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Technical Groups

Ultrafast Dynamics of Molecules in Strong Laser Fields

Featuring Jian Wu, East China Normal University
10 February 2022

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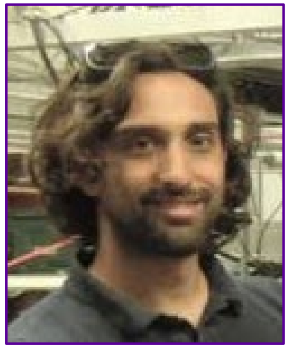

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About Our Technical Group

Our technical group provides a focus for activities related to the development and application of high-intensity lasers as well as novel XUV and x-ray sources

Our mission is to connect the 780+ members of our community through technical events, webinars, networking events, and social media.

Our past activities have included:

- High-Harmonic Sources for Material Development and Metrology in the Semiconductor Industry
- Frontiers of Ultrafast X-Ray Spectroscopy and Imaging Virtual Seminar
- Seeing Electrons In Action Webinar
- Panel discussions at CLEO: 2019 and CLEO: 2016

Connect with our Technical Group

Join our online community to stay up to date on our group's activities. You also can share your ideas for technical group events or let us know if you're interested in presenting your research.

Ways to connect with us:

- Our website at www.optica.org/OH
- On LinkedIn at www.linkedin.com/groups/8356401/
- On Facebook at www.facebook.com/OpticaShortWavelengthTG/
- On Twitter at #OSAOH
- Email us at TGactivities@optica.org

Today’s Speaker

Jian Wu

East China Normal University

Jian Wu is the Director and Professor of the State Key Laboratory of Precision Spectroscopy, East China Normal University. His research focuses on the measurement and control of the ultrafast dynamics of molecules in strong laser fields, including the correlated electron-nuclear dynamics in molecular multiphoton energy absorption, the attosecond intramolecular dynamics of electrons such as the electron localization and tunneling, rescattering and recapture, and the molecular vibrational and rotational wave packets such as the molecular echo, all-optical three-dimensional molecular orientation, and molecular ultrafast buffering.

華東師範大學
EAST CHINA NORMAL UNIVERSITY

Ultrafast Dynamics of Molecules in Strong Laser Fields

Jian Wu (吴健)

State Key Laboratory of Precision Spectroscopy
East China Normal University, China

精密光谱科学与技术国家重点实验室
State Key Laboratory of Precision Spectroscopy

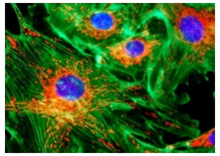
Outline

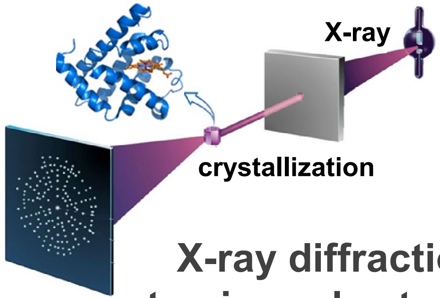
- **Background Introduction**
- **Multiphoton energy absorption: electron-nuclear correlation**
  - ATI & ATD in dissociative ionization of molecules
  - Strong-field Rydberg excitation of molecules
  - An ultrafast stopwatch to clock molecular bond stretching
- **Ro-vibrational dynamics of the nuclear wave-packet**
  - Visualizing unidirectional molecular rotation
  - Echoes of molecules
  - All-optical 3D orientation of molecules
- **Attosecond dynamics of electrons: visualization and control**

Microstructure determines the macroscopic properties of matter

Ultrafast dynamics of the microcosm: attosecond and sub-nanometer spatiotemporal resolution

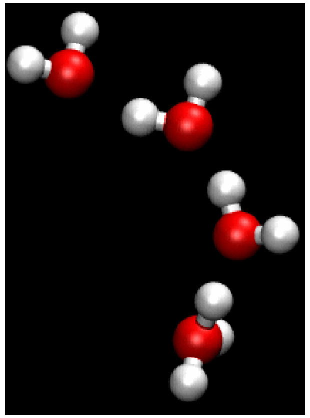
optical microscope:
micron resolution





X-ray diffraction:
atomic scale structure

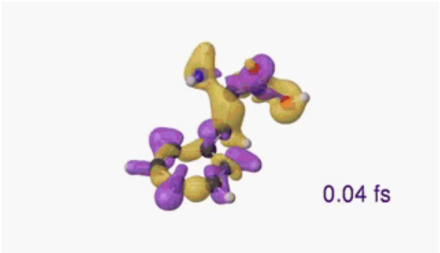
Improving the ability to explore nature



Molecule
restructure & interaction
Nuclear motion：
ps (10^{-12}s)/ fs (10^{-15}s)

steady-state ⟶ dynamics

Electron motion is the key



Science 346, 6207(2014).

Charge, energy & information transport
Electron motion：as (10^{-18}s)

- Reveal novel physical phenomena and mechanisms
- Provide new ideas for novel material and structural design

Ultrafast dynamics of molecules in ultrashort laser pulses

Ultrashort laser pulses:
measurement → physical mechanism → control

Light-molecule interaction

① Photon energy absorption

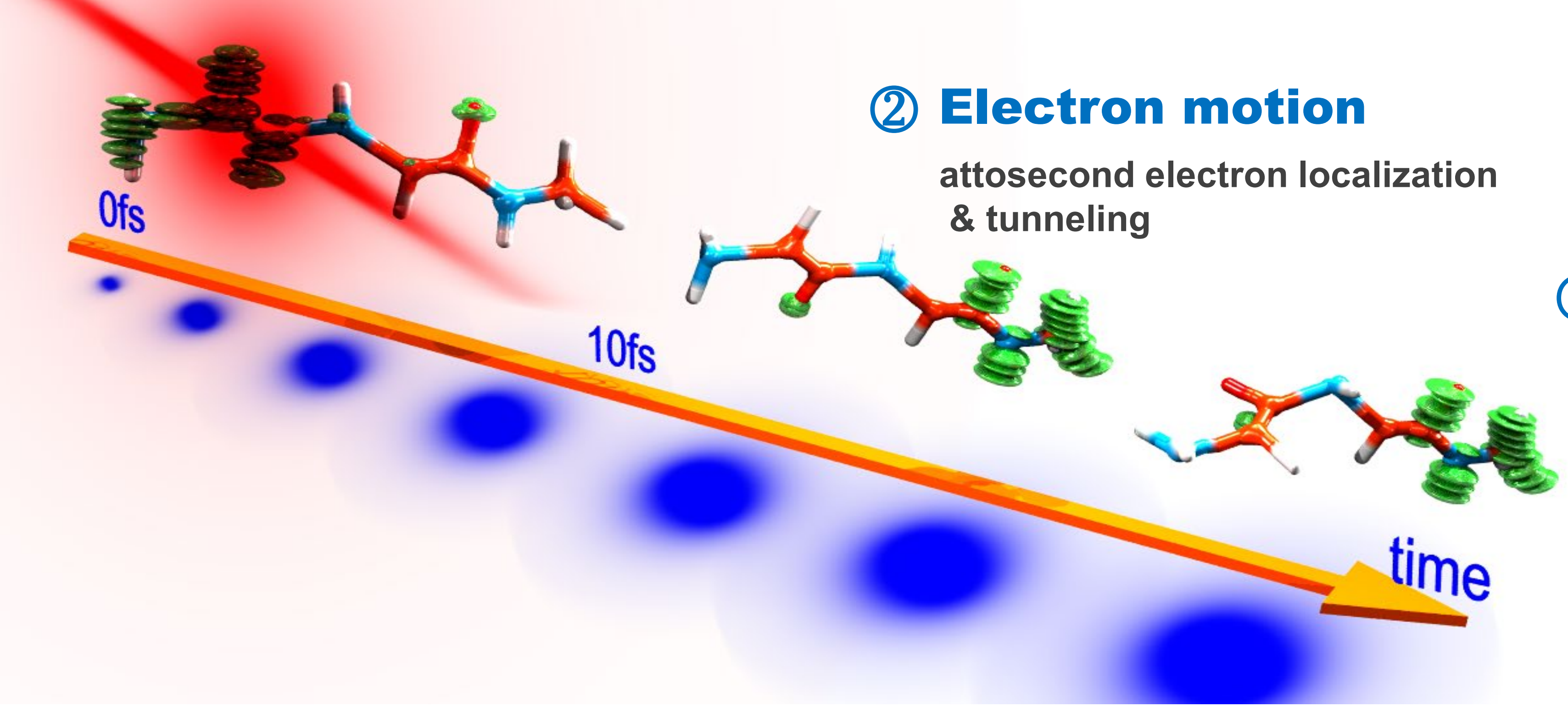
primary stage of light-molecule interaction:
electron-nuclear correlation

② Electron motion

attosecond electron localization
& tunneling

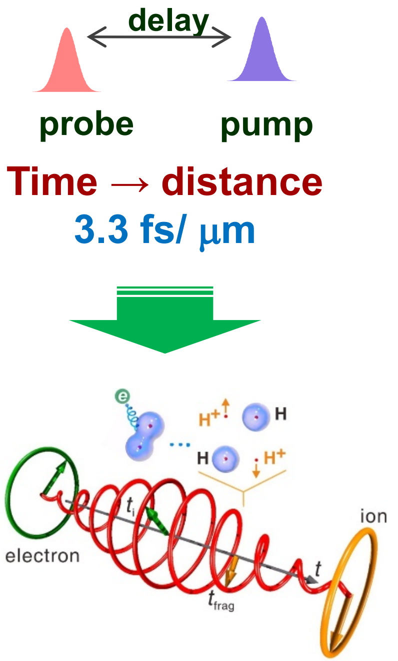
③ Nuclear motion

rotational alignment
vibrational echoes
clocking bond stretching



Experimental techniques

- **Spatial resolution:** momentum space → sub-nanometer (10^{-10} m)
- **Time resolution**： optical field manipulation → attosecond (10^{-18} s)
- **Electron-nuclear correlation:** coincidence measurement → visualization

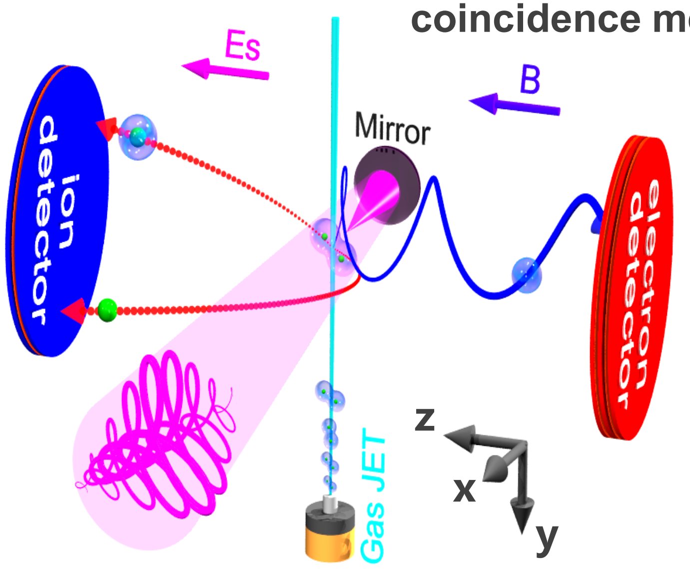


Time → angle

2.6 fs / cycle = 7.3 as / degree

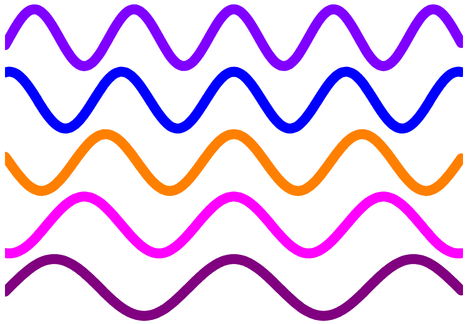
J. Wu et al., Nature Comm. 4, 2177 (2013).

Electron-nuclear coincidence measurement

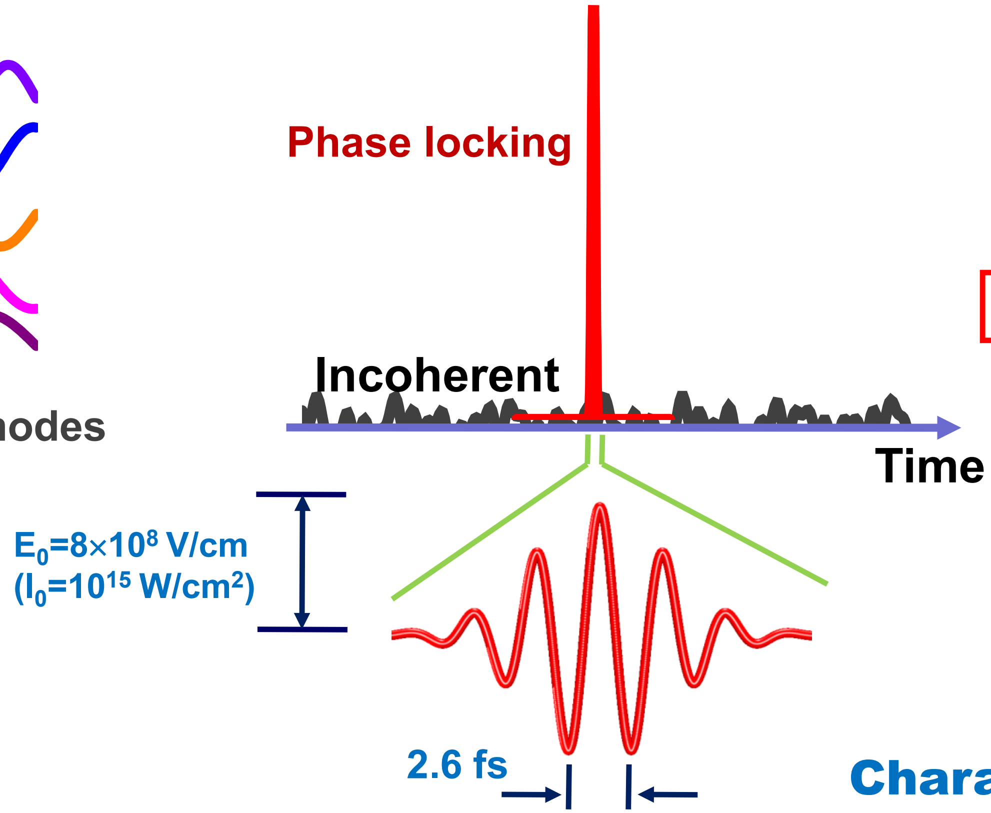


Optical field spatiotemporal manipulation

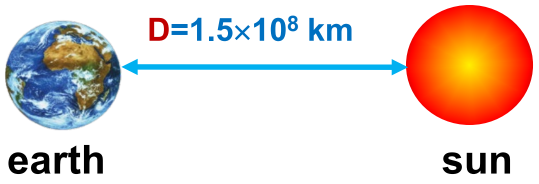
Femtosecond laser pulse



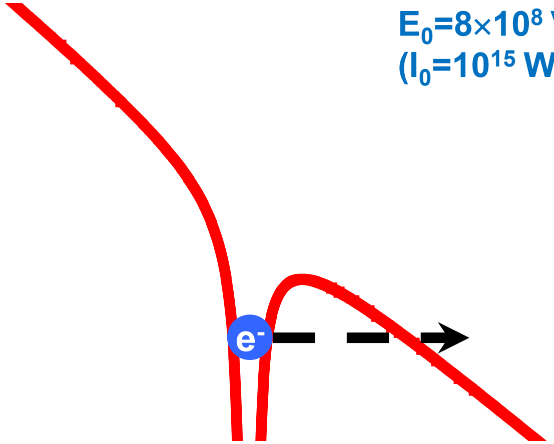
Different frequency modes



$1\ fs = 10^{-15}\ s$



$1\ fs : 1 s = 0.15\ \mu m : D$

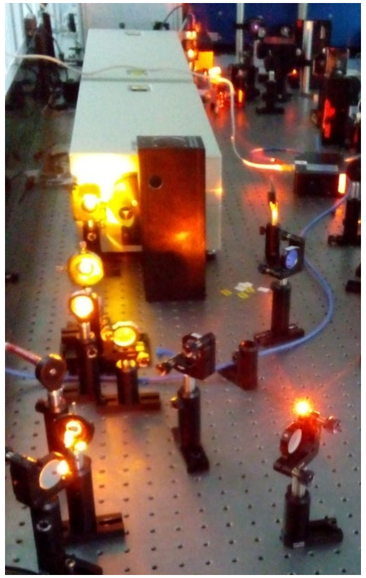


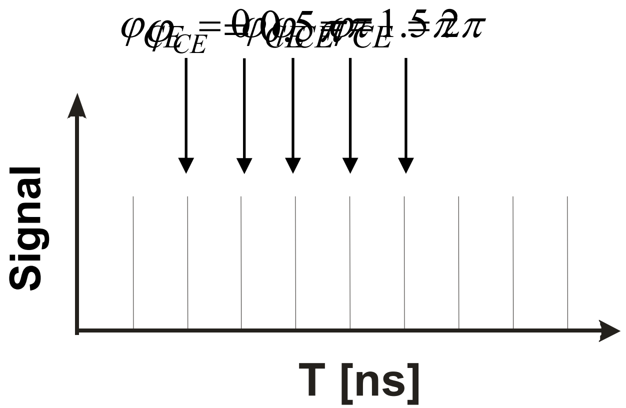
Suppress the potential barrier
Free bound electron

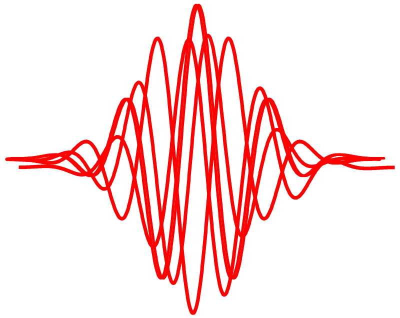
Characters of fs pulse:

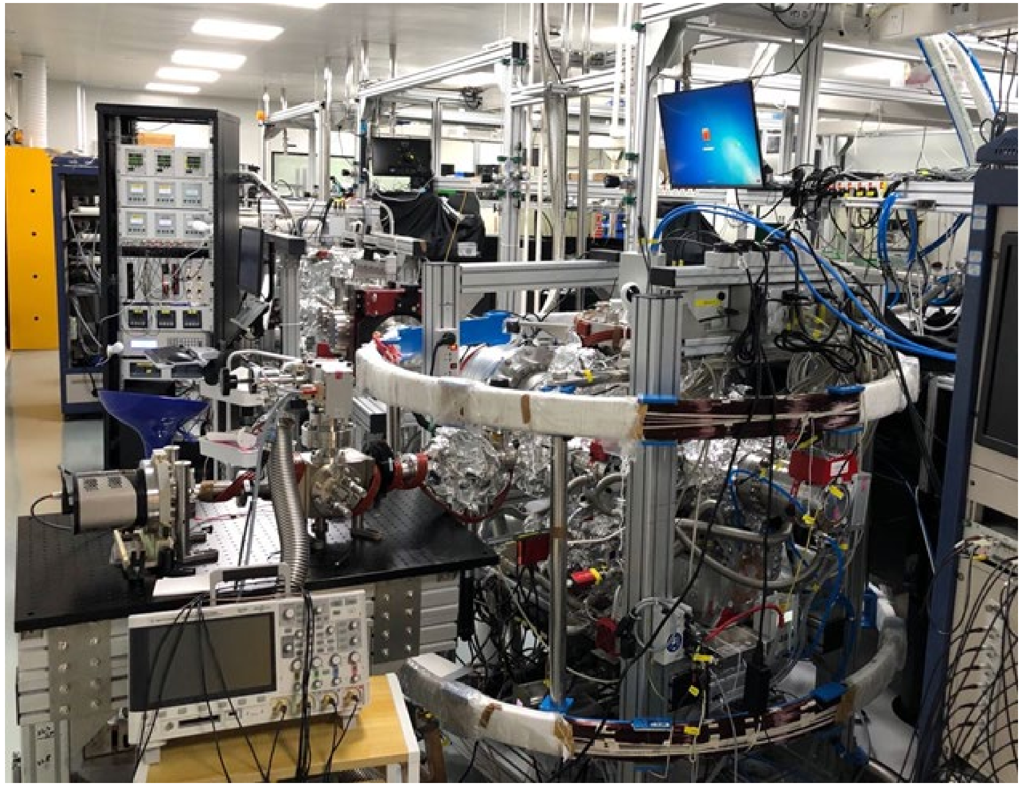
1. Ultrashort time duration
2. Ultrahigh field strength
3. Ultrabroad spectral width

Laser systems and COLTRIMS









COLTRIMS: COLd Target Recoil Ion Momentum Spectroscopy

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  - Echoes of molecules
  - All-optical 3D orientation of molecules
- **Attosecond dynamics of electrons: visualization and control**

ATI & ATD (Above-Threshold Ionization & Dissociation) of molecules

Above-threshold ionization (ATI) of atoms

VOLUME 42, NUMBER 17 PHYSICAL REVIEW LETTERS 23 APRIL 1979

Free-Free Transitions Following Six-Photon Ionization of Xenon Atoms

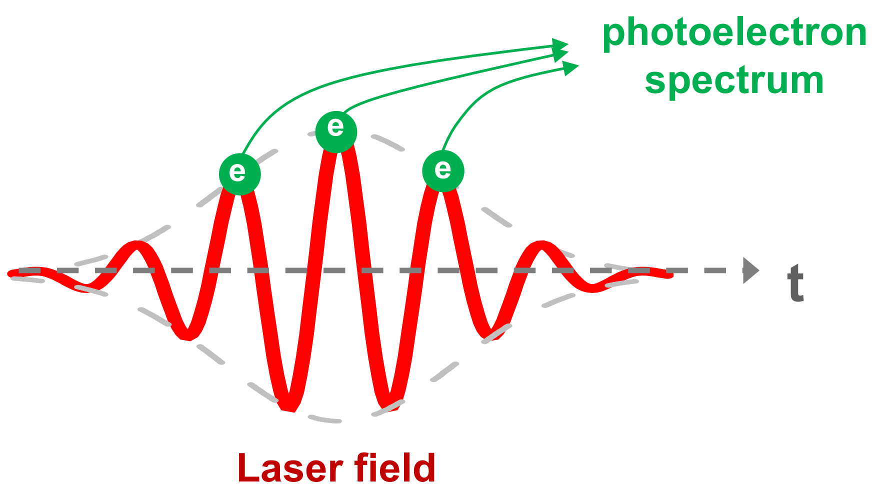
P. Agostini, F. Fabre, G. Mainfray, and G. Petite
Centre d'Etudes Nucléaires de Saclay, Service de Physique Atomique, 91190 Gif-sur-Yvette, France

and

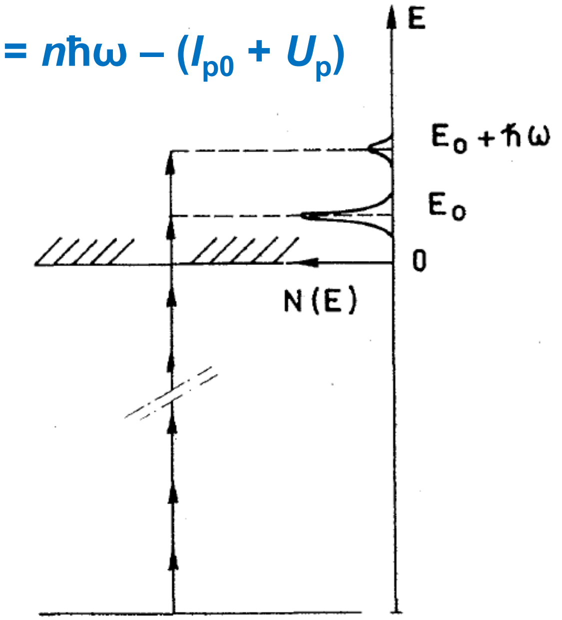
N. K. Rahman
Laboratorio di Chimica Quantistica ed Energetica Molecolare del Consiglio Nazionale delle Ricerche, 56100 Pisa 35, Italy

(Received 29 January 1979)

Quantized nature of light energy:
inter-cycle interference of emitted electron wave packet

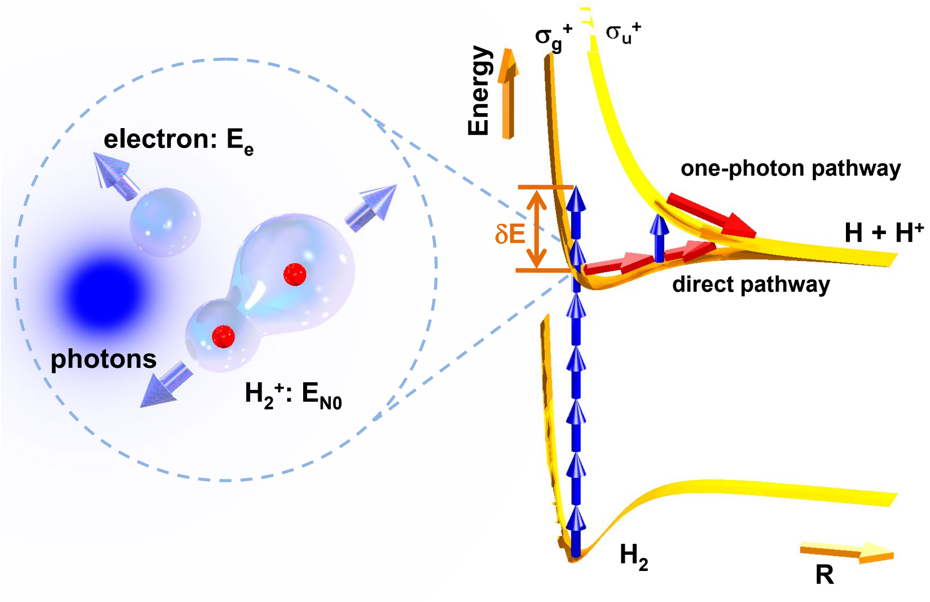


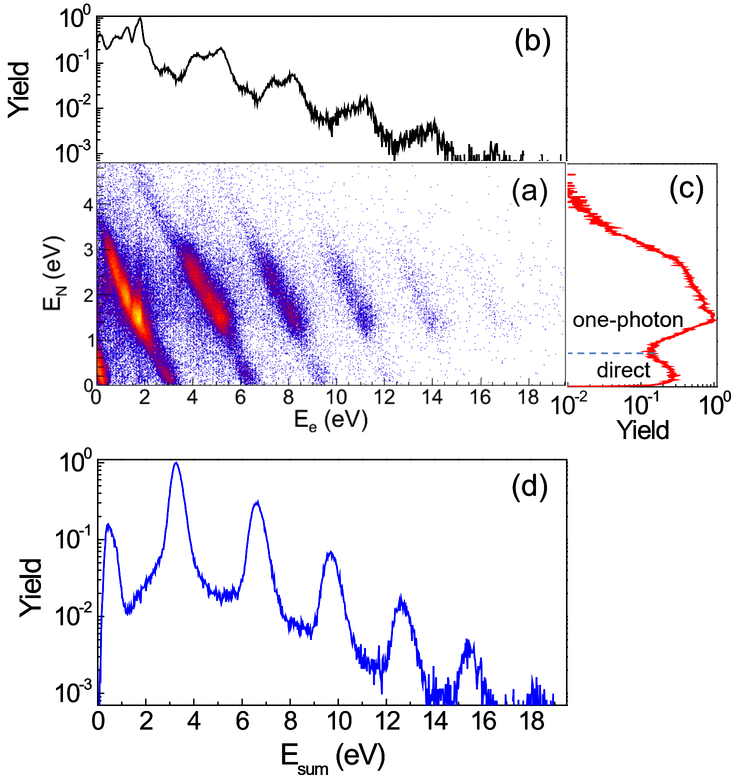
$$E_e = n\hbar\omega - (I_{p0} + U_p)$$



Electron-nuclear sharing of photon energy in a molecule

$E_e + E_N = n\hbar\omega - (I_{p0} + U_p)$





$H_2 + n\hbar\omega \rightarrow H^+ + H + e$

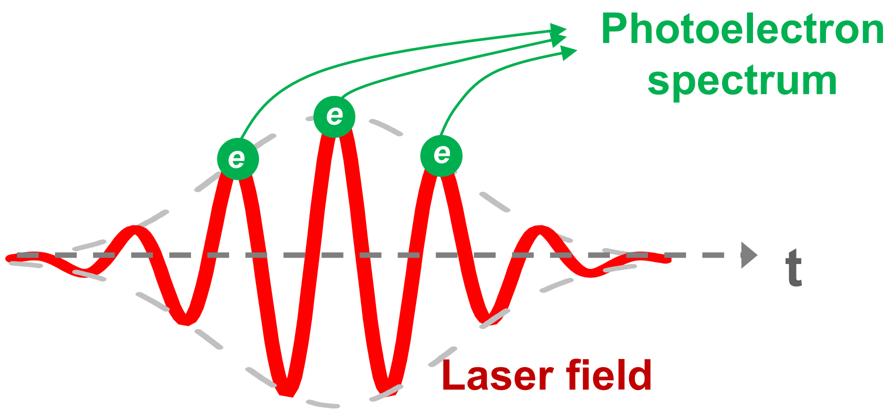
How does a molecule absorb photon energy?

Experimental demonstration: electron-nuclear sharing of photon energy

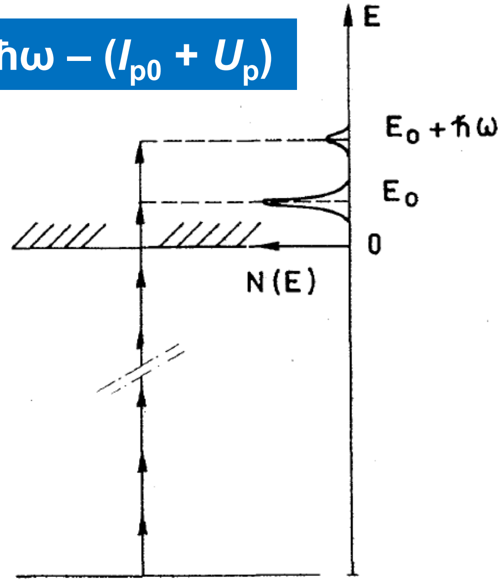
J. Wu *et al.*, Phys. Rev. Lett. 111, 023002 (2013).

Above threshold ionization (ATI)
→ Above threshold dissociation (ATD) of a molecule

Multiphoton absorption by the electron:
Interference of periodically tunneled electron



$E_e = n\hbar\omega - (I_{p0} + U_p)$



Deposition of multiphoton energy into the electron：ATI

Interference of periodically tunneled nuclear wave-packets：
deposition of multiphoton energy into the nuclei

↓

Photon-energy spaced nuclear spectrum : ATD

Electron-nuclear correlation via electron re-scattering

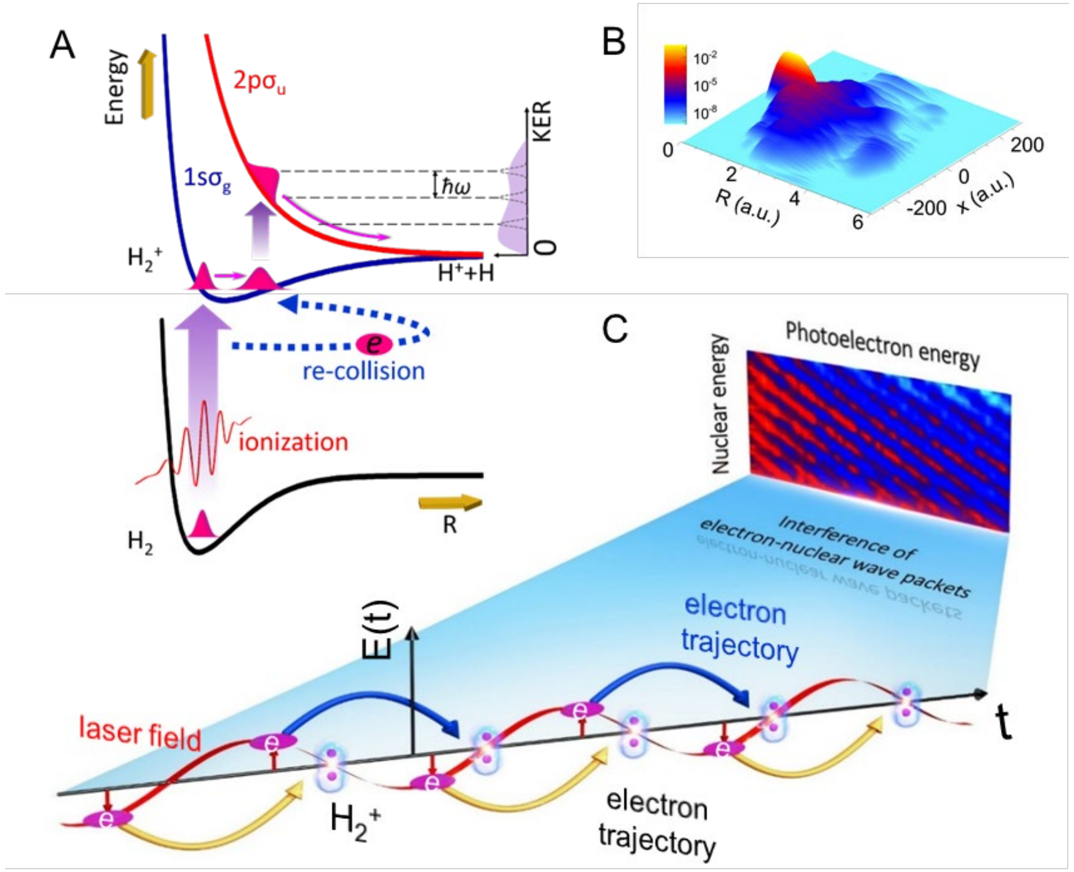
$H_2 + n\hbar\omega \rightarrow H^+ + H + e$

① Electron tunnels & gains energy from the oscillating laser field

② Returning electron transfers the energy to nuclei via the rescattering

Electron-nuclei energy sharing:

$$E_e + E_N = n\hbar\omega - (I_{p0} + U_p)$$

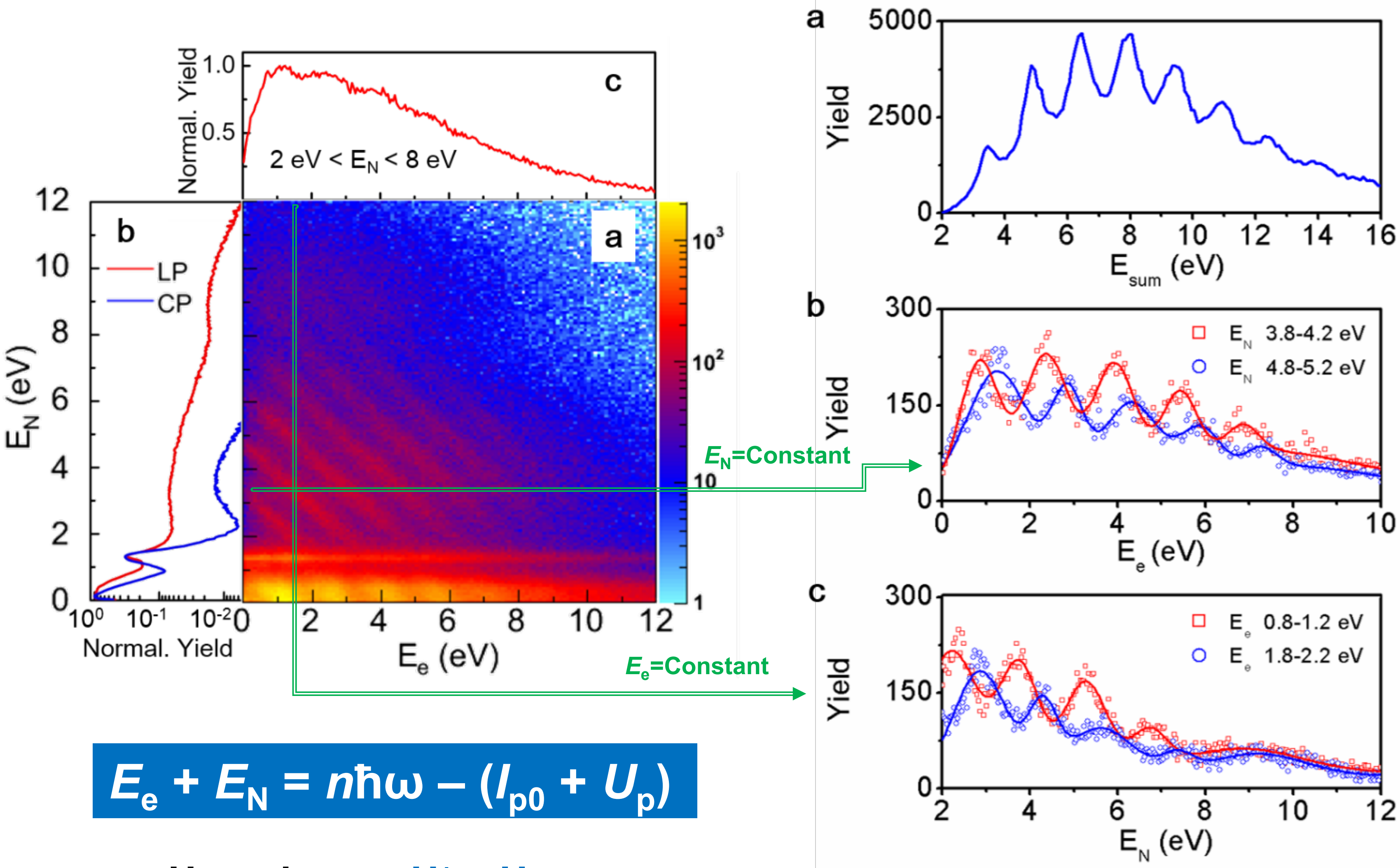


Interference of periodically emitted electron-nuclear wave packets:

molecule as a whole absorbs multiple photon energy → ATI & ATD

P. Lu *et al.*, PNAS 115, 2049 (2018).

Experimental observation of high-order ATD



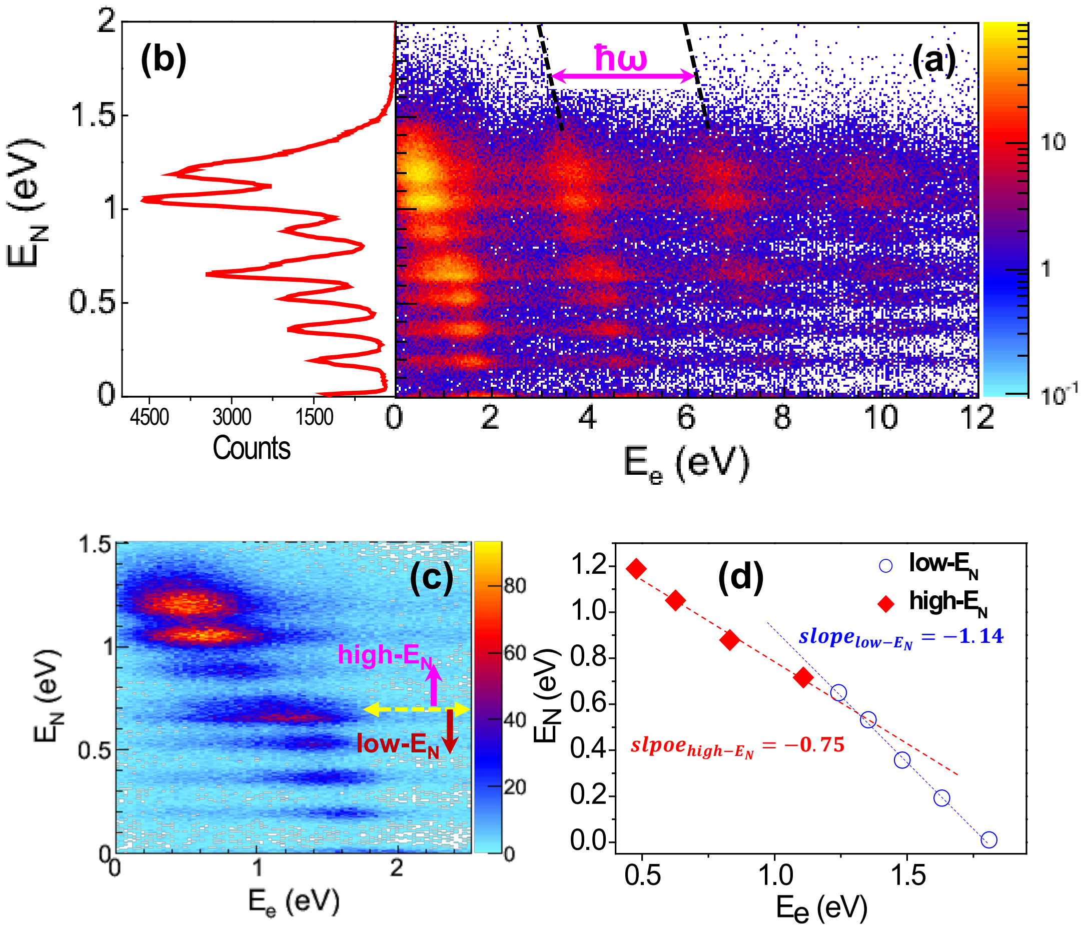
$$E_e + E_N = n\hbar\omega - (I_{p0} + U_p)$$

$H_2 + n\hbar\omega \rightarrow H^+ + H + e$

P. Lu *et al.*, PNAS 115, 2049 (2018).

Electron-nuclear sharing of photon energy: multielectron system (orbital &vibrational)

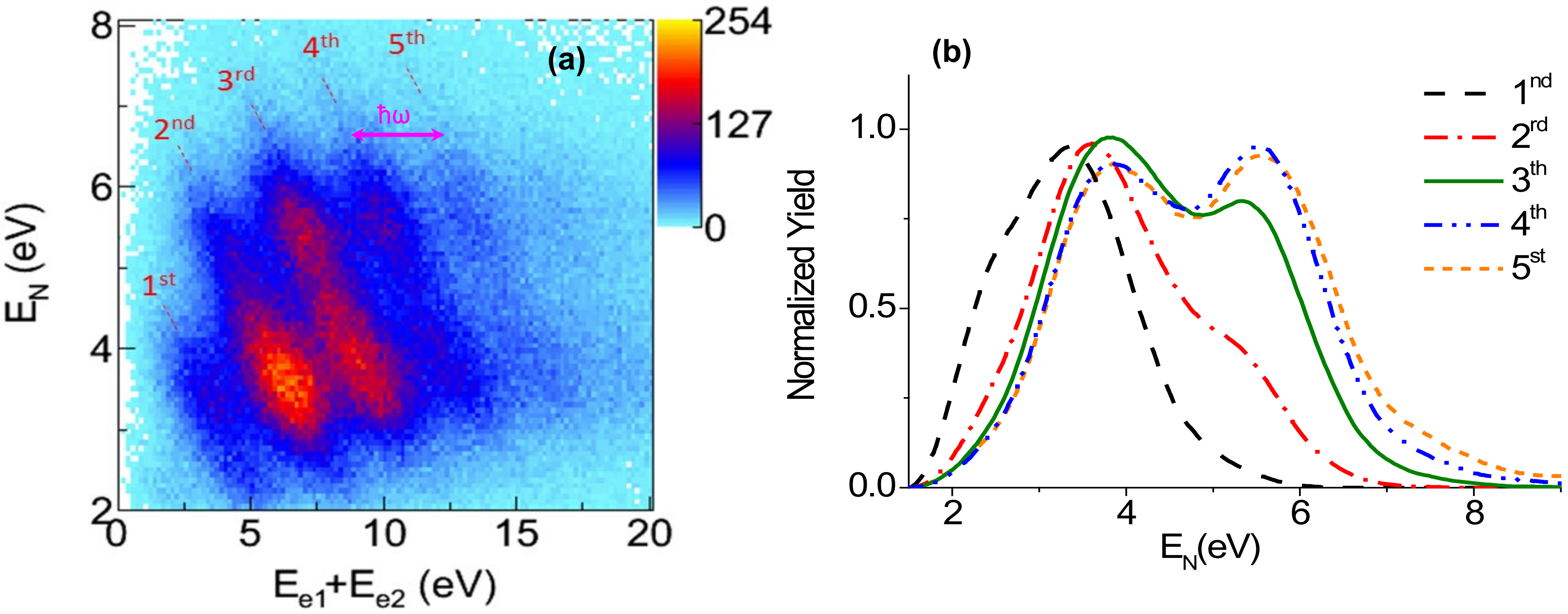
$CO + n\hbar\omega \rightarrow C^+ + O + e$ 7.6×10^{13} W/cm^2 @ 395 nm



W. Zhang *et al.*, Phys. Rev. Lett. 117, 103002 (2016).

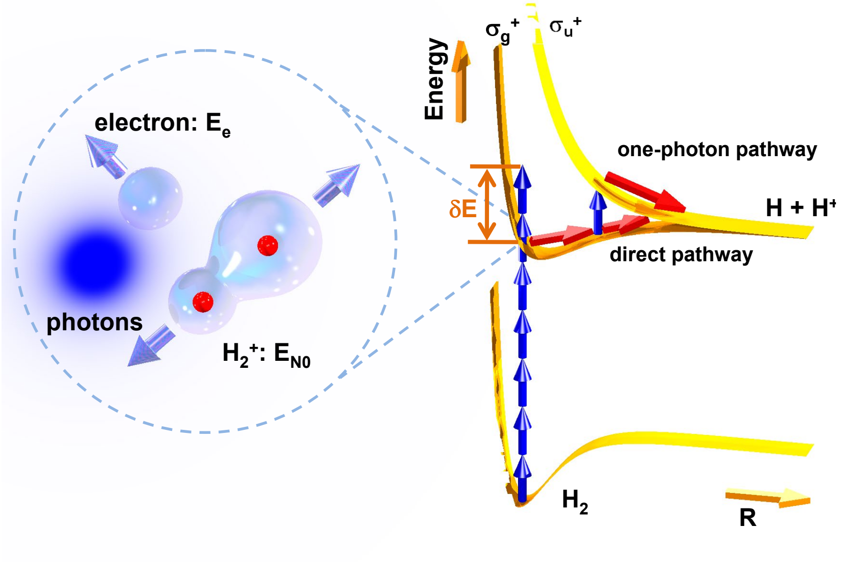
Electron-nuclear correlated above-threshold double ionization (ATDI)

$H_2 + n\hbar\omega \rightarrow H^+ + H^+ + e_1 + e_2$



P. Lu *et al.,* Phys. Rev. A 95, 033404 (2017).

Multiphoton energy absorption by a molecule



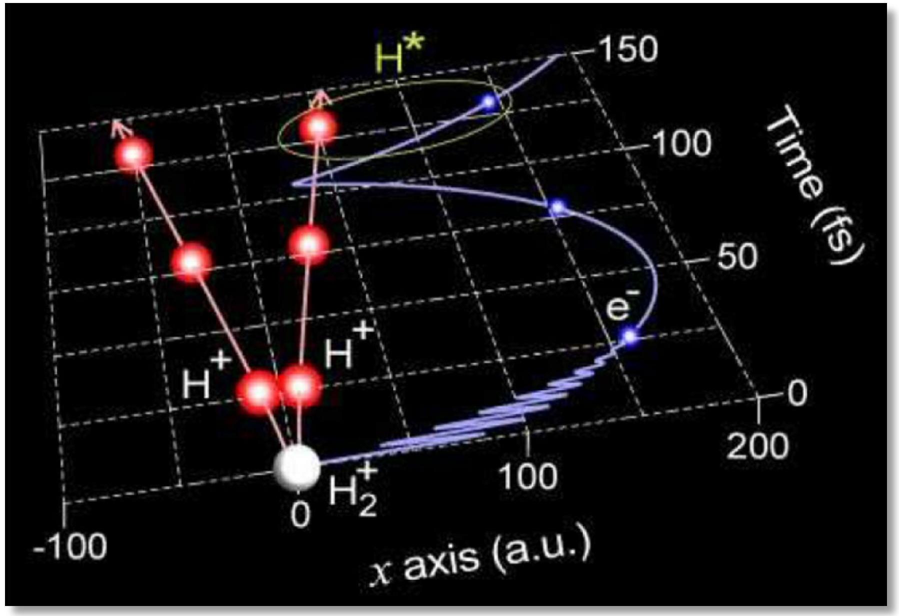
- First ATI photoelectron spectrum in strong-field ionization of atoms (PRL1979).
- In 2013, observation of electron-nuclear sharing of multiphoton photon energy in a molecule: Phys. Rev. Lett.111, 023002 (2013).
- In 2016, important role of the vibrational motion: Phys. Rev. Lett. 117, 103002 (2016).
- In 2018, direct observation of high-order ATD: PNAS 115, 2049 (2018).

Mechanism: electron absorbs photon energy, transfers the energy to nuclei via their correlated interaction.

Strong-field Rydberg excitation of molecules

Frustrated ionization: Rydberg excitation

Dissociative Frustrated Double Ionization (FDI): tunneled electron is recaptured by the ionic fragments during the breaking of molecules.



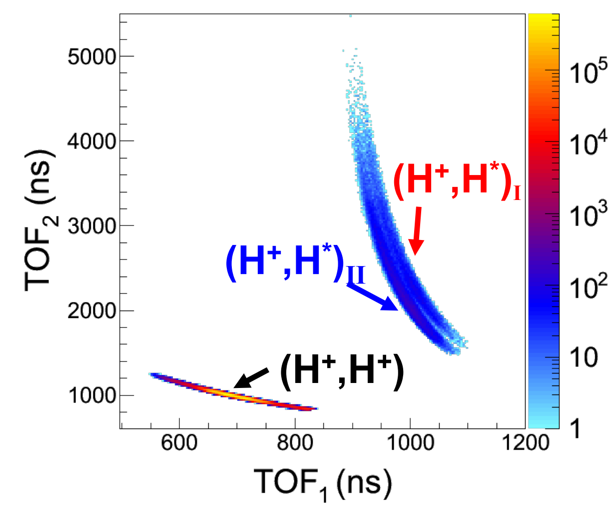
B. Manschwetus *et al.*, Phys. Rev. Lett. 102, 113002 (2009).

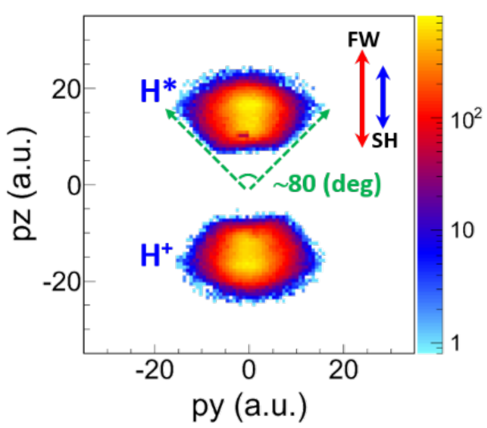
Dynamics of FDI of molecules:

- When and where the dissociative FDI occurs?
- Steering the electron recapture to a desired ionic core?
- Electron-nuclear correlation?

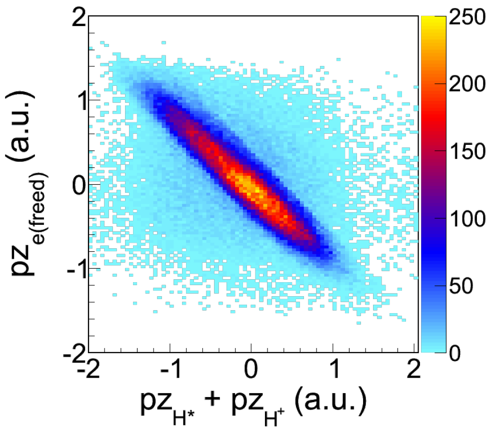
Dissociative FDI of H_2: full coincidence measurements

Photoion-photoion coincidence (PIPICO)

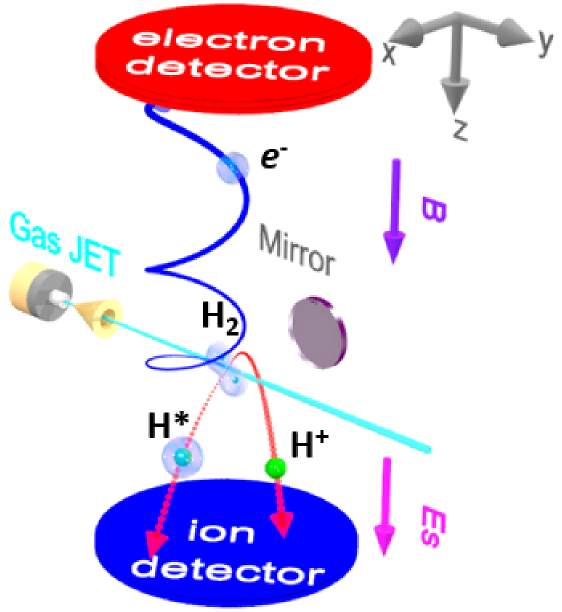




Momentum distribution



Coincidence spectrum



$H_2 + m\hbar\omega$

$\rightarrow H^+ + H^+ + 2e$ (double ionization)

$\rightarrow H^+ + H^* + e$ (FDI: Rydberg excitation)

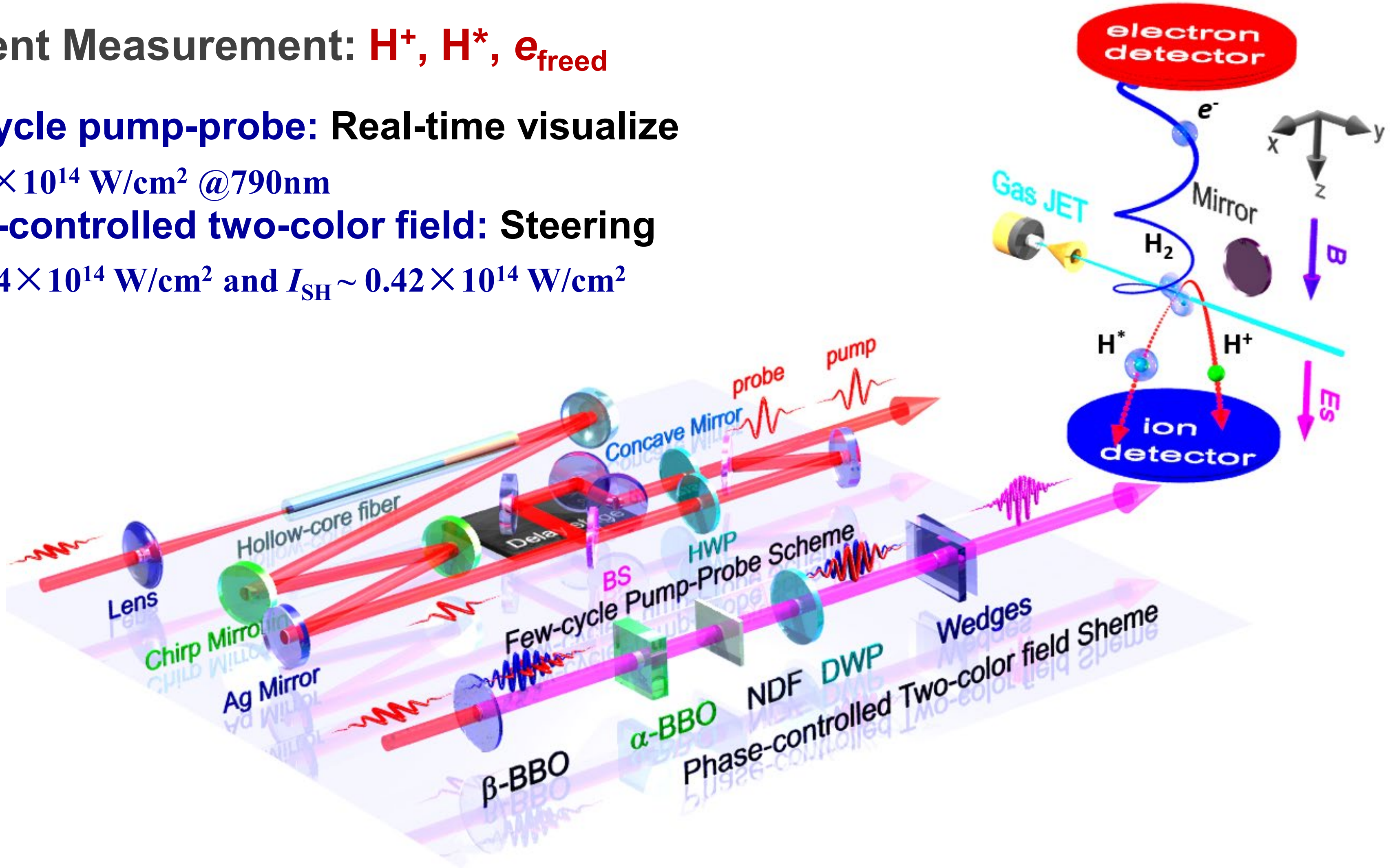
W. Zhang *et al.*, Phys. Rev. Lett. 119, 253202 (2017).

Dissociative FDI of H_2: full coincidence measurements

- Coincident Measurement: H^+, H^*, e_{freed}
  - **Few-cycle pump-probe:** Real-time visualize

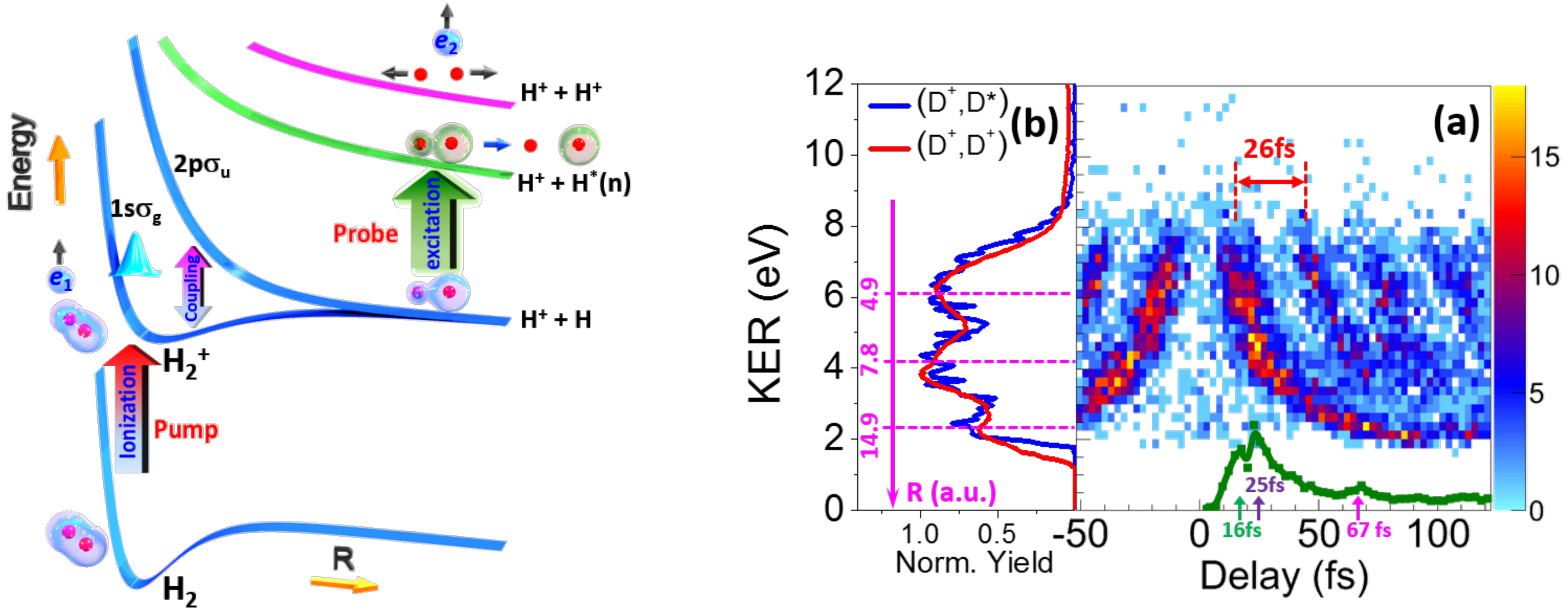
    7fs, 6.4×10^{14} W/cm^2 @790nm
  - **Phase-controlled two-color field:** Steering

    I_{FW} ~ 2.4×10^{14} W/cm^2 and I_{SH} ~ 0.42×10^{14} W/cm^2



W. Zhang *et al.*, Phys. Rev. Lett. 119, 253202 (2017).

Visualizing dissociative FDI of H_2

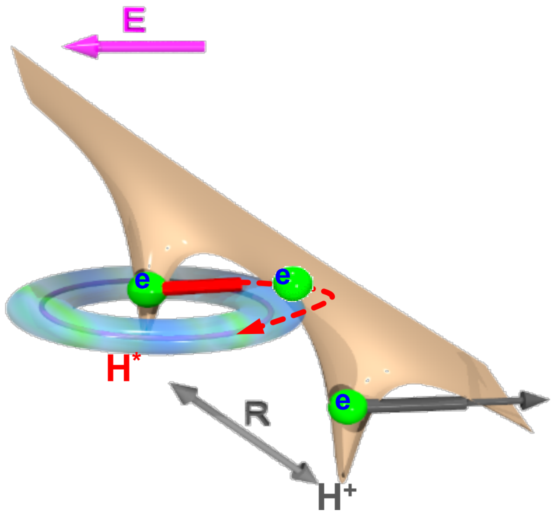


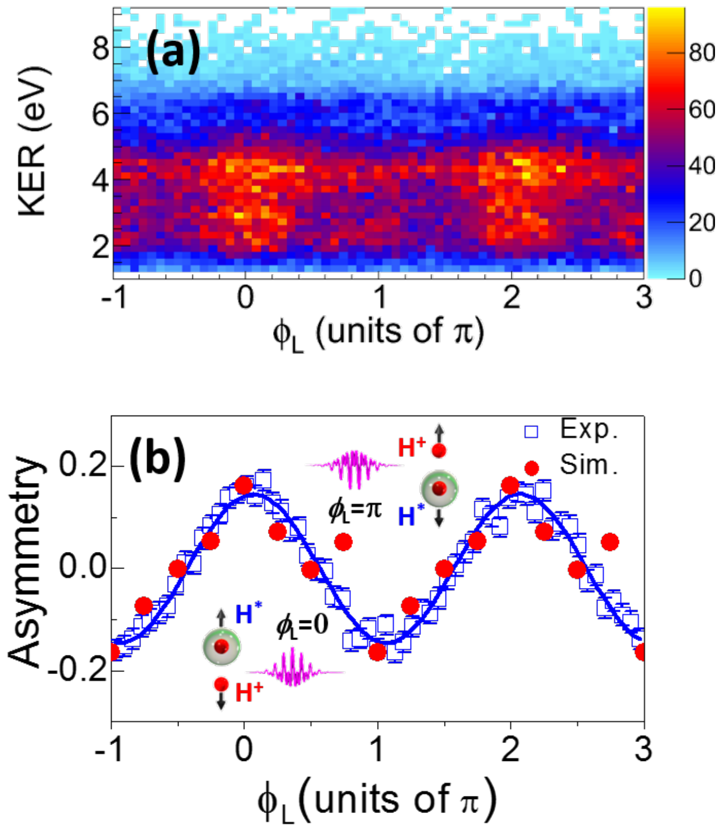
Three internuclear distances (R) of the stretching molecular ion are observed to enhance the dissociative FDI at different instants (τ)

(τ, *R*) ~ (16 fs, 4.9 a.u.), (25 fs, 7.8 a.u.), (67 fs, 14.9 a.u.)

W. Zhang *et al.*, Phys. Rev. Lett. 119, 253202 (2017).

Steering electron recapture





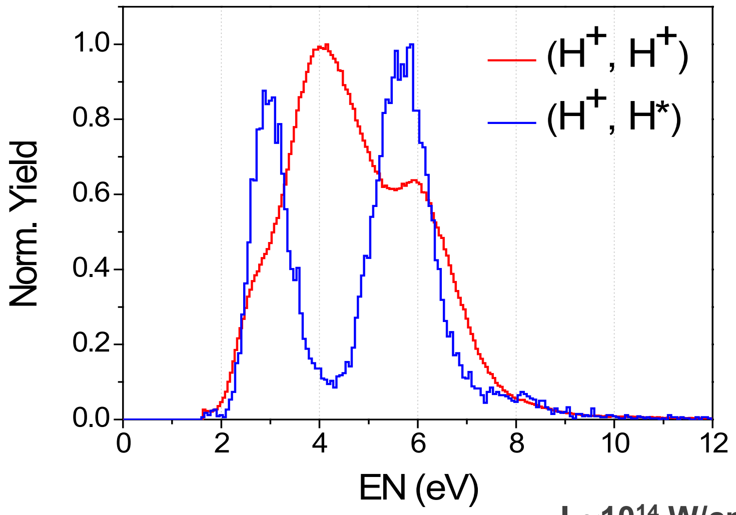
The Rydberg atom is favored to emit to the direction of the maximum of the asymmetric optical field.

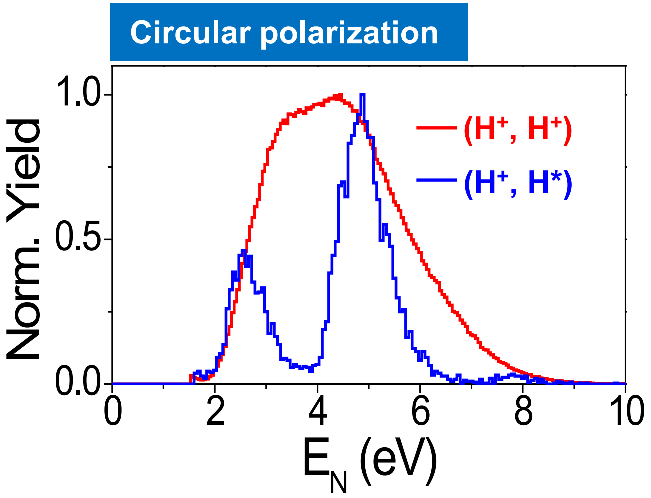
W. Zhang *et al.*, Phys. Rev. Lett. 119, 253202 (2017).

Dissociative FDI of molecules in strong laser fields

Electron recapture

- Similar KER spectrum as double ionization
- Suppression in circular polarization



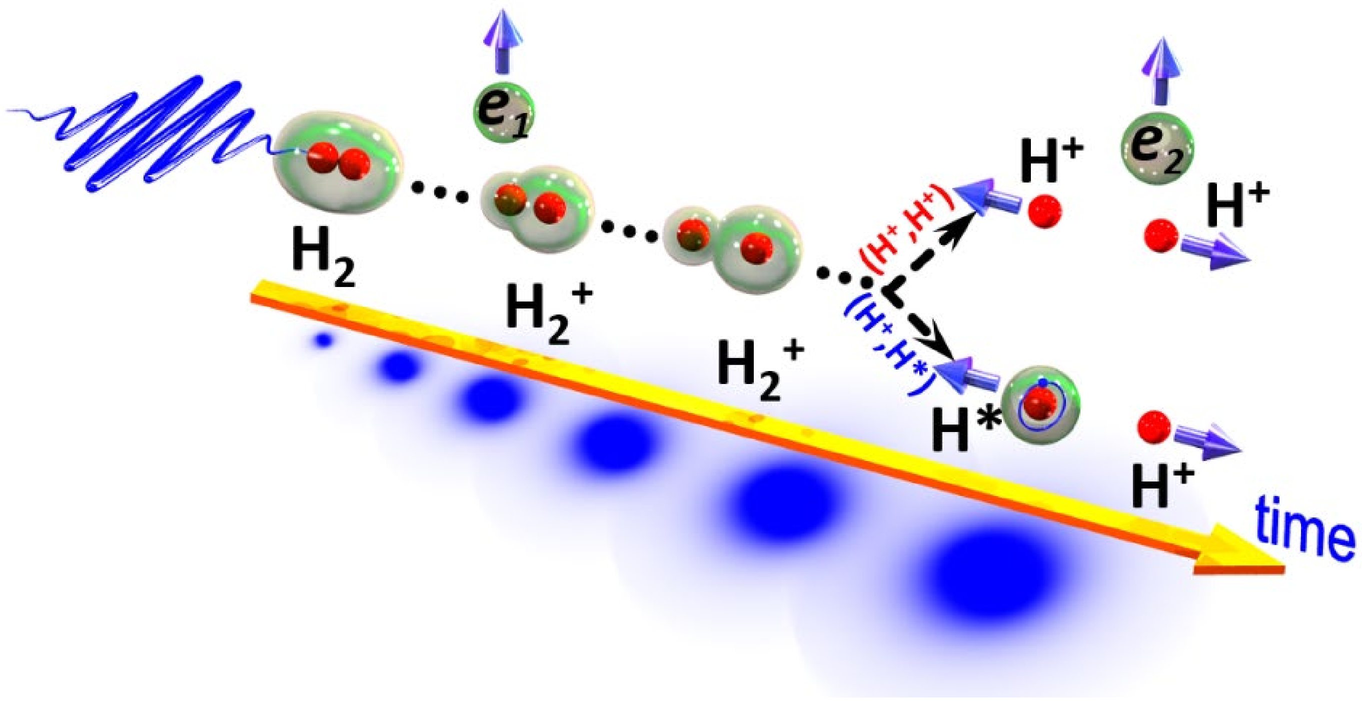


I_0~10^{14} W/cm^2 @ 400 nm

Observation of different KER spectra: (H^+,H^*) vs. (H^+,H^+)

Alternative routes for FDI?

Multiphoton routes: dissociative DI vs. FDI of molecules

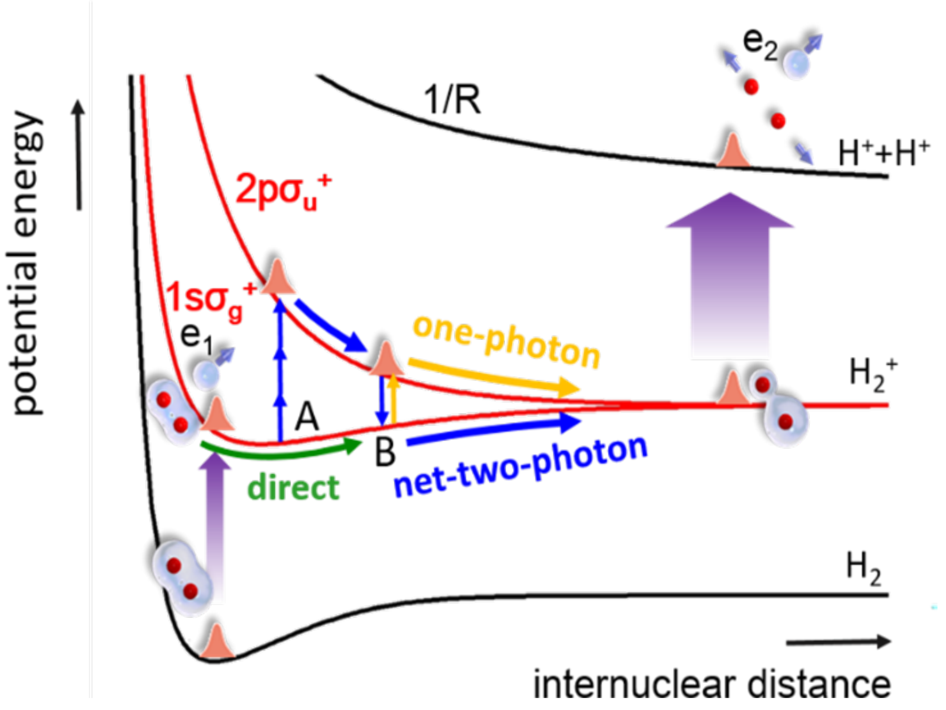


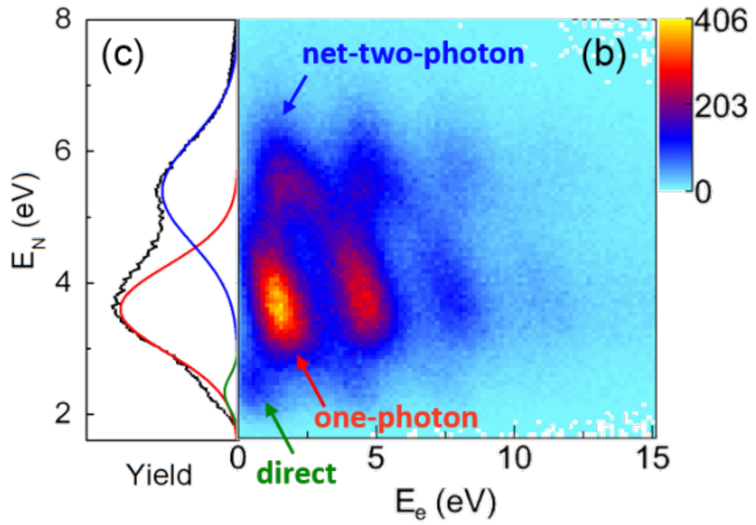
Dissociative double ionization : $H_2 + m\hbar\omega \rightarrow H^+ + H^+ + 2e$

Dissociative frustrated double ionization: $H_2 + m\hbar\omega \rightarrow H^+ + H^* + e$

Multiphoton routes for dissociative DI of molecules: above-threshold double ionization (ATDI)

$H_2 + n\hbar\omega \rightarrow H^+ + H^+ + 2e$



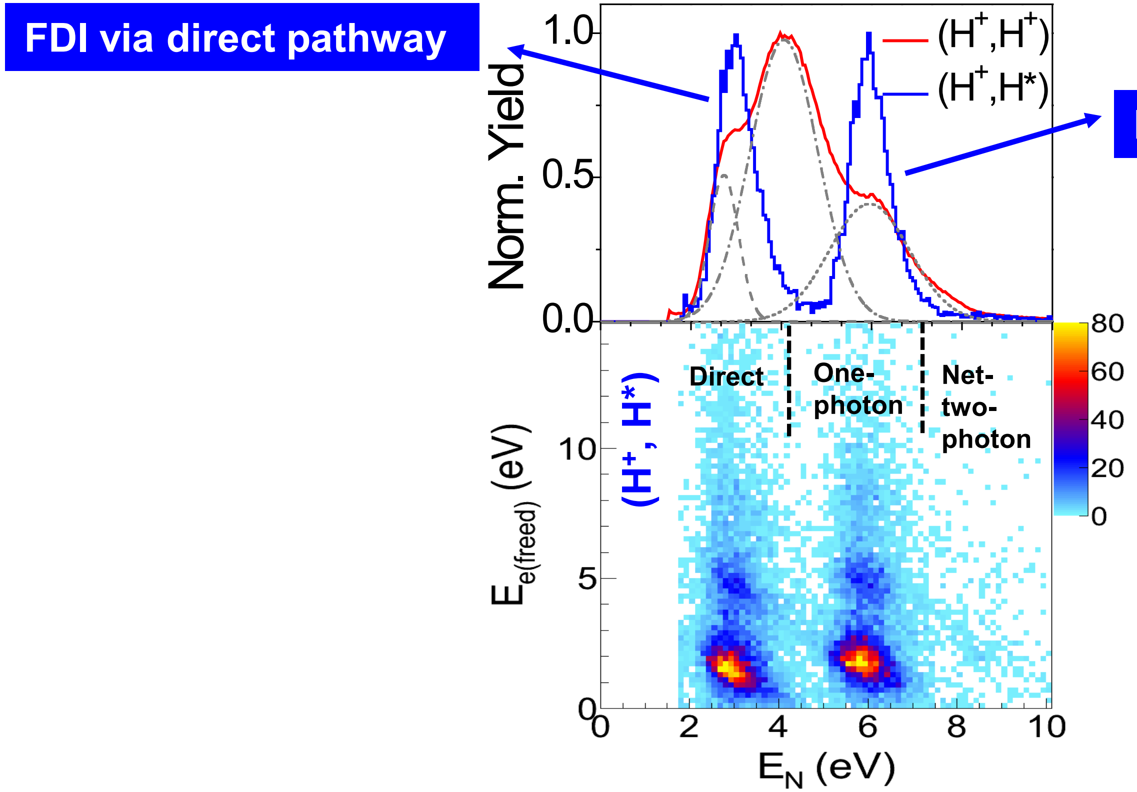


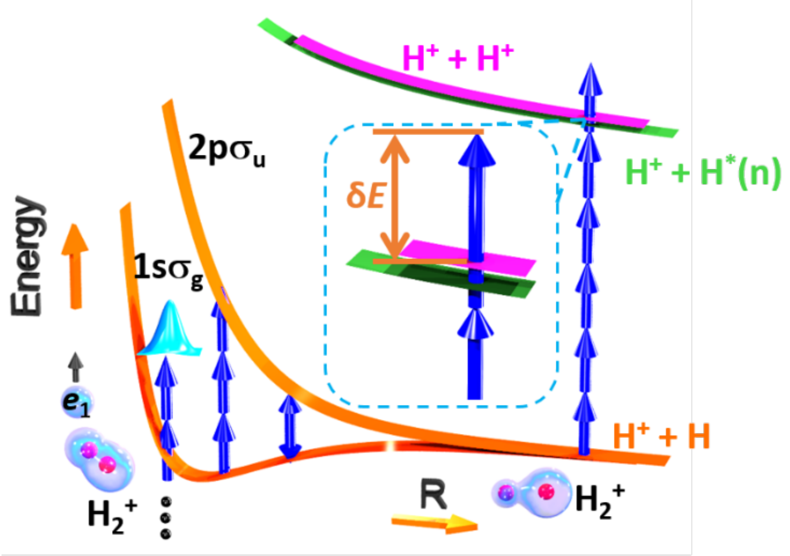
Photon number resolving, enhancing, and suppressing pathways towards CREI.

$H_2 + n\hbar\omega \rightarrow H_2^+ + e$ ……
$H_2^+ + n\hbar\omega \rightarrow H^+ + H^+ + e$

P. Lu *et al.*, Phys. Rev. A 95, 033404 (2017).

Multiphoton routes: dissociative FDI of molecules





Dissociative DI: $H_2 + m\hbar\omega \rightarrow H^+ + H^+ + 2e$

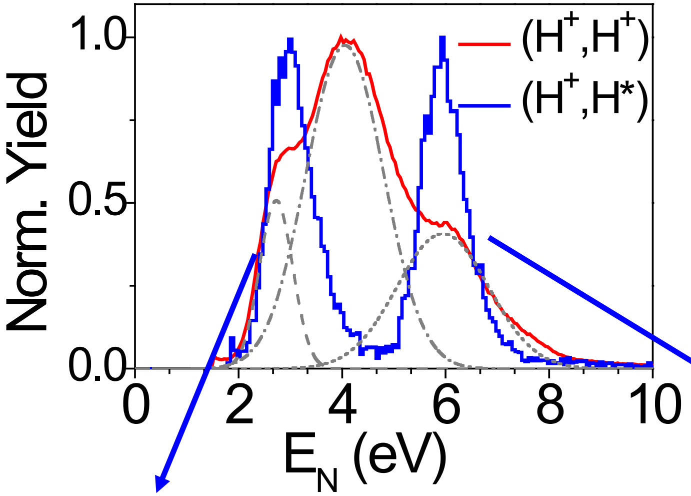
$m\hbar\omega = E_{sum} = E_{H+} + E_{H+} + E_{e1} + E_{e2}$, where $\delta E \sim E_{e2}$

Dissociative FDI: $H_2 + m\hbar\omega \rightarrow H^+ + H^* + e$

$m\hbar\omega = E_{sum} = E_{H+} + E_{H*} + E_e$, where $\delta E \sim E_{H+} + E_{H*}$

W. Zhang *et al.*, Nature Communications 10, 757 (2019).

E_{sum} spectra of DI vs. FDI

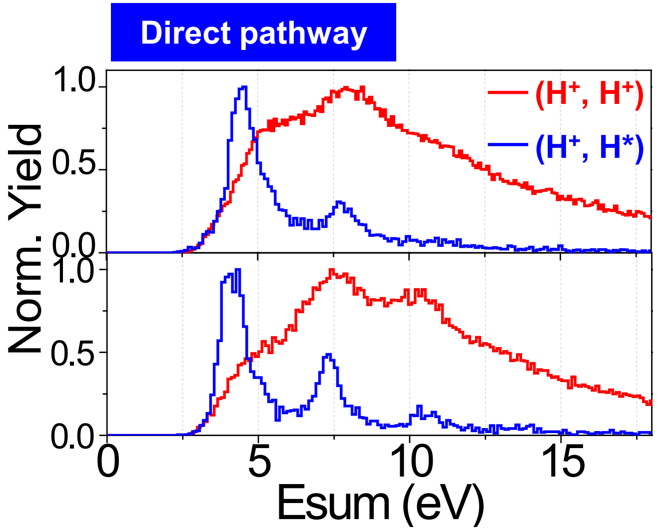


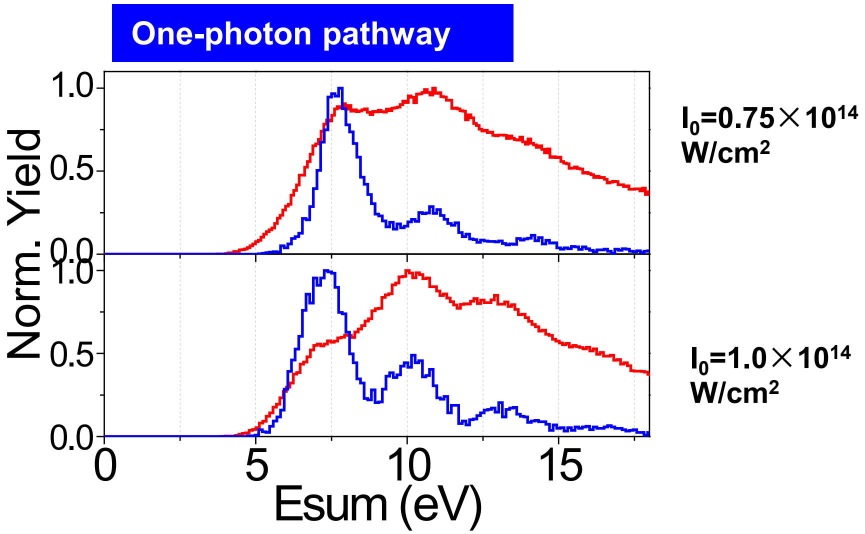
Dissociative DI: $H_2 + m\hbar\omega \rightarrow H^+ + H^+ + 2e$

$m\hbar\omega = E_{sum} = E_{H+} + E_{H+} + E_{e1} + E_{e2}$

Dissociative FDI: $H_2 + m\hbar\omega \rightarrow H^+ + H^* + e$

$m\hbar\omega = E_{sum} = E_{H+} + E_{H^*} + E_{e}$

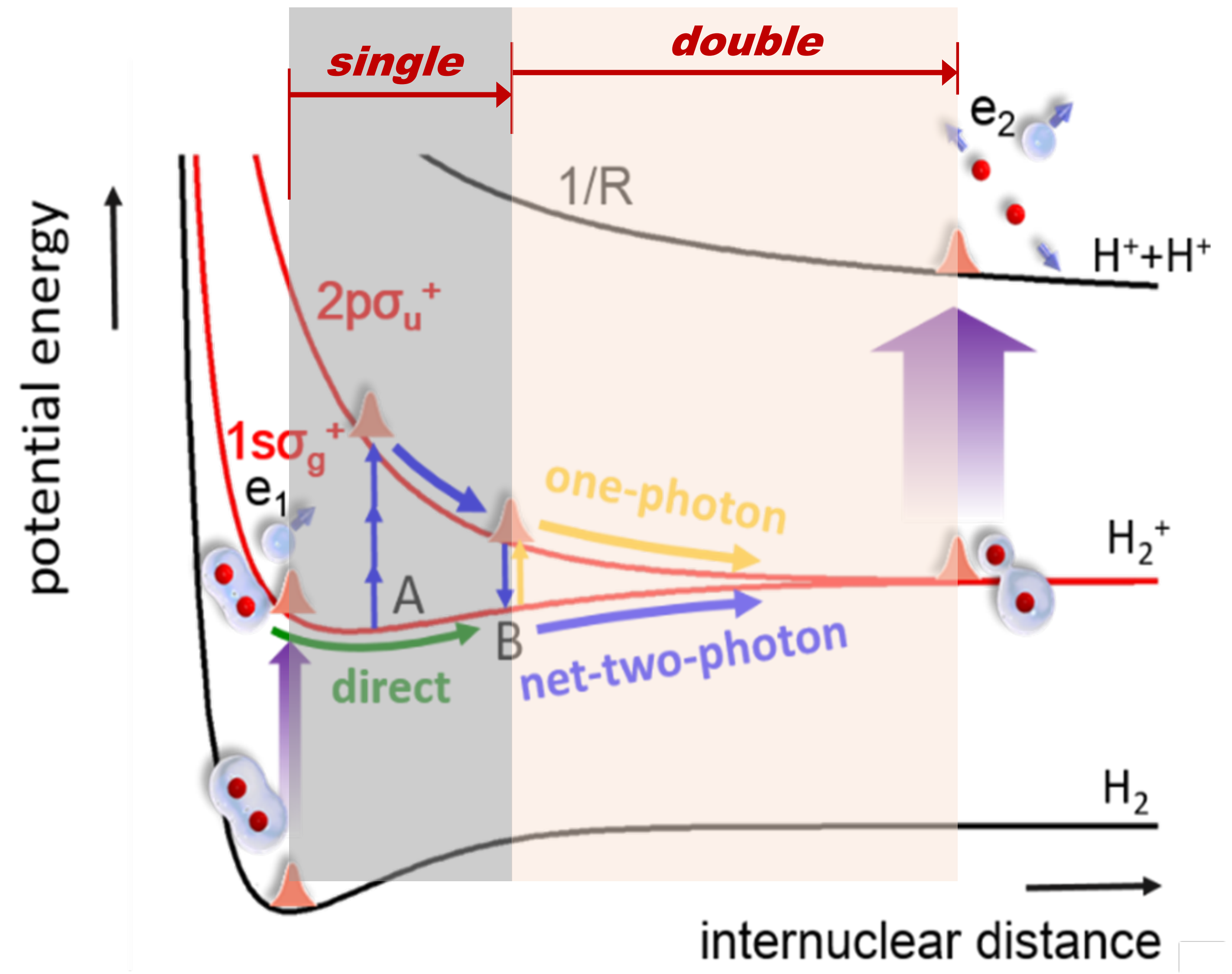




W. Zhang *et al.,* Nature Communications 10, 757 (2019).

An ultrafast stopwatch to clock molecular bond stretching

Dissociative *single* & *double* ionization of H_2

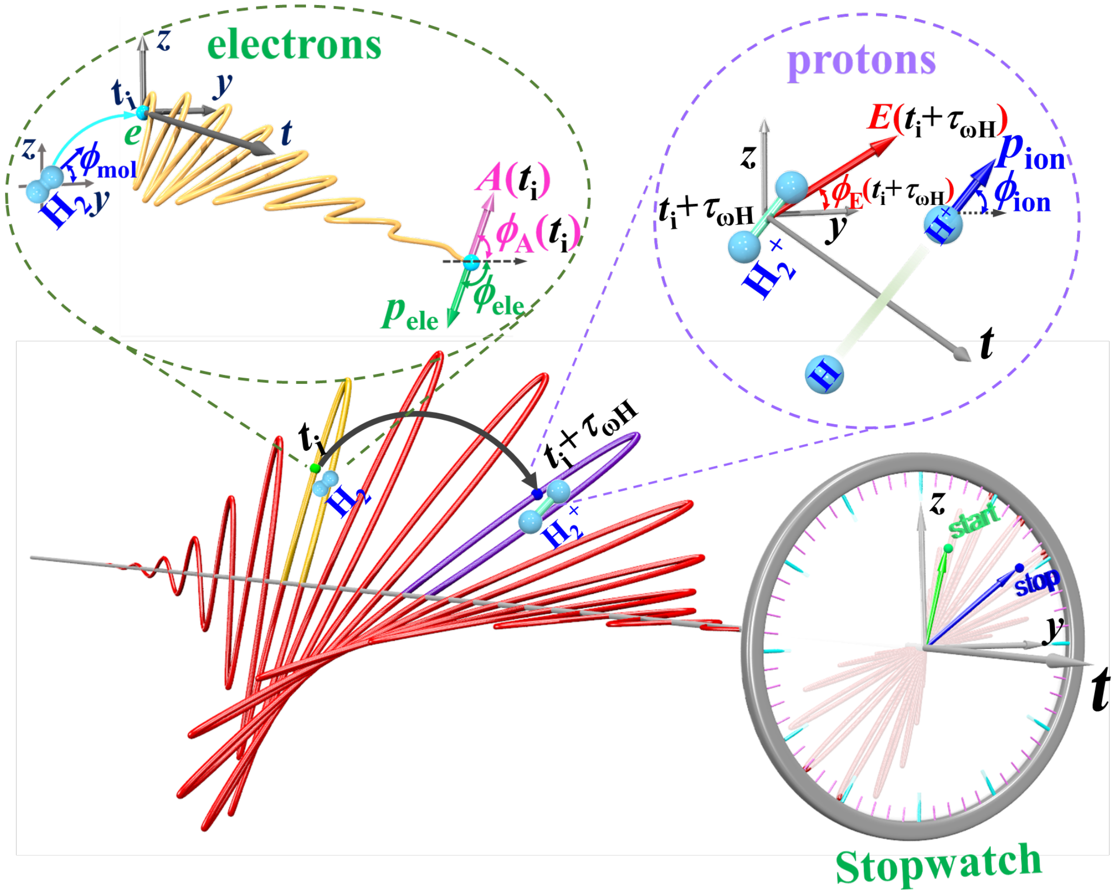


$H_2 + m\hbar\omega \rightarrow H_2^+ + e_1$

$H_2^+ + p\hbar\omega \rightarrow H + H^+$

$H_2^+ + q\hbar\omega \rightarrow H^+ + H^+ + e_2$

Ultrafast stopwatch by polarization-skewed laser pulse

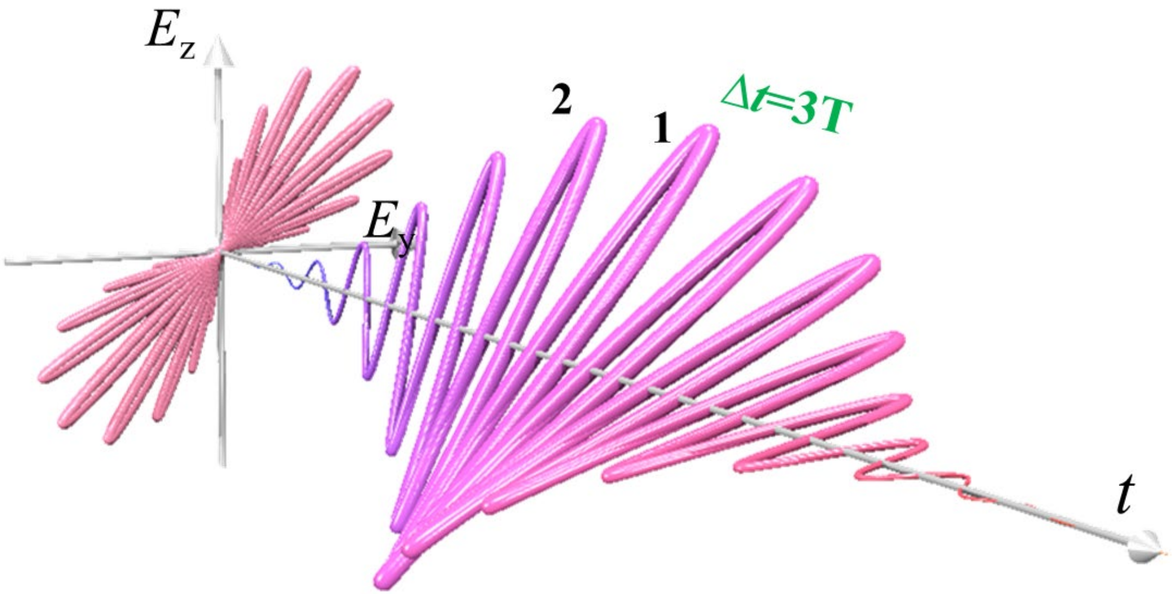


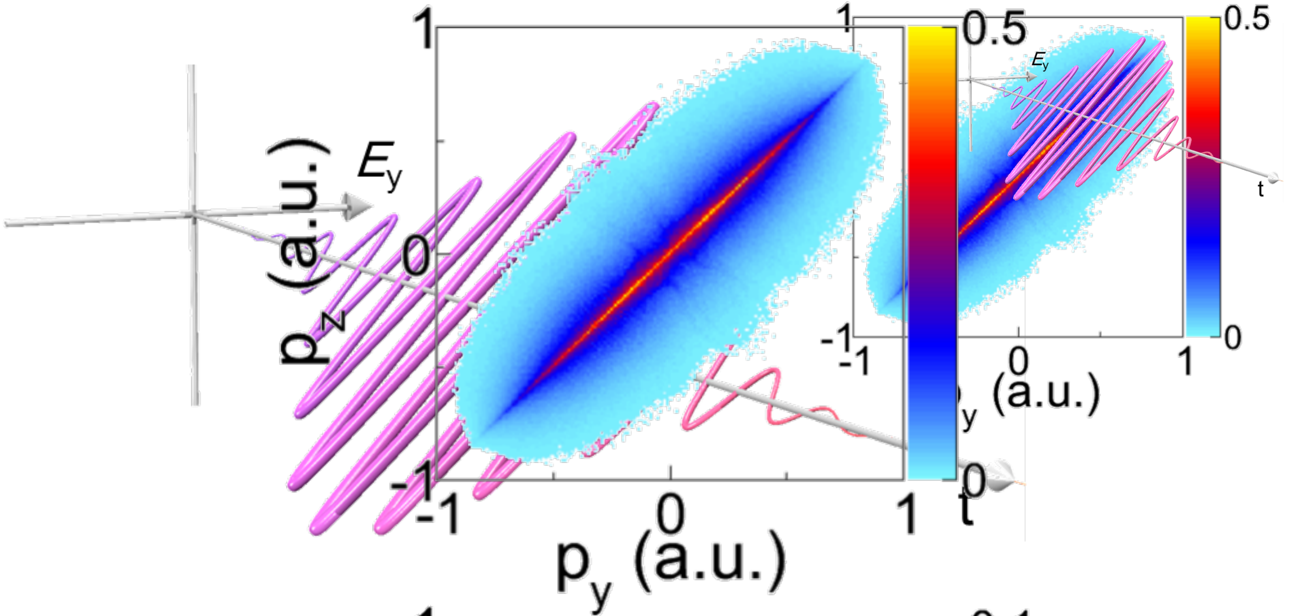
Q. Ji *et al*, Phys. Rev. A 96, 053423 (2017).
Q. Ji *et al*, Phys. Rev. Lett. 123, 233202 (2019).
S. Pan *et al*, Phys. Rev. Lett. 126, 063201 (2021).

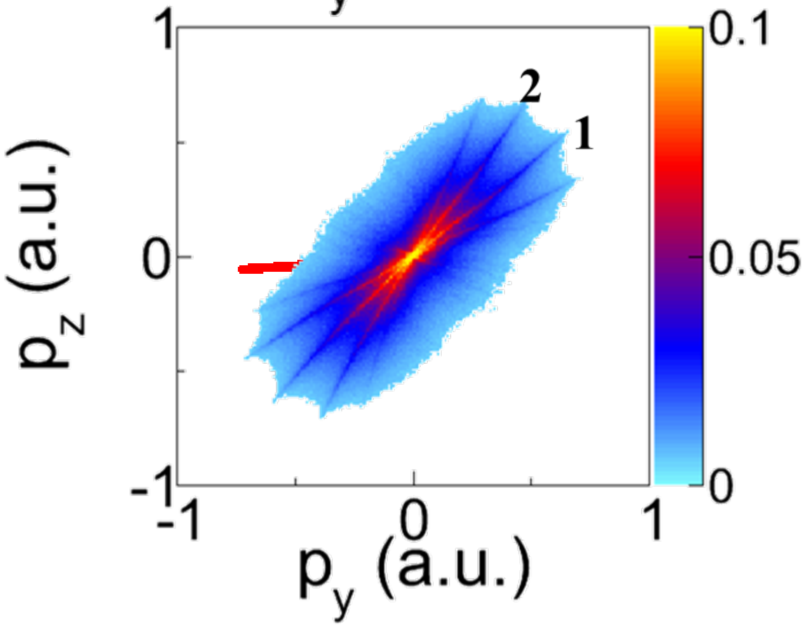
The polarization-skewed laser pulse

multicycle laser pulse

polarization-skewed laser pulse

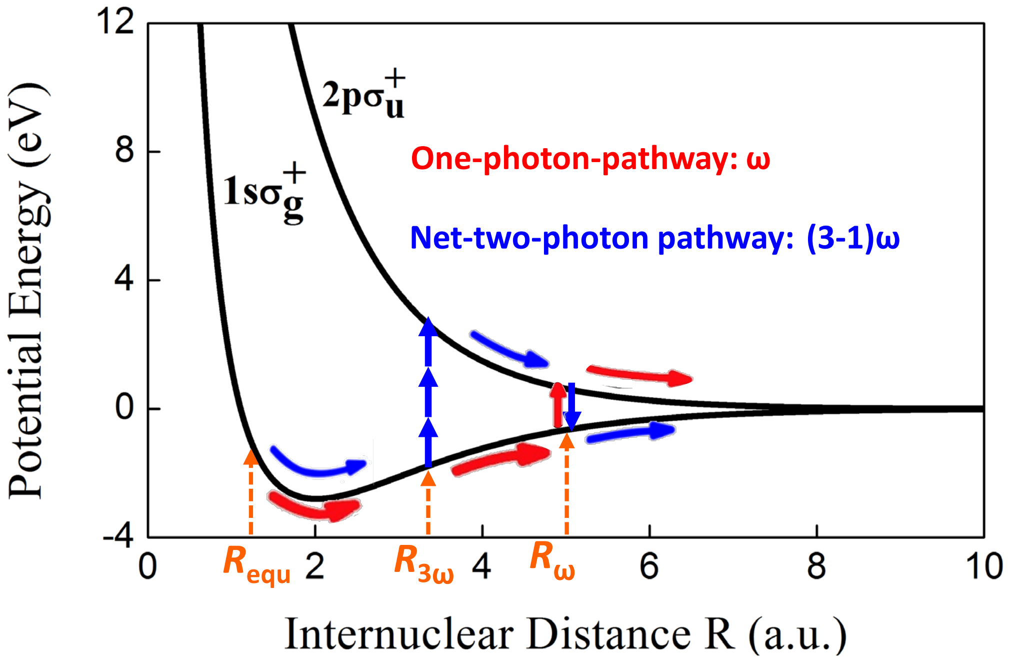


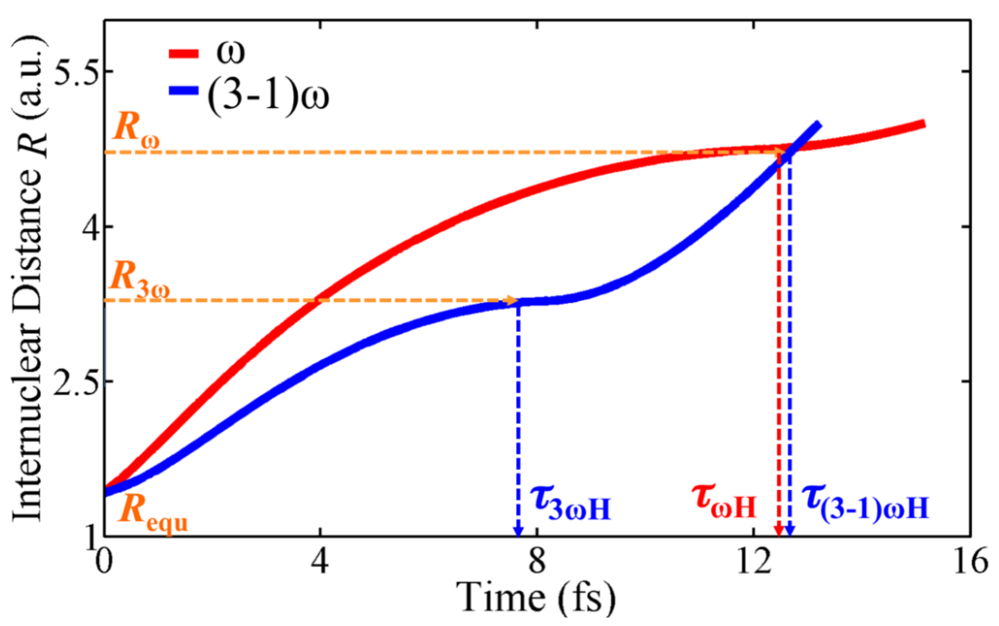


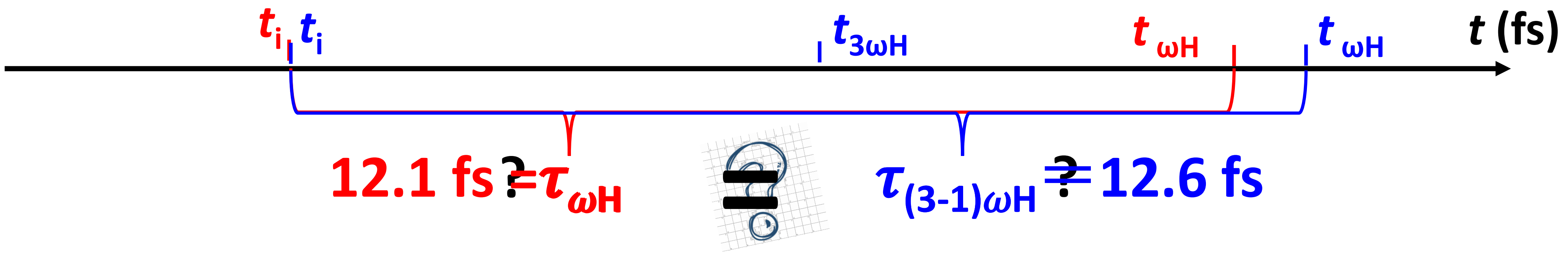


Q. Ji *et al*, Phys. Rev. A 96, 053423 (2017).

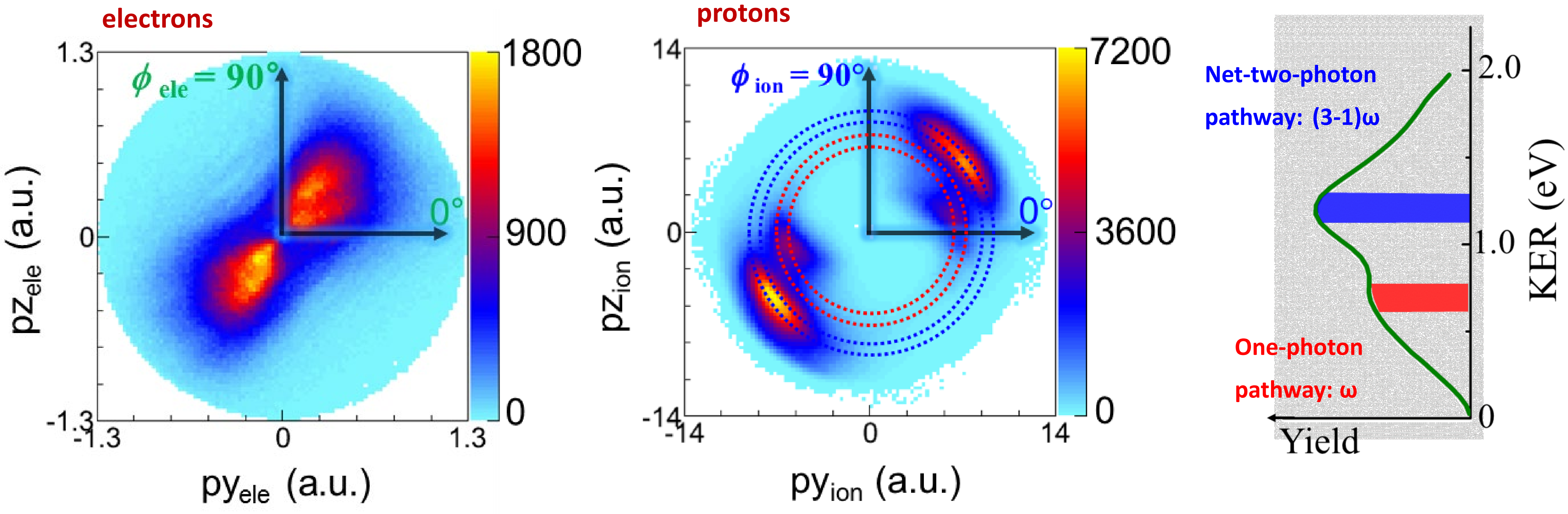
Clocking dissociative single ionization of H_2





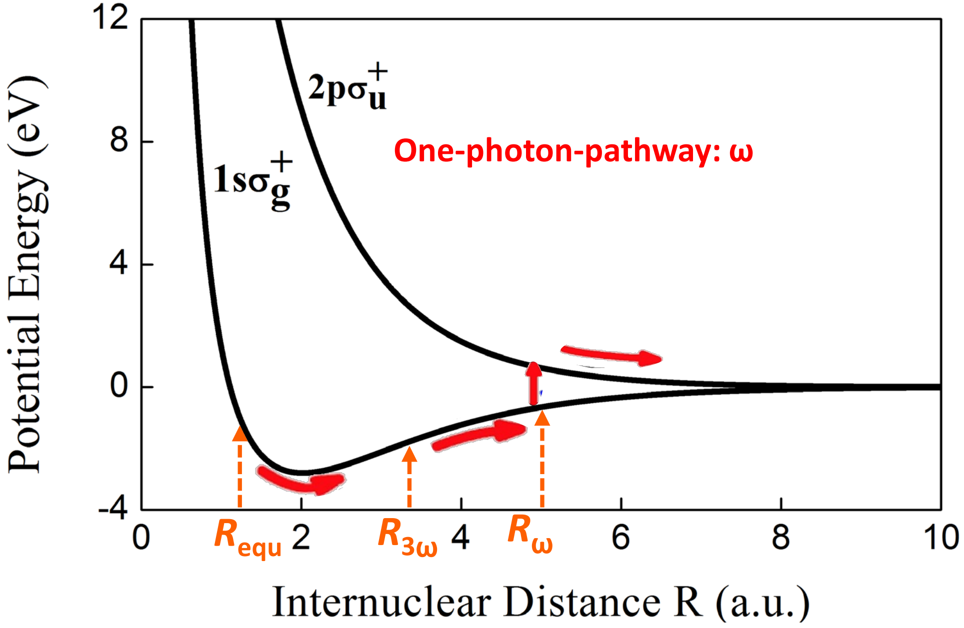


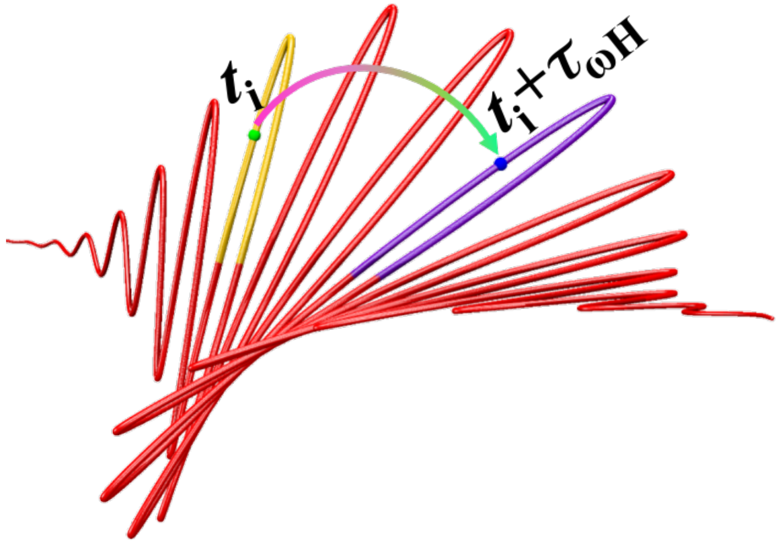
Extraction bond-stretching time: experiments



- KER of proton: distinguish various pathways
- Electron & proton angular distributions: timing using the stopwatch of a PS pulse

Extraction $\tau_{\omega H}$ of the one-photon pathway





$$P_{id}(t_i, \phi_{mol}) \propto P_i(t_i, \phi_{mol}) P_{\omega d}(t_i+\tau_{\omega H}, \phi_{mol})$$

ϕ_{mol}:

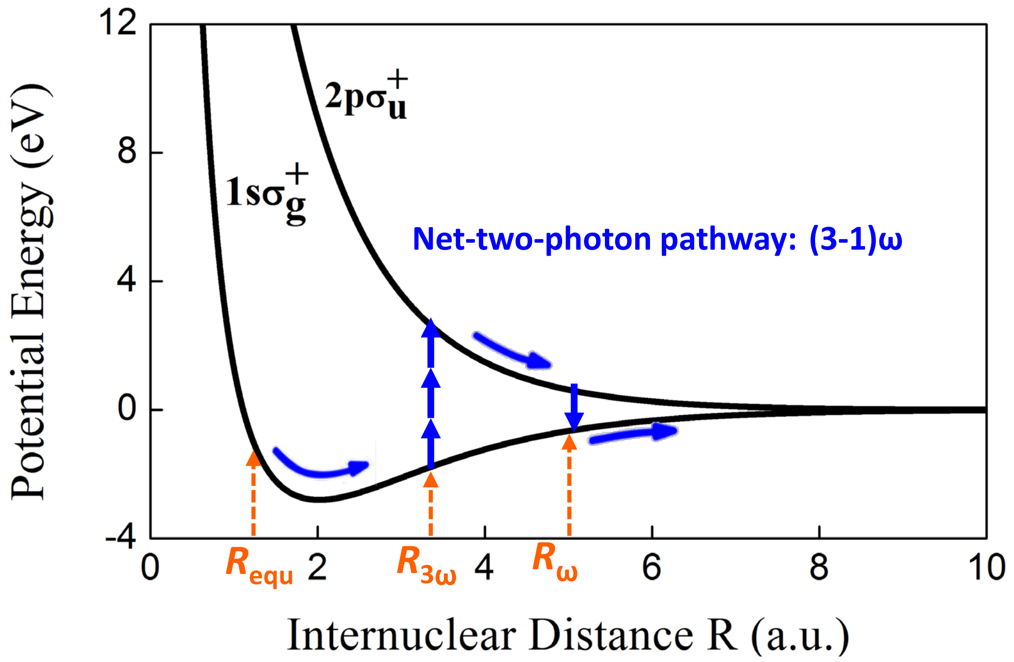
$$P_i(t) \sim \exp\left(-\frac{(t-t_{si})^2}{\sigma_{si}^2}\right).$$

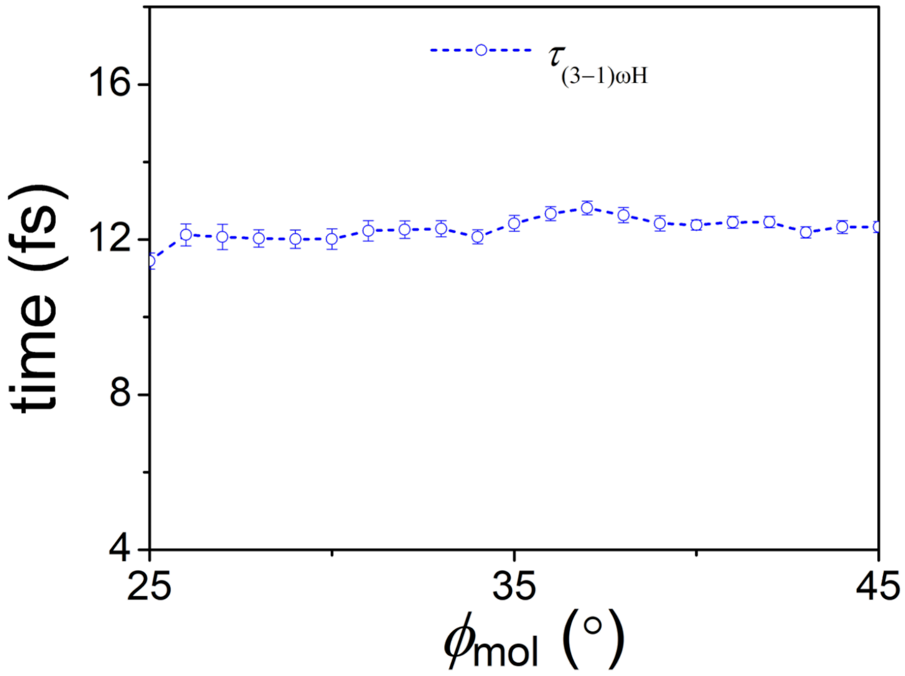
$$P_{\omega d}(t) \sim \exp\left(-\frac{(t-t_{s\omega d})^2}{\sigma_{s\omega d}^2}\right).$$

$$P_{id}(t) \sim \exp\left(-\frac{(t-t_{sid})^2}{\sigma_{sid}^2}\right).$$

$$\tau_{\omega H} = (t_{s\omega d} - t_{sid}) + (t_{si} - t_{sid})\frac{\sigma_{s\omega d}^2}{\sigma_{si}^2}$$

Extraction $\tau_{(3\text{-}1)\omega H}$ of the net-two-photon pathway

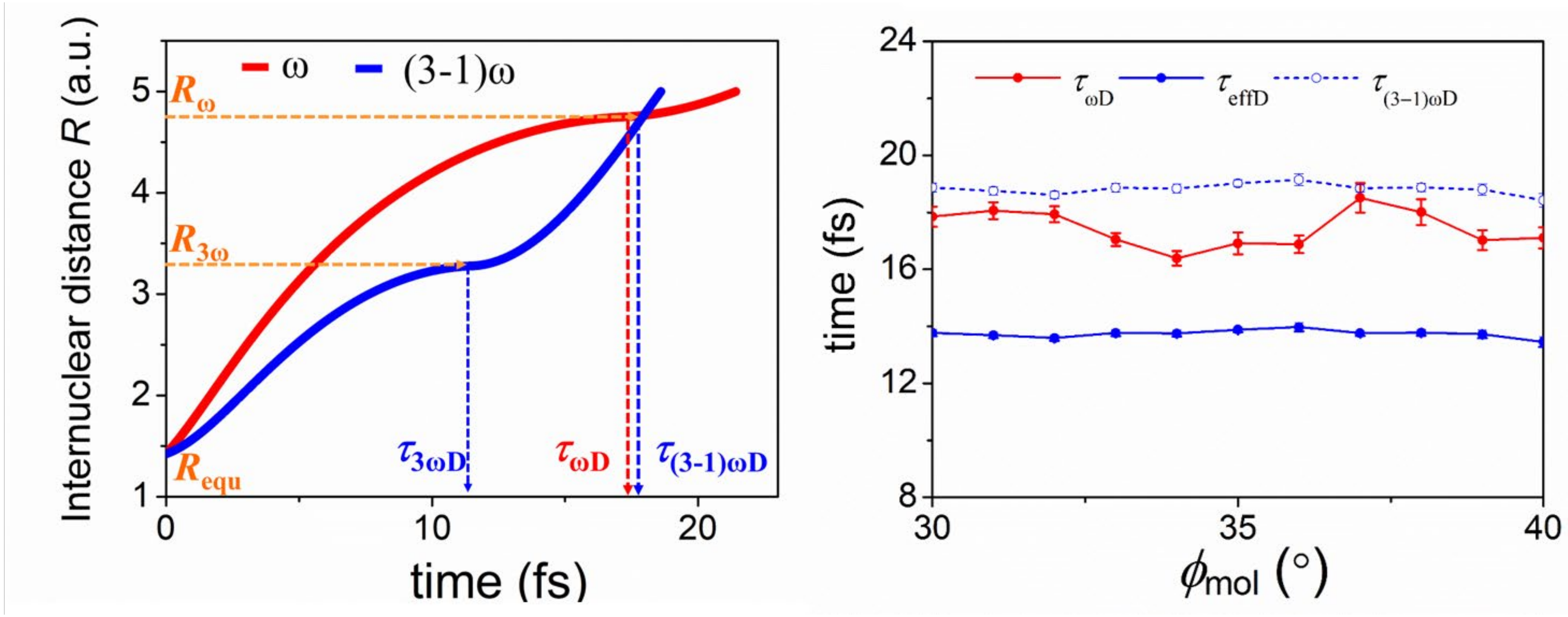




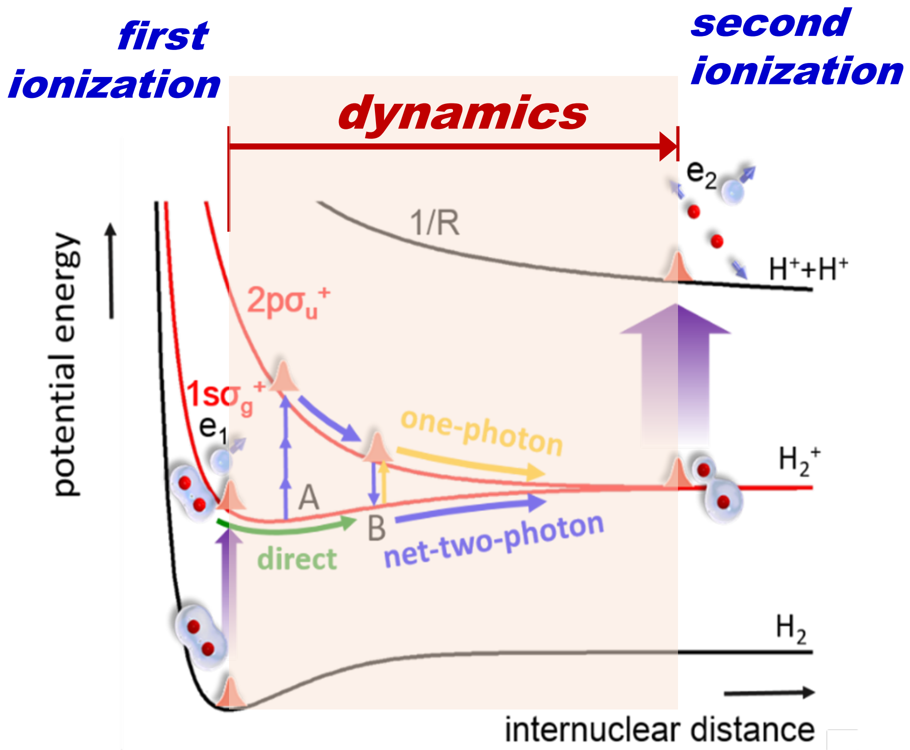
$\tau_{(3\text{-}1)\omega H}$ =12.3 ± 0.3fs = $\tau_{\omega H}$ =12.8 ± 0.6fs

The bond-stretching time of D_2^+

$$\tau_{\omega D} = \sqrt{2}\ \tau_{\omega H} \qquad \tau_{(3\text{-}1)\omega D} = \sqrt{2}\ \tau_{(3\text{-}1)\omega H}$$



Clocking dissociative double ionization of H_2



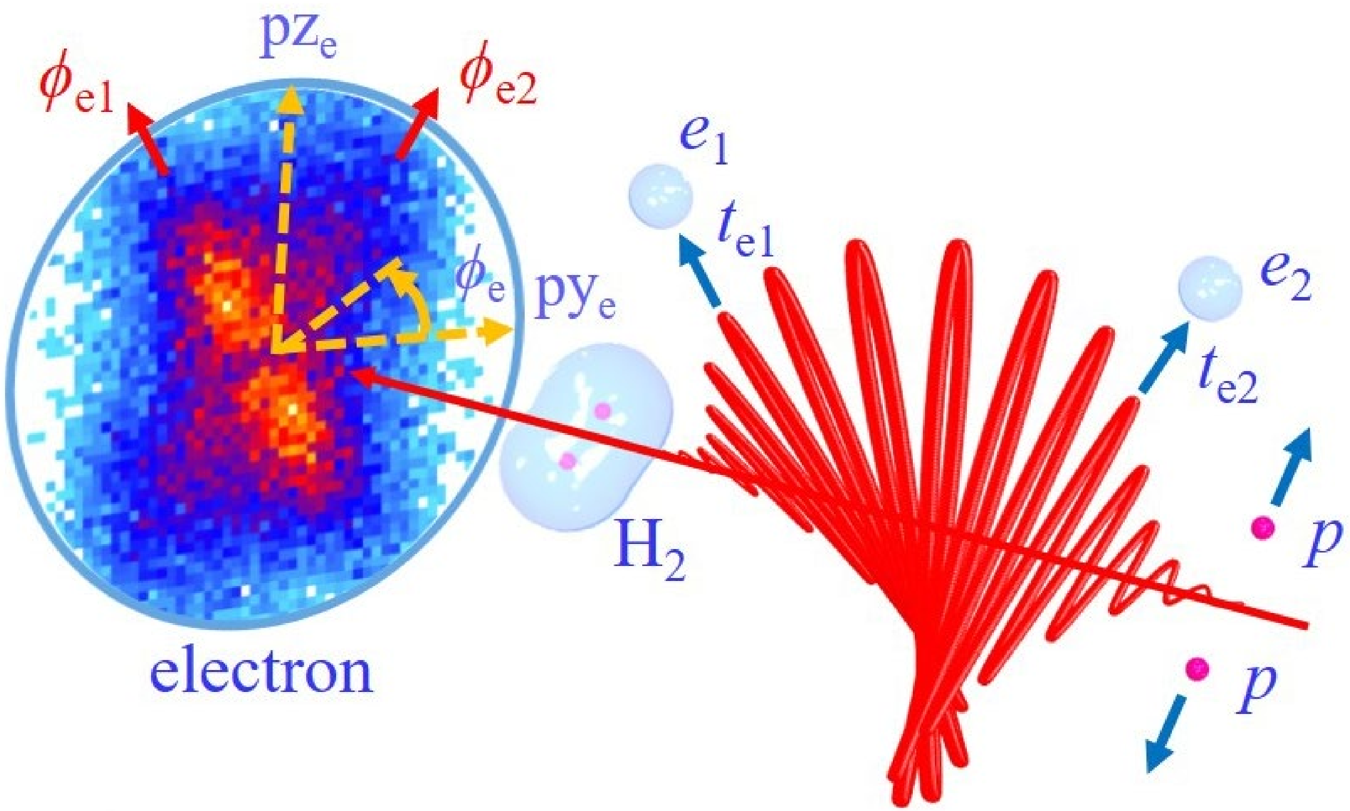
$H_2 + m\hbar\omega \rightarrow H_2^+ + e_1$

$H_2^+ + q\hbar\omega \rightarrow H^+ + H^+ + e_2$

Dynamics of ATDI of molecules:

- Only analyzed by the KER spectra
- Real-time observation?
- Time interval between two ionization steps?

Ultrafast stopwatch: clocking ADTI of H_2



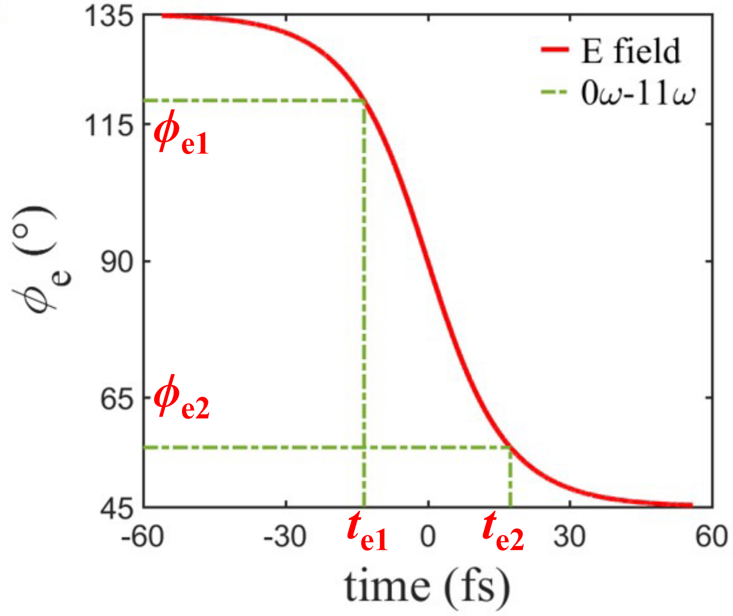
$H_2 + m\hbar\omega \rightarrow H_2^+ + e_1$

$H_2^+ + q\hbar\omega \rightarrow H^+ + H^+ + e_2$

$$\phi_{e1} \rightarrow t_{e1}$$

$$\phi_{e2} \rightarrow t_{e2}$$

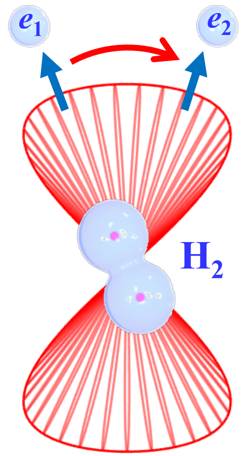
$$\Delta t = t_{e2} - t_{e1}$$

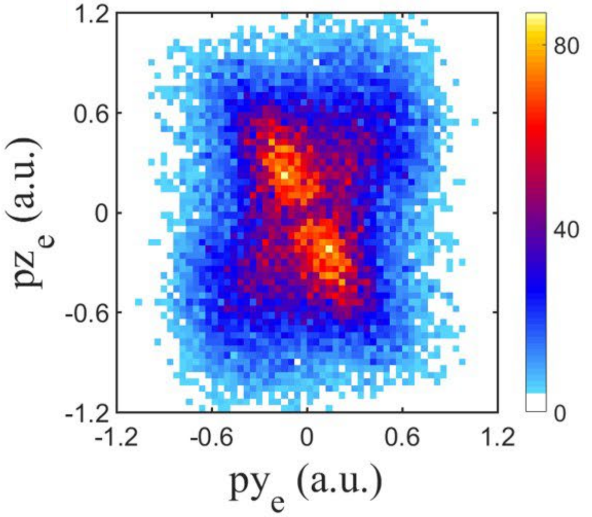


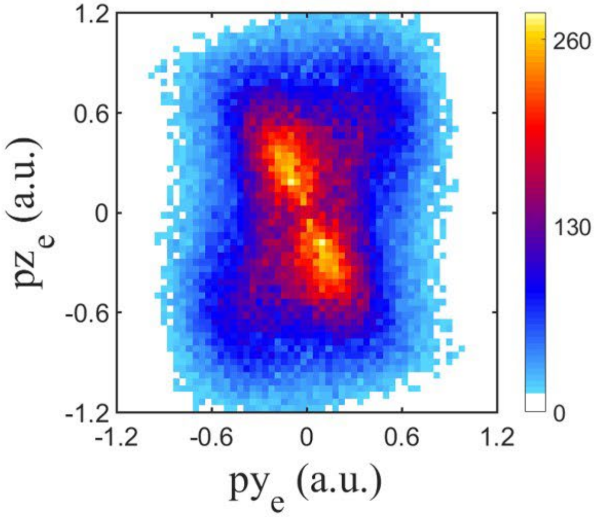
S. Pan *et al*, Phys. Rev. Lett. 126, 063201 (2021).

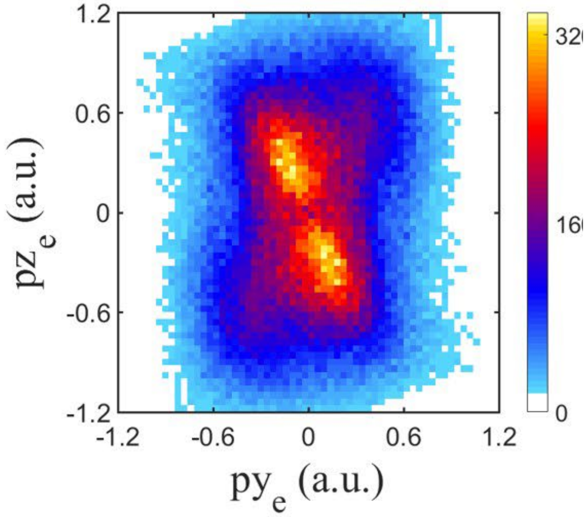
Pathway-resolved momentum distribution of e_1 & e_2

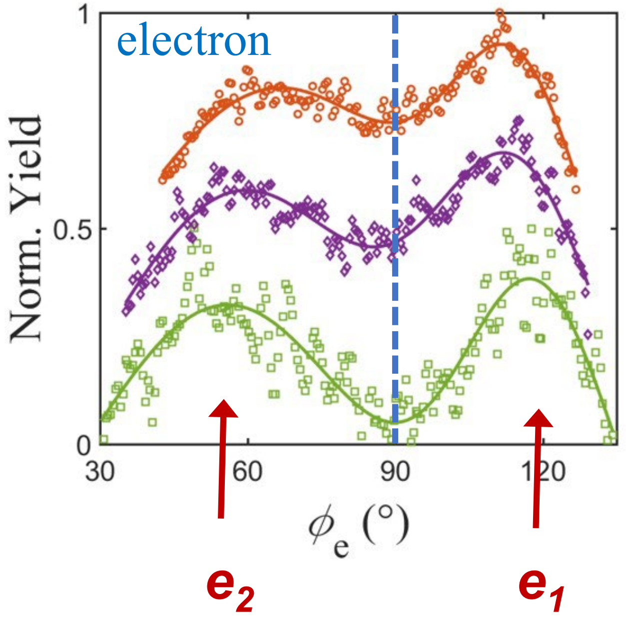
Direct pathway	One-photon pathway	Net-two-photon pathway
0ω-11ω	1ω-11ω	2ω-11ω
30.9 fs (exp.)	25.6 fs (exp.)	21.1 fs (exp.)
29.1 fs (sim.)	25.6 fs (sim.)	19.8 fs (sim.)





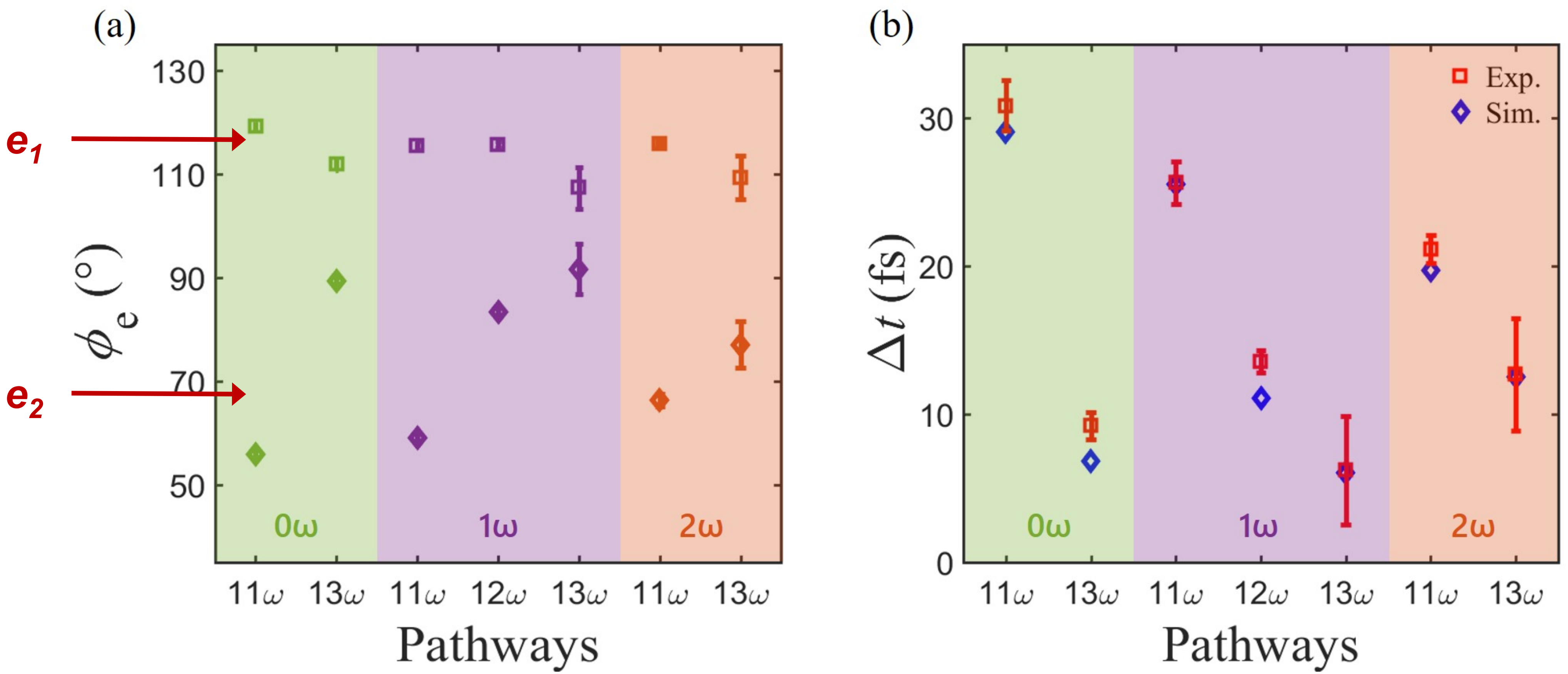






S. Pan *et al*, Phys. Rev. Lett. 126, 063201 (2021).

Measured vs. simulated timings of ATDI

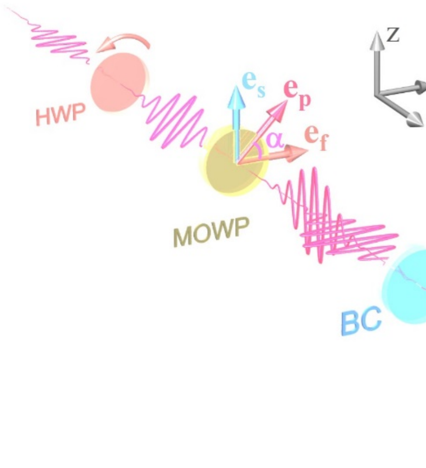


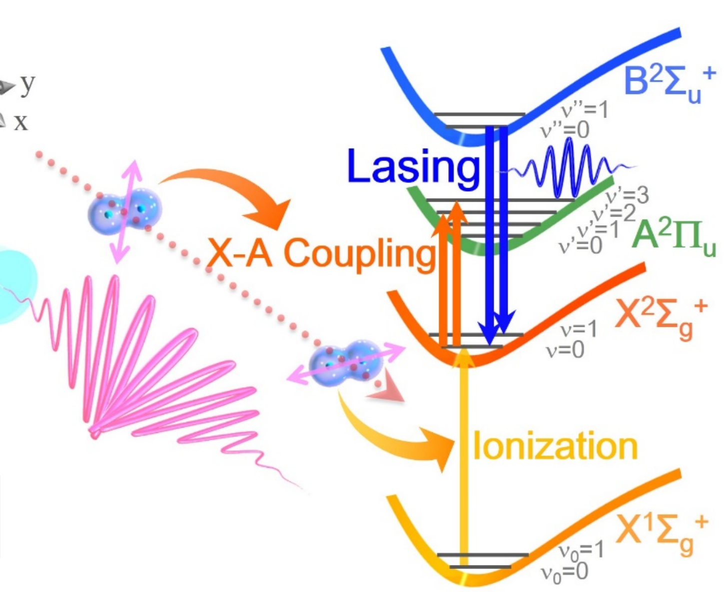
Experimentally resolved time intervals agree well with classically simulated results.

S. Pan *et al*, Phys. Rev. Lett. 126, 063201 (2021).

华東師範大學
EAST CHINA NORMAL UNIVERSITY

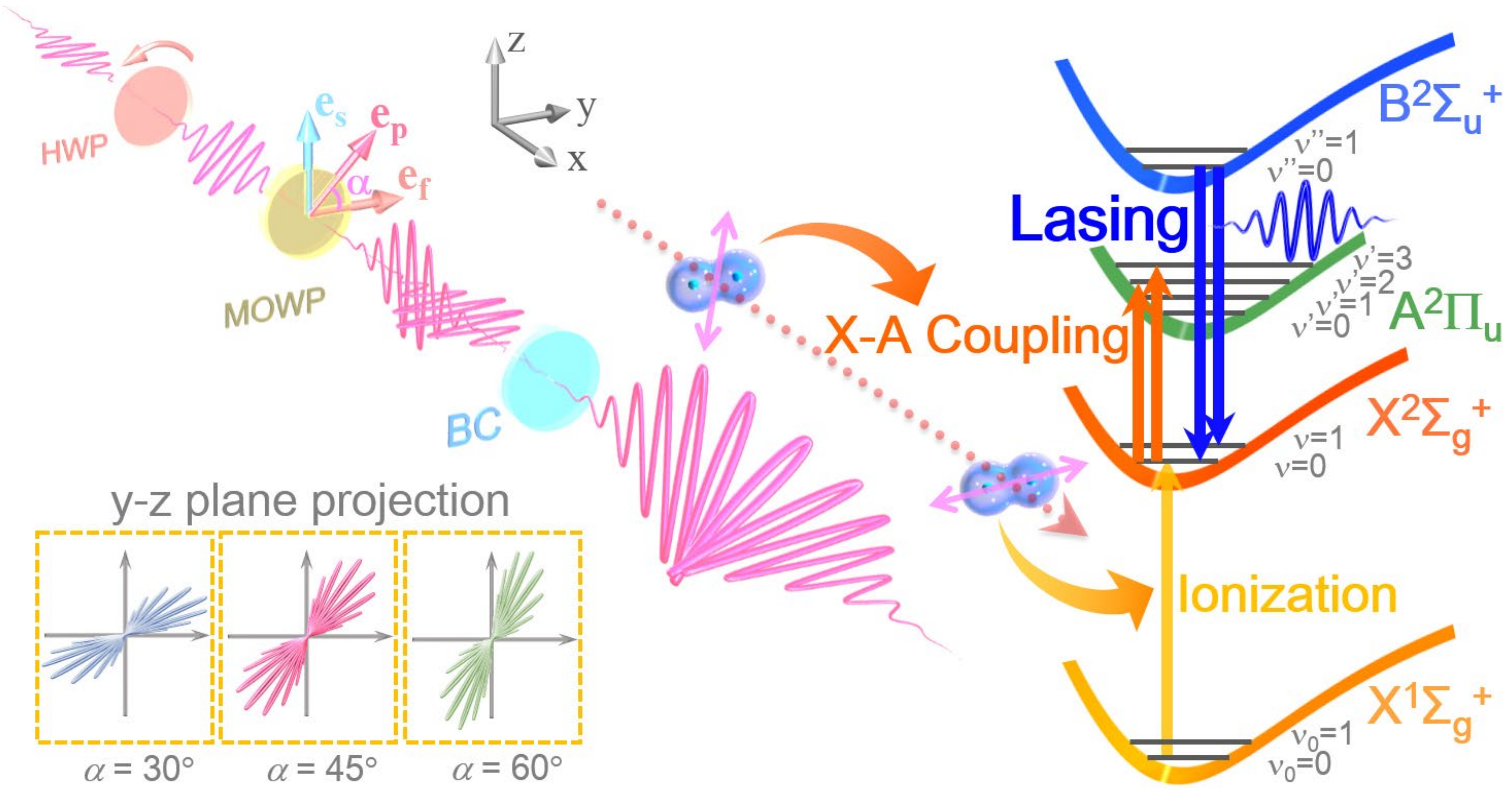
Optimization of N_2^+ lasing by waveform-controlled polarization-skewed pulses





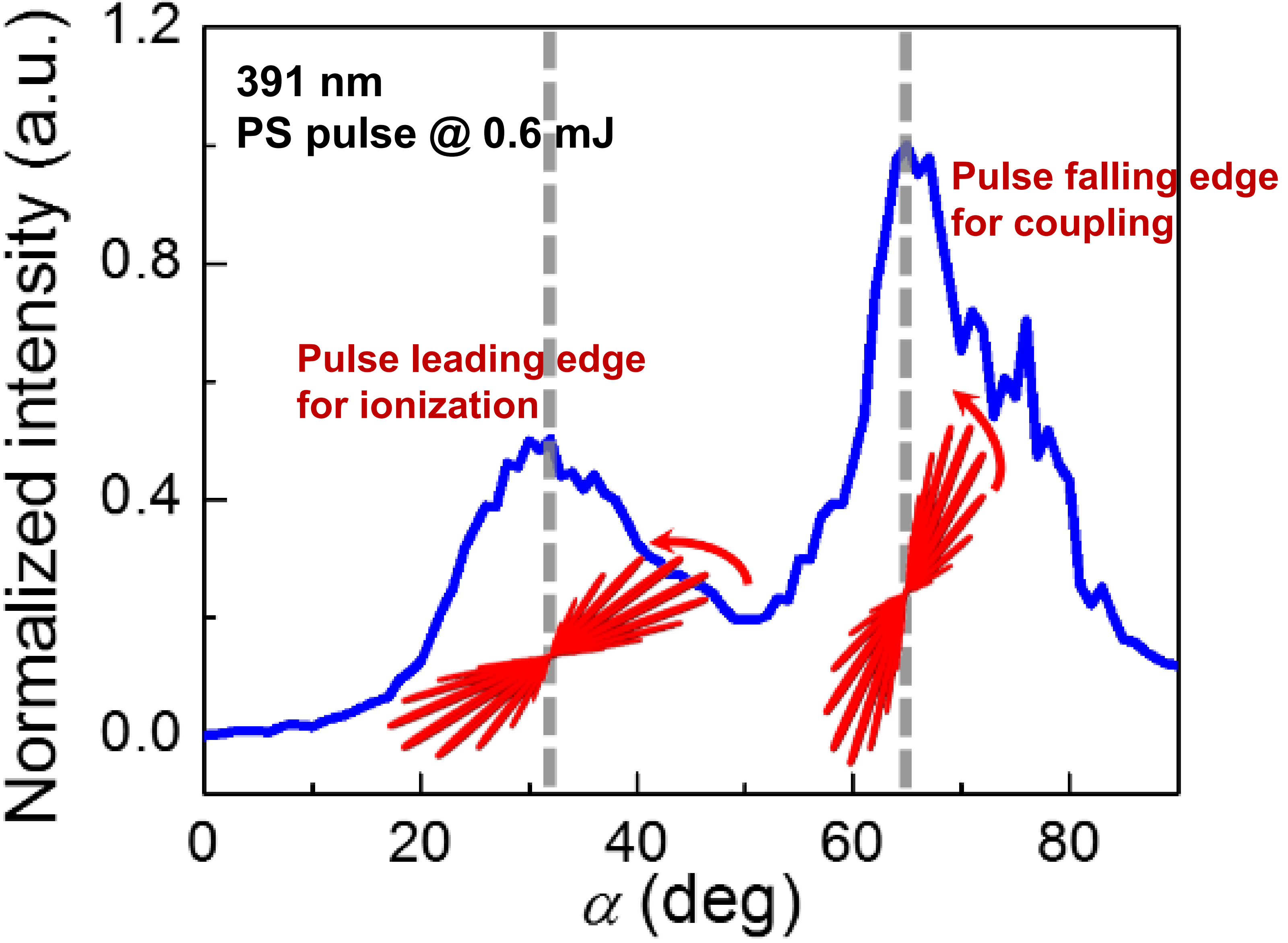
femtosecond filamentation in air

Spatiotemporal waveform shaped PS pulse for N_2^+ lasing

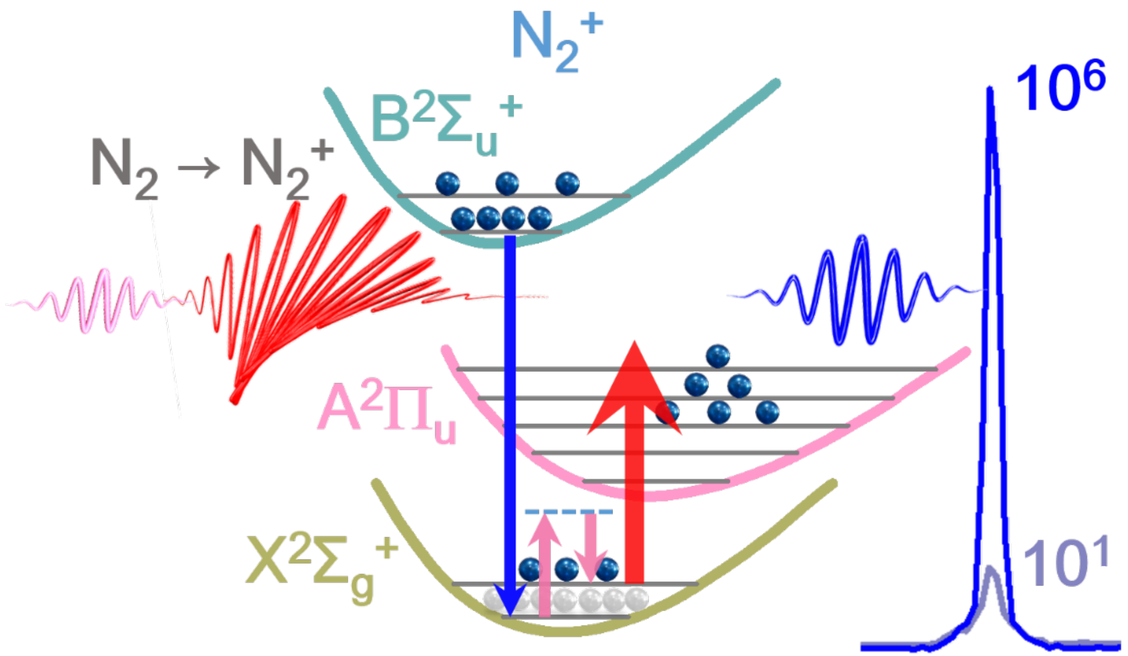


H. Li *et al*, Phys. Rev. Lett. 125, 053201 (2020).
H. Li *et al*, Opt. Lett. 45, 6591 (2020).

Optimizing 391 nm lasing by PS pulse

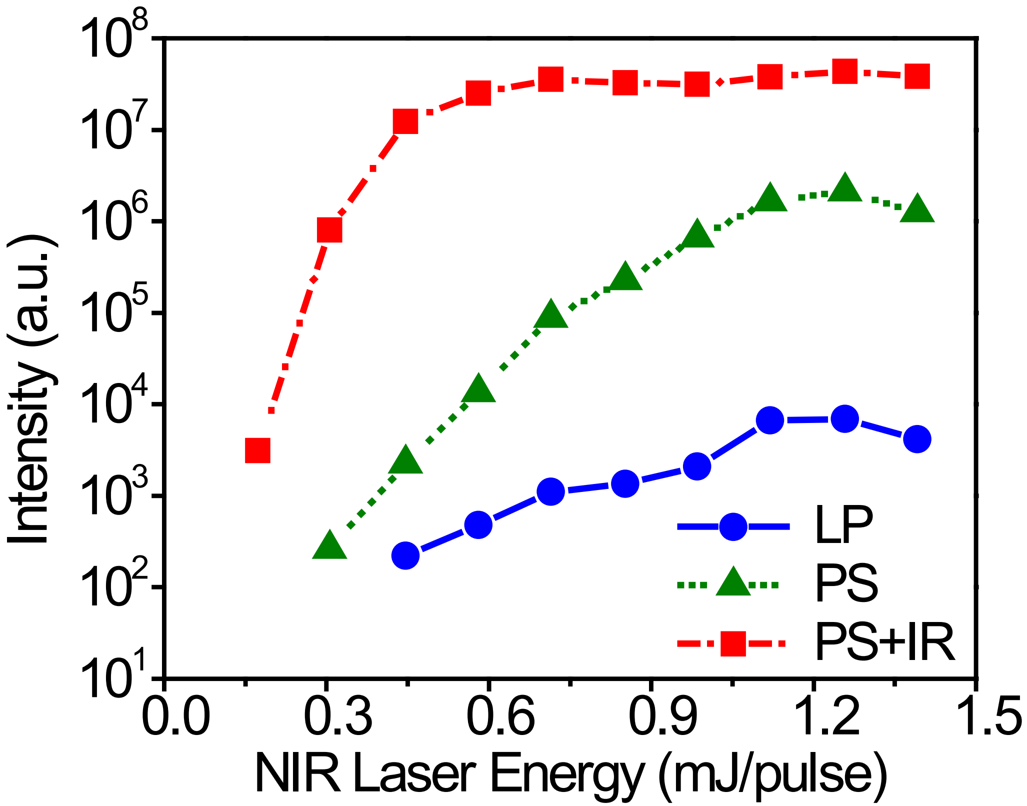


Enhanced N_2^+ lasing by pulse shaping



$$X^2\Sigma_g^+ (v = 1) \leftarrow X^2\Sigma_g^+ (v = 0)$$

$$A^2\Pi_u (v = 2,3,4) \leftarrow X^2\Sigma_g^+ (v = 1)$$



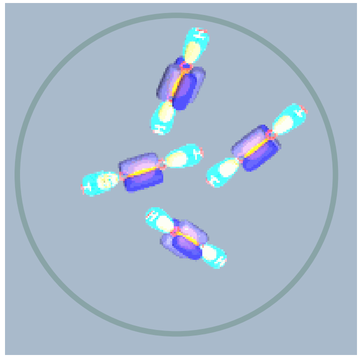
H. Li *et al.*, Phys. Rev. Lett. 125. 053201 (2020).

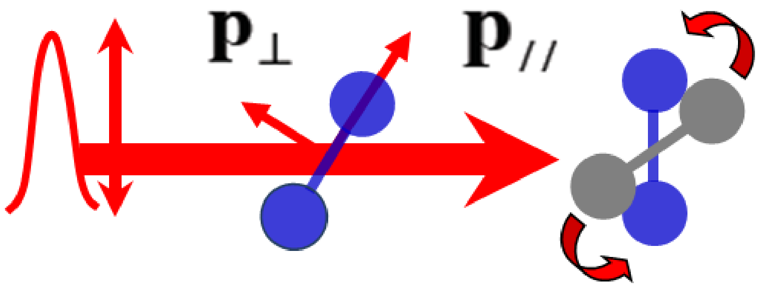
Outline

- **Background Introduction**
- **Multiphoton energy absorption: electron-nuclear correlation**
  - ATI & ATD in dissociative ionization of molecules
  - Strong-field Rydberg excitation of molecules
  - An ultrafast stopwatch to clock molecular bond stretching
- **Ro-vibrational dynamics of the nuclear wave-packet**
  - Visualizing unidirectional molecular rotation
  - Echoes of molecules
  - All-optical 3D orientation of molecules
- **Attosecond dynamics of electrons: visualization and control**

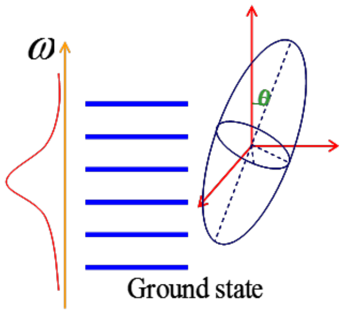
Visualizing unidirectional molecular rotation

Laser induced alignment of molecules

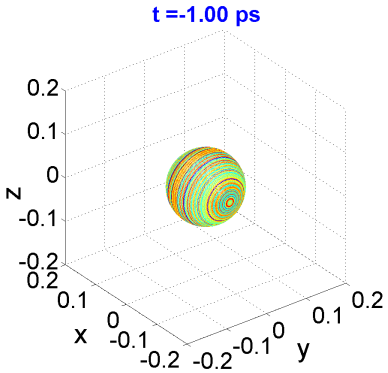




molecular alignment: impulsive



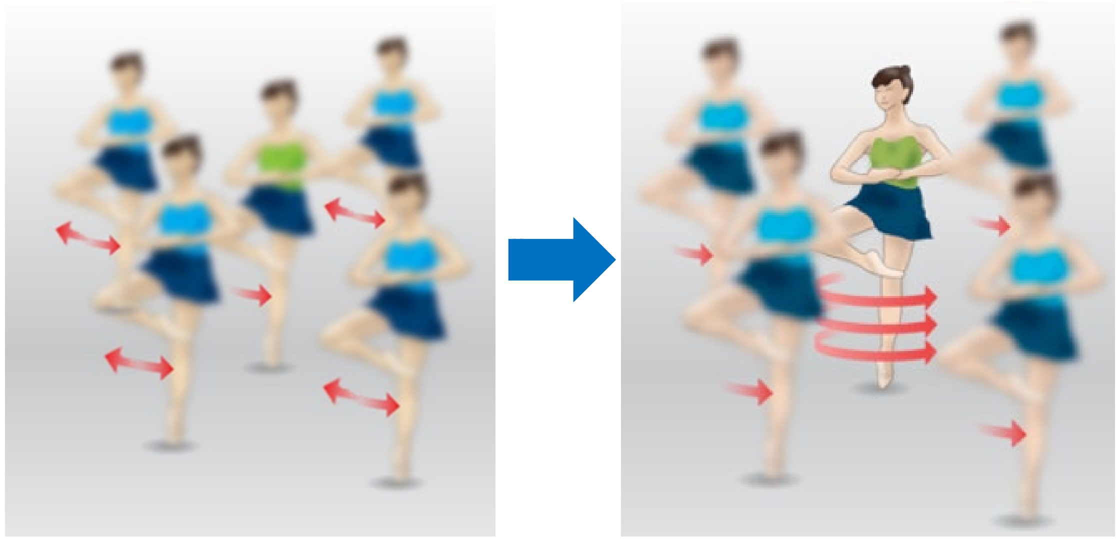
Impulsive rotational
Raman excitation

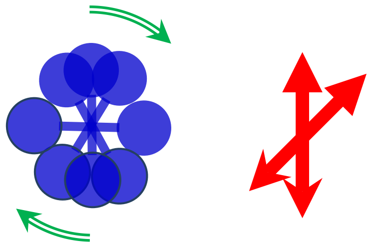


Quantum dynamics:
rotational wave packet

Molecular unidirectional rotation

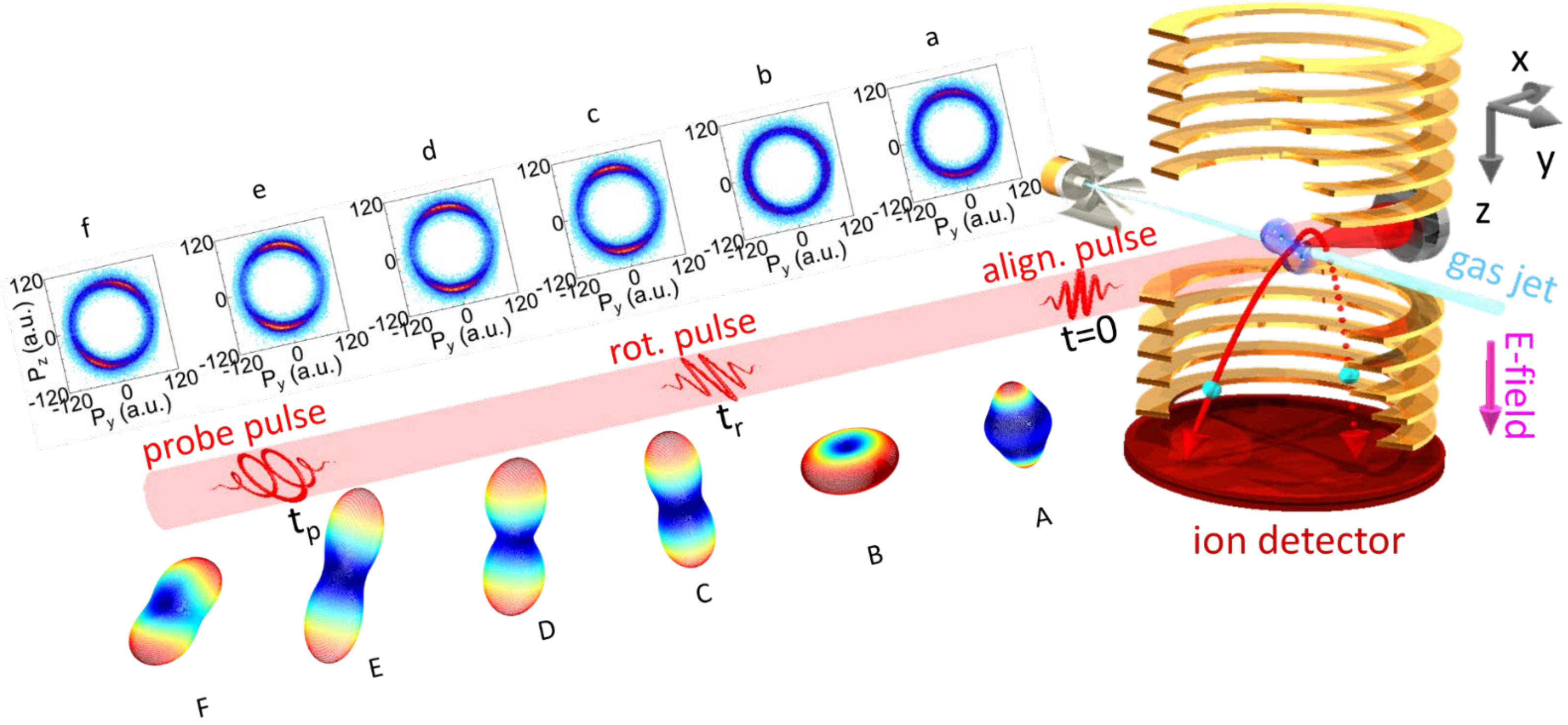
Molecular unidirectional rotation (UDR): molecular super-rotor





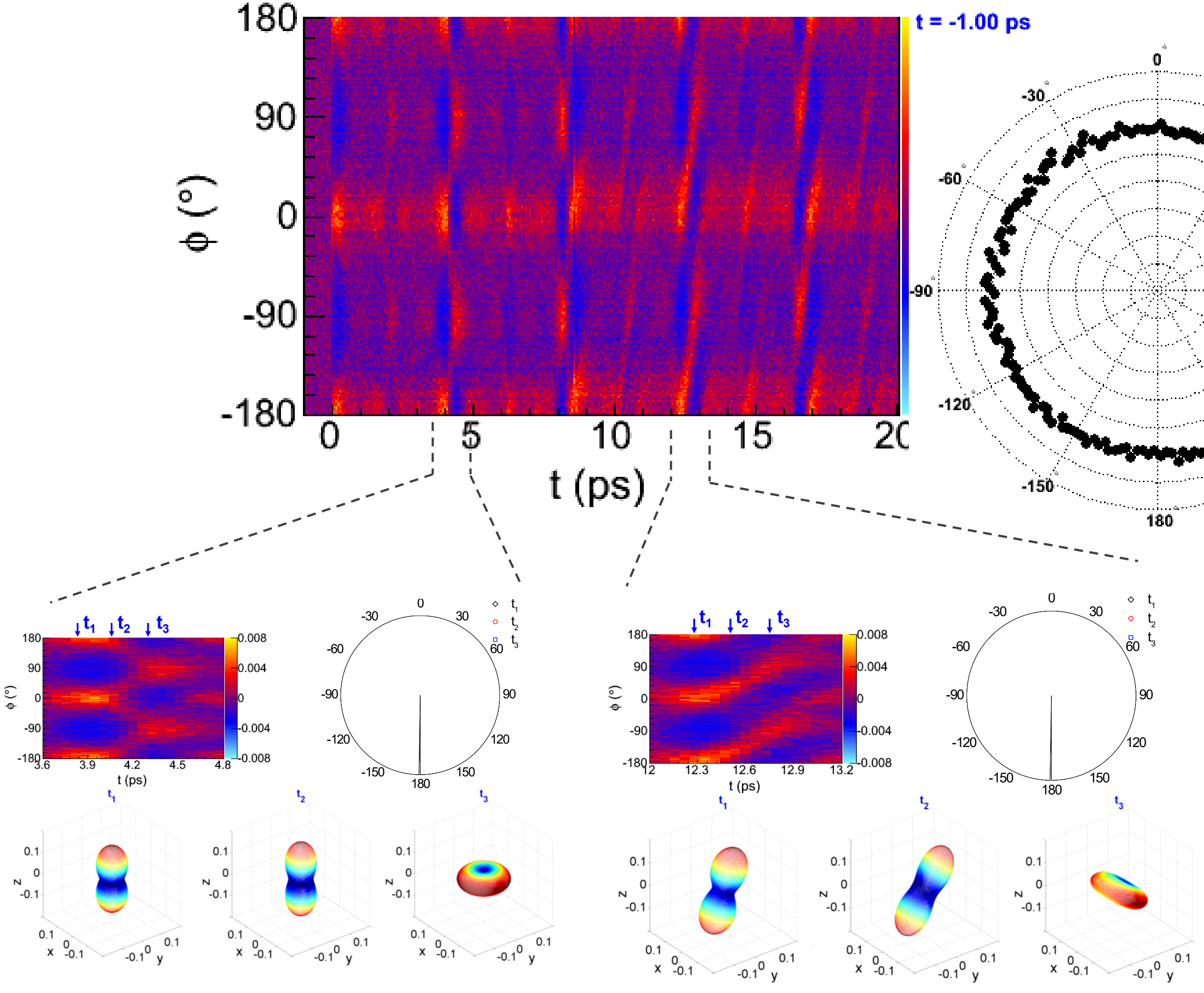
Visualizing rotational wave-packet

Femtosecond coincidence imaging:
Coulomb explosion of molecule



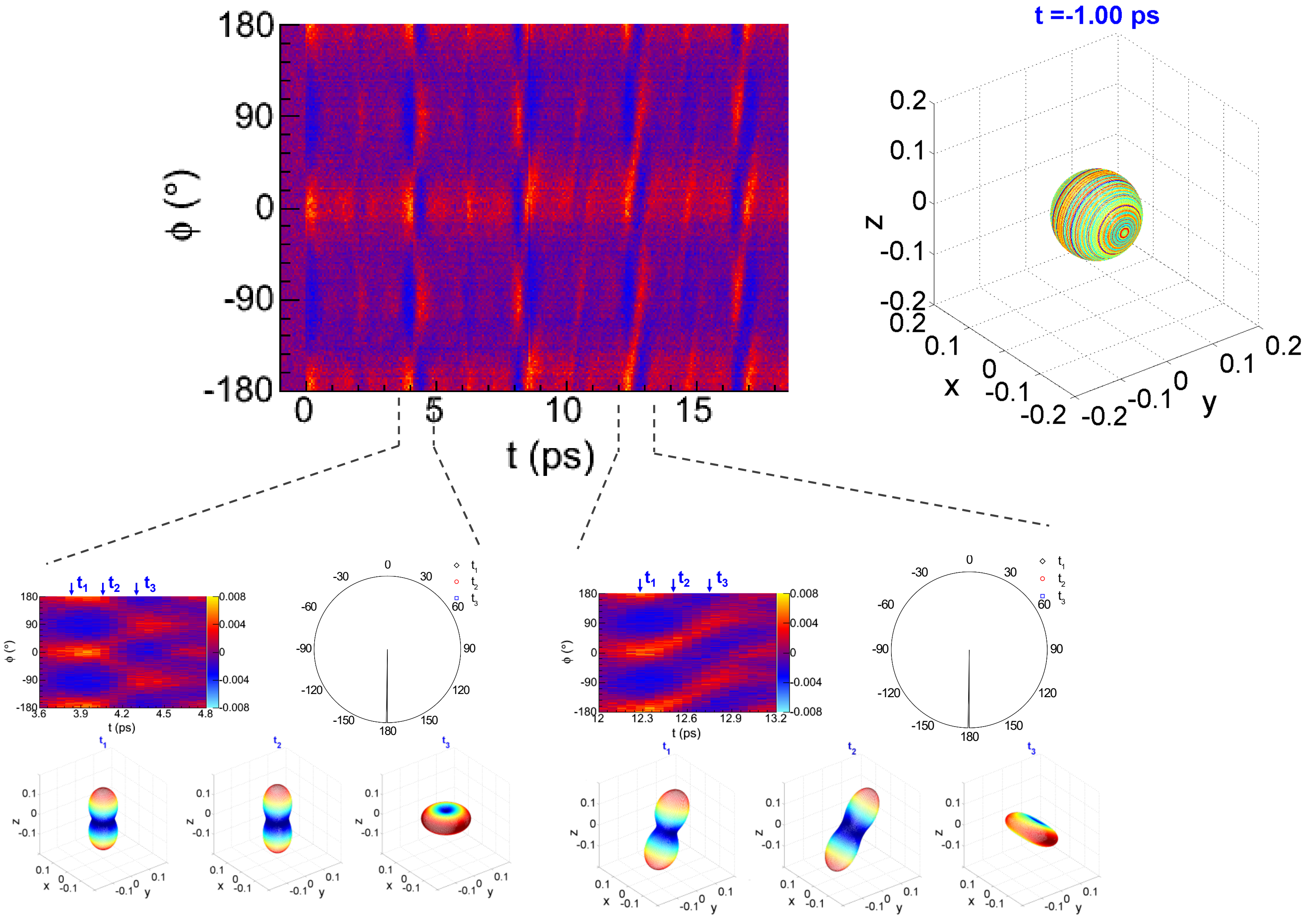
K. Lin *et al.*, PRA 92, 013410 (2015).

Visualizing molecular angular distribution: UDR



K. Lin *et al.*, PRA 92, 013410 (2015).

Visualizing molecular angular distribution: UDR

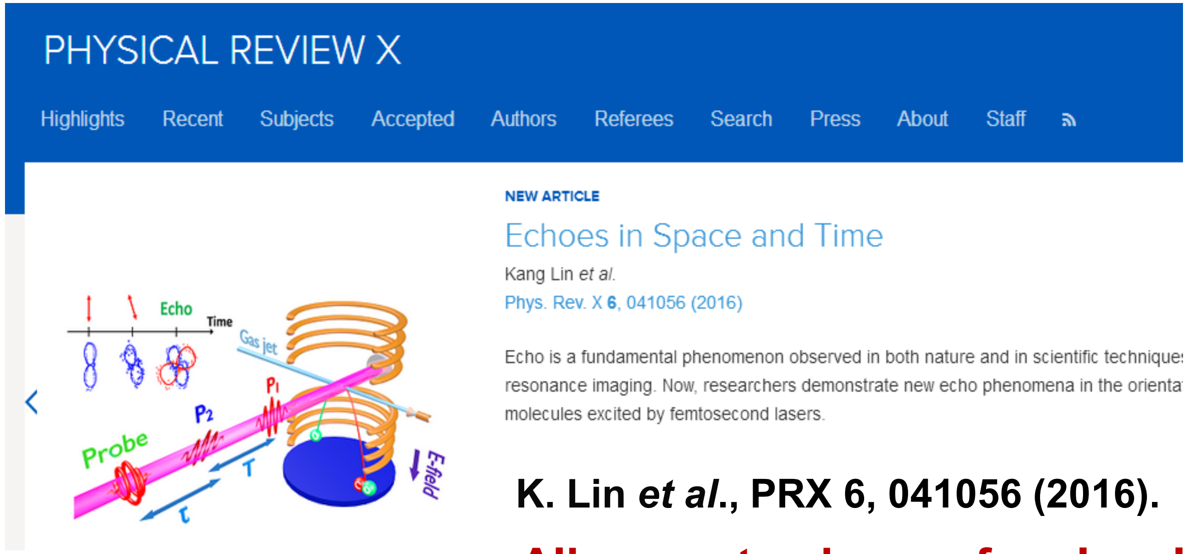


K. Lin *et al.*, PRA 92, 013410 (2015).

Echoes of molecules: rotational & vibrational excitation

Alignment echoes of molecules

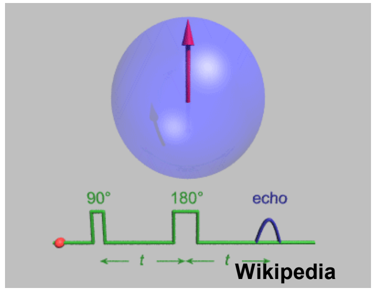
Echo in mountains:
acoustic pulse mirrored by rocks



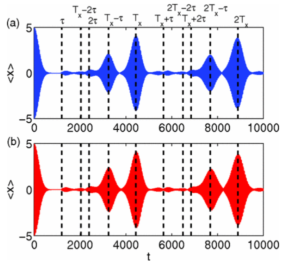
K. Lin *et al.*, PRX 6, 041056 (2016).

Alignment echoes of molecules

Spin echo

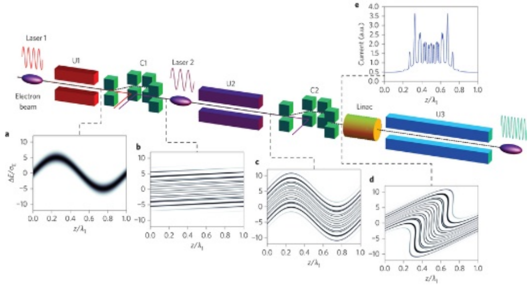


Cold atoms



Phys. Rev. A 86, 023613 (2012).

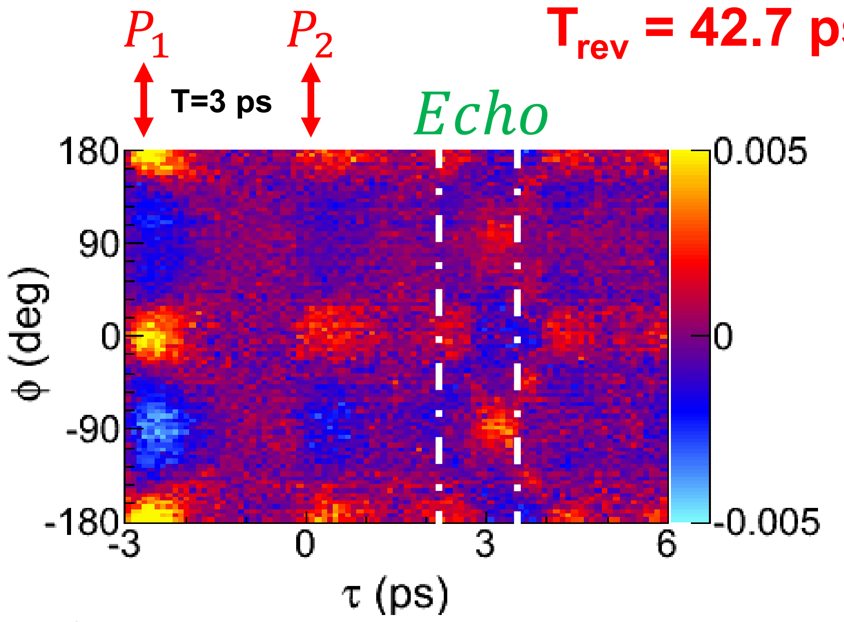
Free-electron lasers

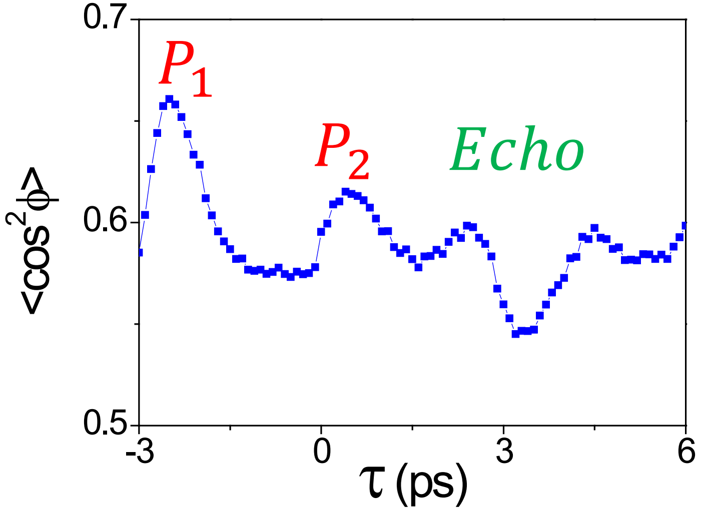


Nature Photonics 10, 512 (2016).

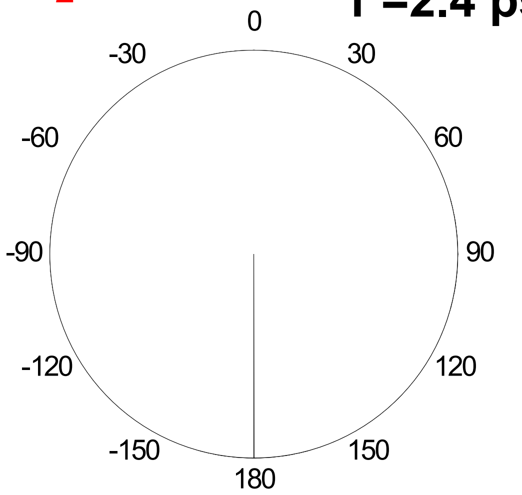
Alignment echoes of molecules: full echoes

T_{rev} = 42.7 ps , CO_2

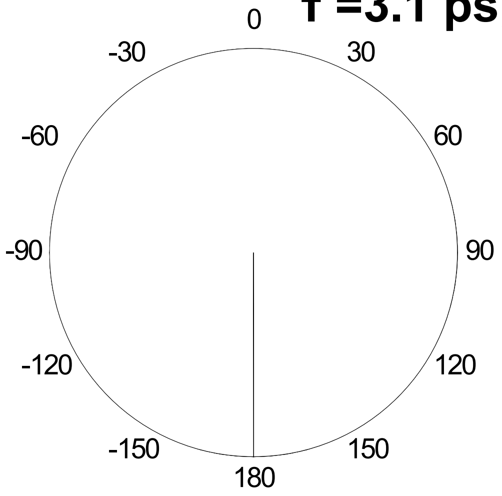




τ =2.4 ps, alignment

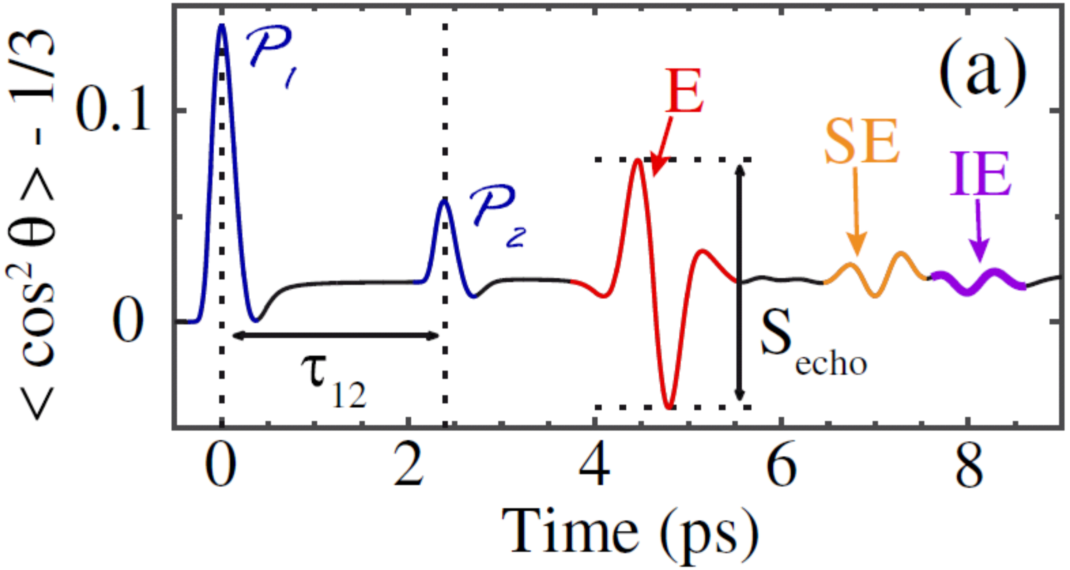


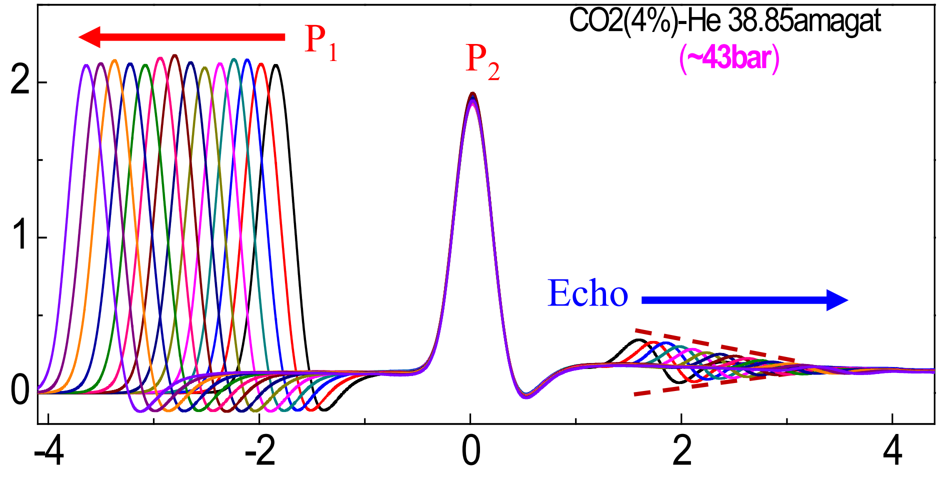
τ =3.1 ps, anti-alignment



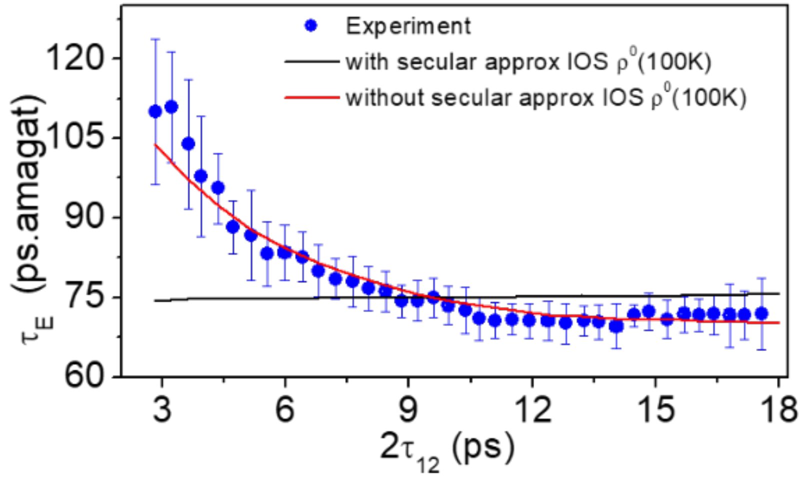
K. Lin *et. al.* Phys. Rev. X 6, 041056 (2016).

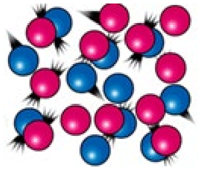
Collisional decay of rotational echoes: dense target





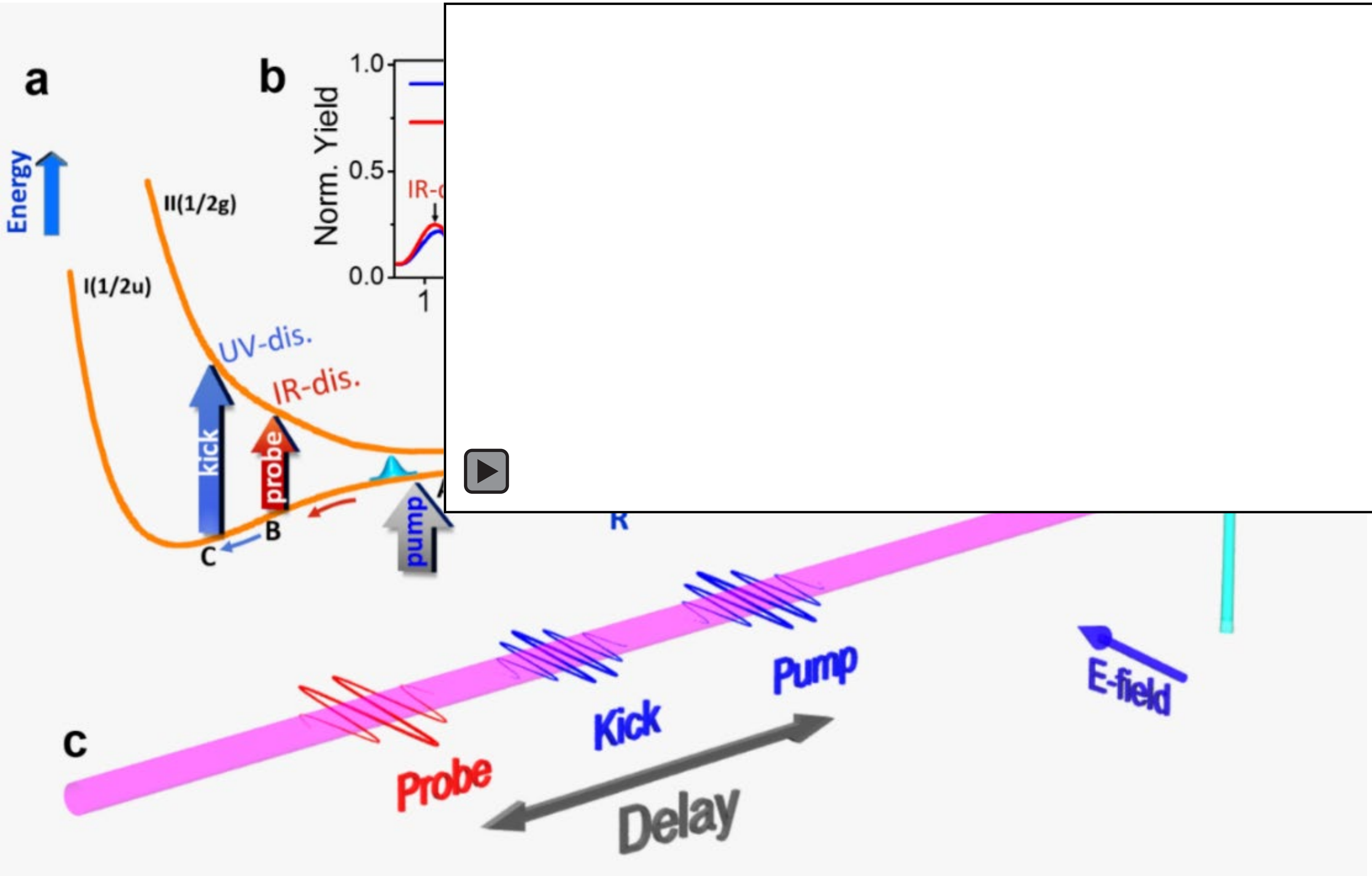
- Timing of the echo is controllable
- Collisional molecular dynamics in dense gas





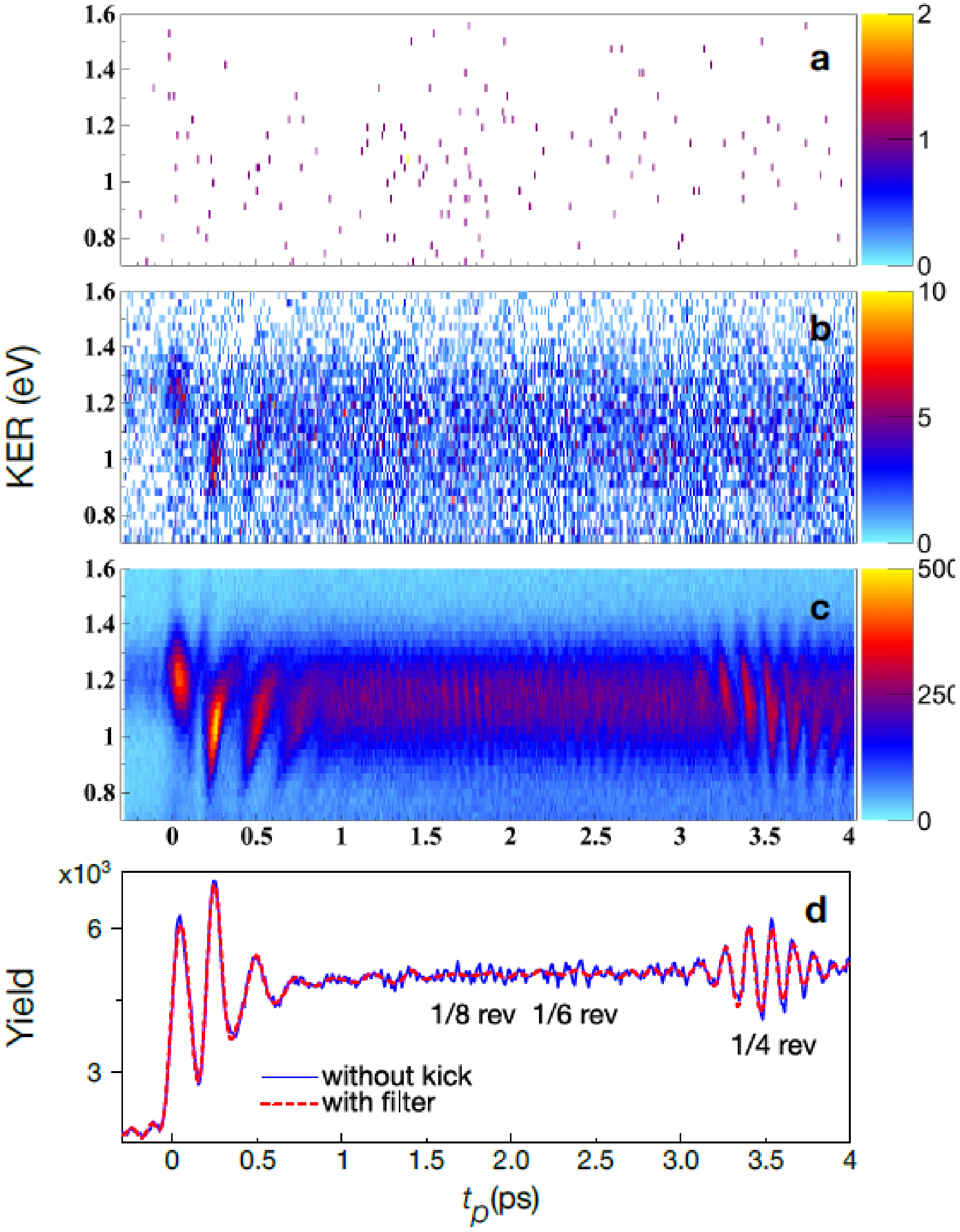
H. Zhang *et. al.* Phys. Rev. Lett. 122, 193401 (2019).
J. Ma *et al.*, Nature Communications 10, 5780 (2019).

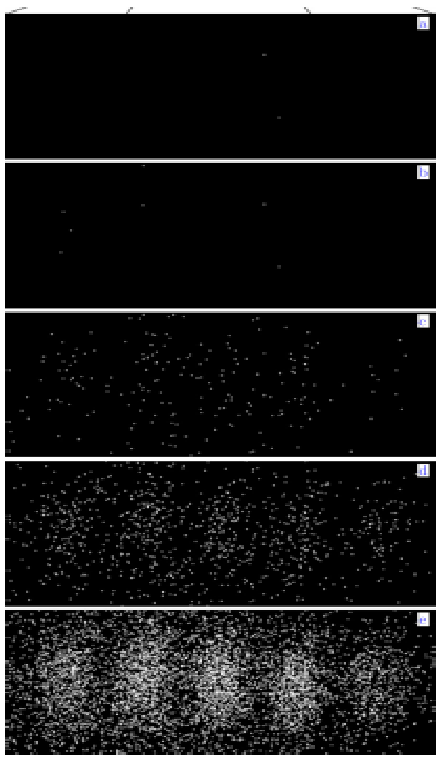
Echoes of vibrational excitation



J. Qiang *et al.,* Nature Physics 16, 328 (2020).
J. Wu *et al.,* Phys. Rev. Lett. 110, 033005 (2013).

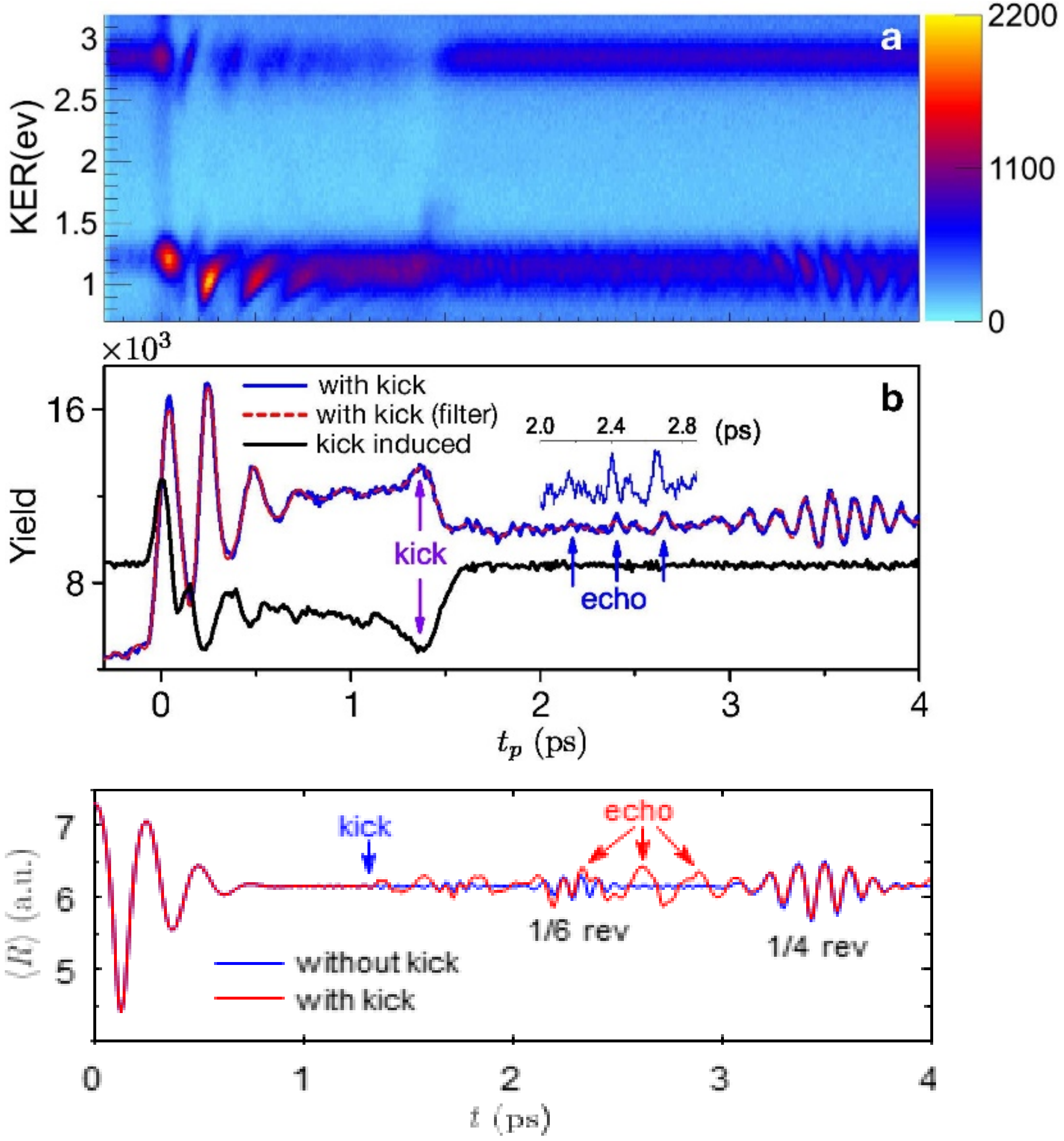
Vibrational wave-packet dynamics





single electron experiment

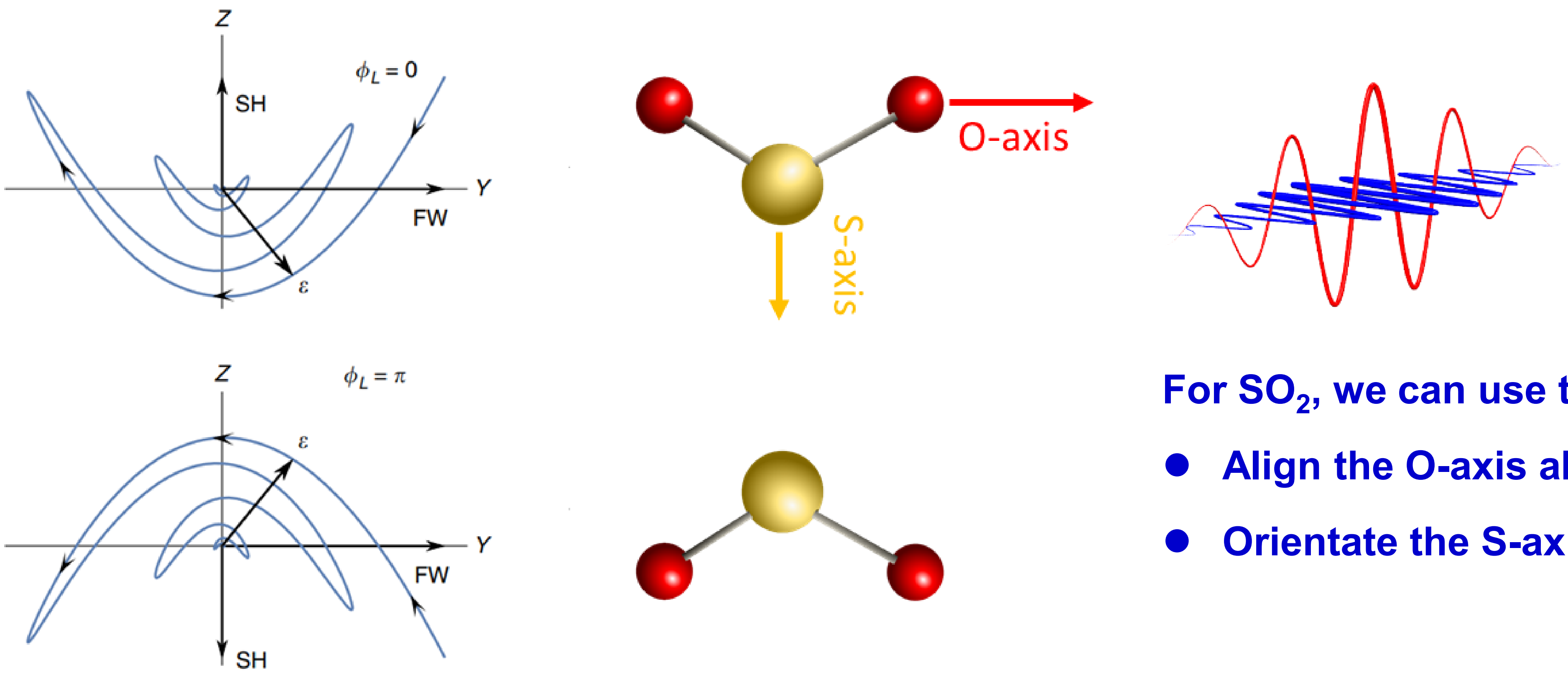
Echoes of vibrational excitation in a single molecule



J. Qiang *et al.*, Nature Physics 16, 328 (2020).

All-optical 3D orientation of molecules

All optical field-free 3D orientation: fixing a molecule in space

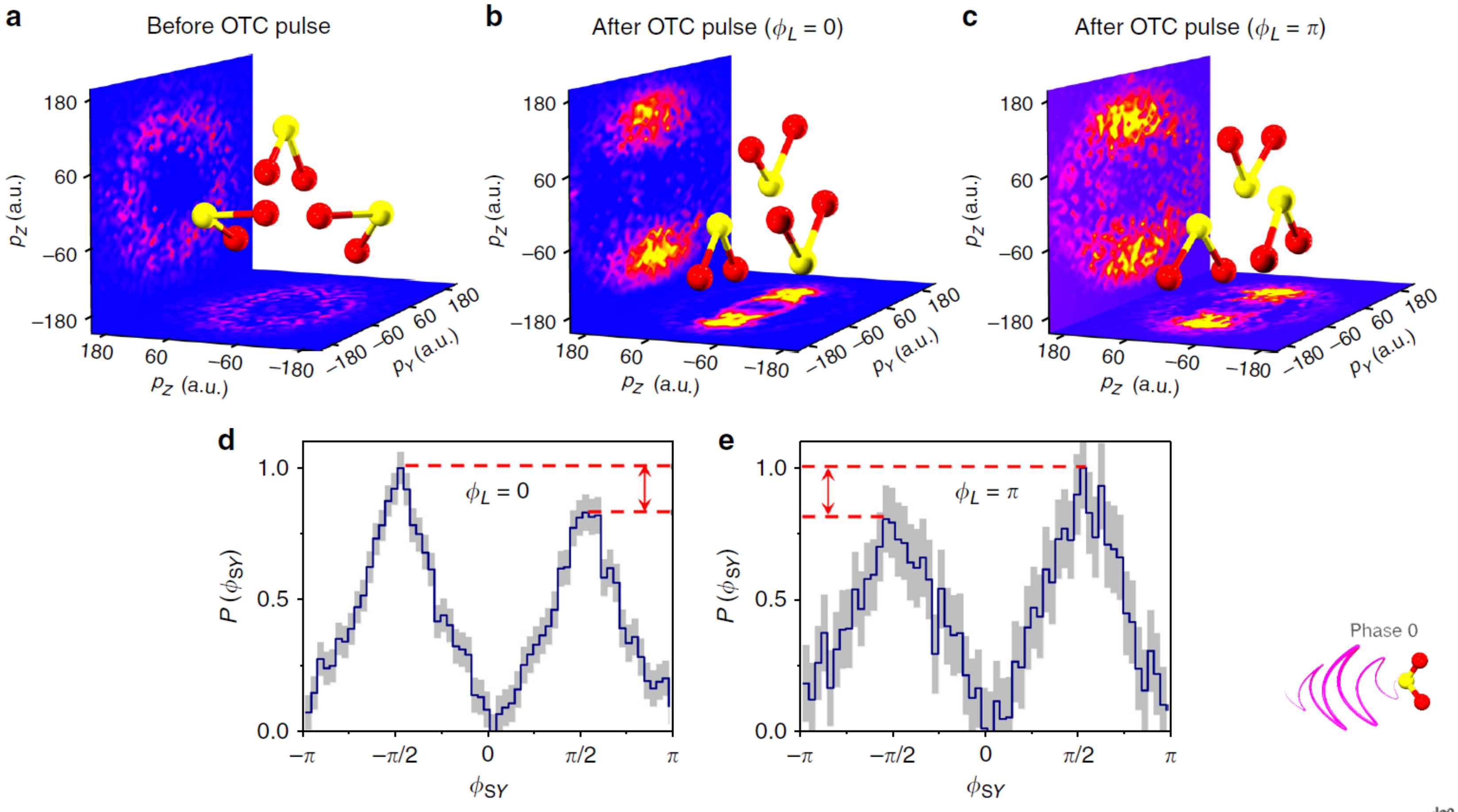


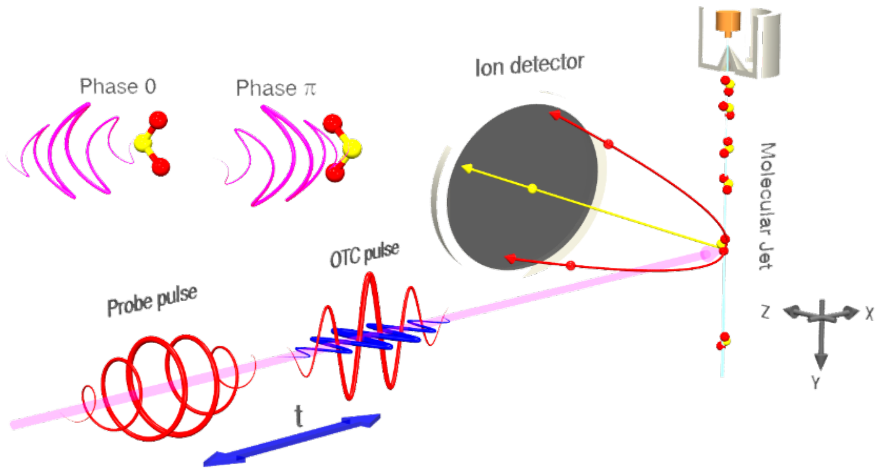
For SO_2, we can use the OTC fields to

- **Align the O-axis along FW**
- **Orientate the S-axis along SH**

K. Lin *et al.*, Nat. Commun. 9, 5134 (2018).

All optical field-free 3D orientation



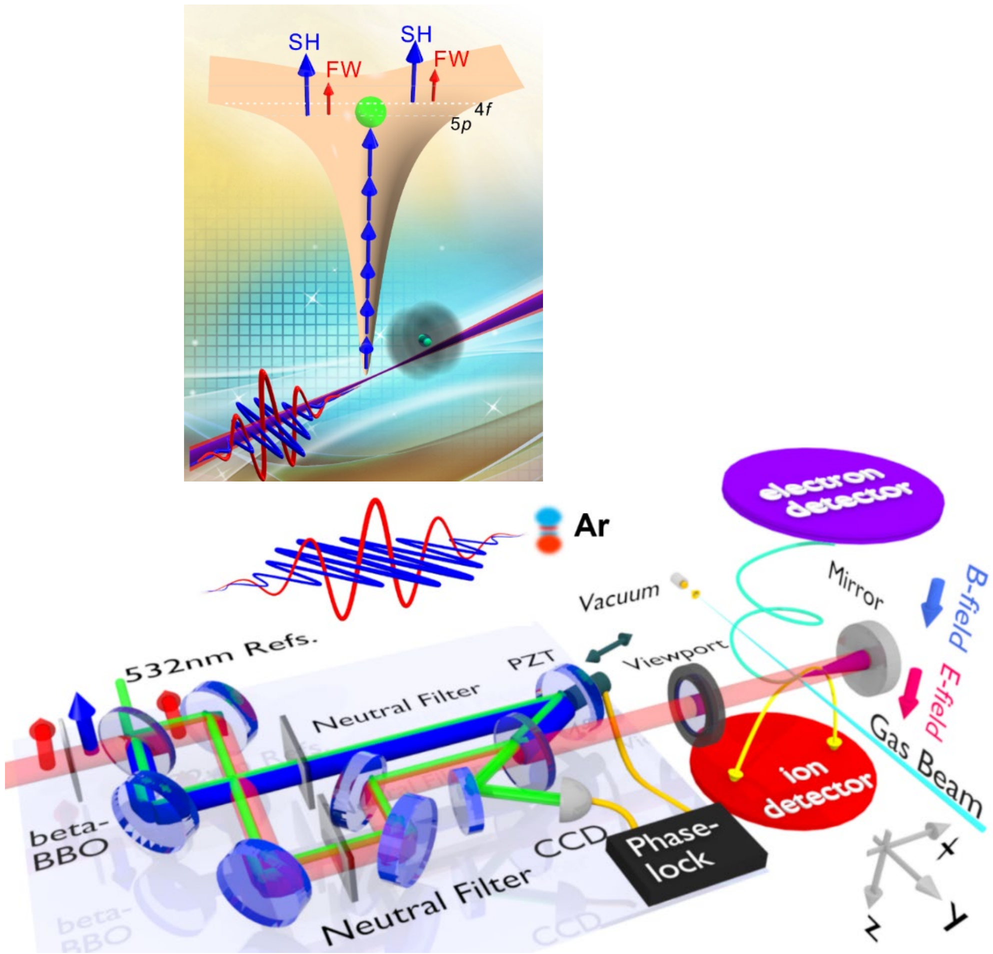


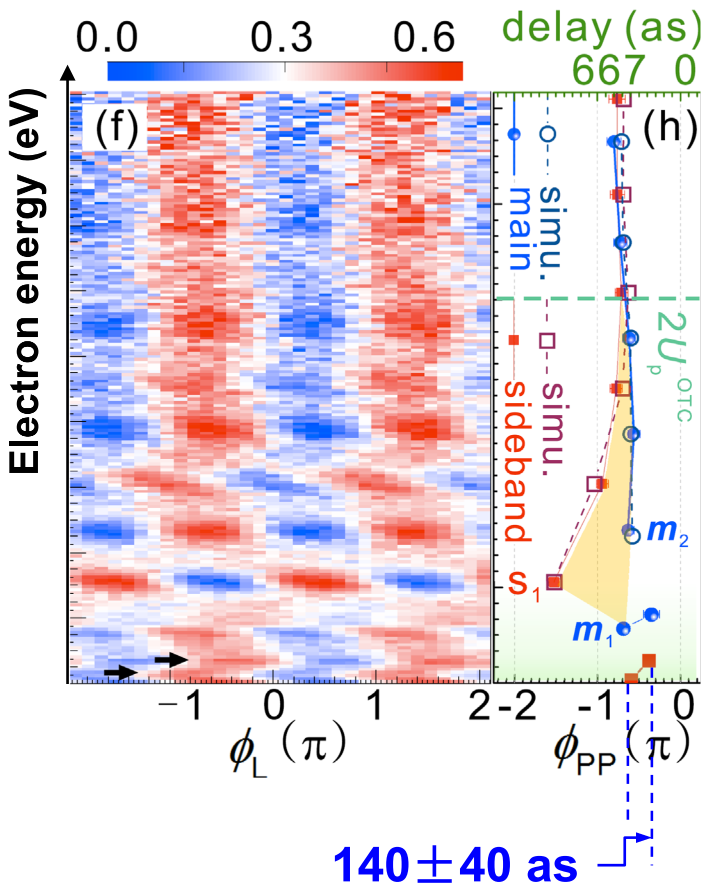
K. Lin *et al.*, Nat. Commun. 9, 5134 (2018).

Outline

- **Background Introduction**
- **Multiphoton energy absorption: electron-nuclear correlation**
  - ATI & ATD in dissociative ionization of molecules
  - Strong-field Rydberg excitation of molecules
  - An ultrafast stopwatch to clock molecular bond stretching
- **Ro-vibrational dynamics of the nuclear wave-packet**
  - Visualizing unidirectional molecular rotation
  - Echoes of molecules
  - All-optical 3D orientation of molecules
- **Attosecond dynamics of electrons: visualization and control**

Phase-of-phase (multiphoton) attoclock:
Freeman resonance time delay

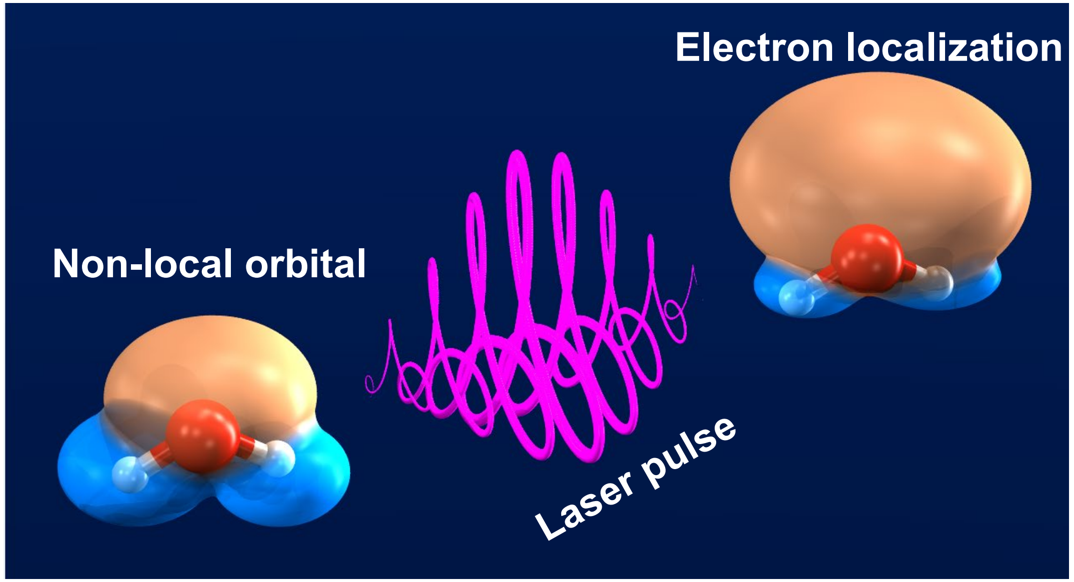


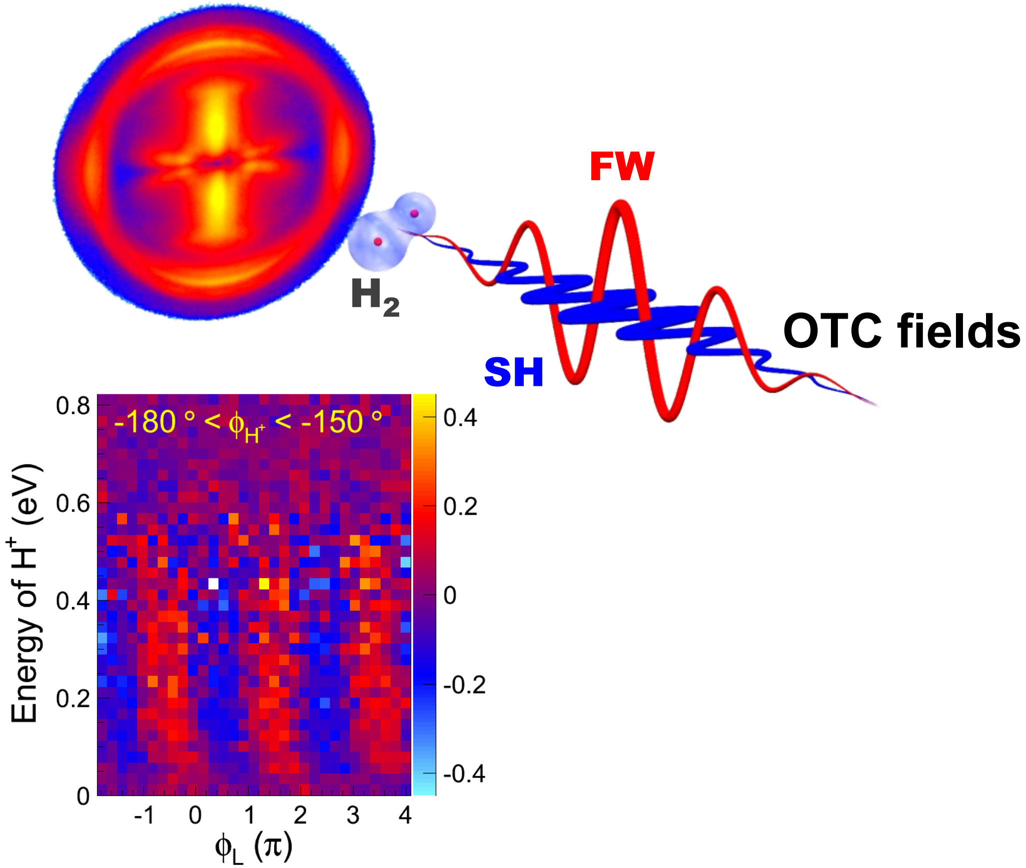


X. Gong *et al.*, Phys. Rev. Lett. 118, 143203 (2017).

Phase-controlled orthogonally polarized two-color (OTC) laser fields

2D control of the electron localization in molecules

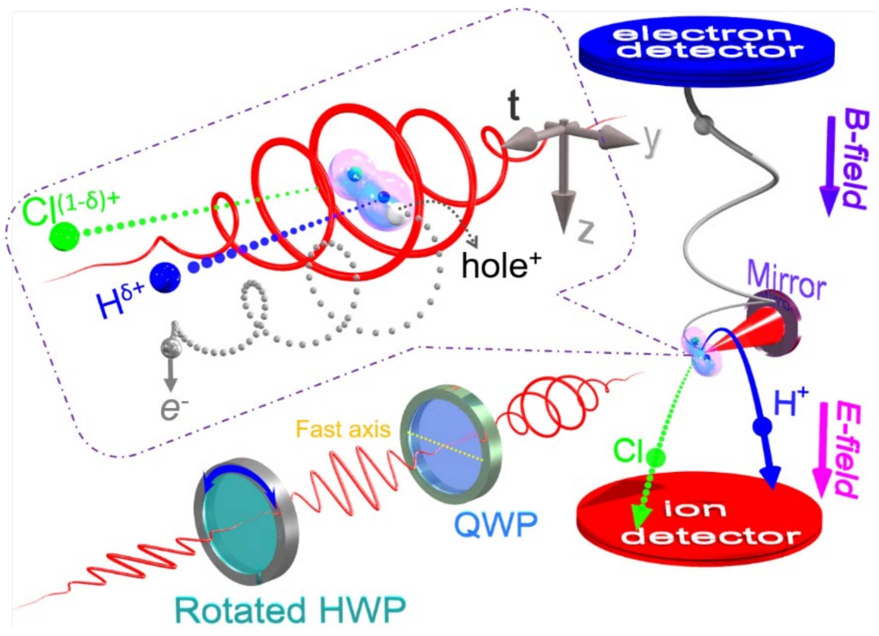


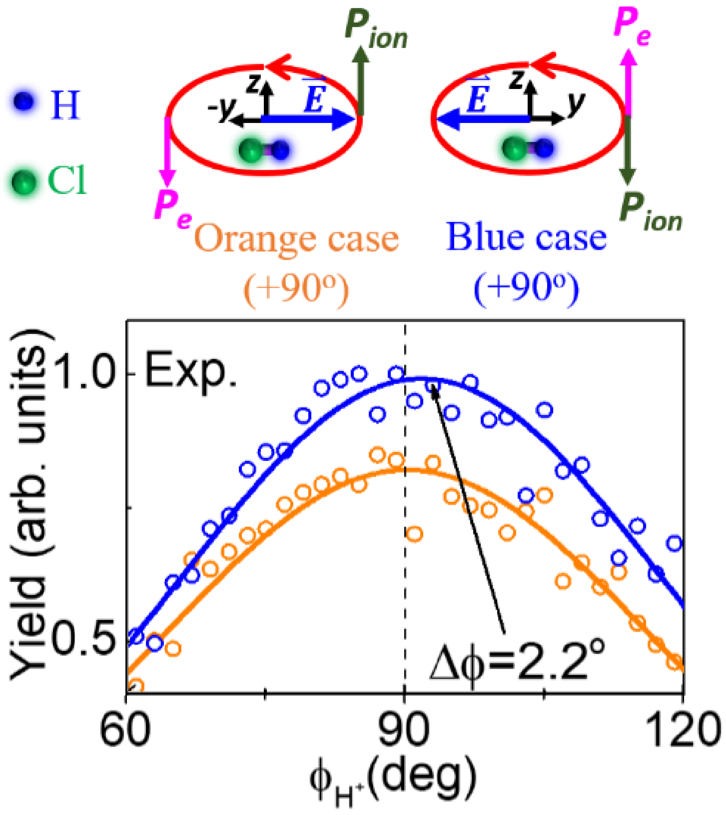


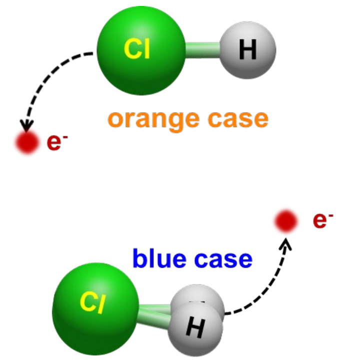
X. Gong *et al.*, Phys. Rev. Lett. 113, 203001 (2014).

Transient Valence Charge Localization in Strong-Field Dissociative Ionization of Molecules

Tunneling-site-sensitive ultrafast dynamics of molecules





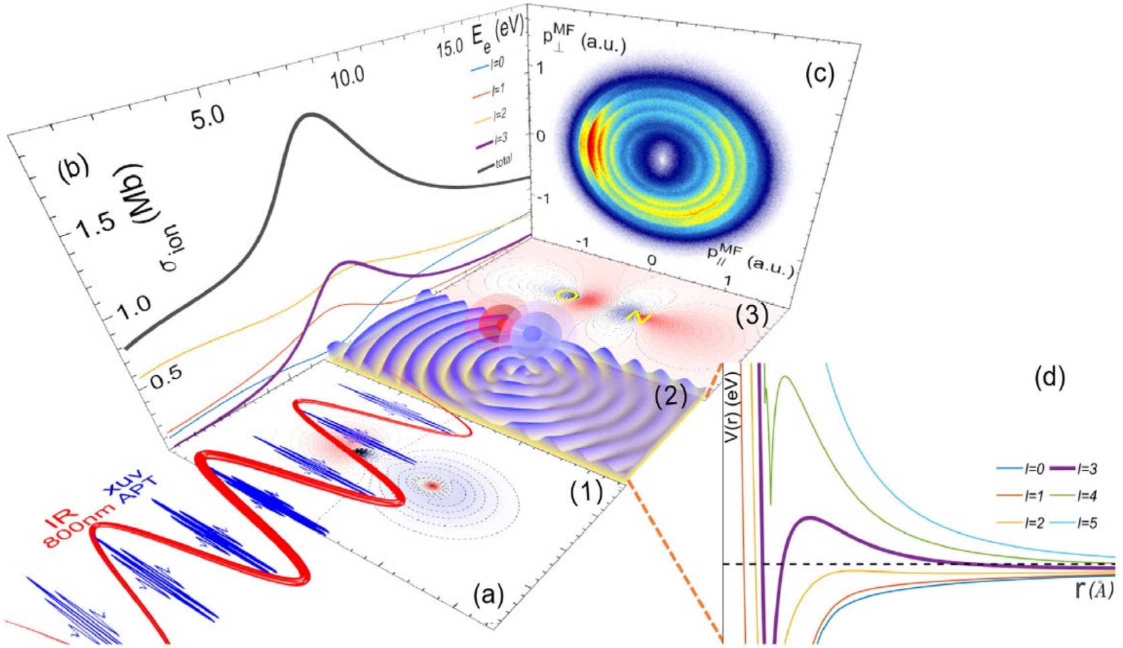


- **Where does the e⁻ recoil go?**
- **The center of mass of HCl^+? If so, H gets 1/36, and Cl gets 35/36.**

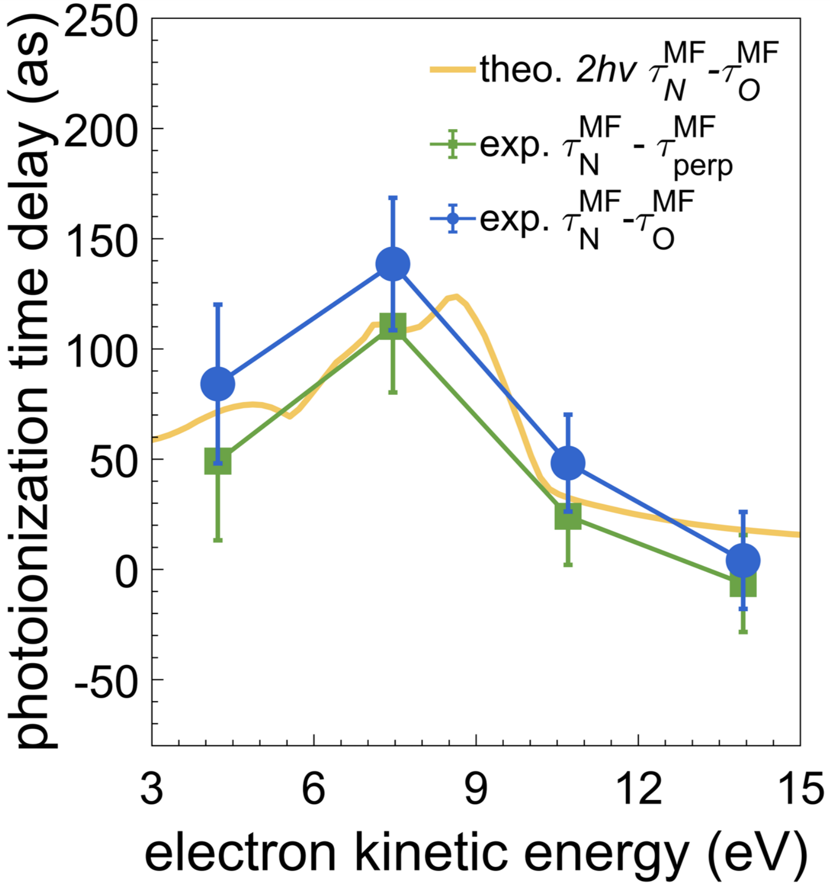
When the electron tunnels out with an exit near H which digs a transient electron hole on this site, a positive transient charge is localized on H, leading to a much larger laser impulse to the H^+ fragments as compared to the mass-dominated scenario.

J. Ma *et al*, Phys. Rev. Lett. 127, 183201 (2021).

Asymmetric Attosecond Photoionization in Molecular Shape Resonance



Emission site-resolved photoemission



The asymmetric photoemission time delay between the N end & O end: ~150 as

X. Gong *et al*, Phys. Rev. X 12, 011002 (2022).

Summary

Ultrafast Dynamics of Molecules in Strong Laser Fields

measurement → physical mechanism → control

- **Multiphoton energy absorption: electron-nuclear correlation**
  - ATI & ATD in dissociative ionization of molecules
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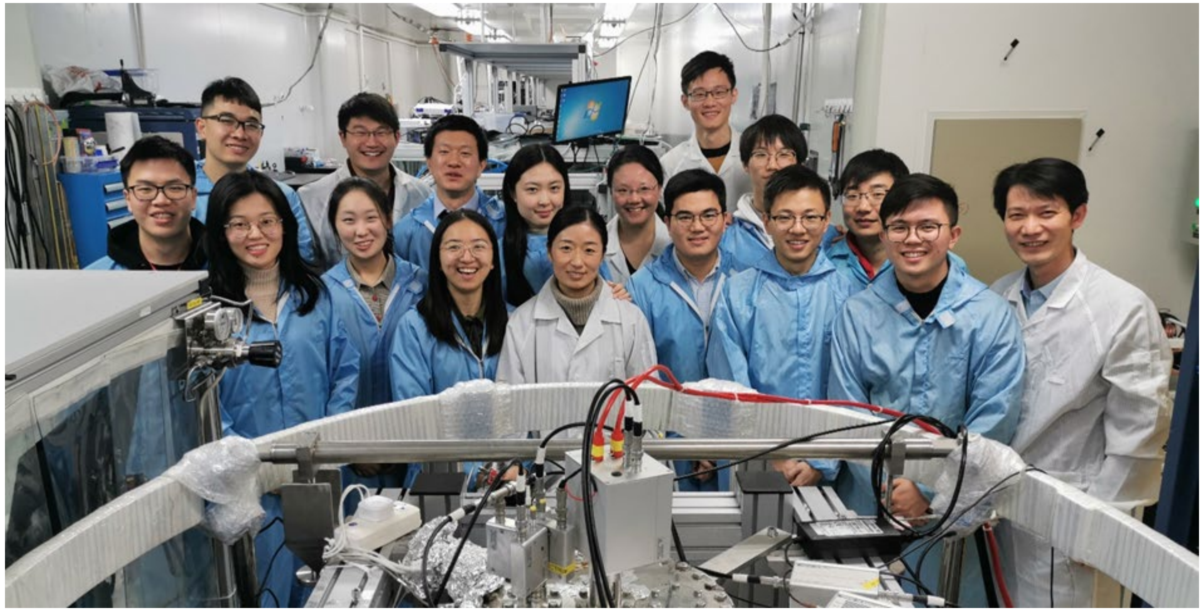

Thank you for your attention!

For more details:
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