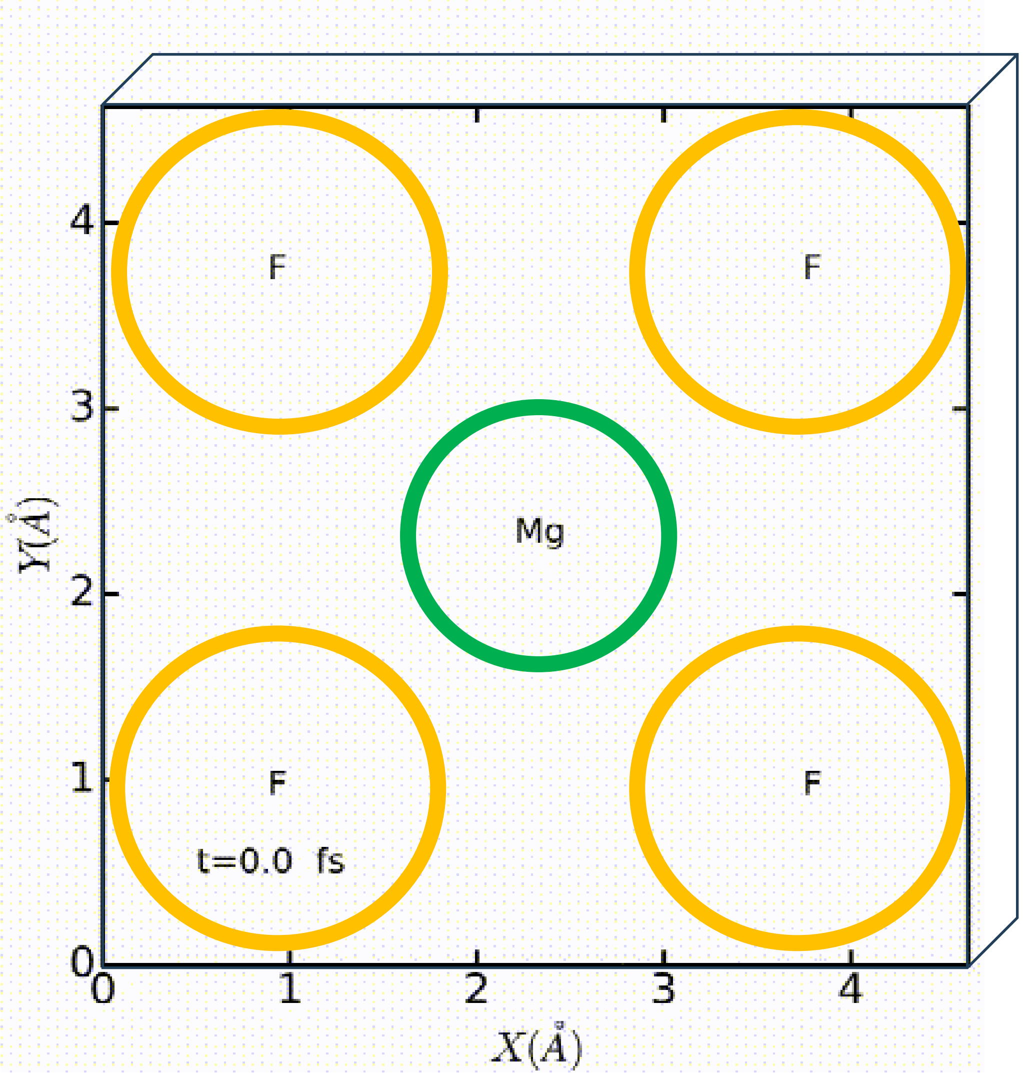
# 例：　固体价电子成像

## 固体电子的实时动态



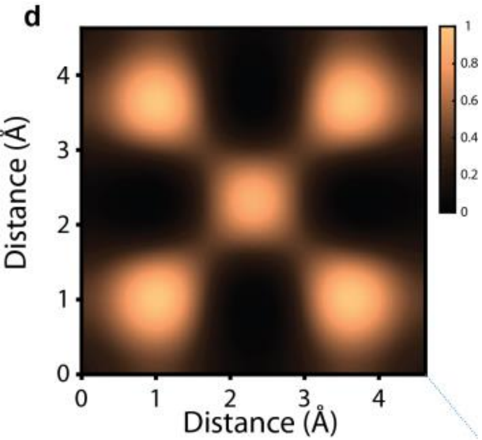


# 固体电子的实时动态



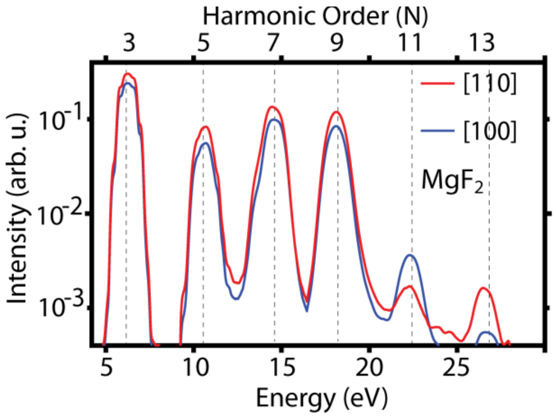
# 固体电子分布 ↔ 高次谐波谱

实 验



→

←



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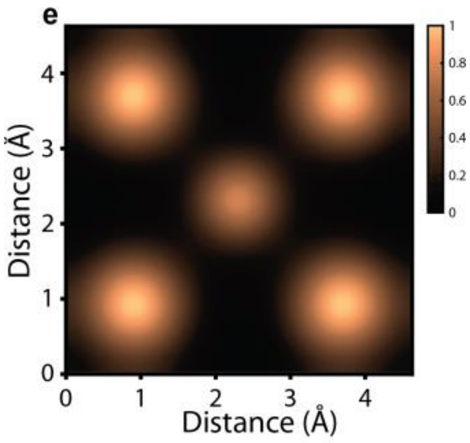
←

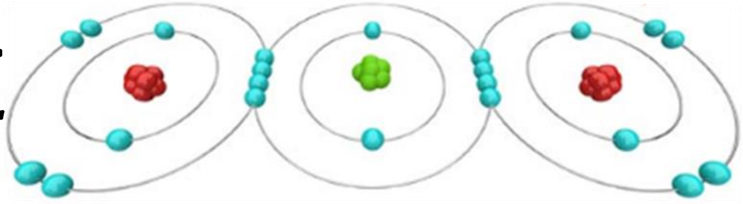
理 论



- 只对价电子敏感
- 同时测量幅度和相位
- 空间精度：皮米
- 时间分辨？

H. Lakhotia, H. Y. Kim, M. Zhan, S. Hu, S. Meng, E. Goulielmakis, Nature 583, 55 (2020).

# 量子力学基本问题



## 薛定谔方程（1926）

$$i\hbar\frac{\partial}{\partial t}\Psi(r,R,t) = \hat{H}_{tot}\Psi(r,R,t),\quad \hat{H}_{\text{tot}}(\boldsymbol{r},\boldsymbol{R},t)=\sum_{\alpha}\frac{P_\alpha^2}{2M_\alpha}+\sum_{i}\frac{p_i^2}{2m}+\sum_{i<j}\frac{e^2}{|\boldsymbol{r}_i-\boldsymbol{r}_j|}-\sum_{i,\alpha}\frac{Z_\alpha e^2}{|\boldsymbol{r}_i-\boldsymbol{R}_\alpha|}+\sum_{\alpha,\beta}\frac{Z_\alpha Z_\beta e^2}{|\boldsymbol{R}_\alpha-\boldsymbol{R}_\beta|}+U_{\text{ext}}(\boldsymbol{r},\boldsymbol{R},t)$$

## 玻恩-奥本海默近似（1927）

假定1. 电子波函数和原子核波函数分离： $\hat{H}_{\text{BO}}(\boldsymbol{r};\boldsymbol{R})\varphi(\boldsymbol{r};\boldsymbol{R}) = E_{\text{BO}}(\boldsymbol{R})\varphi(\boldsymbol{r};\boldsymbol{R})$

假定2. 电子始终处于基态上（无动力学）： $M_\alpha\ddot{\boldsymbol{R}} = -\nabla_R E_{\text{BO}}^{(0)}(\boldsymbol{R})$

**取得巨大成功：晶体结构预测、电子能带计算等，但不能处理激发态**

## 超越玻恩-奥本海默近似

（玻恩-黄展开，1954；严格因子化Gross，2010）

$$\Psi(r,R,t)\equiv\Phi_R(r,t)\Xi(R,t)\cong\Phi_{R_0(t)}(r)\ \delta(R(t)-R_0(t))\Rightarrow\Phi_{R(t)}(r,\boldsymbol{t})\ \Xi(R(t)-R_0(t))$$

**优点：波函数的动力学演化，结合第一性原理计算**

# 发展第一性原理激发态动力学方法

- ***模型哈密顿量 → 第一性原理计算***

$$\hat{H}_{\mathrm{tot}}(\boldsymbol{r},\boldsymbol{R},t)=\sum_{\alpha}\frac{P_\alpha^2}{2M_\alpha}+\sum_i\frac{p_i^2}{2m}+\sum_{i<j}\frac{e^2}{|\boldsymbol{r}_i-\boldsymbol{r}_j|}-\sum_{i,\alpha}\frac{Z_\alpha e^2}{|\boldsymbol{r}_i-\boldsymbol{R}_\alpha|}+\sum_{\alpha,\beta}\frac{Z_\alpha Z_\beta e^2}{|\boldsymbol{R}_\alpha-\boldsymbol{R}_\beta|}+U_{\mathrm{ext}}(\boldsymbol{r},\boldsymbol{R},t)$$

- ***电子运动：实时演化多体波函数（TDDFT）***

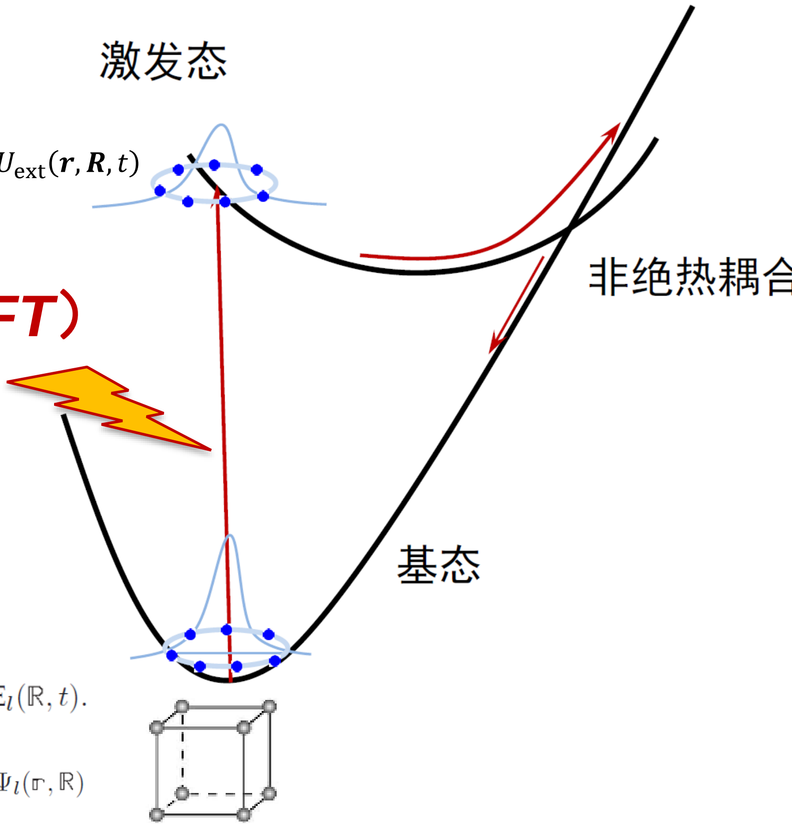
$$i\hbar\frac{\partial\phi_j(\mathbf{r},t)}{\partial t}=\left(-\frac{\hbar^2}{2m}\nabla_{\mathbf{r}}^2+v_s[\rho](\mathbf{r},t)\right)\phi_j(\mathbf{r},t).$$

- ***原子核运动：非绝热效应***

$$i\frac{\partial}{\partial t}\Xi_k(\mathbb{R},t)=\left[-\sum_{j=1}^{N_n}\frac{\nabla_{\mathbf{R}_j}^2}{2M_j}+E_k(\mathbb{R})\right]\Xi_k(\mathbb{R},t)-\sum_{l=0}^{\infty}\sum_{j=1}^{N_n}\frac{1}{2M_j}\left[\boldsymbol{\tau}_{kl}^j\cdot\nabla_{\mathbf{R}_j}+\widetilde{\tau}_{kl}^j\right]\Xi_l(\mathbb{R},t).$$

**非绝热耦合系数：** $\boldsymbol{\tau}_{kl}^j=\int d\mathbb{r}\ \Psi_k^*(\mathbb{r},\mathbb{R})\nabla_{\mathbf{R}_j}\Psi_l(\mathbb{r},\mathbb{R})$

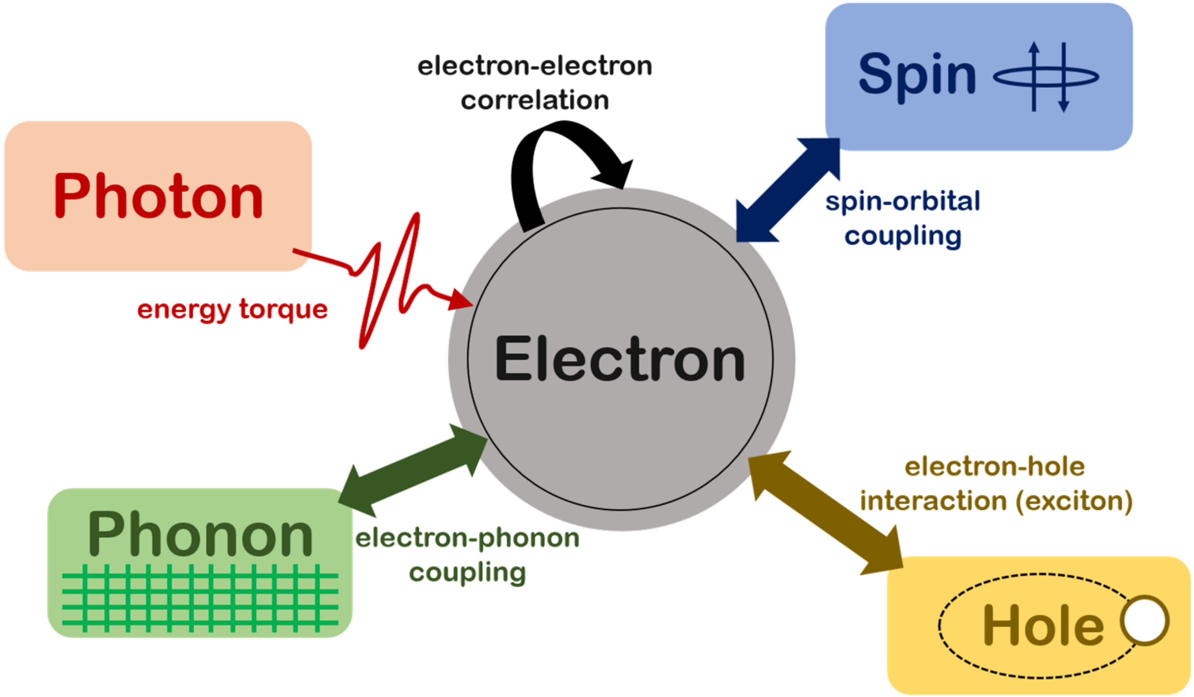
$$\widetilde{\tau}_{kl}^j=\int d\mathbb{r}\ \Psi_k^*(\mathbb{r},\mathbb{R})\nabla_{\mathbf{R}_j}^2\Psi_l(\mathbb{r},\mathbb{R})$$



- **非绝热**（电子跃迁）
- **非微扰**（实时）
- **非平衡**（外场）

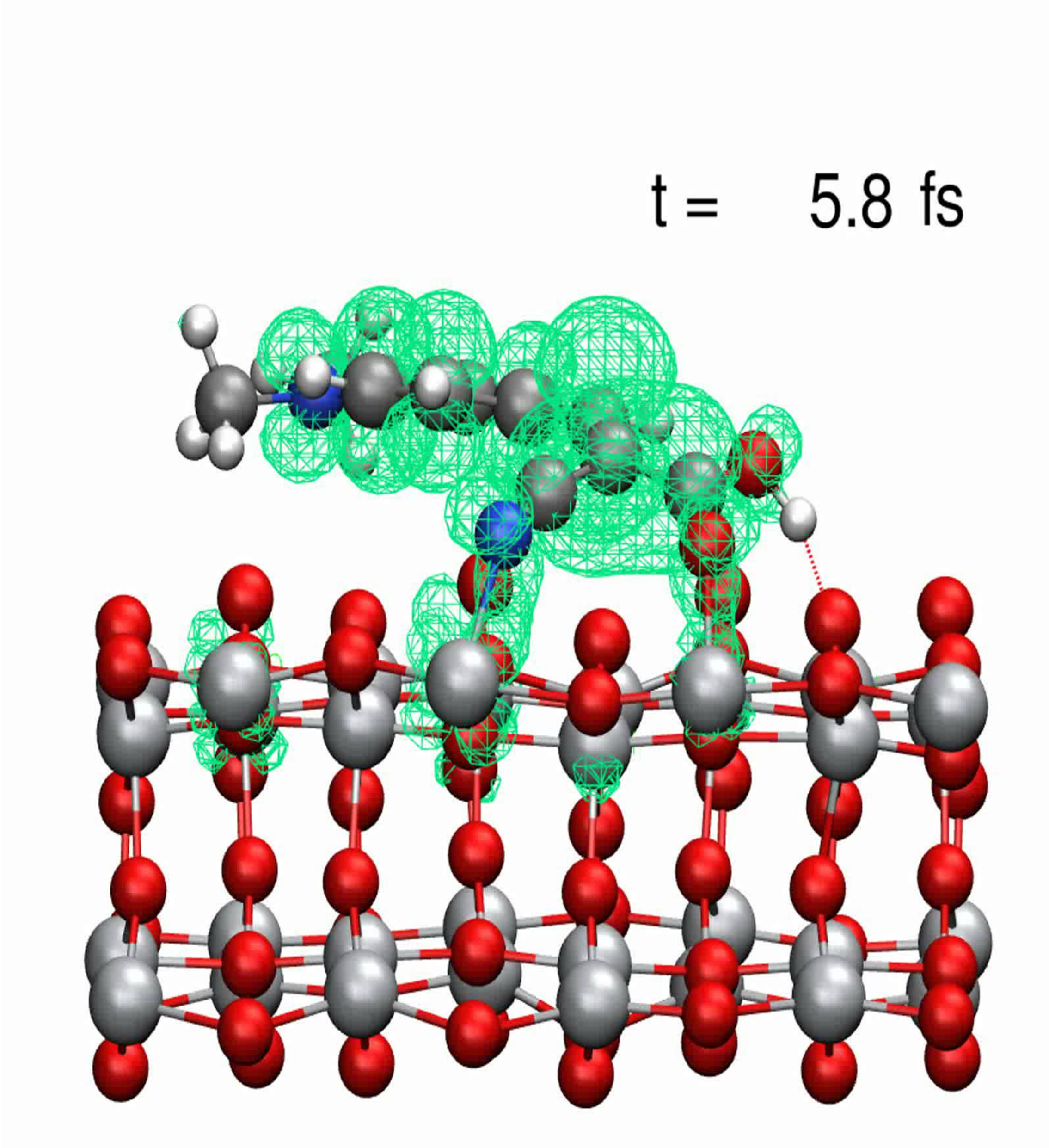
# 凝聚态体系中的阿秒动力学

超快凝聚态动力学中复杂的多体相互作用



S. Hu et al., Chin. Phys. Lett. 40 117801(2023).

# Electron Injection Dynamics

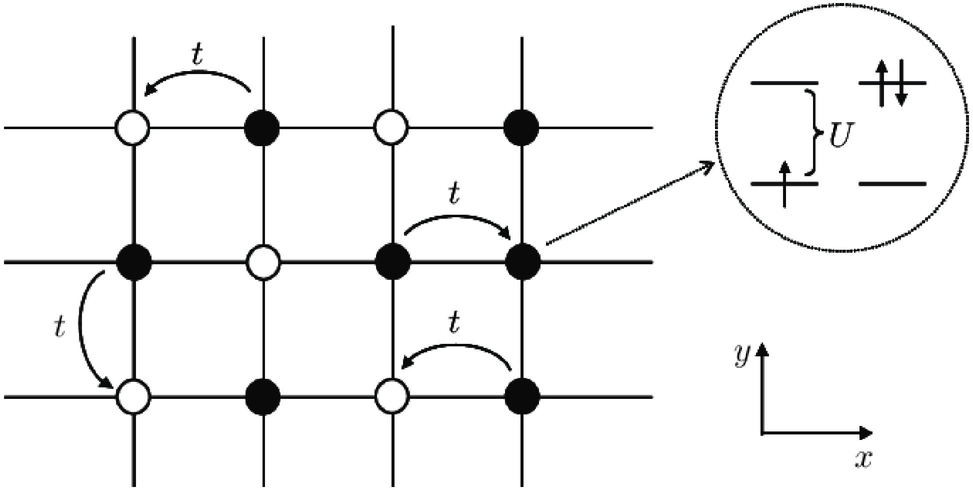


$$\Phi_{\text{inject}} = 1 \Big/ \left(1 + \frac{\tau_{\text{inj}}}{\tau_{\text{relax}}}\right)$$

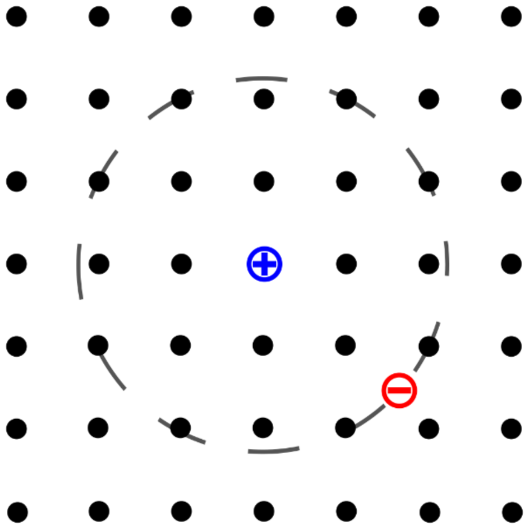
# 凝聚态体系中的阿秒动力学

## 阿秒库伦关联动力学

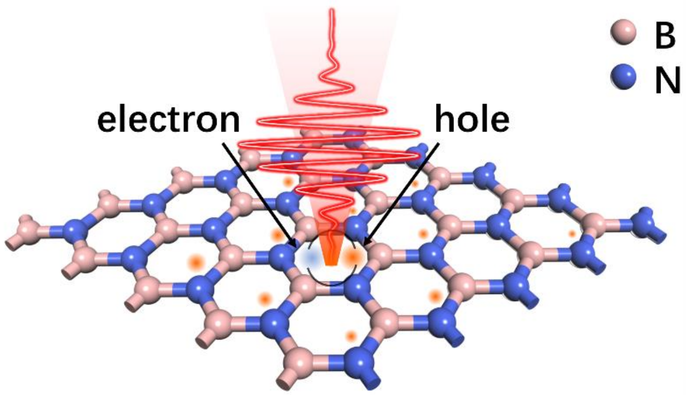
电子-电子关联动力学

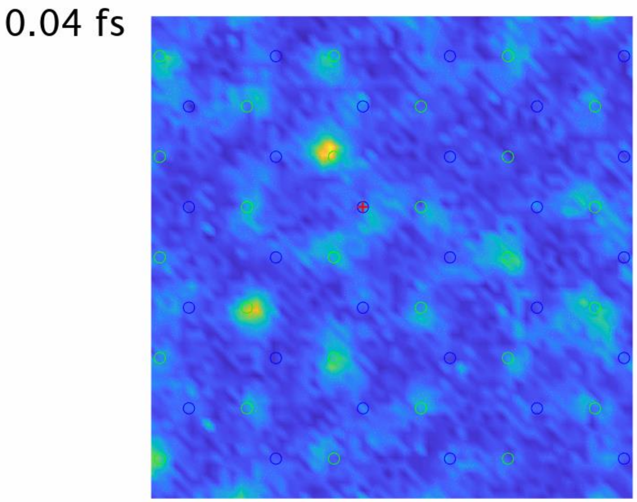


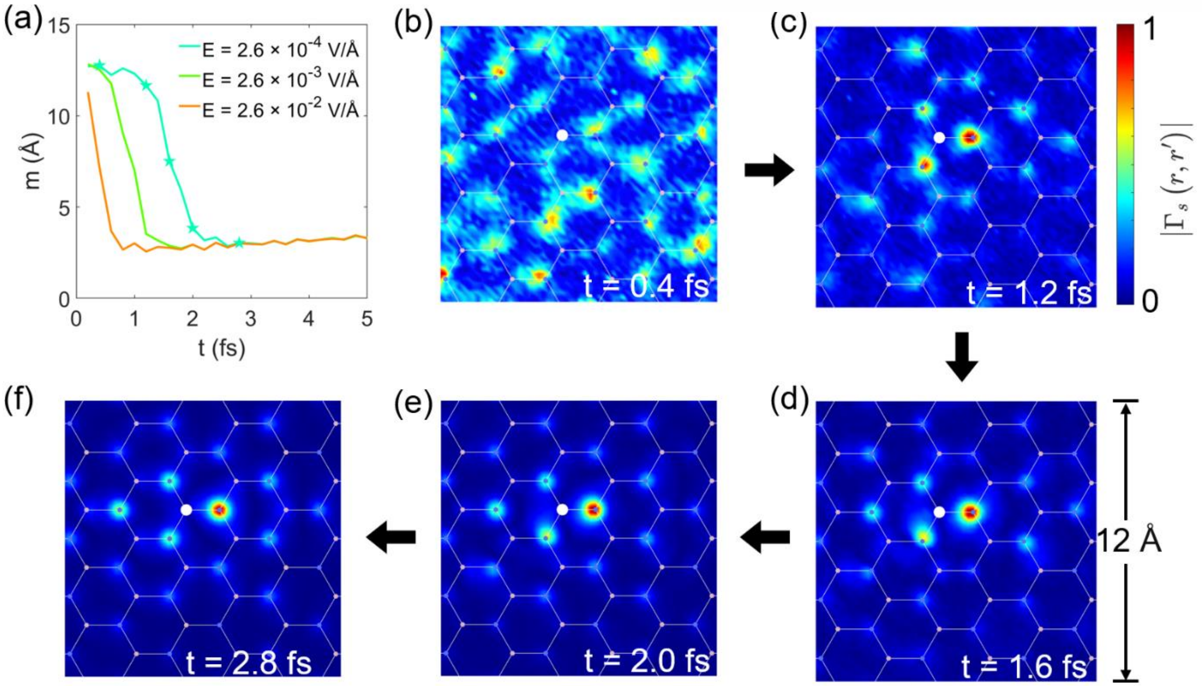
电子-空穴关联 (激子) 动力学



# 光激发单层BN：激子的形成过程







Qing Chen, Sheng Meng et al. (2024).

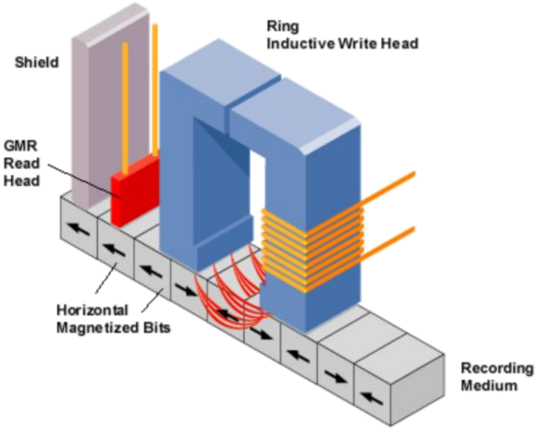
# 凝聚态体系中的阿秒动力学

## 阿秒磁性动力学

### 磁存储

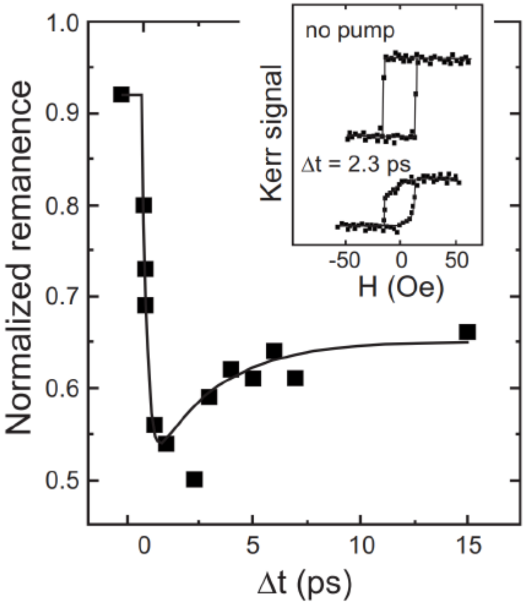
### 强磁场产生

### 磁性调控



时间极限： 2 ps

### 光致退磁



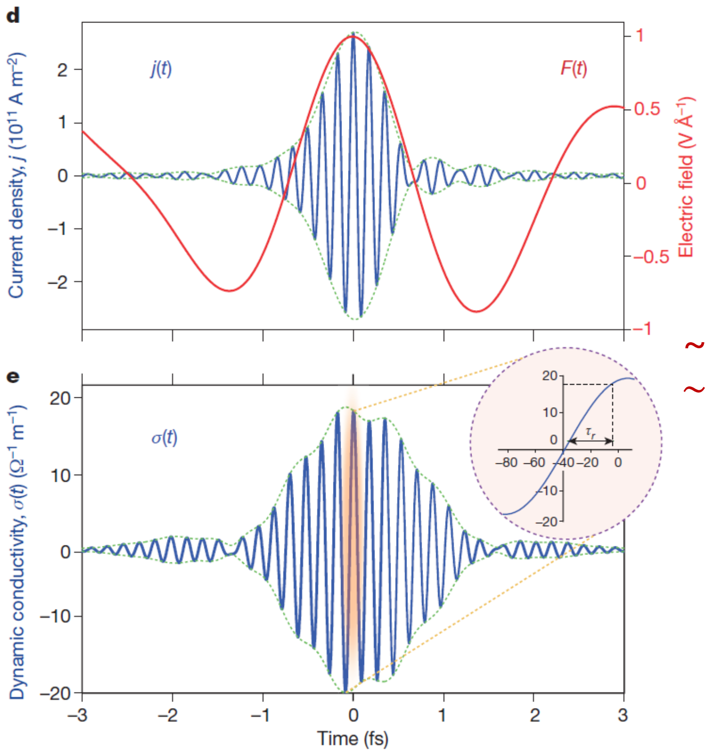
时间 < 1 ps

E. Beaurepaire et al., Phys. Rev. Lett. 76 , 4250 (1996).
I. Tudosa et al., Nature 428 , 831 (2004).

# 凝聚态体系中的阿秒动力学

凝聚态体系阿秒动力学的应用

## 阿秒电子计量学



~ 30 as

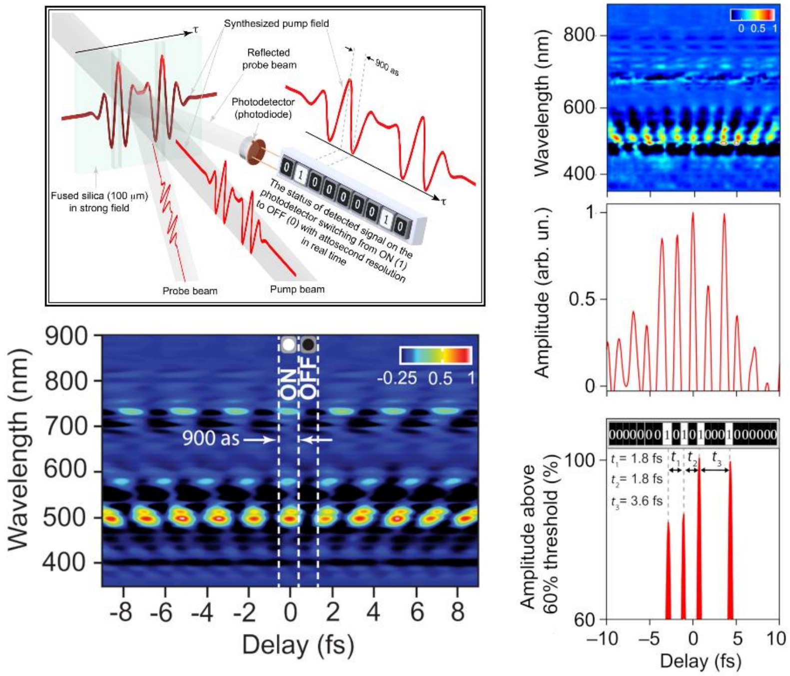
~ 8 PHz

M. Garg et al., Nature 538, 359–363 (2016).

## 超快数据编码

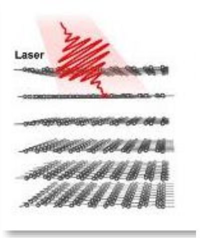
~ 900 as

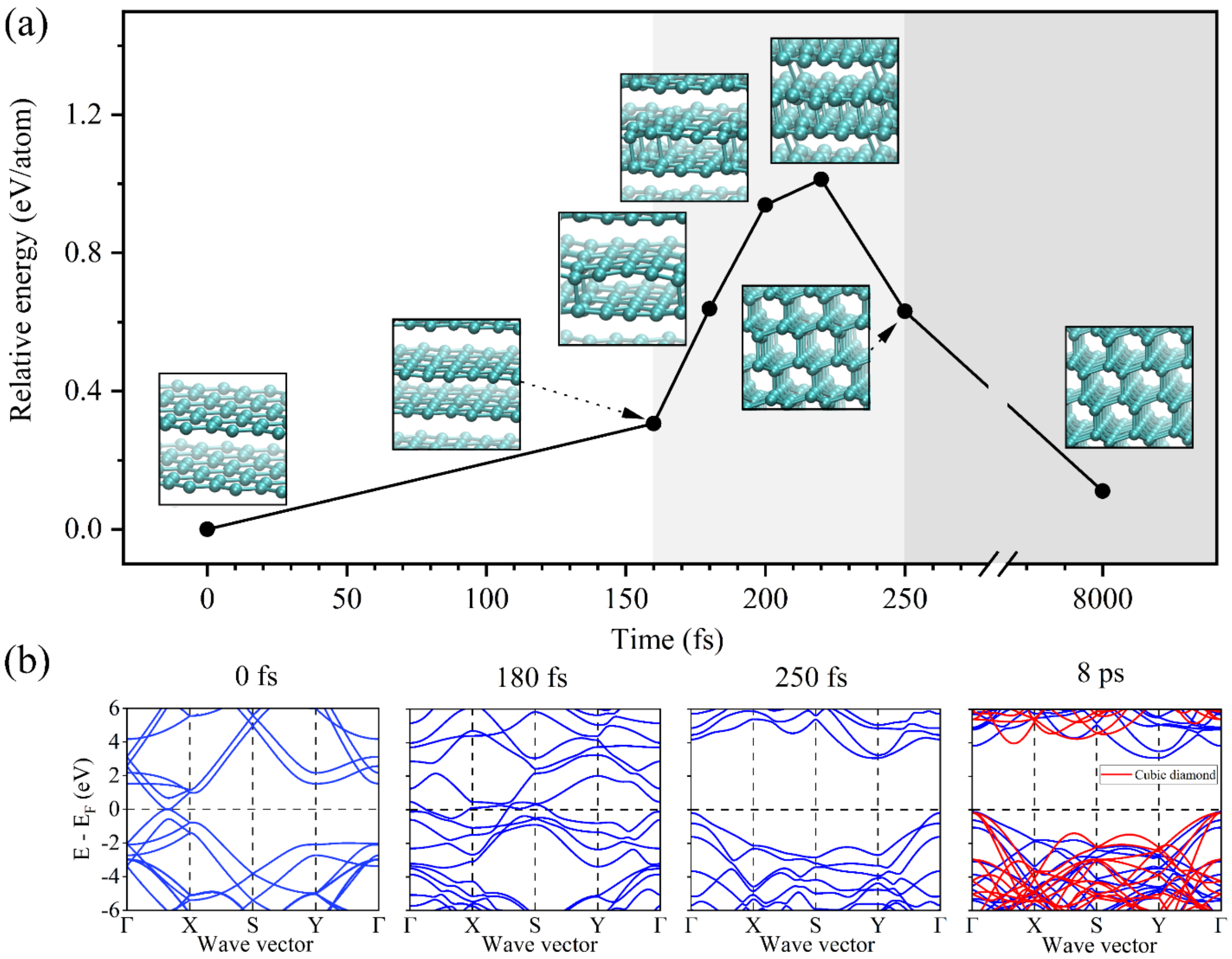
~ 1 PHz



D. Hui et al., Sci. Adv. 9 eadf1015 (2023).

# 超快激光调控相变

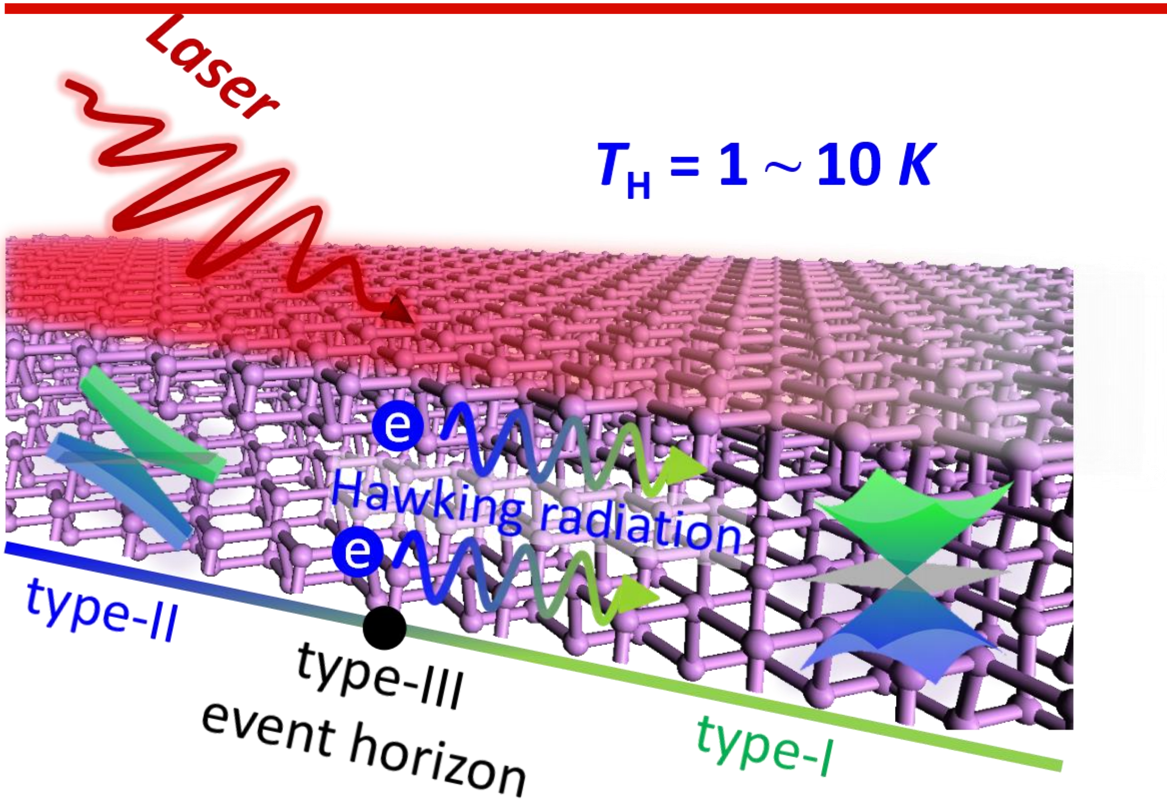


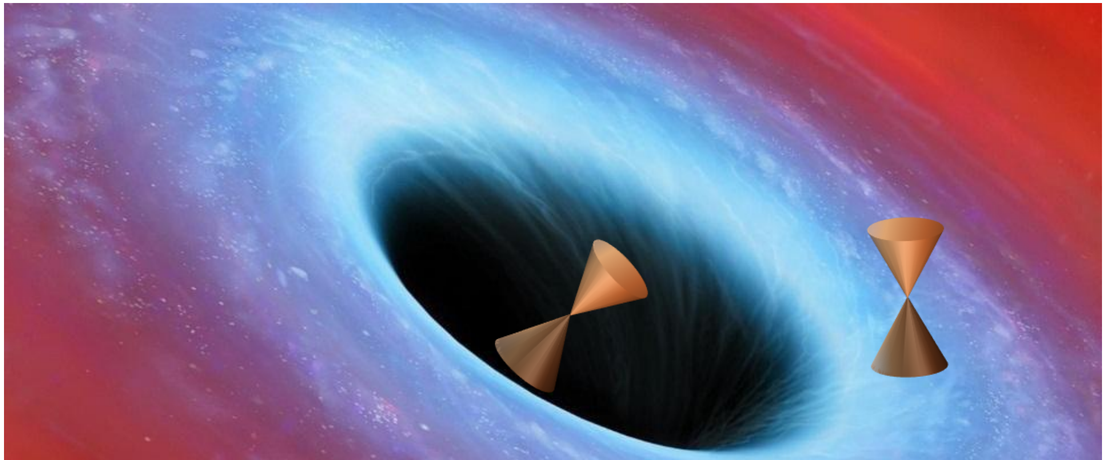


C.C. Song, M.X. Guan, Y.Z. Jia, D.Q. Chen, J.Y. Xu, C. Zhang*, S. Meng*. npj Comput. Mater. 9, 76 (2023)

Y.Z. Jia et al. (2024)

# Simulating Black Hole and Hawking Radiation





H Liu et al., Phys. Rev. Lett. 120, 237403 (2018)
H Liu et al., Chin. Phys. Lett. 37, 067101 (2020)

# OUTLINE

I. What is atto?

II. Brief history of “time” (ultrafast studies)

III. The rise of attosecond science

IV. Attoscience in condensed matter
  - development of attosecond technology
  - attosecond dynamics & applications

V. Advanced Attosecond Laser Facility

VI. Outlook

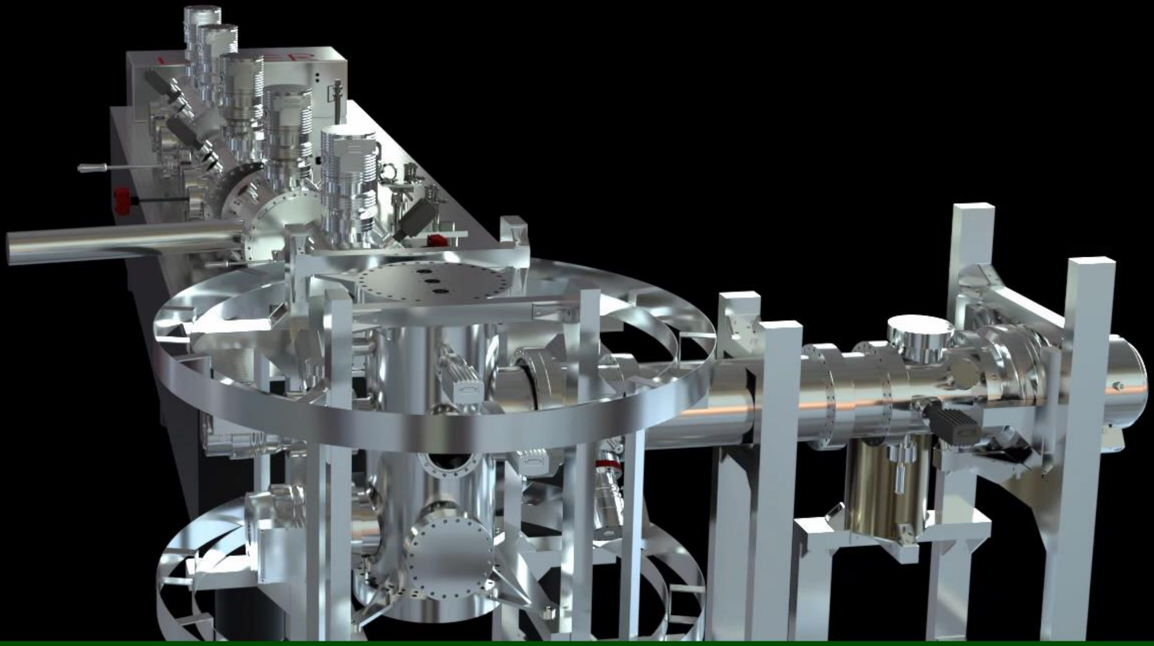
# 先进阿秒激光大科学装置

**阿秒脉冲：**
追踪和控制
电子运动

固体物理学
纳米技术
原子物理学
医学
物理化学
X射线/粒子源
分子生物学
信息技术
生物医学
计量学

将为多学科领域研究提供前所未有的探测和操控手段

# 阿秒束线与应用终端简介



# 预期可以解决的重大科学问题

**束线和终端相互配合，为物理学、化学、材料科学、信息科学、能源科学以及生命科学中的基础研究突破提供支撑**

| 束线 | 极紫外阿秒激光<br>5条束线 | 软X射线阿秒激光<br>3条束线 | 太赫兹辐射源<br>2条束线 |
|---|---|---|---|
| 重点领域 | 物理、化学、能源 | 损伤、加工、军工 | 生物、药物、信息 |
| 重大科学问题 | 固体电子学<br>阿秒磁学<br>原子分子物理<br>量子相干过程 | 含能材料<br>辐照损伤<br>瞬态成像 | 光化学反应<br>库珀对 |
| 对应终端 | 1 ~ 11 | 12 ~ 18 | 19 ~ 22 |

# OUTLINE

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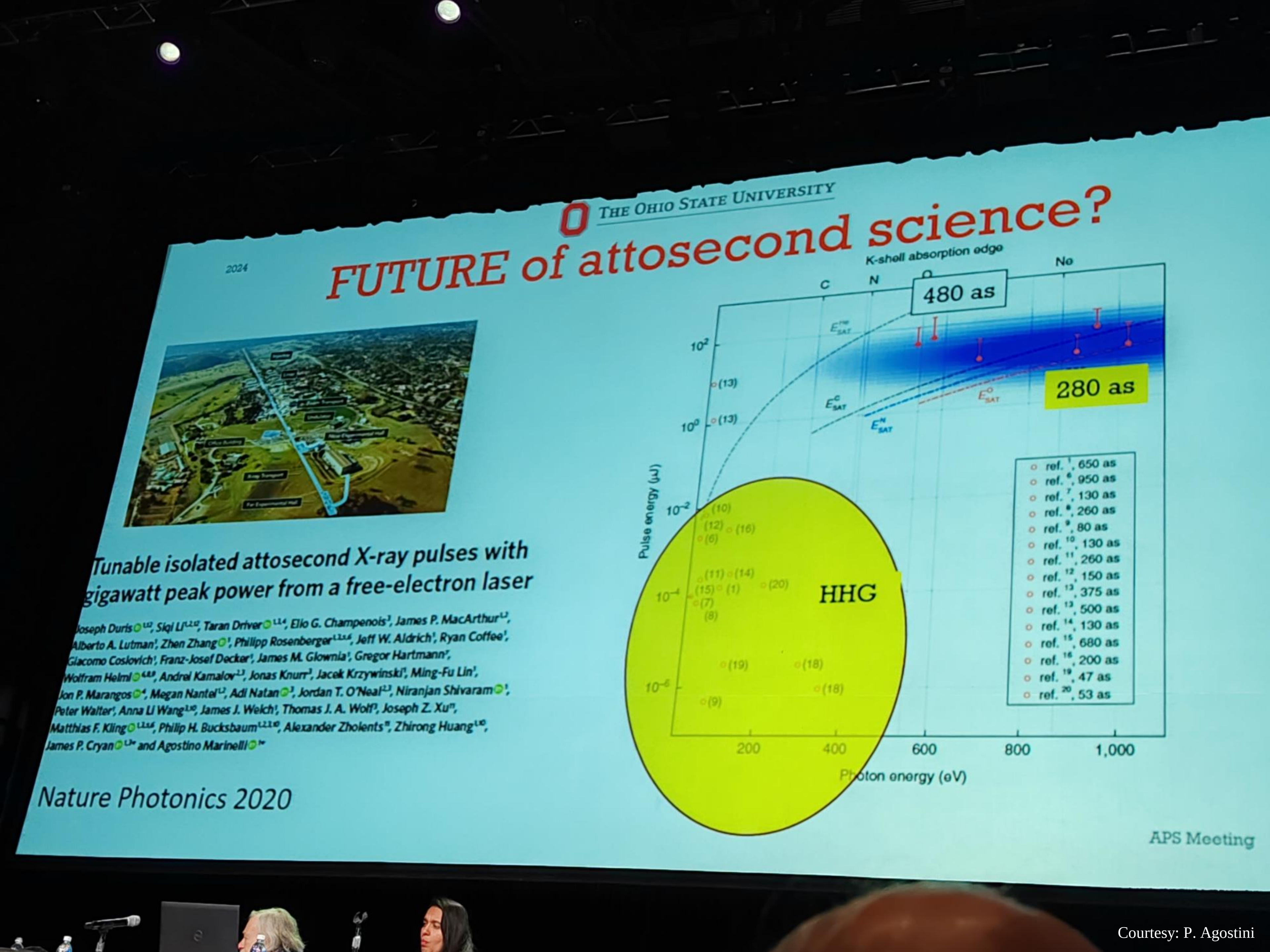


THE OHIO STATE UNIVERSITY

2024

# FUTURE of attosecond science?

**Tunable isolated attosecond X-ray pulses with gigawatt peak power from a free-electron laser**

Joseph Duris[1,12], Siqi Li[1,2,12], Taran Driver[1,3,4], Elio G. Champenois[3], James P. MacArthur[1,2], Alberto A. Lutman[1], Zhen Zhang[1], Philipp Rosenberger[1,3,5,6], Jeff W. Aldrich[1], Ryan Coffee[1], Giacomo Coslovich[1], Franz-Josef Decker[1], James M. Glownia[1], Gregor Hartmann[7], Wolfram Helml[4,8,9], Andrei Kamalov[2,3], Jonas Knurr[3], Jacek Krzywinski[1], Ming-Fu Lin[1], Jon P. Marangos[4], Megan Nantel[1,2], Adi Natan[3], Jordan T. O'Neal[2,3], Niranjan Shivaram[1], Peter Walter[1], Anna Li Wang[3,10], James J. Welch[1], Thomas J. A. Wolf[3], Joseph Z. Xu[11], Matthias F. Kling[1,3,5,6], Philip H. Bucksbaum[1,2,3,10], Alexander Zholents[11], Zhirong Huang[1,10], James P. Cryan[1,3*] and Agostino Marinelli[1*]

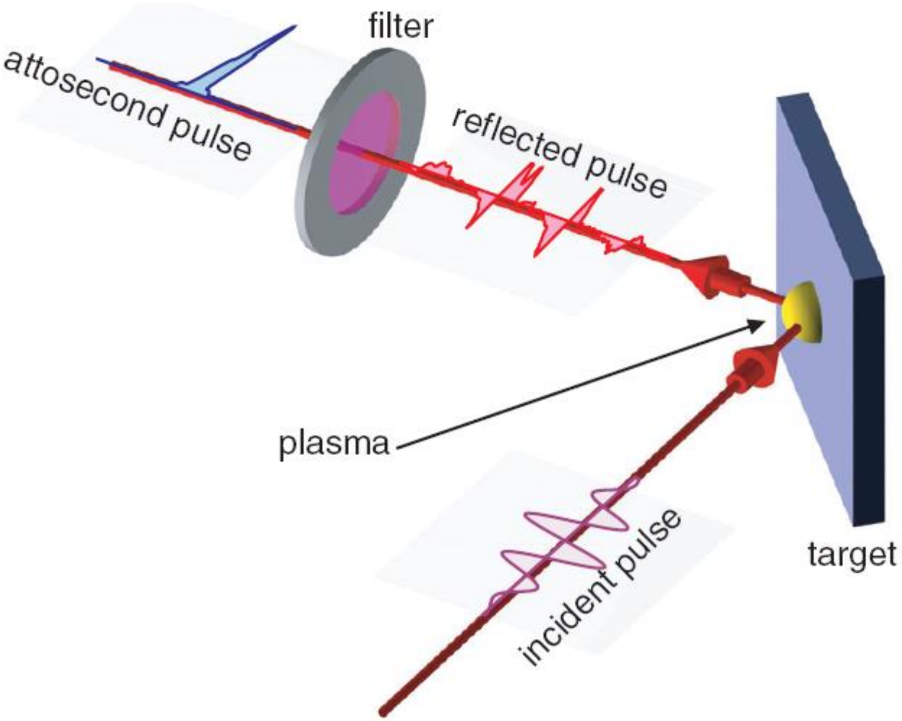
*Nature Photonics 2020*

APS Meeting

Courtesy: P. Agostini

# 更短阿秒、仄秒光脉冲产生—相对论振镜实验方案

**通过高能量激光与固体密度等离子体相互作用，能够产生10阿秒以下甚至仄秒（$10^{-21}$s）的超短脉冲，并且所获得的脉冲能量更高，是未来超快激光进一步推进的重要方向**



**焦点＜1μm**

**要实现0.3阿秒，需要峰值功率密度**P= $6 \times 10^{20}$ W/$cm^2$,**需要激光20fs，1拍瓦**

**要实现3仄秒，需要峰值功率密度**P= $1.38 \times 10^{22}$ W/$cm^2$,**需要激光10fs，2拍瓦**

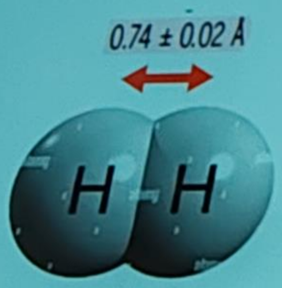
**要获得更短更强的阿秒、仄秒脉冲，需要不断提高激光峰值功率**

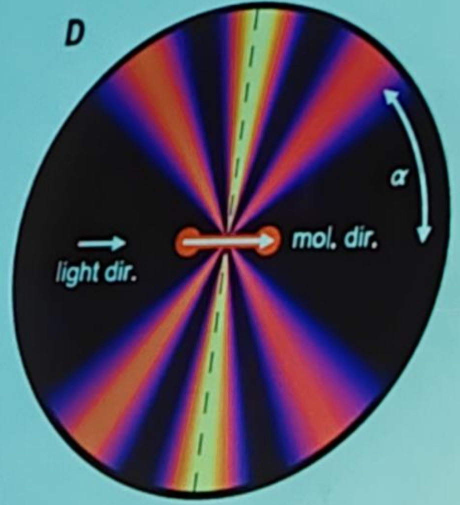
The Ohio State University

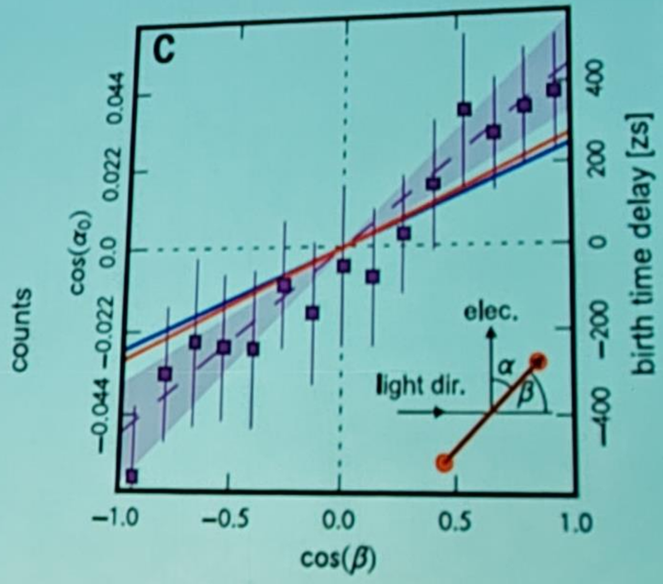
# Zeptosecond birth time delay in molecular photoionization

Sven Grundmann[1]*, Daniel Trabert[1], Kilian Fehre[1], Nico Strenger[1], Andreas Pier[1], Leon Kaiser[1], Max Kircher[1], Miriam Weller[1], Sebastian Eckart[1], Lothar Ph. H. Schmidt[1], Florian Trinter[1,2,3], Till Jahnke[1]*, Markus S. Schöffler[1], Reinhard Dörner[1]*

*Science* **370**, 339–341 (2020)







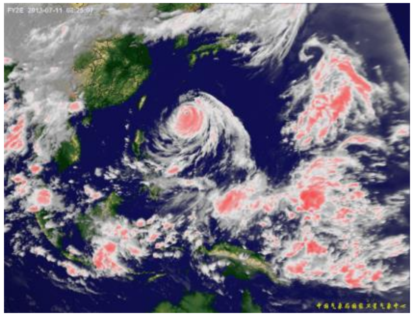
$\Delta t = 247$ zs

# 总　结

## 阿秒激光—观察世界的一个新视角

《星月夜》

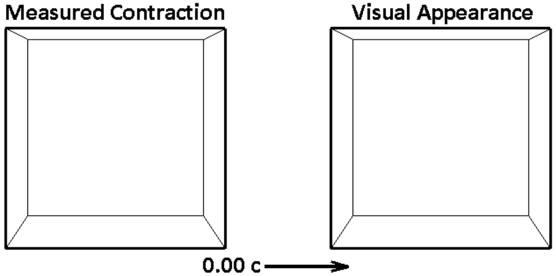
—— 梵高



来源：中国气象局

《花底草帽女子》

—— 毕加索



From zh.wikipedia.org

# Atomscale Energy Conversion and Quantum Dynamics