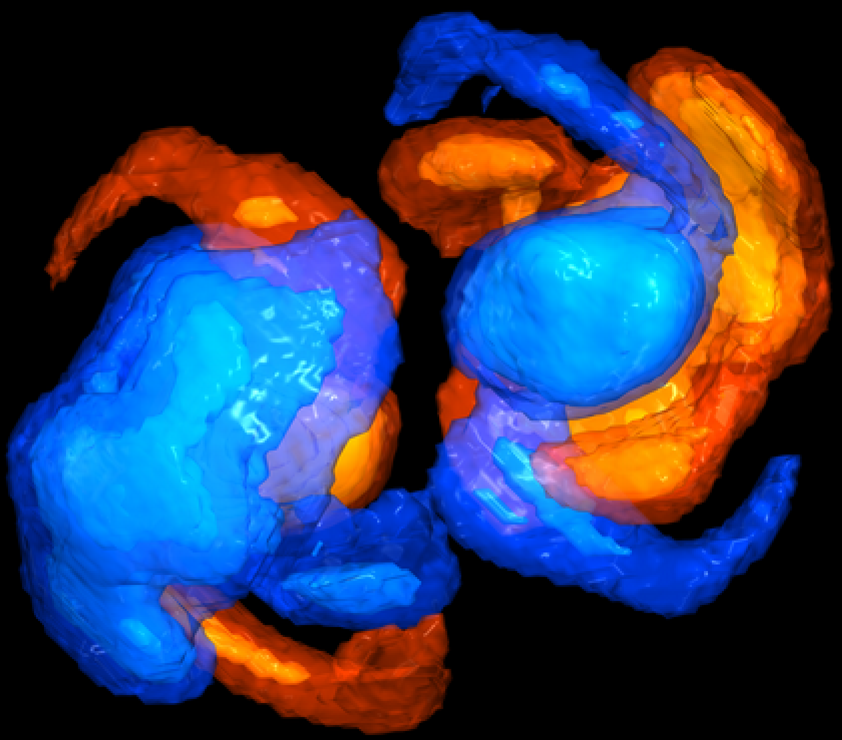
Chirality at CELIA
and Chirality in strong laser fields

Yann Mairesse,
Centre Lasers Intenses et Applications
Université de Bordeaux – CEA – CNRS
http://harmodyn.celia.u-bordeaux.fr



Chirality at CELIA

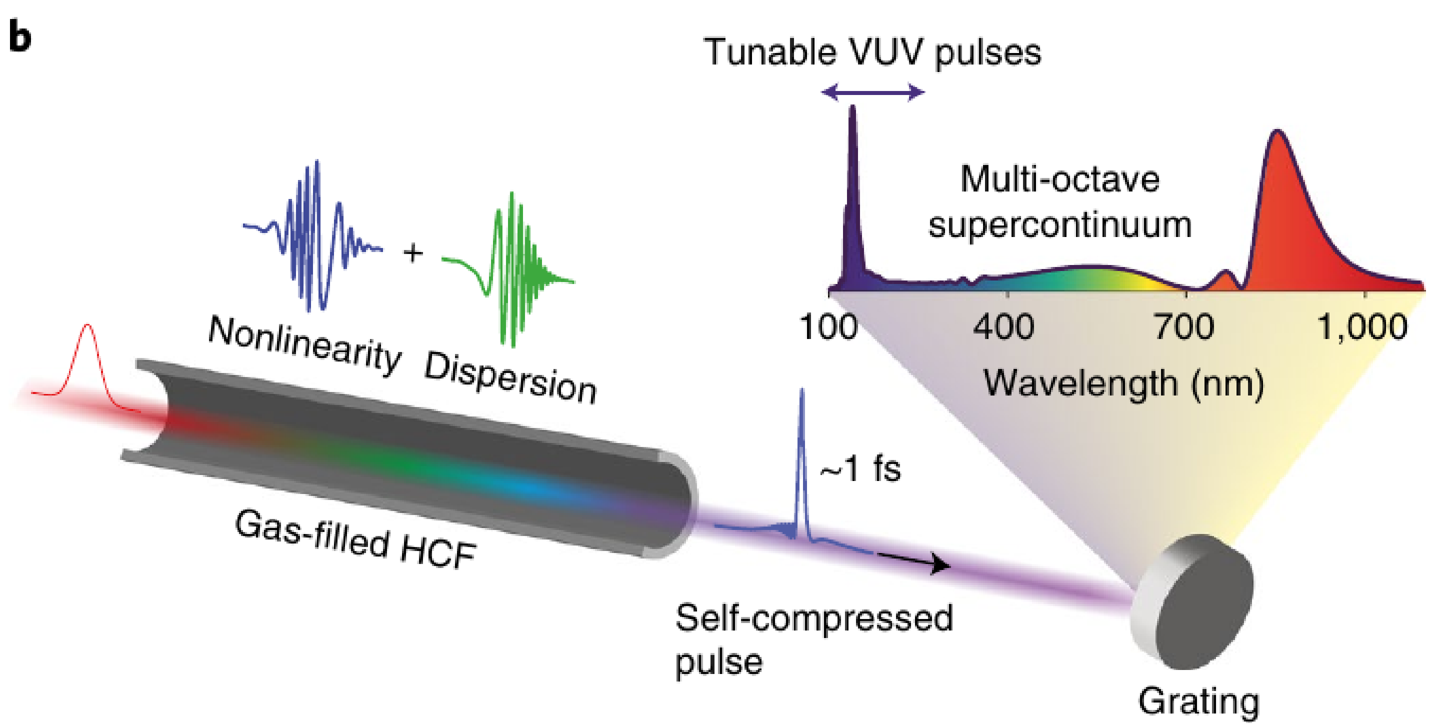
Groupe Optique Lasers Femtosecondes – GOLF

Mid infrared sources for chiral strong field physics

Jean-Christophe Delagnes, Stéphane Petit

Generation of ultrabroadband radiation (from VUV to visible) from soliton dynamics with tunable polarization state and Orbital Angular Momentum

Dominique Descamps, Stéphane Petit



nature photonics
ARTICLES
https://doi.org/10.1038/s41566-019-0416-4

High-energy pulse self-compression and ultraviolet generation through soliton dynamics in hollow capillary fibres

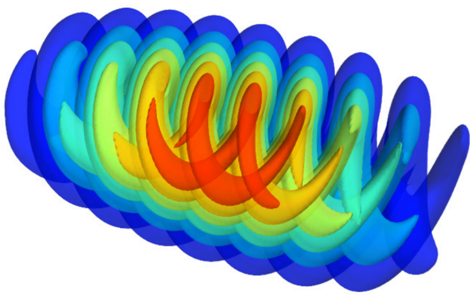
John C. Travers*, Teodora F. Grigorova, Christian Brahms and Federico Belli

Chirality at CELIA

Groupe Interaction, Fusion par Confinement Inertiel, Astrophysique – IFCIA

Irradiation of dielectric materials by intense femtosecond OAM laser beams

Benoit Chimier, Rachel Nuter, collaboration with Etienne Brasselet



Electric field in an OAM laser pulse

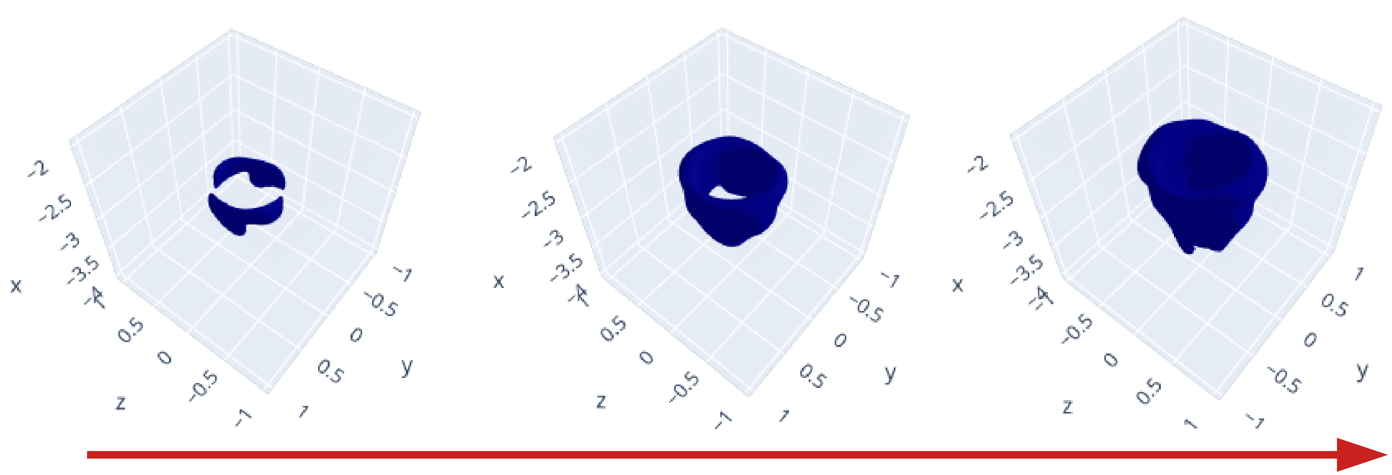
OAM laser beam are characterized by a helical wave front with an optical vortex in center

Intensity ~ 10^{13}-10^{15} W/cm²

Modeled by numerical algorithm able to inject any kind of exotic laser beams in Maxwell solver (JPC 321, 1110 (2016))

Simulations of the interaction between dielectric material and OAM laser beam performed by using Maxwell-Fluid code ARCTIC : Maxwell solver coupled to electron dynamics (APA 127, 334 (2021))

Energy deposition in bulk of fused silica (laser pulse comes from the top)



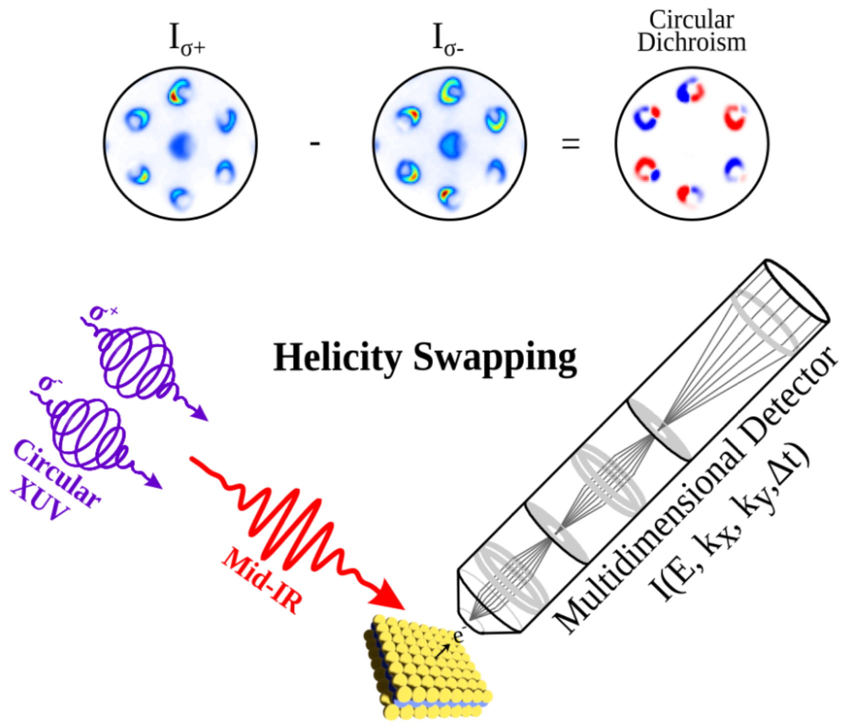
Chirality at CELIA

Groupe Harmoniques XUV et Applications – HXUV

Time-resolved XUV circular dichroism in angle-resolved photoemission spectroscopy (ARPES) to probe ultrafast dynamics in quantum materials

Samuel Beaulieu, Jérôme Gaudin

Dichroism originating from Berry phase



Photoelectron circular dichroism and novel probes of ultrafast chirality

Chiral femtochemistry – See Valérie Blanchet's poster

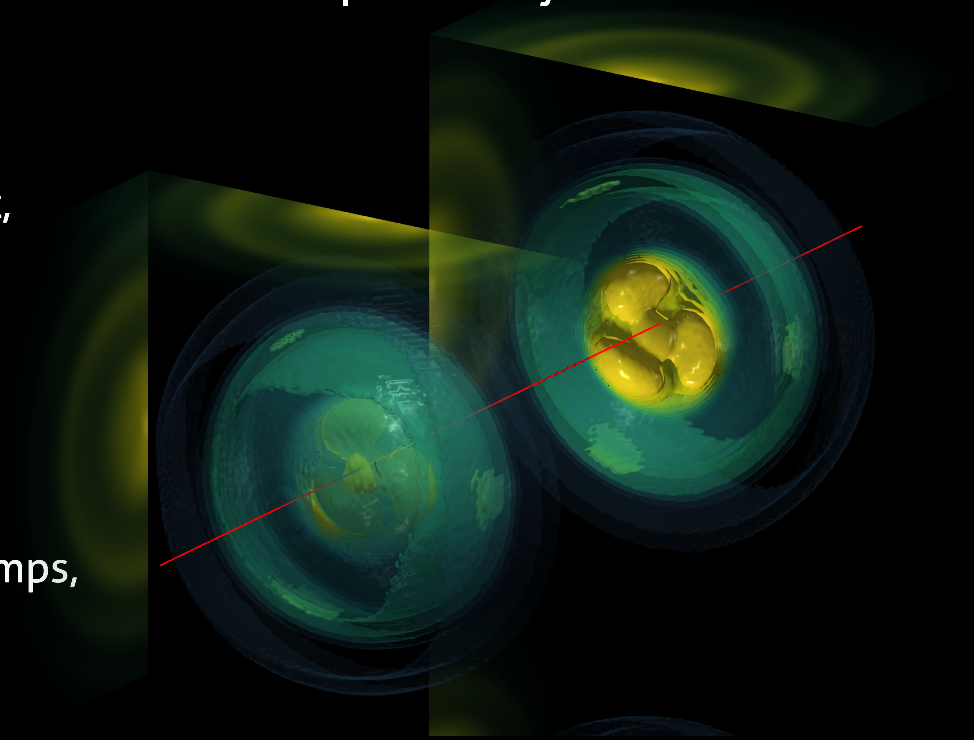
Fast and accurate measurement of enantiomeric excess – See Antoine Comby's poster

Strong fields and attoseconds – See Debobrata Rajak's poster and this talk

Probing chiral nanomaterials – CHIMERA project with CBMN, ISM, LCP-A2MC Metz

Chirality in strong laser fields

Yann Mairesse,
CELIA, Université de Bordeaux – CEA – CNRS
http://harmodyn.celia.u-bordeaux.fr



CELIA Bordeaux
Experiments :
Samuel Beaulieu,
Sandra Beauvarlet,
Etienne Bloch,
Antoine Comby,
Debobrata Rajak,
Valérie Blanchet,
Yann Mairesse
Laser Team :
Dominique Descamps,
Stéphane Petit

Theory :
Alex Clergerie,
Sylvain Larroque,
Baptiste Fabre,
Bernard Pons

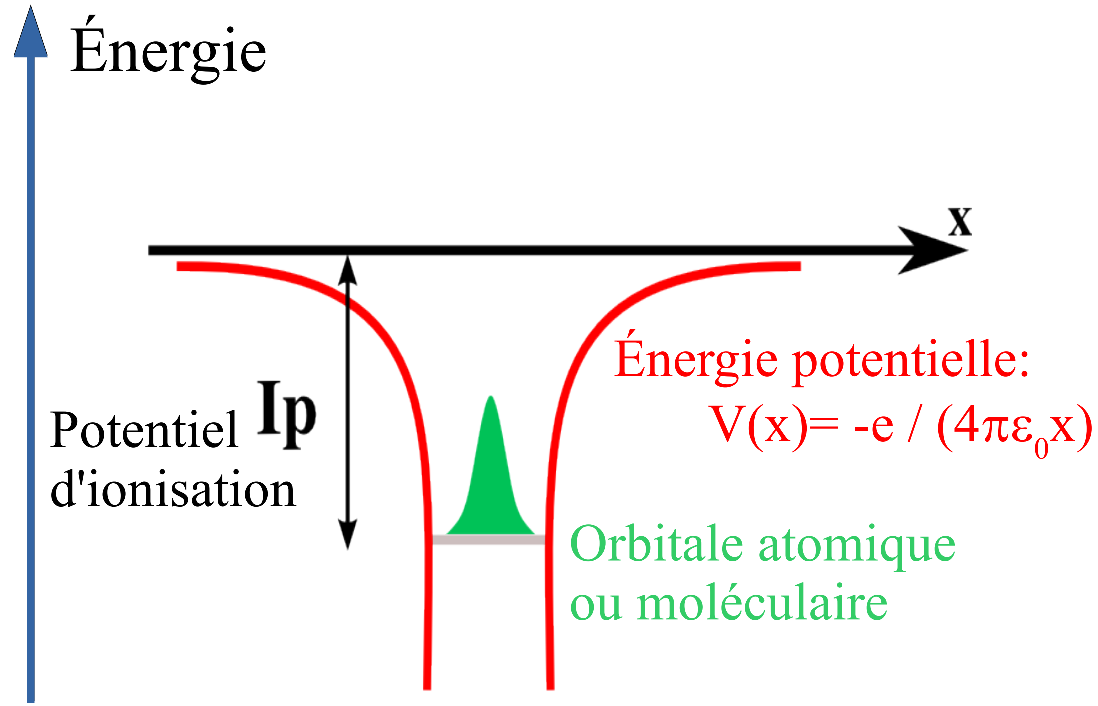
Weizmann Rehovot
Shaked Rozen
Ayelet Julie Uzan
Nirit Dudovich

LCPMR Paris
Richard Taïeb

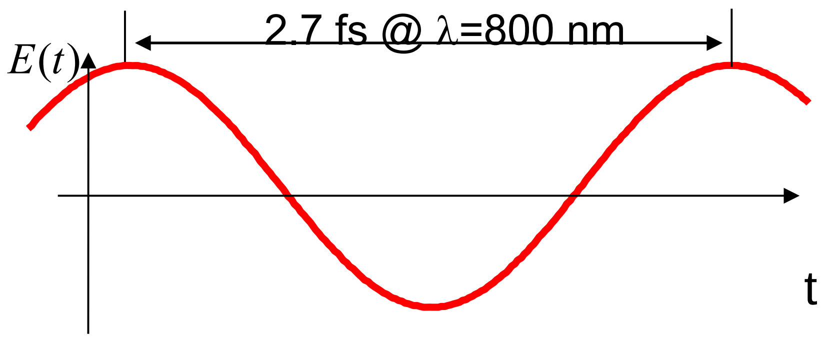
cnrs dépasser les frontières
université de BORDEAUX
cea
RÉGION Nouvelle-Aquitaine
erc European Research Council
Established by the European Commission

This project has received funding from the European Research Council (ERC) under the European Union's Horizon 2020 research and innovation programme (grant agreement No 682978)

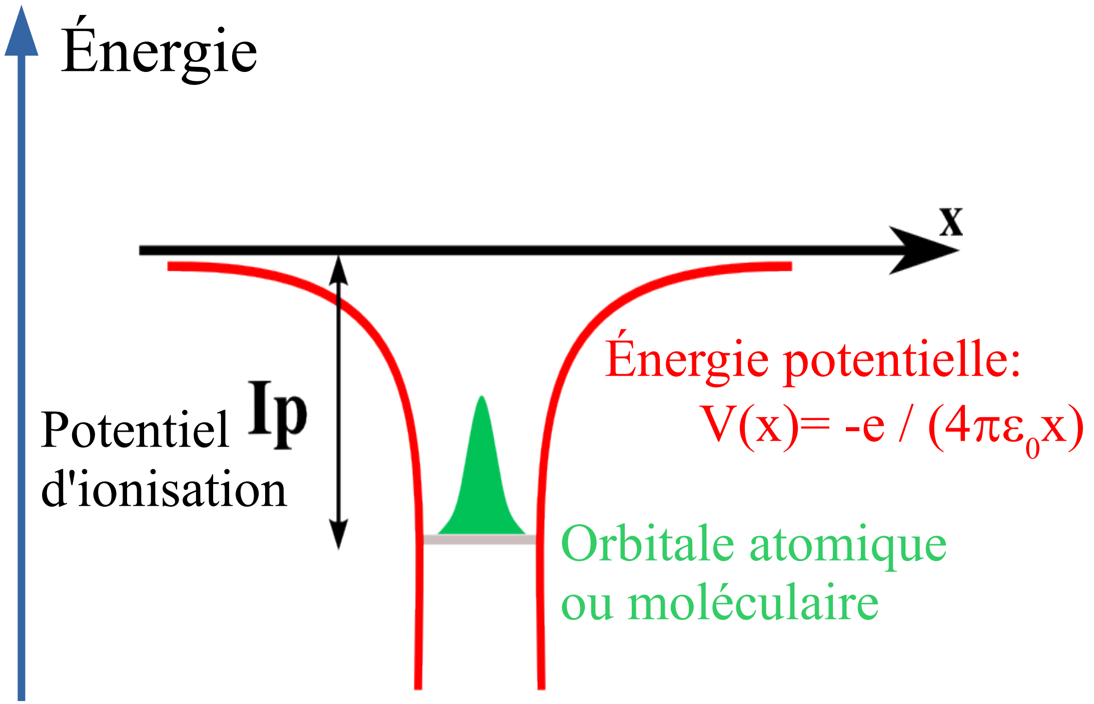
Strong field processes



Strong field processes



Infrared laser field



Value of laser electric field ?

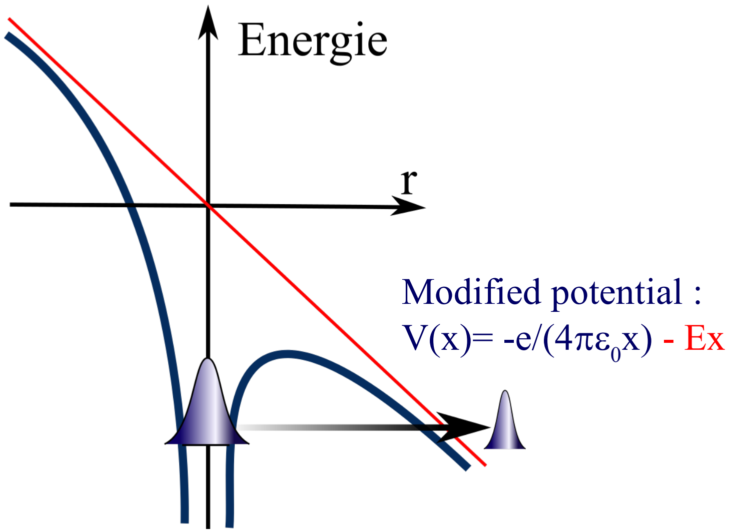
$I = 10^{14}$ W/cm² → $E = 2.8\times10^{10}$ V/m

Coulomb electric field ?

First Bohr orbit of the hydrogen atom → $E = 5.1\times10^{11}$ V/m

→ It is possible to reach electric fields values similar to the atomic binding potential with fs laser pulses

Strong field ionization

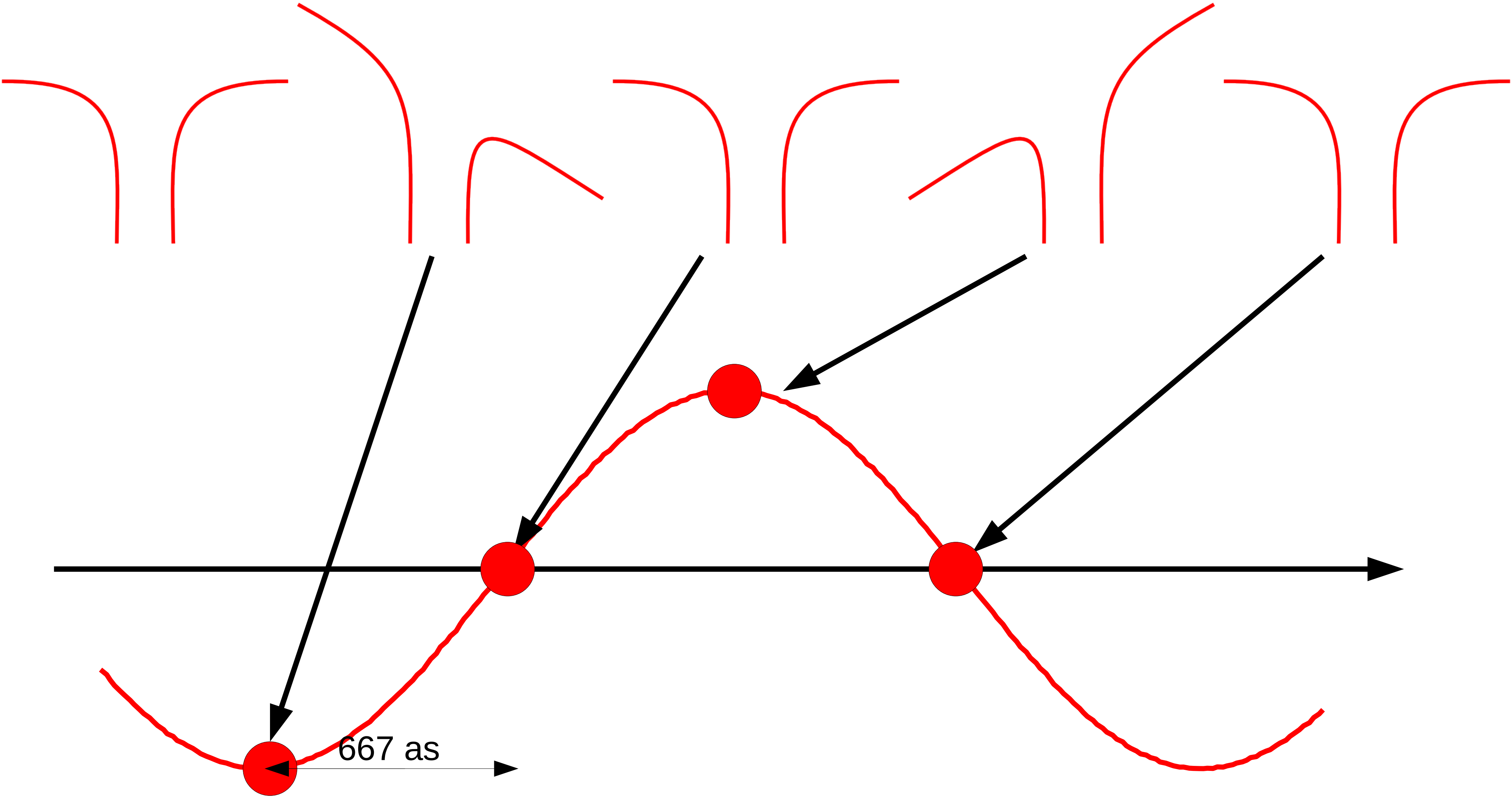


The strong laser field lowers the barrier

The electron can tunnel out
→ Tunnel ionization

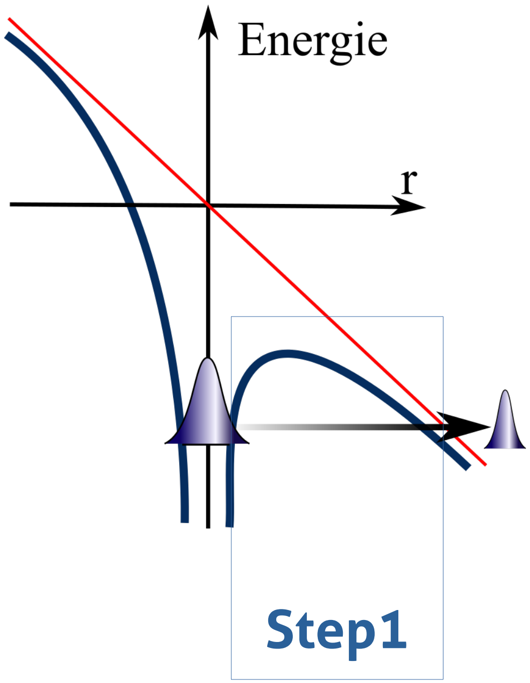
What happens next ?

Coulomb potential in a strong laser field



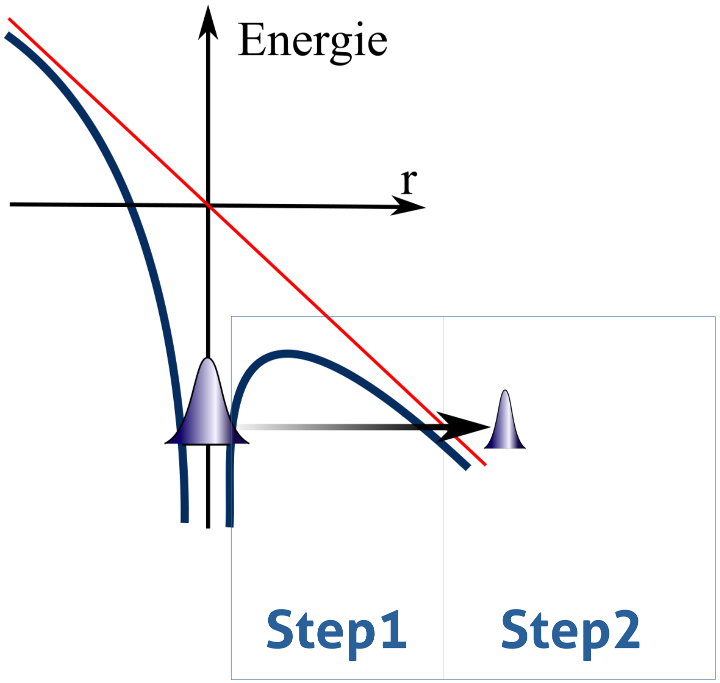
Tunnel ionization restricted to a few 100 as
Followed by acceleration by the laser field

Coulomb potential in a strong laser field



Step1 : classically forbidden region – sub-barrier tunneling

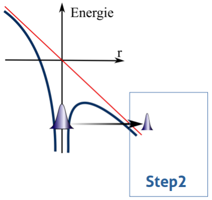
Coulomb potential in a strong laser field

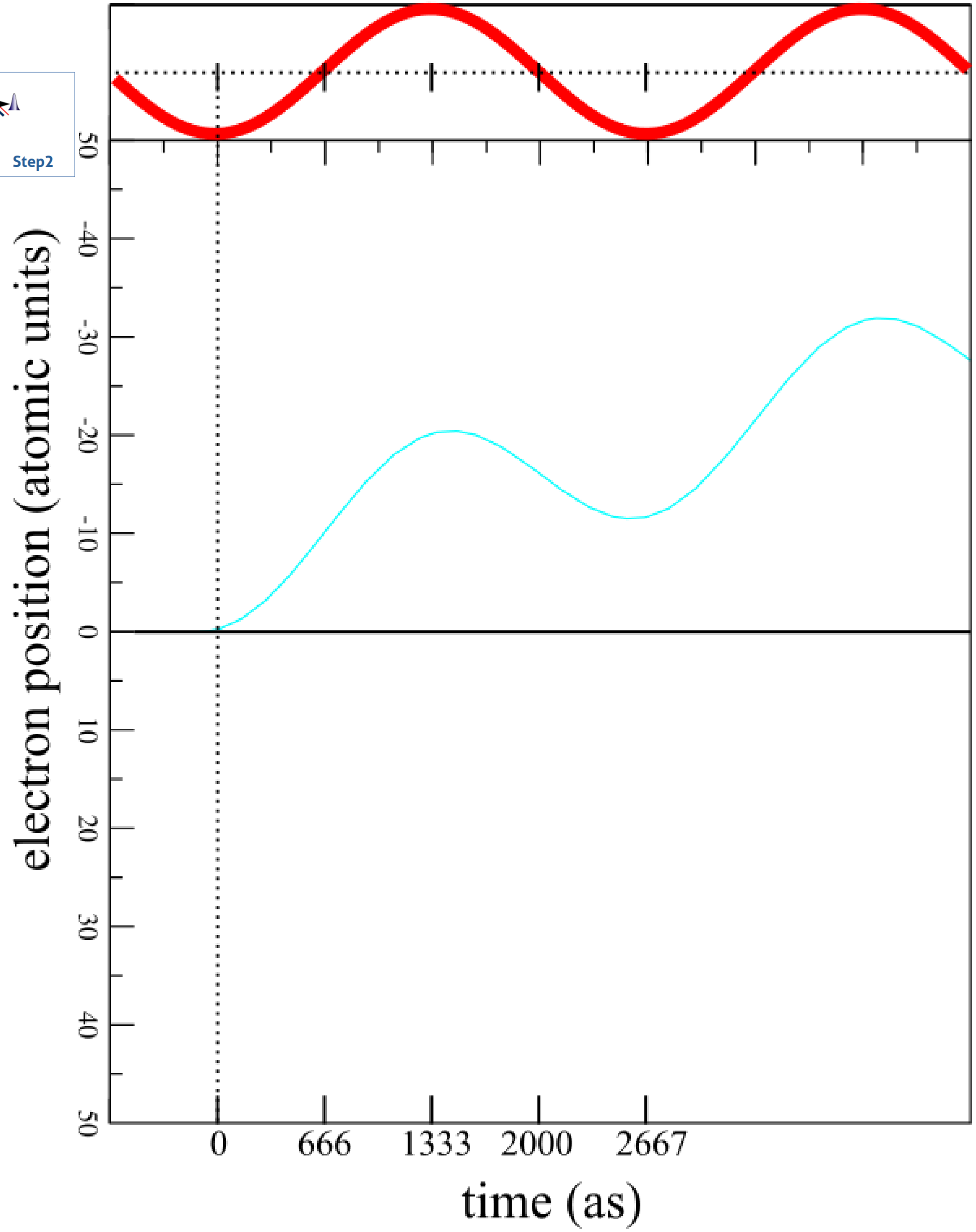


Step1 : classically forbidden region – sub-barrier
tunneling

Step 2 : post-tunneling dynamics – continuum
acceleration by the laser field

Electron trajectories in a strong laser field





Laser electric field

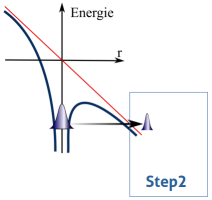
Classical electron trajectory
Strong field approximation :
neglect the influence of ionic potential

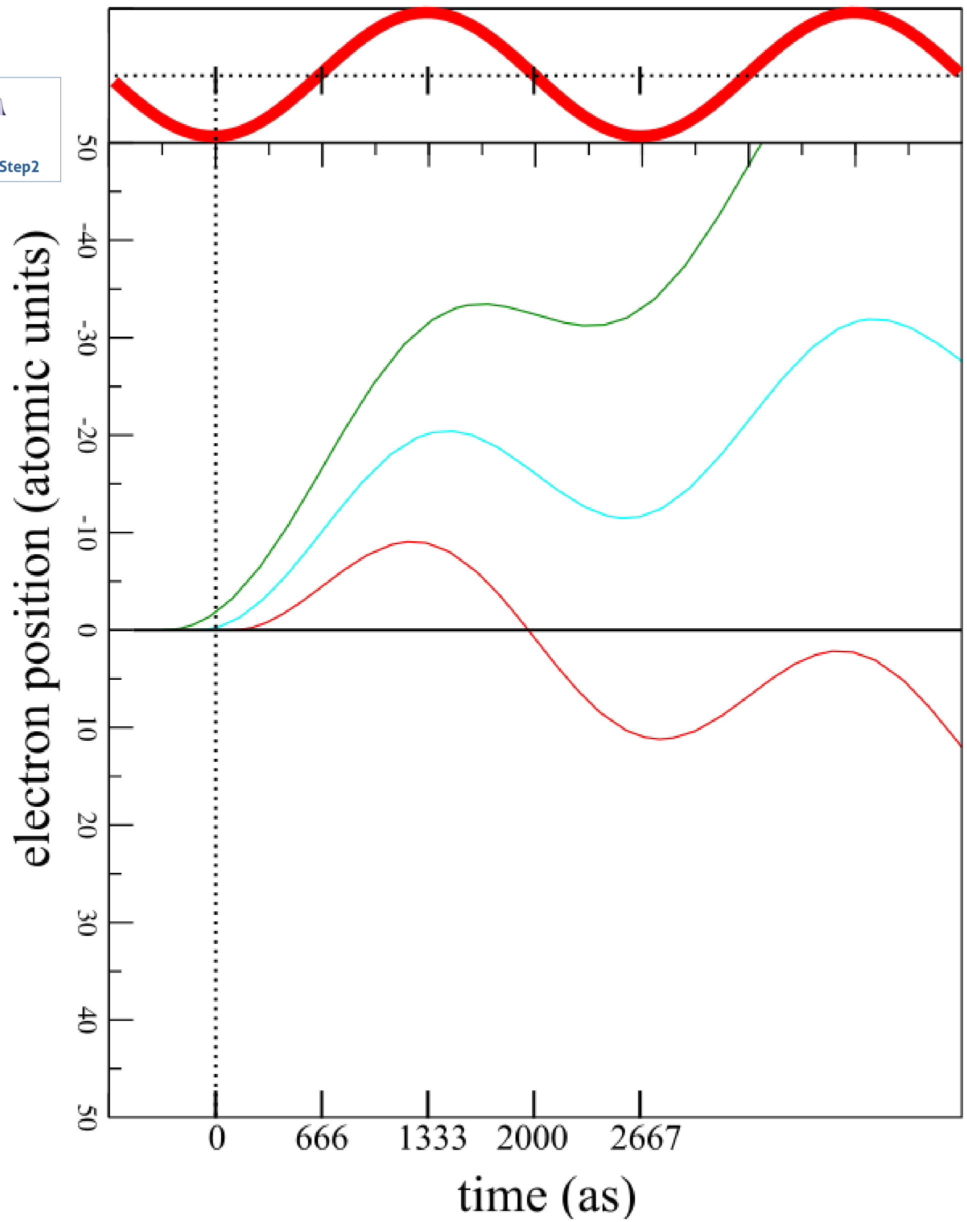
PHYSICAL REVIEW A **80**, 023402 (2009)

One-electron atom in a strong and short laser pulse: Comparison of classical and quantum descriptions

P. Botheron and B. Pons
CELIA, Université de Bordeaux-I–CNRS–CEA, 351 Cours de la Libération, F-33405 Talence, France
(Received 13 May 2009; published 3 August 2009)

Electron trajectories in a strong laser field





Laser electric field

Classical electron trajectory
Strong field approximation :
neglect the influence of ionic potential

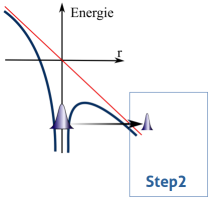
Motion depends on initial conditions
ie ionization time of the electron

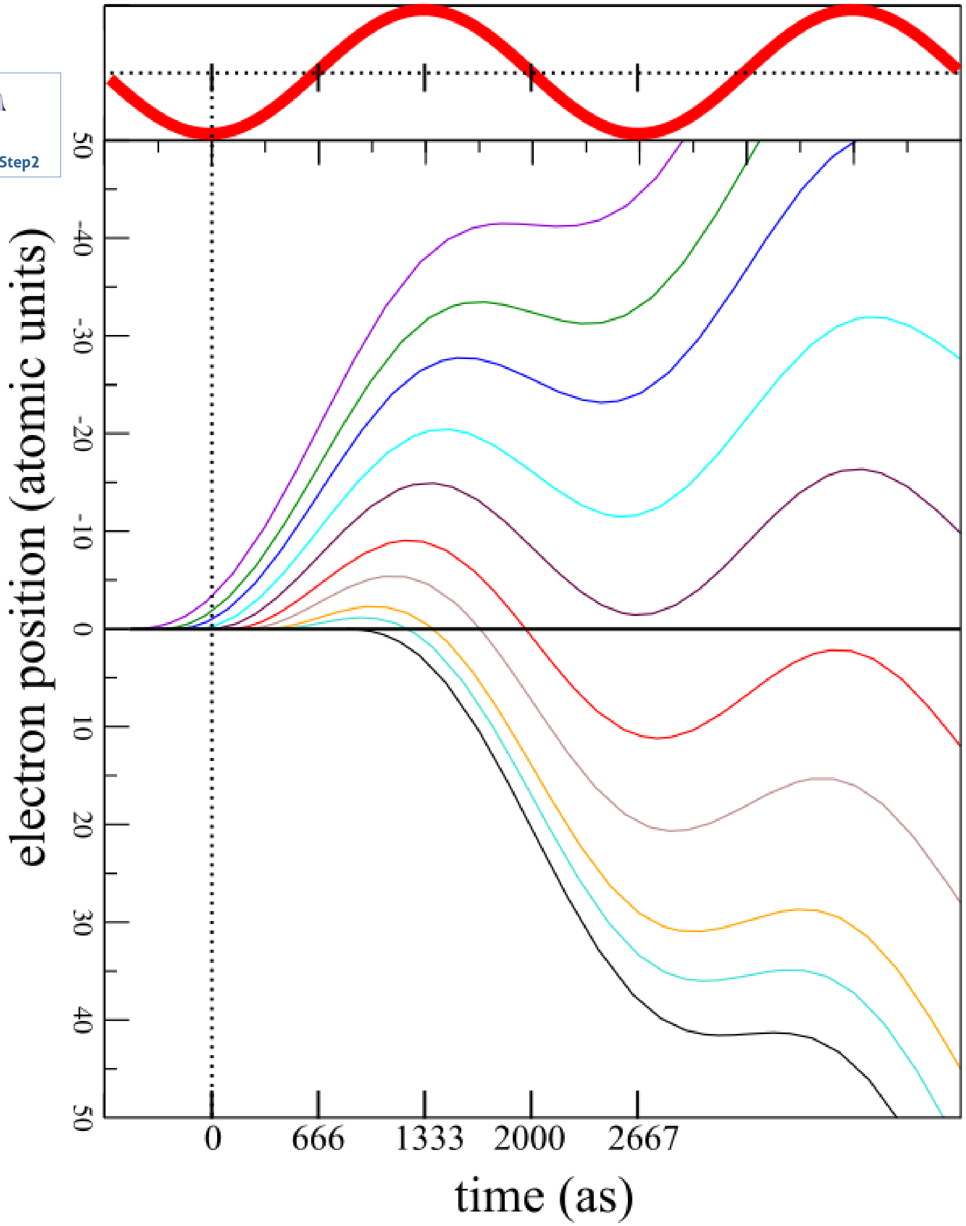
PHYSICAL REVIEW A **80**, 023402 (2009)

One-electron atom in a strong and short laser pulse: Comparison of classical and quantum descriptions

P. Botheron and B. Pons
CELIA, Université de Bordeaux-I–CNRS–CEA, 351 Cours de la Libération, F-33405 Talence, France
(Received 13 May 2009; published 3 August 2009)

Electron trajectories in a strong laser field





Laser electric field

Classical electron trajectory
Strong field approximation :
neglect the influence of ionic potential

Motion depends on initial conditions
ie ionization time of the electron

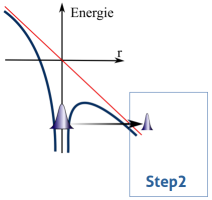
Different trajectories :
- ‘Direct trajectories’
  → direct escape
- ’Indirect trajectories’
  → come back to ion before escaping

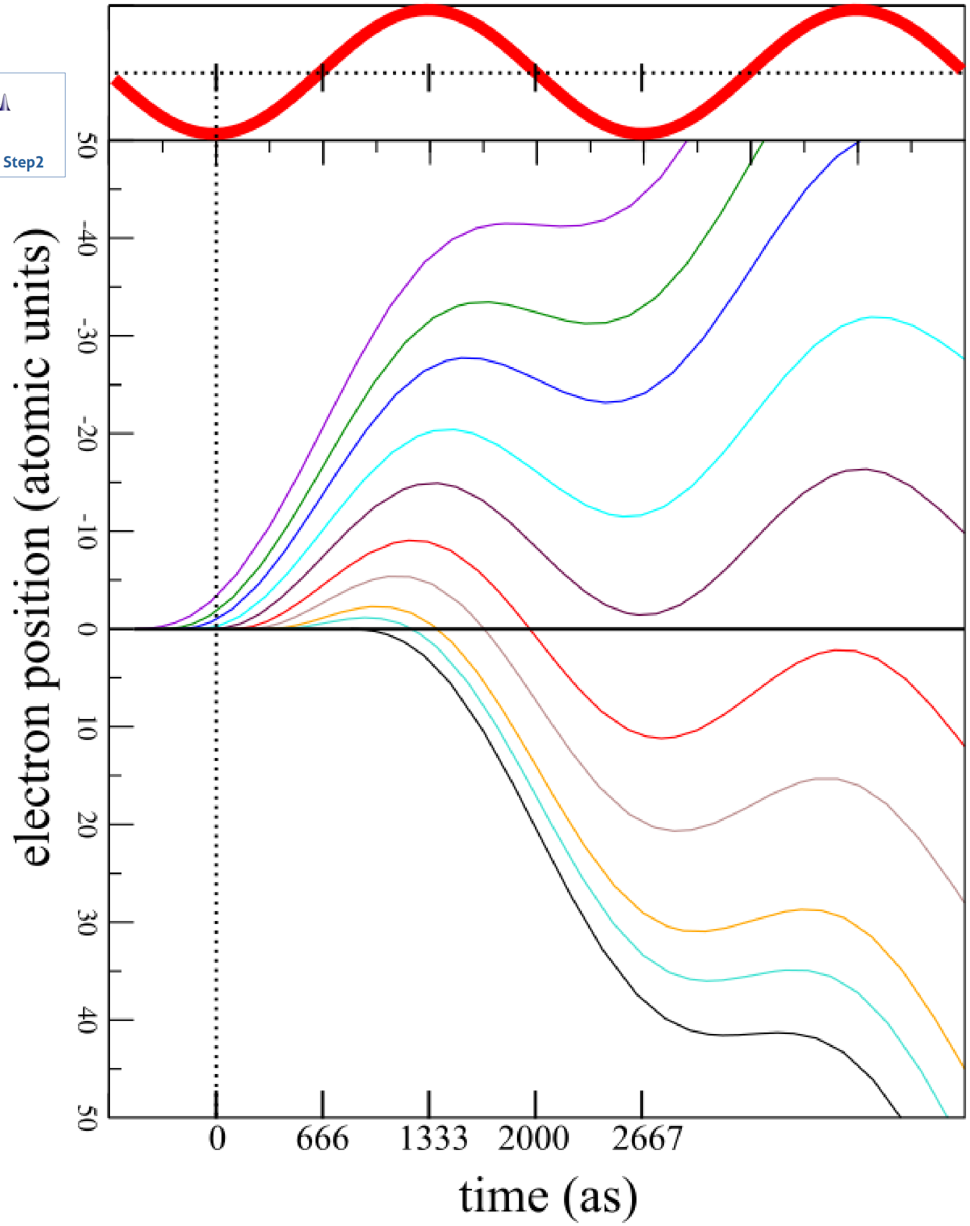
PHYSICAL REVIEW A **80**, 023402 (2009)

One-electron atom in a strong and short laser pulse: Comparison of classical and quantum descriptions

P. Botheron and B. Pons
CELIA, Université de Bordeaux-I–CNRS–CEA, 351 Cours de la Libération, F-33405 Talence, France
(Received 13 May 2009; published 3 August 2009)

Electron trajectories in a strong laser field





Laser electric field

Classical electron trajectory
Strong field approximation :
neglect the influence of ionic potential

Motion depends on initial conditions
ie ionization time of the electron

Different trajectories :
'Direct trajectories'
→ direct escape
'Indirect trajectories'
→ come back to ion before escaping

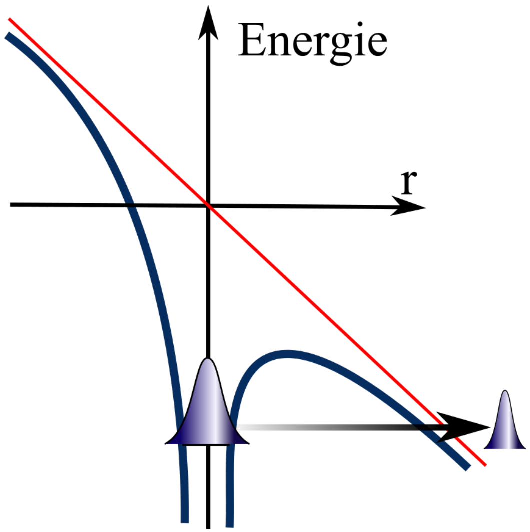
Validated by quantum calculations
Bohmian trajectories

PHYSICAL REVIEW A **80**, 023402 (2009)

One-electron atom in a strong and short laser pulse: Comparison of classical and quantum descriptions

P. Botheron and B. Pons
CELIA, Université de Bordeaux-I–CNRS–CEA, 351 Cours de la Libération, F-33405 Talence, France
(Received 13 May 2009; published 3 August 2009)

Electron dynamics in a strong laser field

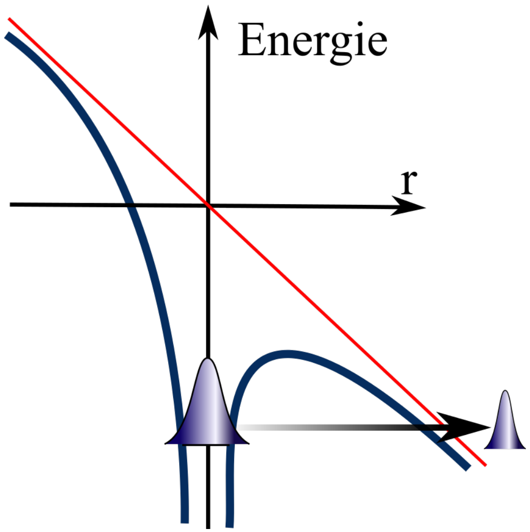


Acceleration of the electron by the laser field

→ kinetic energy gain (typ. several 10 eV, λ~ few Angstroms)

Above-threshold ionization (ATI)

Electron dynamics in a strong laser field

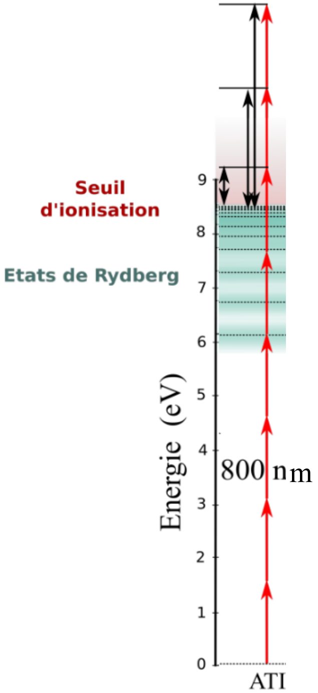


Acceleration of the electron by the laser field

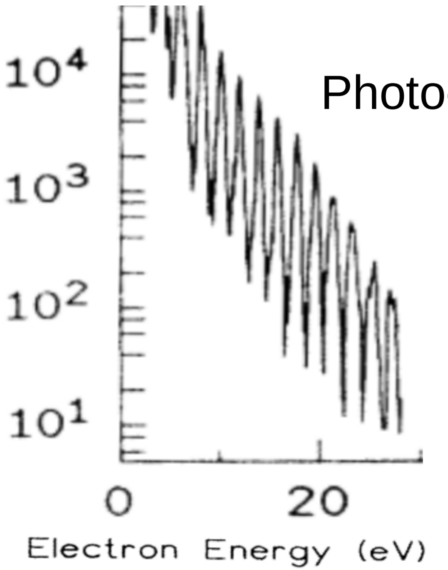
→ kinetic energy gain (typ. several 10 eV, λ~ few Angstroms)

Above-threshold ionization (ATI)

Frequency-domain picture of Above-threshold ionization :

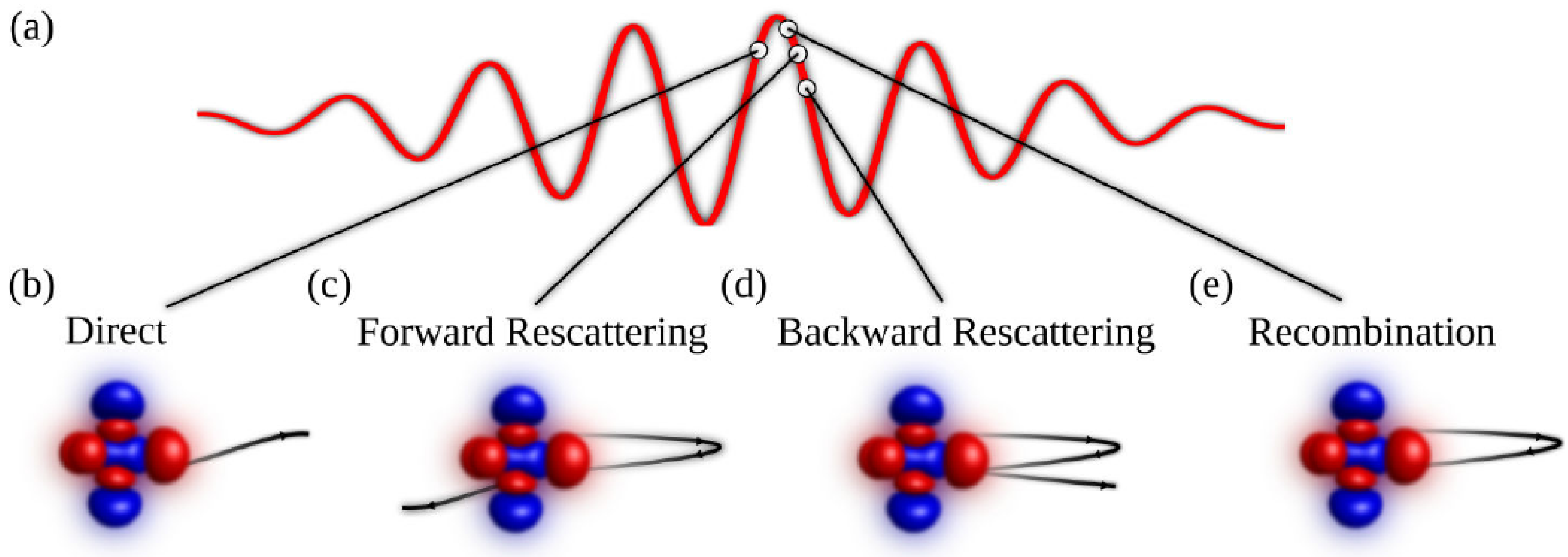


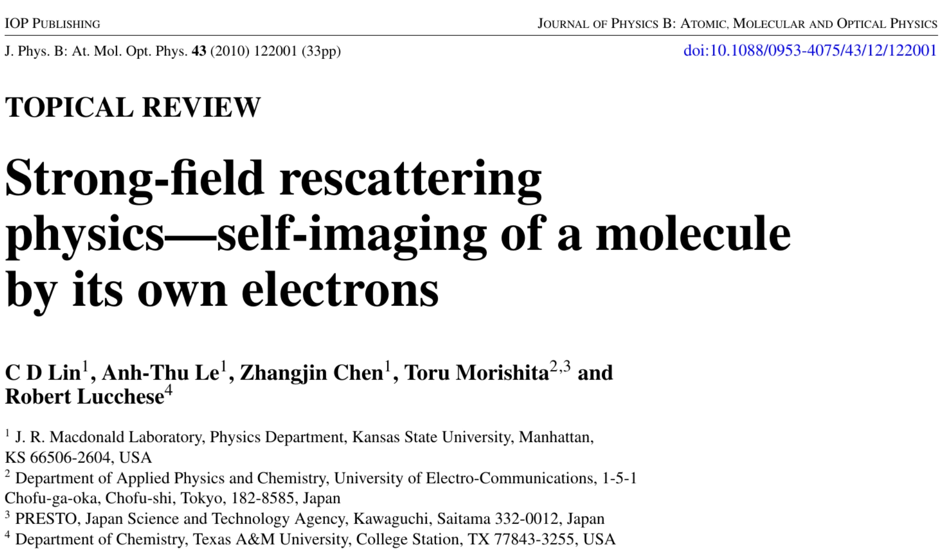
Spectrum = series of peaks, separated by one laser photon energy (the process repeats every laser period)



Strong field processes

Used for many applications in molecular imaging




IOP PUBLISHING JOURNAL OF PHYSICS B: ATOMIC, MOLECULAR AND OPTICAL PHYSICS

J. Phys. B: At. Mol. Opt. Phys. **43** (2010) 122001 (33pp) doi:10.1088/0953-4075/43/12/122001

TOPICAL REVIEW

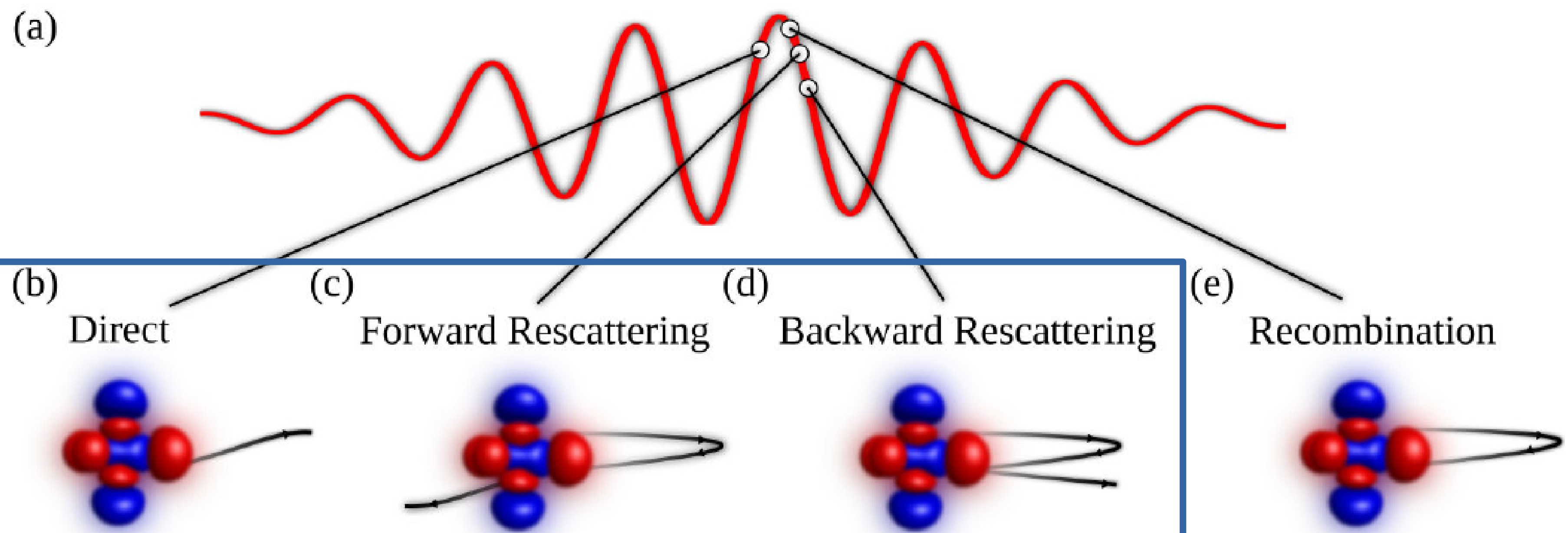
Strong-field rescattering physics—self-imaging of a molecule by its own electrons

C D Lin[1], Anh-Thu Le[1], Zhangjin Chen[1], Toru Morishita[2,3] and Robert Lucchese[4]

[1] J. R. Macdonald Laboratory, Physics Department, Kansas State University, Manhattan, KS 66506-2604, USA
[2] Department of Applied Physics and Chemistry, University of Electro-Communications, 1-5-1 Chofu-ga-oka, Chofu-shi, Tokyo, 182-8585, Japan
[3] PRESTO, Japan Science and Technology Agency, Kawaguchi, Saitama 332-0012, Japan
[4] Department of Chemistry, Texas A&M University, College Station, TX 77843-3255, USA

Strong field processes

Used for many applications in molecular imaging



Angle-resolved photoelectron spectroscopy

IOP Publishing — Journal of Physics B: Atomic, Molecular

J. Phys. B: At. Mol. Opt. Phys. **43** (2010) 122001 (33pp) — doi:10.1088/0953-4

TOPICAL REVIEW

Strong-field rescattering physics—self-imaging of a molecule by its own electrons

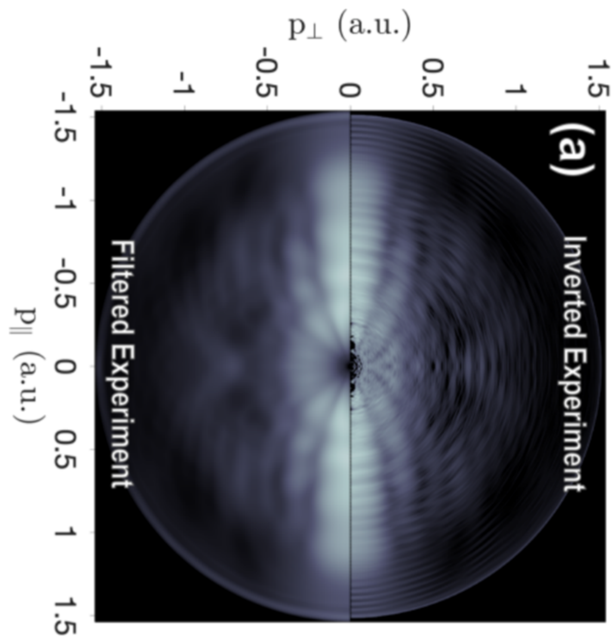
C D Lin[1], Anh-Thu Le[1], Zhangjin Chen[1], Toru Morishita[2,3] and Robert Lucchese[4]

[1] J. R. Macdonald Laboratory, Physics Department, Kansas State University, Manhattan, KS 66506-2604, USA

[2] Department of Applied Physics and Chemistry, University of Electro-Communications, 1-5-1 Chofu-ga-oka, Chofu-shi, Tokyo, 182-8585, Japan

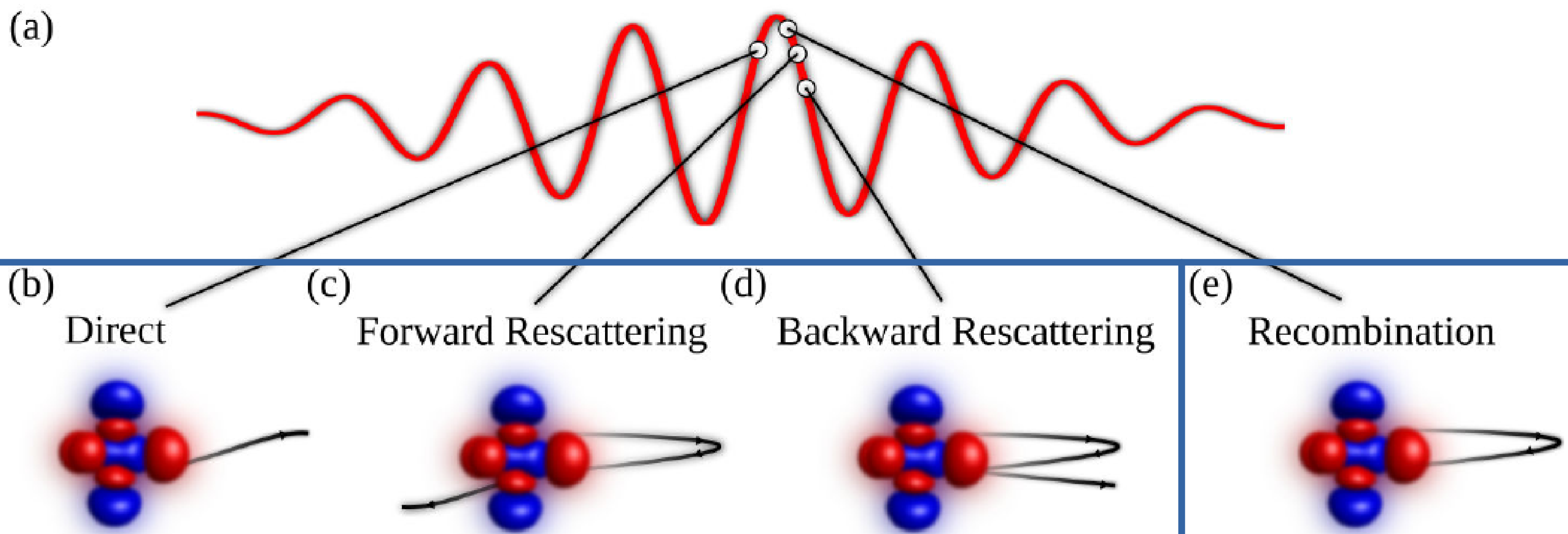
[3] PRESTO, Japan Science and Technology Agency, Kawaguchi, Saitama 332-0012, Japan

[4] Department of Chemistry, Texas A&M University, College Station, TX 77843-3255, USA



Strong field processes

Used for many applications in molecular imaging



Angle-resolved photoelectron spectroscopy

XUV spectroscopy
High-order harmonic generation
(attosecond pulses)

IOP PUBLISHING JOURNAL OF PHYSICS B: ATOMIC, MOLECULAR AND OPTICAL PHYSICS

J. Phys. B: At. Mol. Opt. Phys. **43** (2010) 122001 (33pp) doi:10.1088/0953-4075/43/12/122001

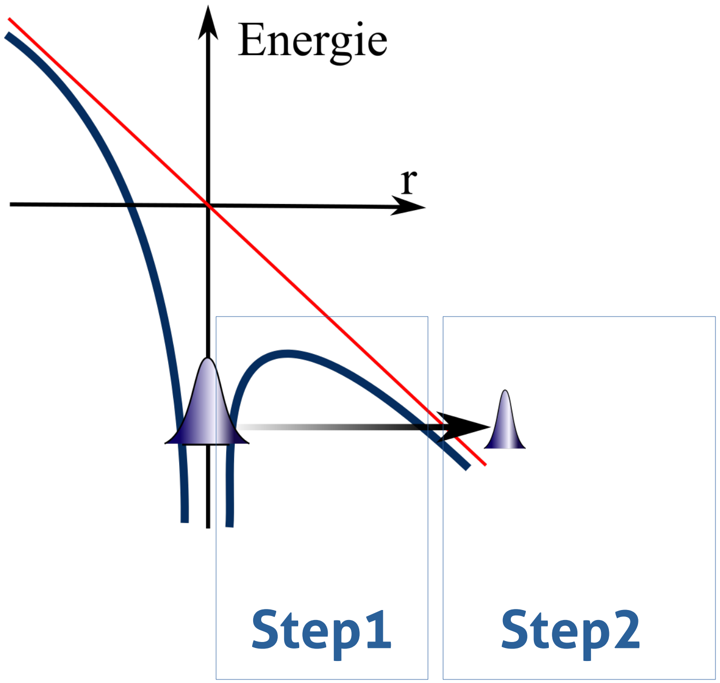
TOPICAL REVIEW

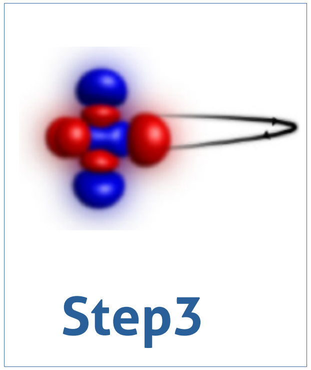
Strong-field rescattering physics—self-imaging of a molecule by its own electrons

C D Lin[1], Anh-Thu Le[1], Zhangjin Chen[1], Toru Morishita[2,3] and Robert Lucchese[4]

[1] J. R. Macdonald Laboratory, Physics Department, Kansas State University, Manhattan, KS 66506-2604, USA
[2] Department of Applied Physics and Chemistry, University of Electro-Communications, 1-5-1 Chofu-ga-oka, Chofu-shi, Tokyo, 182-8585, Japan
[3] PRESTO, Japan Science and Technology Agency, Kawaguchi, Saitama 332-0012, Japan
[4] Department of Chemistry, Texas A&M University, College Station, TX 77843-3255, USA

Strong field processes



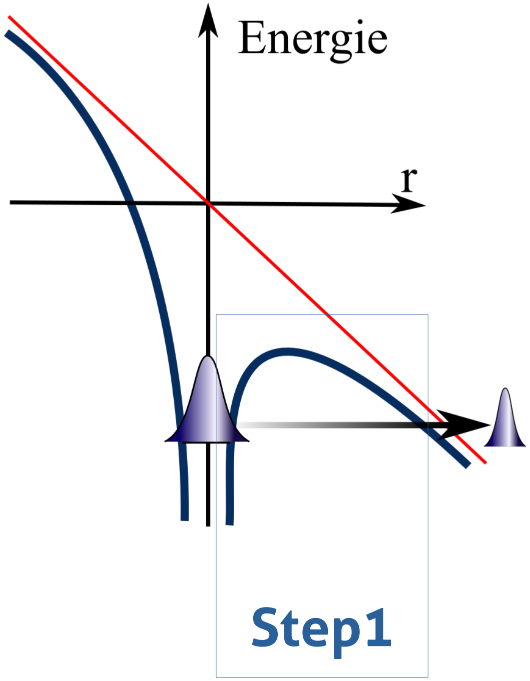


Step1 : classically forbidden region – sub-barrier
tunneling

Step 2 : post-tunneling dynamics – continuum
acceleration by the laser field

Step 3 : re-collision

Structural sensitivity of strong field processes



Step1 : classically forbidden region – sub-barrier

tunneling

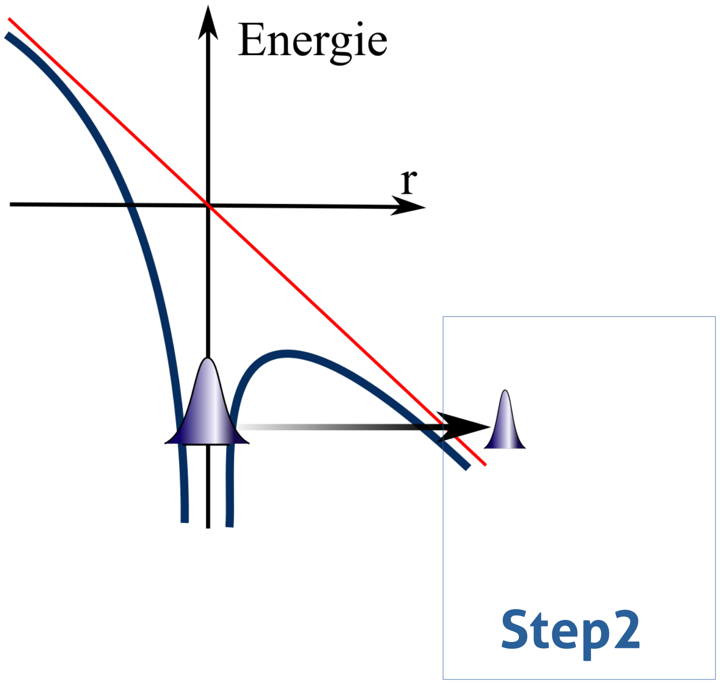
Phase shift of the emerging wavepacket

Step 2 : post-tunneling dynamics – continuum

acceleration by the laser field

Step 3 : re-collision

Structural sensitivity of strong field processes



Step1 : classically forbidden region – sub-barrier

tunneling

Phase shift of the emerging wavepacket

Step 2 : post-tunneling dynamics – continuum

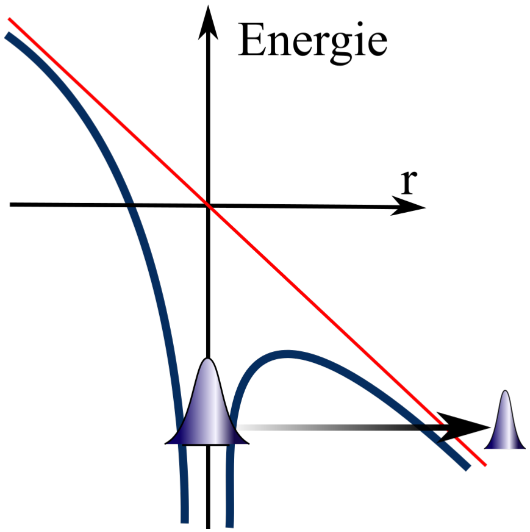
acceleration by the laser field

Scattering off the potential

→ angle-resolved photoelectron spectrum

Step 3 : re-collision

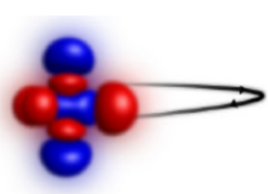
Structural sensitivity of strong field processes



Step1 : classically forbidden region – sub-barrier
tunneling
Phase shift of the emerging wavepacket

Step 2 : post-tunneling dynamics – continuum
acceleration by the laser field
Scattering off the potential
→ angle-resolved photoelectron spectrum

Step 3 : re-collision
Radiative recombination
→ High-harmonic spectroscopy

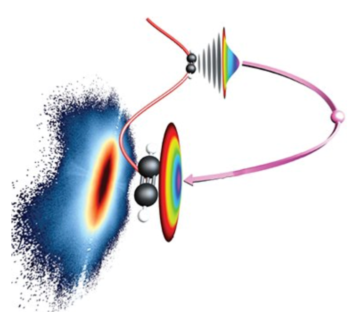


articles

Tomographic imaging of molecular orbitals

J. Itatani[1,2], J. Levesque[1,3], D. Zeidler[1], Hiromichi Niikura[1,4], H. Pépin[3], J. C. Kieffer[3], P. B. Corkum[1] & D. M. Villeneuve[1]

Diffraction of the electron wavepacket by nuclei
Laser-induced electron diffraction



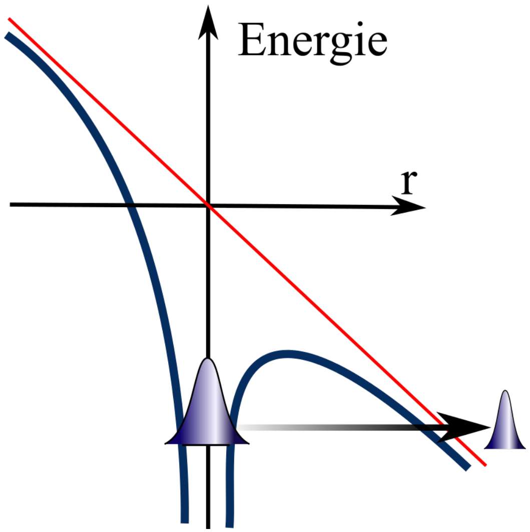
LETTER

doi:10.1038/nature10820

Imaging ultrafast molecular dynamics with laser-induced electron diffraction

Cosmin I. Blaga[1], Junliang Xu[2], Anthony D. DiChiara[1], Emily Sistrunk[1], Kaikai Zhang[1], Pierre Agostini[1], Terry A. Miller[3], Louis F. DiMauro[1] & C. D. Lin[2]

Structural sensitivity of strong field processes



Step1 : classically forbidden region – sub-barrier
tunneling
Phase shift of the emerging wavepacket

Step 2 : post-tunneling dynamics – continuum
acceleration by the laser field
Scattering off the potential
→ angle-resolved photoelectron spectrum

Step 3 : re-collision
Radiative recombination
→ High-harmonic spectroscopy

Diffraction of the electron wavepacket by nuclei
Laser-induced electron diffraction

Chiral response in strong-fields?

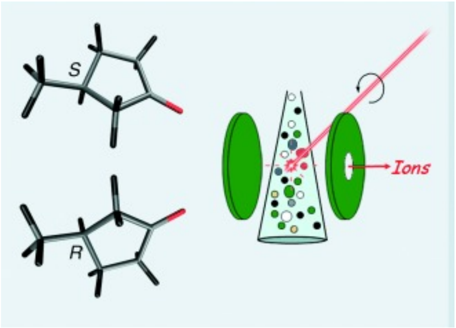
Chiral response in ionization ?

Measurement of mass spectrum

→ Circular Dichroism

Different number of ions produced by left and right CP

Magnetic dipole / electric quadrupole transitions



Circular Dichroism Laser Mass Spectrometry: Differentiation of 3-Methylcyclopentanone Enantiomers

Ulrich Boesl von Grafenstein* and Alexander Bornschlegl[a]

ChemPhysChem **2006**, *7*, 2085 – 2087

REPORT

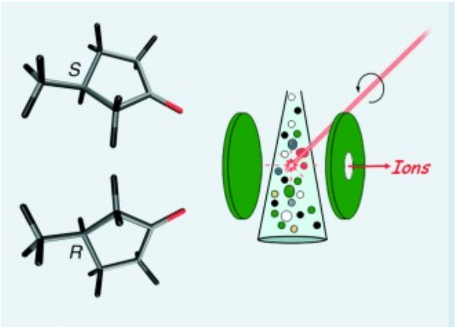
SPECTROSCOPY

Mass-resolved electronic circular dichroism ion spectroscopy

Steven Daly[1], Frédéric Rosu[2], Valérie Gabelica[1]*

Daly *et al.*, *Science* **368**, 1465–1468 (2020)

Chiral response in ionization ?



Measurement of mass spectrum
→ Circular Dichroism

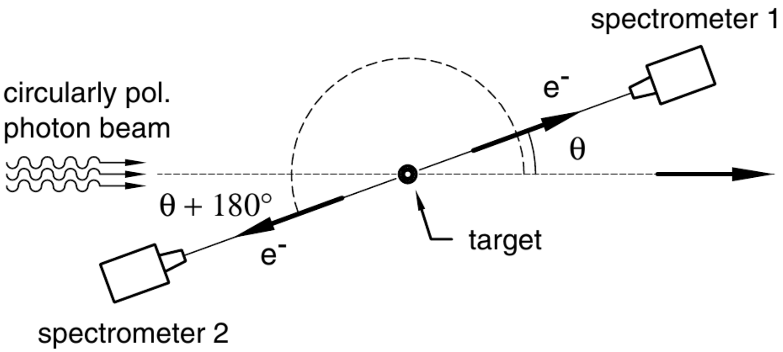
Different number of ions produced by left and right CP

Magnetic dipole / electric quadrupole transitions

Measurement of angular distribution of electrons
→ PhotoElectron Circular Dichroism

Different number of electrons ejected forward and backward by LCP and RCP

Strong effect : electric dipole transition

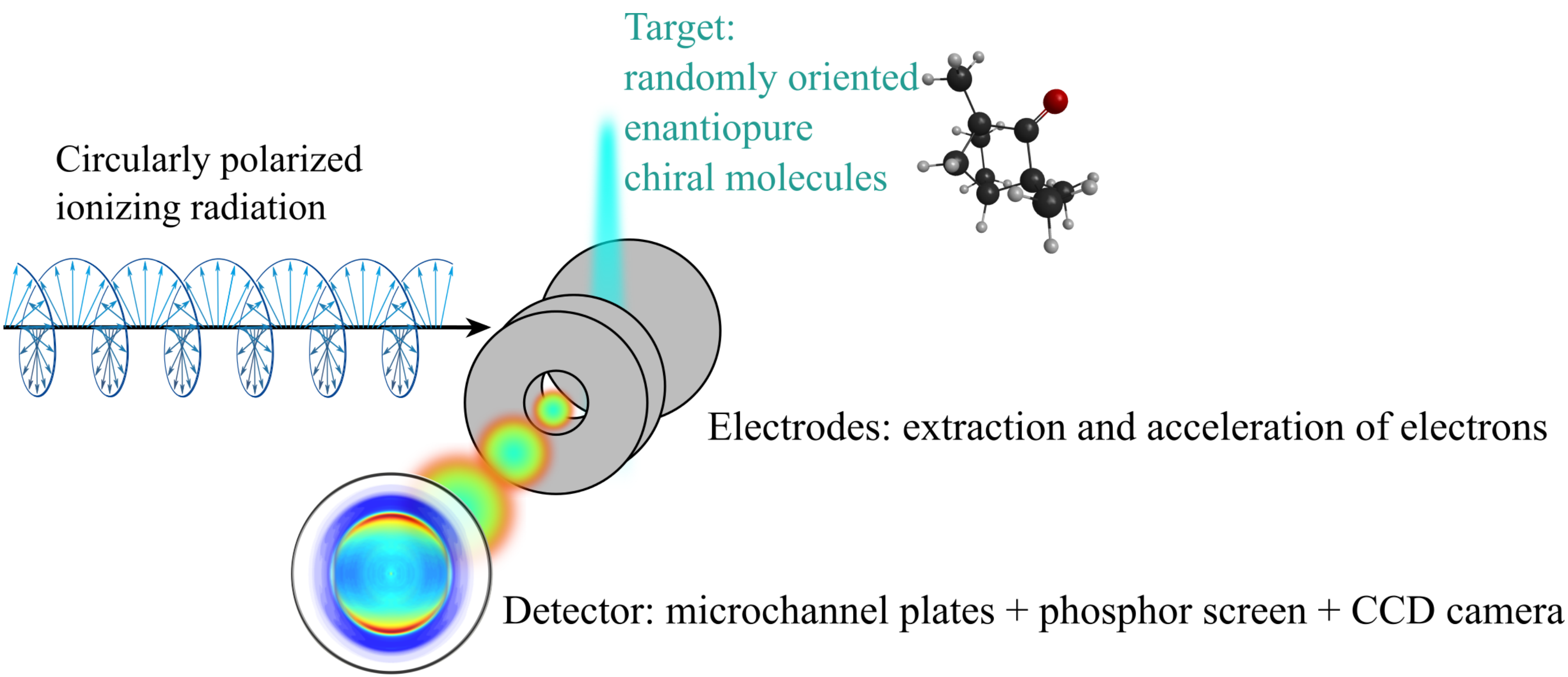


VOLUME 86, NUMBER 7 PHYSICAL REVIEW LETTERS 12 FEBRUARY 2001

Asymmetry in Photoelectron Emission from Chiral Molecules Induced by Circularly Polarized Light

N. Böwering,[1,2] T. Lischke,[1,3] B. Schmidtke,[1] N. Müller,[1] T. Khalil,[1] and U. Heinzmann[1]
[1]*Universität Bielefeld, Fakultät für Physik, 33501 Bielefeld, Germany*
[2]*Max-Born-Institut, Max-Born Strasse 2A, 12489 Berlin, Germany*
[3]*Fritz-Haber-Institut der Max-Planck-Gesellschaft, 14195 Berlin, Germany*
(Received 9 August 2000)

Photoelectron circular dichroism

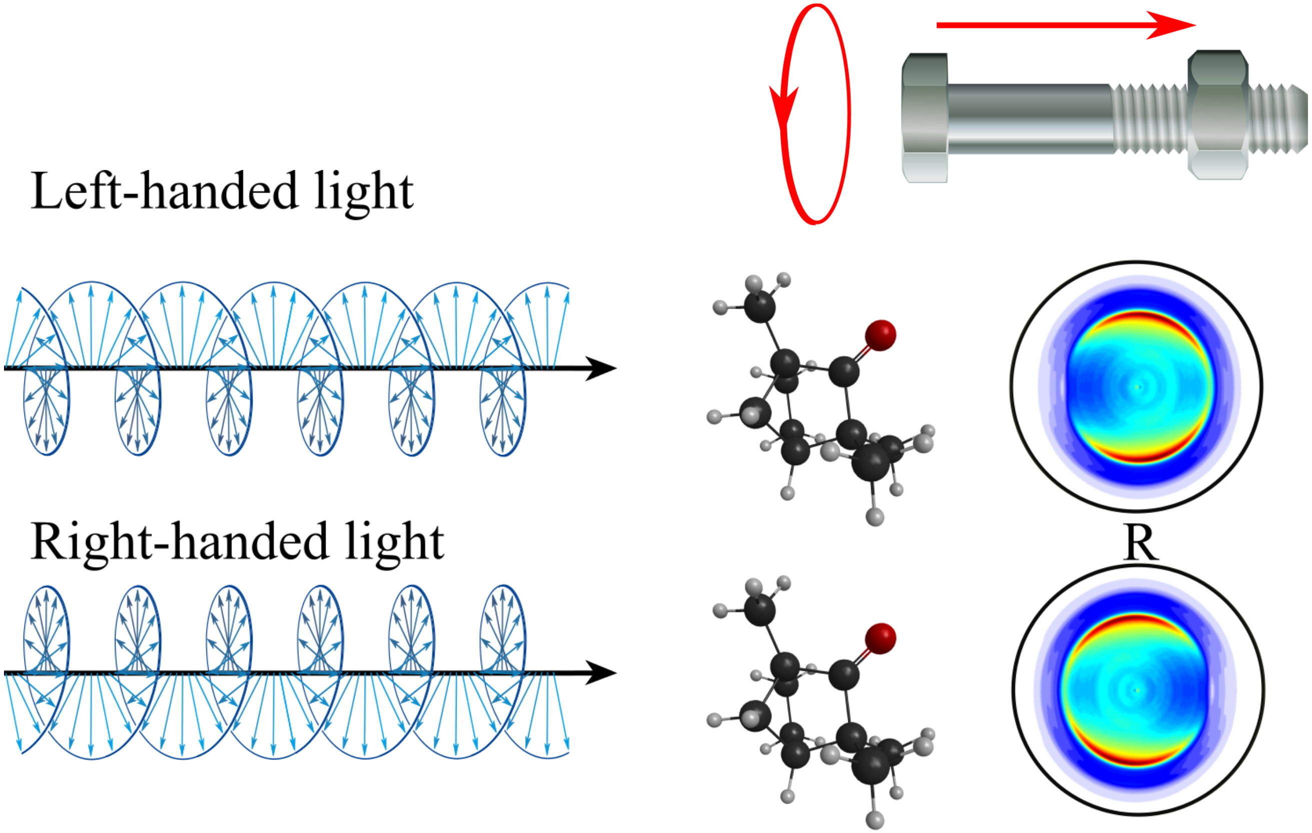


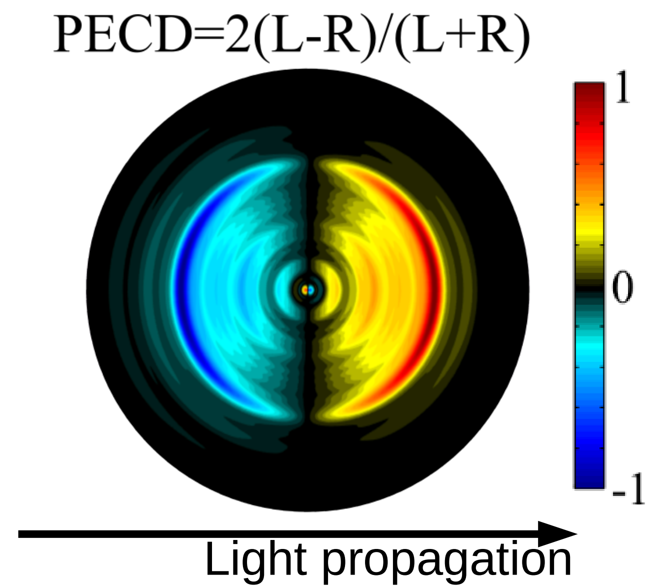
Detector : Velocity Map Imaging spectrometer

→ Projection of the angular distribution of the photoelectron spectrum

Photoelectron circular dichroism

B. Ritchie, PRA 13, 1411 (1976)
I. Powis, JCP 112, 301 (2000)
N. Bowering et al., PRL 86, 1187 (2001)

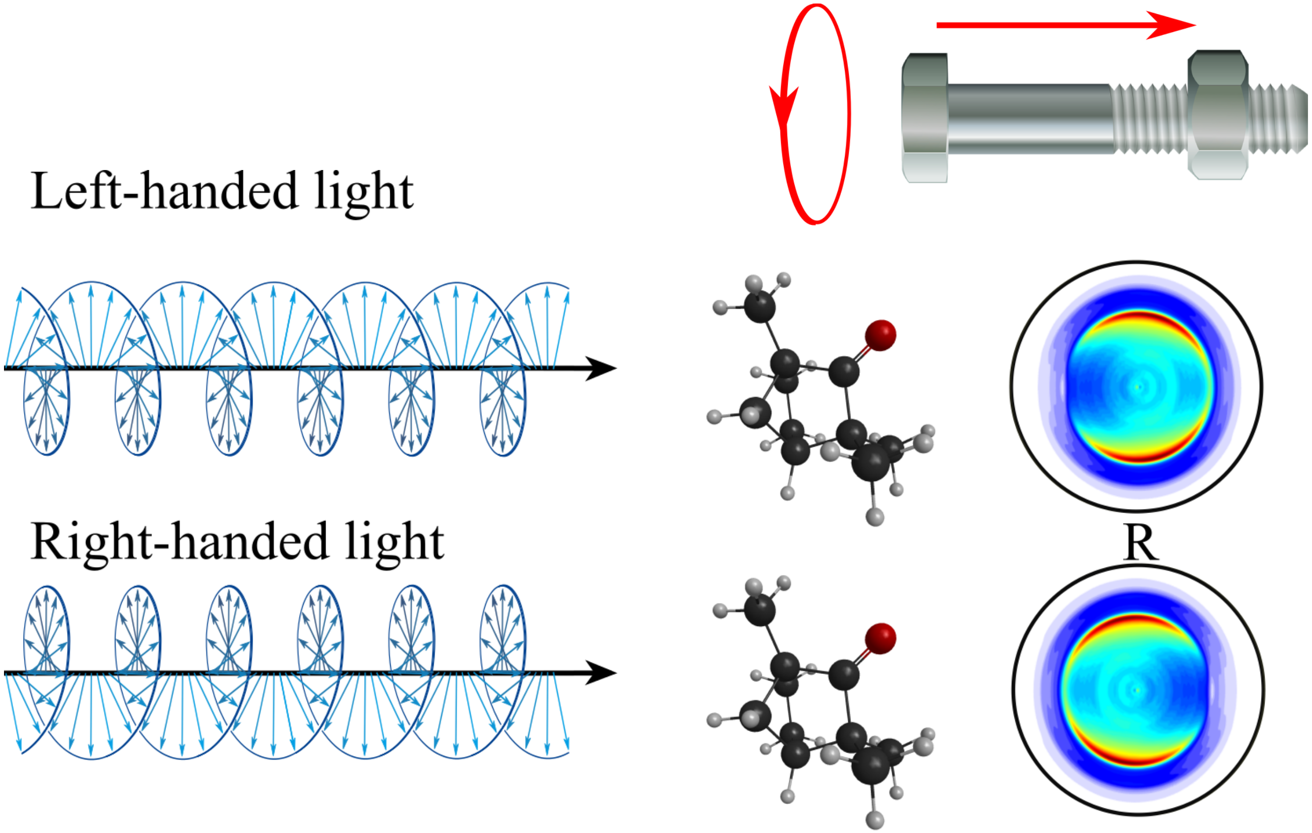


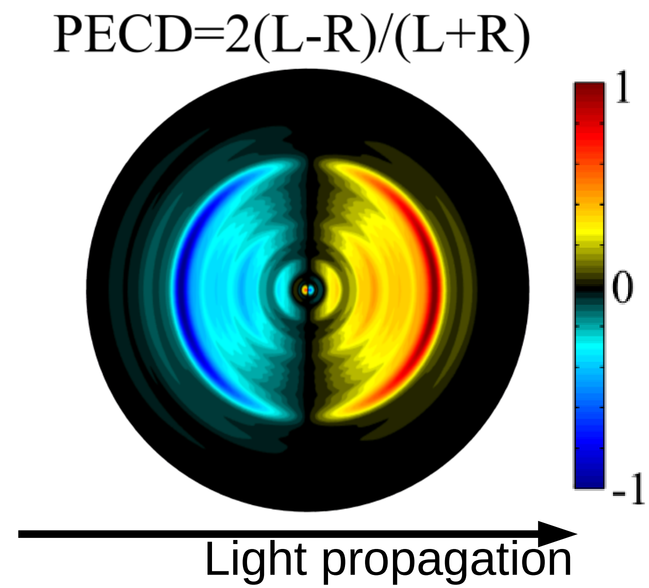


For reviews see M. Janssen and I. Powis, PCCP 16, 856 (2014)
L. Nahon et al., J. Elec. Spec. Rel. Phen. 2014, 322 (2015)

Photoelectron circular dichroism

B. Ritchie, PRA 13, 1411 (1976)
I. Powis, JCP 112, 301 (2000)
N. Bowering et al., PRL 86, 1187 (2001)





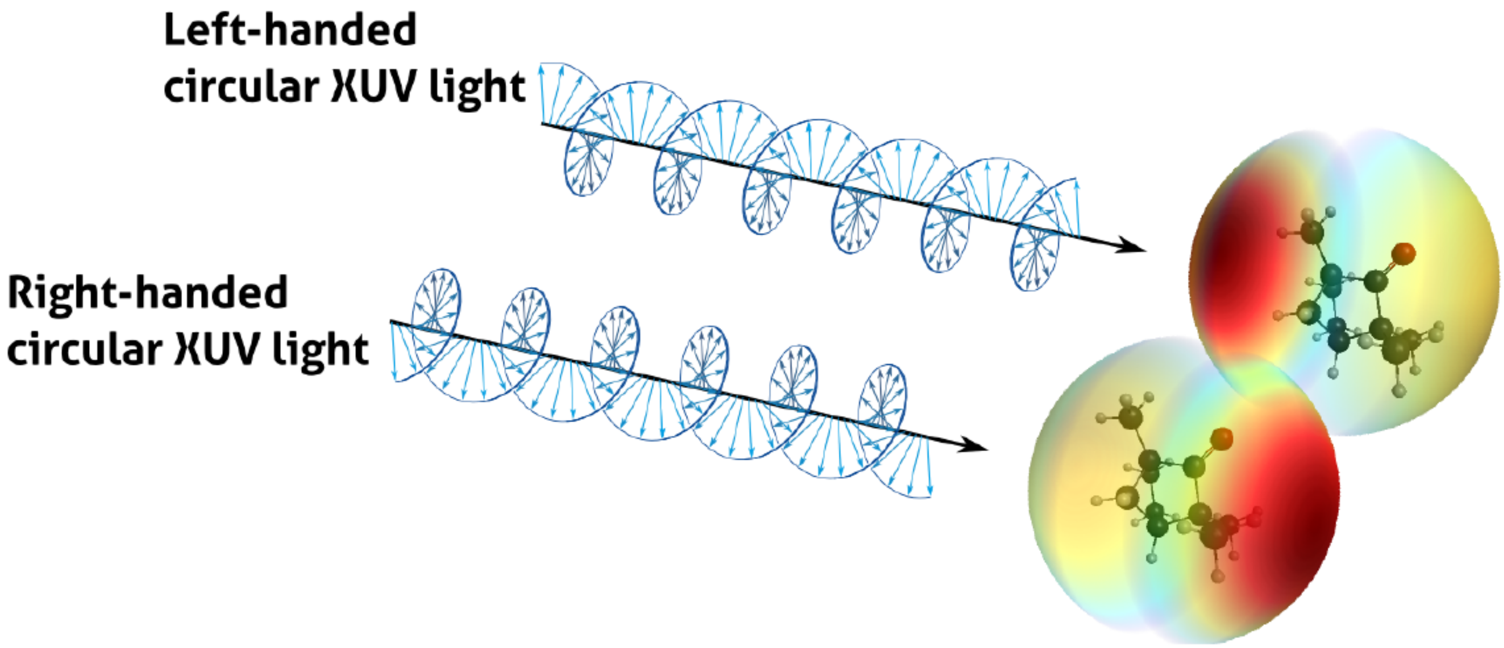
PECD=Pure electric dipole effect → much stronger than most other CDs

Up to 34 % measured

For reviews see M. Janssen and I. Powis, PCCP 16, 856 (2014)
L. Nahon et al., J. Elec. Spec. Rel. Phen. 2014, 322 (2015)

Physical origin of PECD

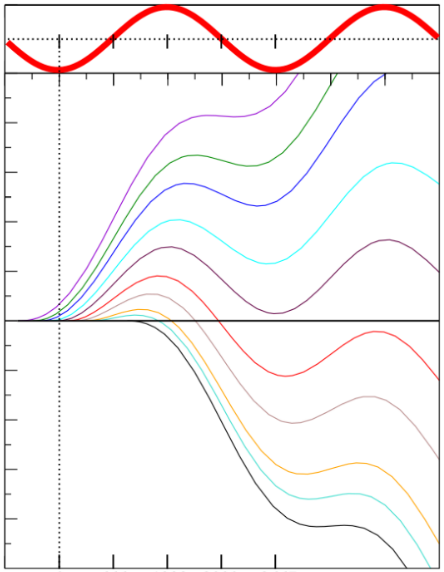
Scattering phases of the outgoing electrons in the chiral potential



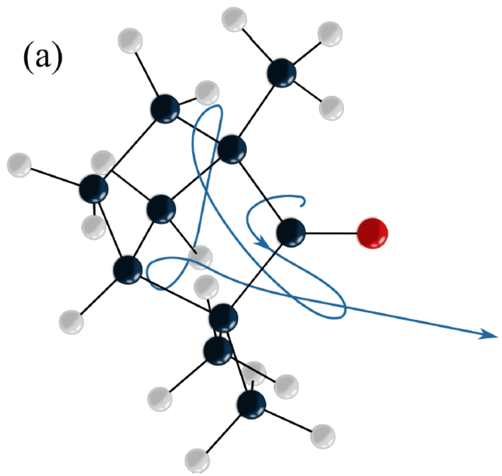
For reviews see M. Janssen and I. Powis, PCCP 16, 856 (2014)
L. Nahon et al., J. Elec. Spec. Rel. Phen. 2014, 322 (2015)

Physical origin of PECD

Classical interpretation ?



1D trajectories, linear polarization, without potential

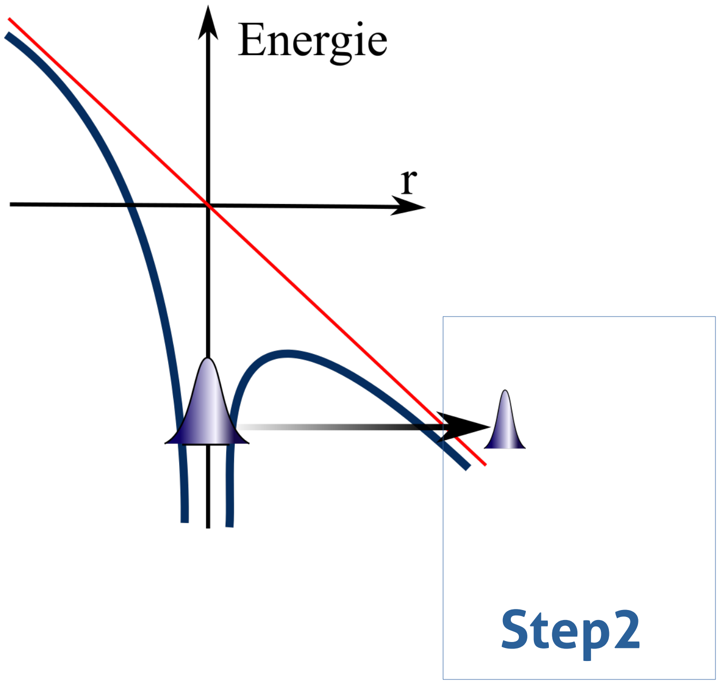


3D trajectories, circular polarization, with chiral potential

→ **PECD**

S. Beaulieu et al., New J. Phys. 102002 (2016)

Chiral sensitivity of strong field processes



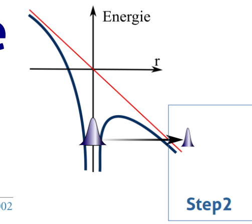
Step1 : classically forbidden region – sub-barrier
tunneling

Step 2 : post-tunneling dynamics – continuum
acceleration by the laser field

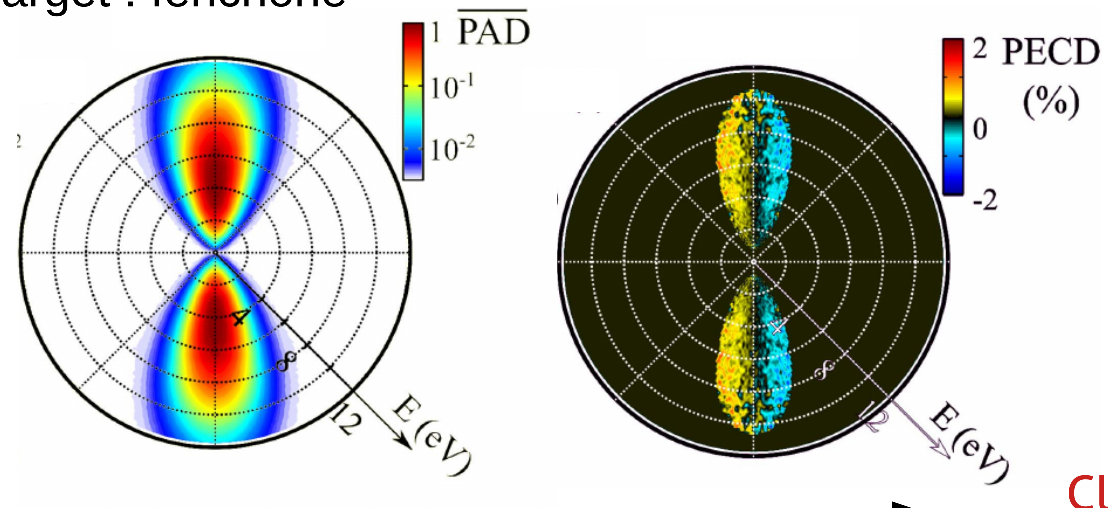
Scattering off the potential
→ angle-resolved photoionization cross section
→ **Photoelectron circular dichroism**

Photoelectron circular dichroism in strong field ionization

Ionization by strong Mid-Infrared (1850 nm) laser pulses – tunneling regime



Target : fenchone



Laser propagation direction

New J. Phys. **18** (2016) 102002 doi:10.1088/1367-2630/18/10/102002

New Journal of Physics
The open access journal at the forefront of physics

Deutsche Physikalische Gesellschaft DPG
IOP Institute of Physics

Published in partnership with: Deutsche Physikalische Gesellschaft and the Institute of Physics

FAST TRACK COMMUNICATION

Universality of photoelectron circular dichroism in the photoionization of chiral molecules

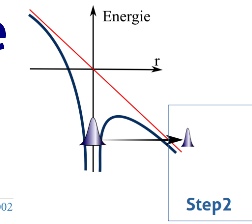
S Beaulieu[1,2], A Ferré[1], R Géneaux[3], R Canonge[1], D Descamps[1], B Fabre[1], N Fedorov[1], F Légaré[2], S Petit[1], T Ruchon[3], V Blanchet[1], Y Mairesse[1] and B Pons[1]

[1] Université de Bordeaux—CNRS—CEA, CELIA, UMR5107, F-33405 Talence, France
[2] ALLS/LSF, INRS-ÉMT, 1650 Boulevard Lionel-Boulet, Varennes, Qc, Canada
[3] LIDYL, CEA, CNRS, Université Paris-Saclay, CEA Saclay F-91191 Gif-sur-Yvette, France

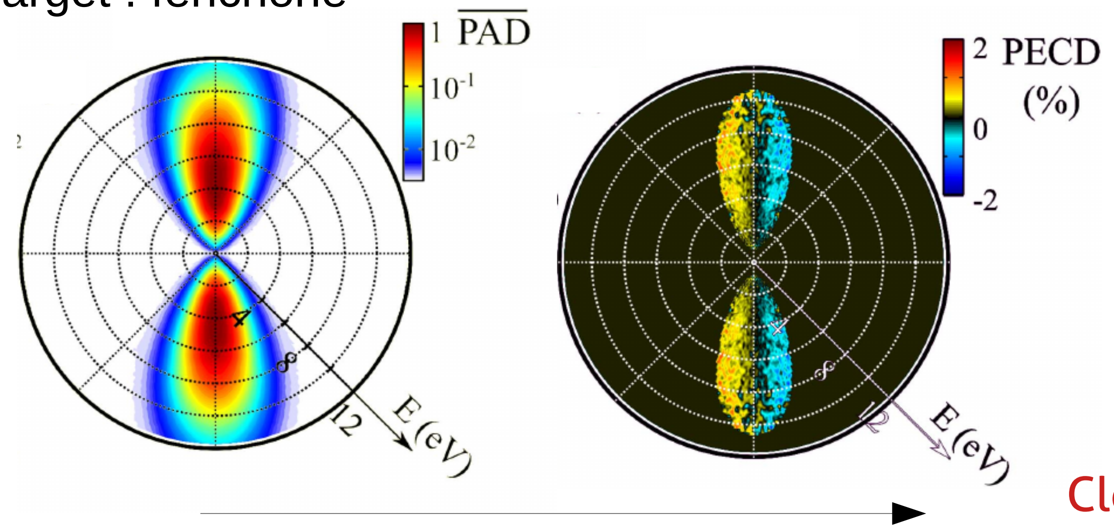
Clear PECD, ~1 %

Photoelectron circular dichroism in strong field ionization

Ionization by strong Mid-Infrared (1850 nm) laser pulses – tunneling regime



Target : fenchone



Laser propagation direction

New J. Phys. 18 (2016) 102002
doi:10.1088/1367-2630/18/10/102002
New Journal of Physics
The open access journal at the forefront of physics
Published in partnership with: Deutsche Physikalische Gesellschaft and the Institute of Physics
FAST TRACK COMMUNICATION

Universality of photoelectron circular dichroism in the photoionization of chiral molecules

S Beaulieu[1,2], A Ferré[1], R Géneaux[3], R Canonge[1], D Descamps[1], B Fabre[1], N Fedorov[1], F Légaré[2], S Petit[1], T Ruchon[3], V Blanchet[1], Y Mairesse[1] and B Pons[1]
[1] Université de Bordeaux—CNRS—CEA, CELIA, UMR5107, F-33405 Talence, France
[2] ALLS/LSF, INRS-ÉMT, 1650 Boulevard Lionel-Boulet, Varennes, Qc, Canada
[3] LIDYL, CEA, CNRS, Université Paris-Saclay, CEA Saclay F-91191 Gif-sur-Yvette, France

Clear PECD, ~1 %

Time-resolved attosecond photoionization of chiral molecules

Few attosecond delay between electron ejected forward and backward

Target : camphor

CHEMICAL PHYSICS

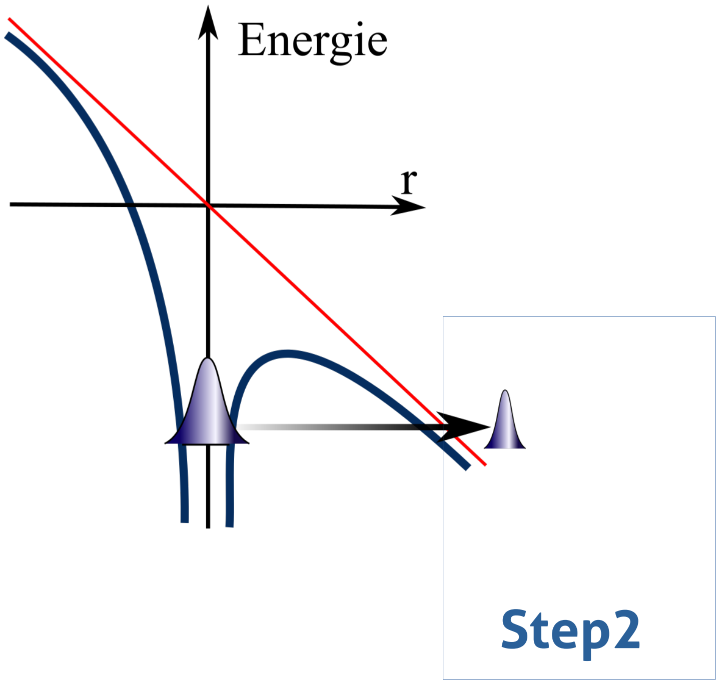
Attosecond-resolved photoionization of chiral molecules

S. Beaulieu,[1,2]* A. Comby,[1] A. Clergerie,[1] J. Caillat,[3] D. Descamps,[1] N. Dudovich,[4] B. Fabre,[1] R. Géneaux,[5] F. Légaré,[2] S. Petit,[1] B. Pons,[1] G. Porat,[4] T. Ruchon,[5] R. Taïeb,[3] V. Blanchet,[1] Y. Mairesse[1]

Beaulieu *et al.*, *Science* **358**, 1288–1294 (2017)

→ Signatures of chiral potential during electron acceleration in the continuum

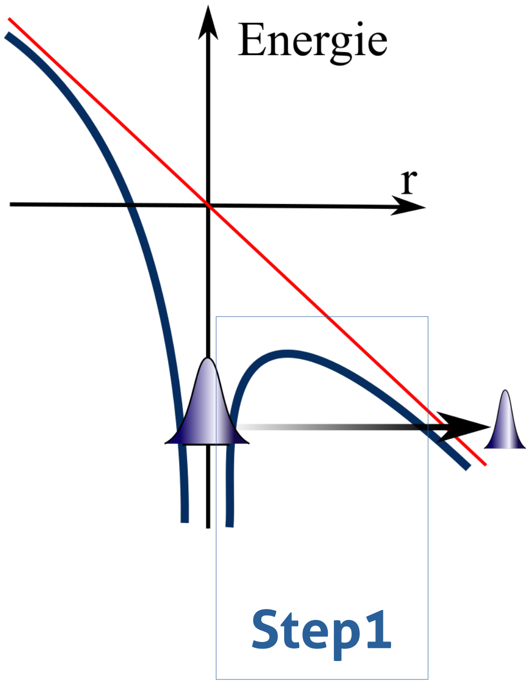
Chiral sensitivity of strong field ionization ?



Scattering off chiral potential in Step 2:
→ imprints a phase and amplitude asymmetry in the photoelectron distribution

Issue : if the electron emerges far from the core, the chiral part of the potential vanishes

Chiral sensitivity of strong field ionization ?



Scattering off chiral potential in Step 2:
→ imprints a phase and amplitude asymmetry in the photoelectron distribution

Issue : if the electron emerges far from the core, the chiral part of the potential vanishes

Scattering off chiral potential in Step 1 ?
→ is there an influence of chirality on the wavepacket emerging from the tunnel ?

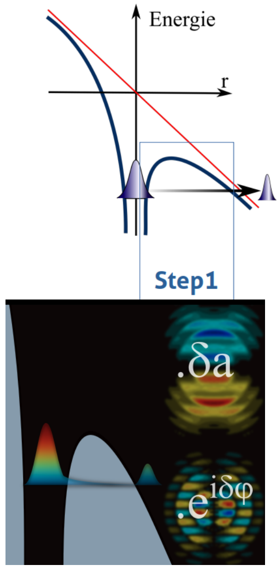
Chiral sensitivity of tunneling

PHYSICAL REVIEW X **VOL..XX,** 000000 (XXXX)

Revealing the Influence of Molecular Chirality on Tunnel-Ionization Dynamics

E. Bloch,[1] S. Larroque,[1] S. Rozen,[2] S. Beaulieu,[1] A. Comby,[1] S. Beauvarlet,[1] D. Descamps,[1] B. Fabre,[1] S. Petit,[1] R. Taïeb,[3] A. J. Uzan,[2] V. Blanchet,[1] N. Dudovich,[2] B. Pons,[1] and Y. Mairesse[1]
[1]*Université de Bordeaux-CNRS-CEA, CELIA, UMR5107, F33405 Talence, France*
[2]*Weizmann Institute of Science, Rehovot, 76100, Israel*
[3]*Sorbonne Université, CNRS, Laboratoire de Chimie Physique-Matière et Rayonnement, LCPMR, F-75005 Paris, France*

(Received 19 May 2021; accepted 13 October 2021)



Sub-barrier dynamics induce a chiroptical response

- No chiral tunneling delay
- Sub-barrier amplitude and phase modulation of the wavepacket

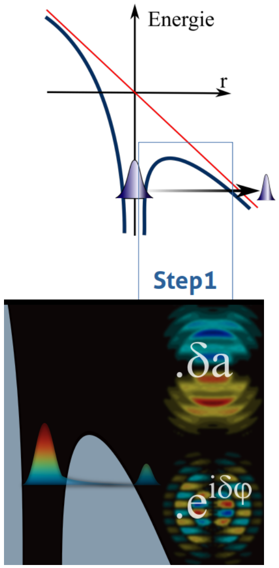
Chiral sensitivity of tunneling

PHYSICAL REVIEW X **VOL..XX,** 000000 (XXXX)

Revealing the Influence of Molecular Chirality on Tunnel-Ionization Dynamics

E. Bloch,[1] S. Larroque,[1] S. Rozen,[2] S. Beaulieu,[1] A. Comby,[1] S. Beauvarlet,[1] D. Descamps,[1]
B. Fabre,[1] S. Petit,[1] R. Taïeb,[3] A. J. Uzan,[2] V. Blanchet,[1] N. Dudovich,[2] B. Pons,[1] and Y. Mairesse[1]
[1]*Université de Bordeaux-CNRS-CEA, CELIA, UMR5107, F33405 Talence, France*
[2]*Weizmann Institute of Science, Rehovot, 76100, Israel*
[3]*Sorbonne Université, CNRS, Laboratoire de Chimie Physique-Matière et Rayonnement, LCPMR, F-75005 Paris, France*

(Received 19 May 2021; accepted 13 October 2021)



Sub-barrier dynamics induce a chiroptical response

- No chiral tunneling delay
- Sub-barrier amplitude and phase modulation of the wavepacket
- Dynamical process : must feel the rotation of the laser field

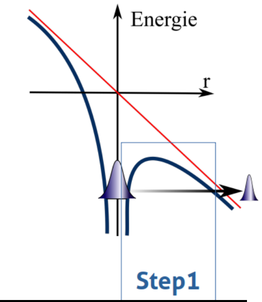
Chiral sensitivity of tunneling

PHYSICAL REVIEW X **VOL..XX,** 000000 (XXXX)

Revealing the Influence of Molecular Chirality on Tunnel-Ionization Dynamics

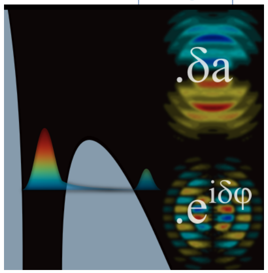
E. Bloch,[1] S. Larroque,[1] S. Rozen,[2] S. Beaulieu,[1] A. Comby,[1] S. Beauvarlet,[1] D. Descamps,[1] B. Fabre,[1] S. Petit,[1] R. Taïeb,[3] A. J. Uzan,[2] V. Blanchet,[1] N. Dudovich,[2] B. Pons,[1] and Y. Mairesse[1]
[1]Université de Bordeaux-CNRS-CEA, CELIA, UMR5107, F33405 Talence, France
[2]Weizmann Institute of Science, Rehovot, 76100, Israel
[3]Sorbonne Université, CNRS, Laboratoire de Chimie Physique-Matière et Rayonnement, LCPMR, F-75005 Paris, France

(Received 19 May 2021; accepted 13 October 2021)



Sub-barrier dynamics induce a chiroptical response

No chiral tunneling delay
Sub-barrier amplitude and phase modulation of the wavepacket
Dynamical process : must feel the rotation of the laser field



Non-adiabatic tunneling

The electron gets excited under the barrier

PHYSICAL REVIEW LETTERS **120,** 013201 (2018)

Under-the-Tunneling-Barrier Recollisions in Strong-Field Ionization

Michael Klaiber,* Karen Z. Hatsagortsyan,† and Christoph H. Keitel
Max-Planck-Institut für Kernphysik, Saupfercheckweg 1, 69117 Heidelberg, Germany

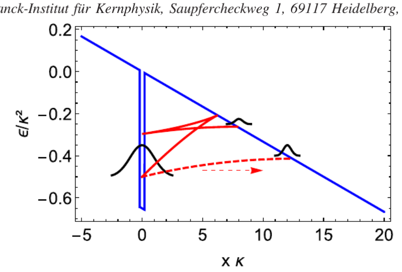


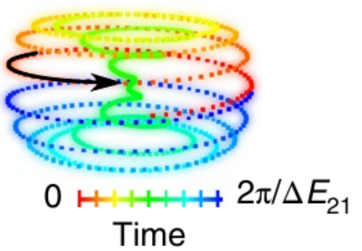
FIG. 1. Schematic picture of laser-induced tunneling ionization: (dashed line) the direct trajectory, and (solid line) the under-the-barrier recolliding trajectory.The Keldysh parameter is $\gamma = 1$, featuring nonadiabatic tunneling, i.e., when the energy is not constant during tunneling.

The excitation process can be chirosensitive

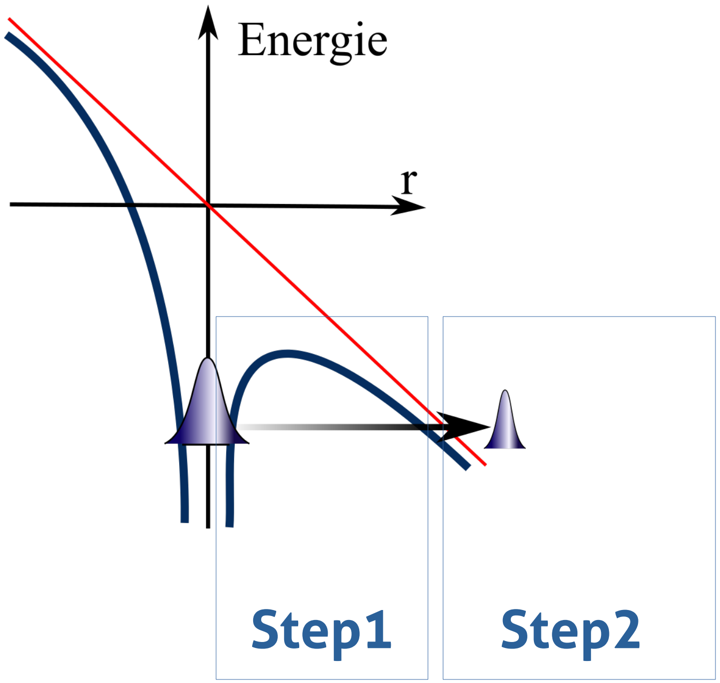
ARTICLES nature physics
https://doi.org/10.1038/s41567-017-0038-z

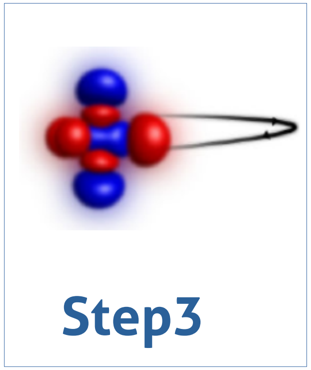
Photoexcitation circular dichroism in chiral molecules

S. Beaulieu[1,2], A. Comby[1], D. Descamps[1], B. Fabre[1], G. A. Garcia[3], R. Géneaux[4], A. G. Harvey[5], F. Légaré[2], Z. Mašín[5], L. Nahon[3], A. F. Ordonez[5,6], S. Petit[1], B. Pons[1]*, Y. Mairesse[1]*, O. Smirnova[5,6]* and V. Blanchet[1]



Chiral sensitivity of strong field processes



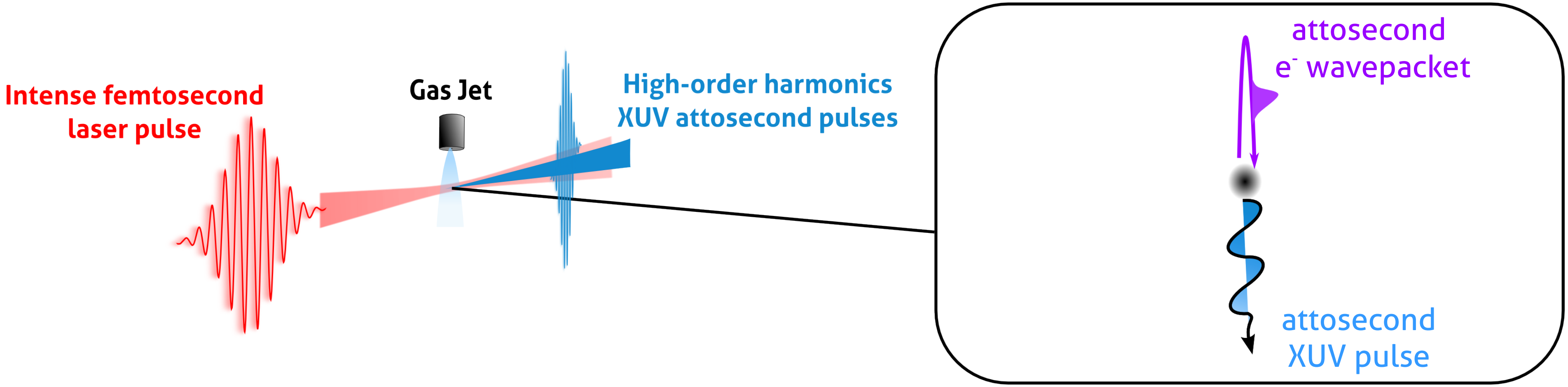


Step1 : classically forbidden region – sub-barrier
→ Photoelectron circular dichroism

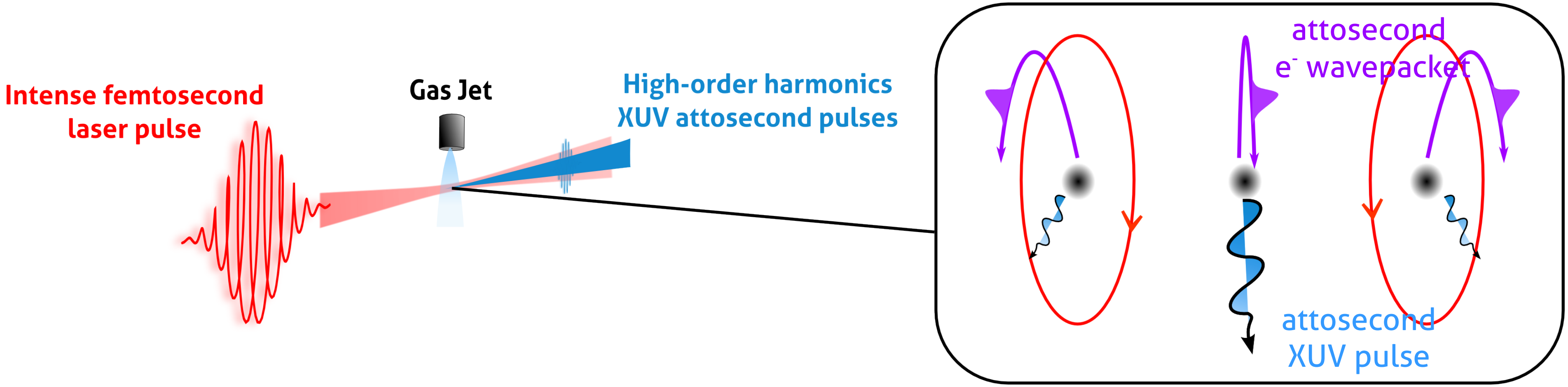
Step 2 : post-tunneling dynamics – continuum
→ Photoelectron circular dichroism

Step 3 : re-collision ?

High-order harmonic generation



High-order harmonic generation



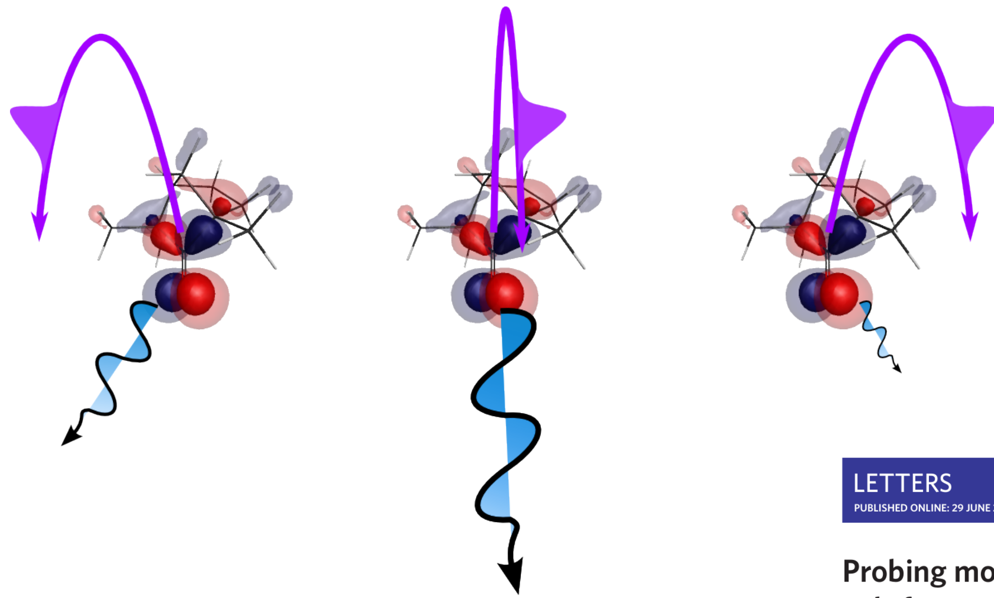
Recollision probability decreases with ellipticity

Sensitivity to chirality ?

Chiral high-harmonic generation

Strong difference (1%) in the emission from left and right elliptical light

Chiral recollision



LETTERS
PUBLISHED ONLINE: 29 JUNE 2015 | DOI: 10.1038/NPHYS3369
nature physics

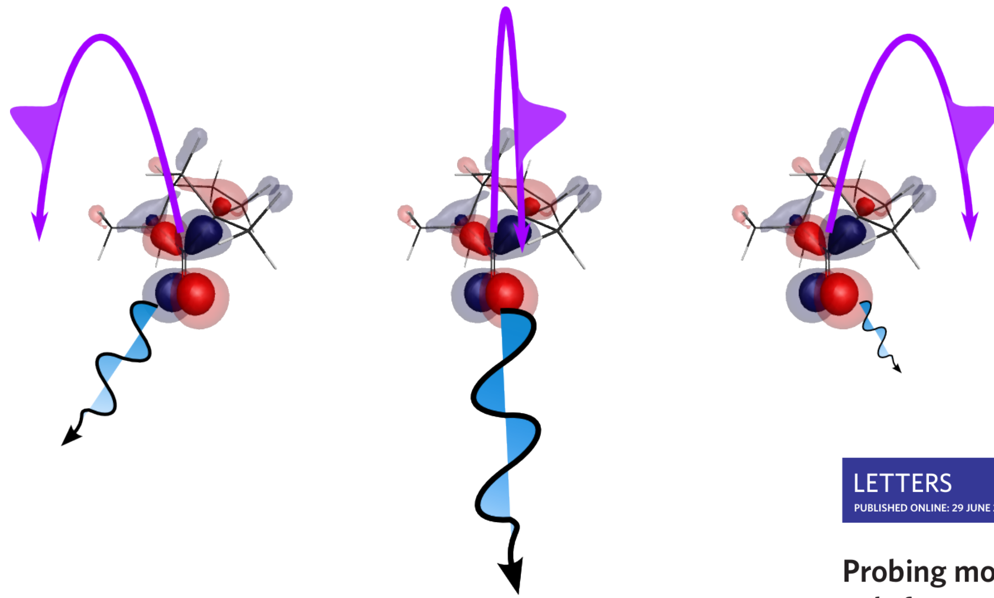
Probing molecular chirality on a sub-femtosecond timescale

R. Cireasa[1,2], A. E. Boguslavskiy[3,4,5], B. Pons[6], M. C. H. Wong[3], D. Descamps[6], S. Petit[6], H. Ruf[6], N. Thiré[1,7], A. Ferré[6], J. Suarez[8], J. Higuet[6], B. E. Schmidt[7], A. F. Alharbi[3,9], F. Légaré[7], V. Blanchet[1,6], B. Fabre[6], S. Patchkovskii[4,10], O. Smirnova[10]*, Y. Mairesse[6]* and V. R. Bhardwaj[3]*

Chiral high-harmonic generation

Strong difference (1%) in the emission from left and right elliptical light

Chiral recollision



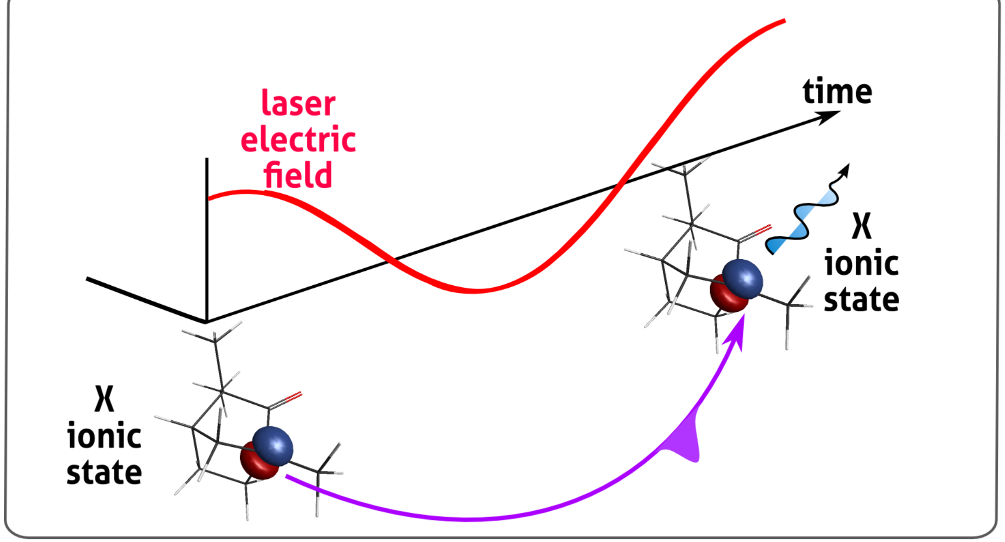
LETTERS
PUBLISHED ONLINE: 29 JUNE 2015 | DOI: 10.1038/NPHYS3369
nature physics

Probing molecular chirality on a sub-femtosecond timescale

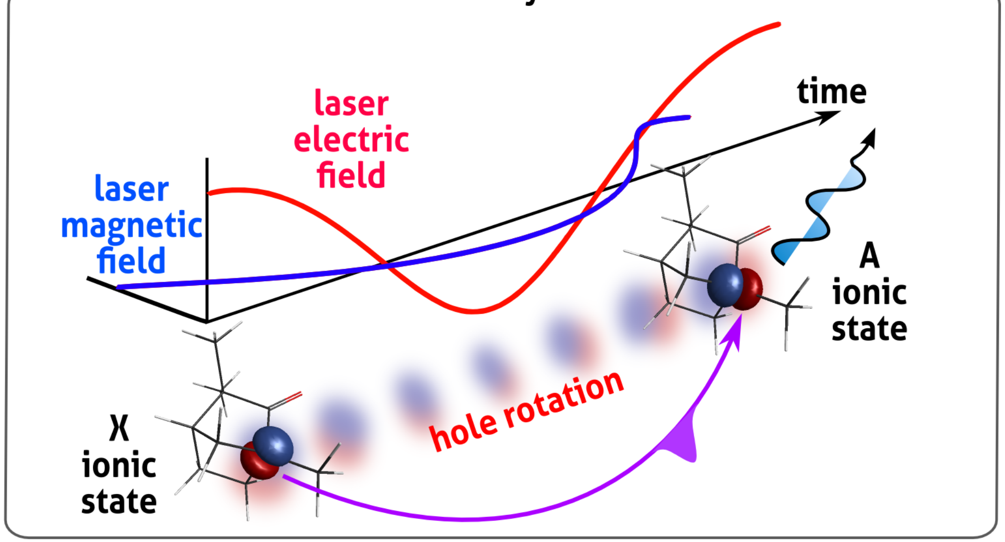
R. Cireasa[1,2], A. E. Boguslavskiy[3,4,5], B. Pons[6], M. C. H. Wong[3], D. Descamps[6], S. Petit[6], H. Ruf[6], N. Thiré[1,7], A. Ferré[6], J. Suarez[8], J. Higuet[6], B. E. Schmidt[7], A. F. Alharbi[3,9], F. Légaré[7], V. Blanchet[1,6], B. Fabre[6], S. Patchkovskii[4,10], O. Smirnova[10]*, Y. Mairesse[6]* and V. R. Bhardwaj[3]*

Origin:

"Direct" recollision



Attosecond chiral hole dynamics



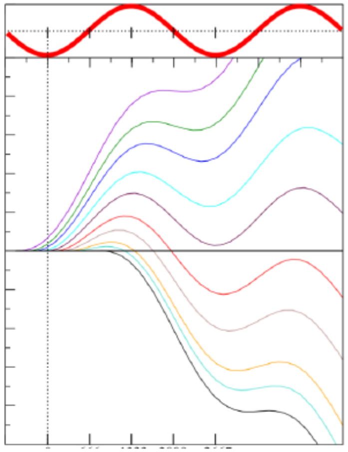
Attosecond rotation of the hole under the laser magnetic field

Optimizing the strong-field chiral response

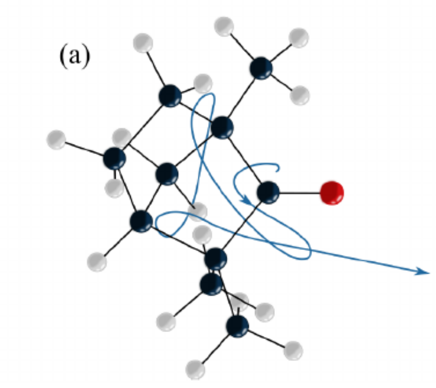
Key idea of strong-field physics :

electron trajctories can be manipulated by shaping the laser field

→ Can we shape trajectories optimizing the chiral response ?
Can we get the electrons to rotate but also recollide ?



1D trajectories, linear polarization, without potential



3D trajectories, circular polarization, with chiral potential

→ PECD

Bi-circular bichromatic fields

Bicircular fields :

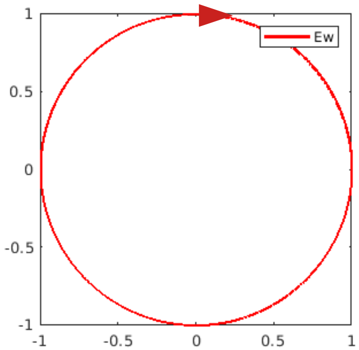
Superposition of fundamental and second harmonic fields

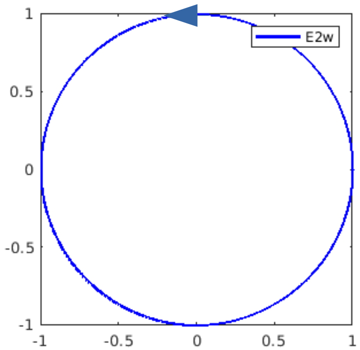
Intensity ratio r

Circularly polarized (helicity σ)

$$E_x = \sqrt{I_\omega}\sigma \left[\sin(\omega t) + \sqrt{r}\sin(2\omega t)\right]$$

$$E_y = \sqrt{I_\omega} \left[\cos(\omega t) + \sqrt{r}\cos(2\omega t)\right]$$





Bi-circular bichromatic fields

Bicircular fields :

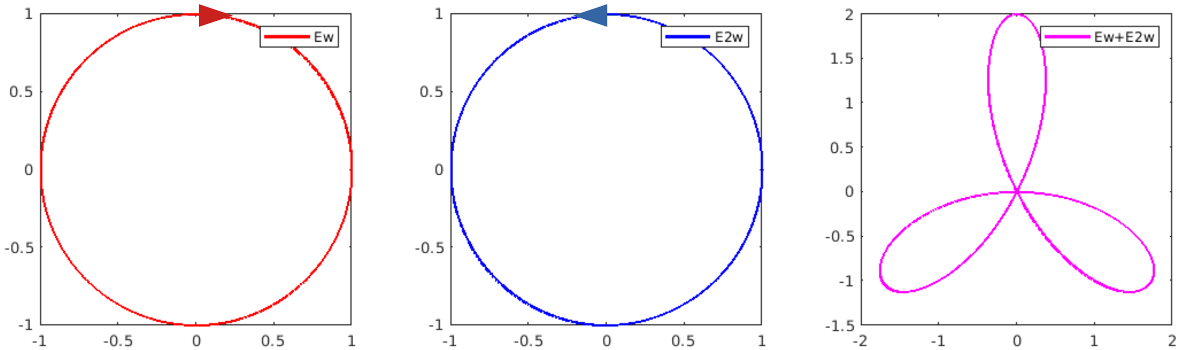
Superposition of fundamental and second harmonic fields

Intensity ratio r

Circularly polarized (helicity σ)

$$E_x = \sqrt{I_\omega}\sigma\left[\sin(\omega t) + \sqrt{r}\sin(2\omega t)\right]$$

$$E_y = \sqrt{I_\omega}\left[\cos(\omega t) + \sqrt{r}\cos(2\omega t)\right]$$

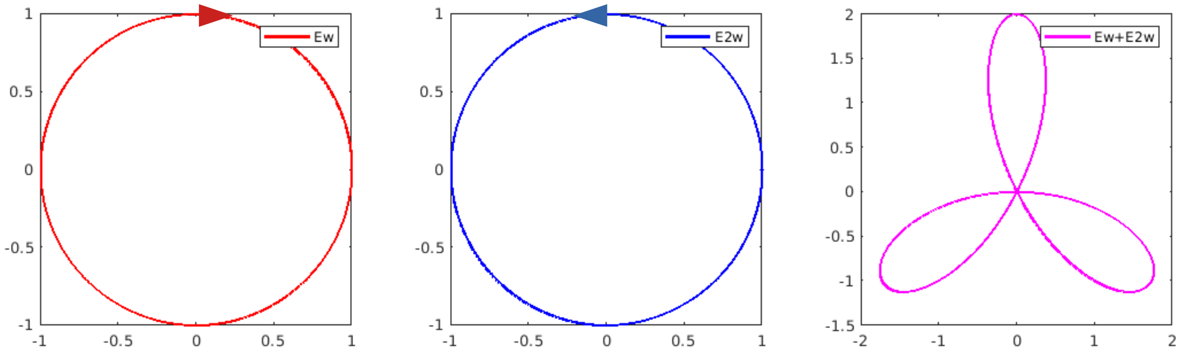


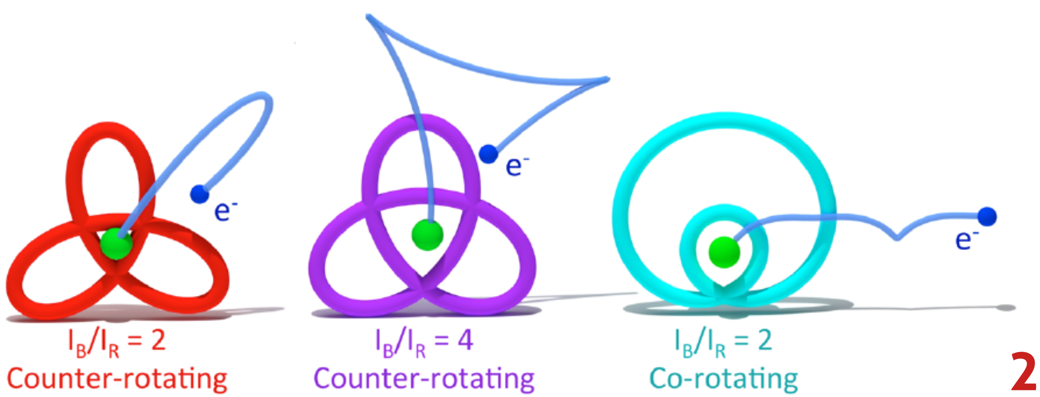
Bi-circular bichromatic fields

Bicircular fields :

Superposition of fundamental and second harmonic fields
Intensity ratio r
Circularly polarized (helicity σ)

$$E_x = \sqrt{I_\omega}\sigma\left[\sin(\omega t) + \sqrt{r}\sin(2\omega t)\right]$$
$$E_y = \sqrt{I_\omega}\left[\cos(\omega t) + \sqrt{r}\cos(2\omega t)\right]$$





2D manipulation of the electron trajectories

PHYSICAL REVIEW A **93**, 053406 (2016)

Controlling electron-ion rescattering in two-color circularly polarized femtosecond laser fields

Christopher A. Mancuso,[1,*] Daniel D. Hickstein,[1] Kevin M. Dorney,[1] Jennifer L. Ellis,[1] Elvedin Hasović,[2] Ronny Knut,[1] Patrik Grychtol,[1] Christian Gentry,[1] Maithreyi Gopalakrishnan,[1] Dmitriy Zusin,[1] Franklin J. Dollar,[1,†] Xiao-Min Tong,[3] Dejan B. Milošević,[2,4,5] Wilhelm Becker,[5] Henry C. Kapteyn,[1] and Margaret M. Murnane[1]

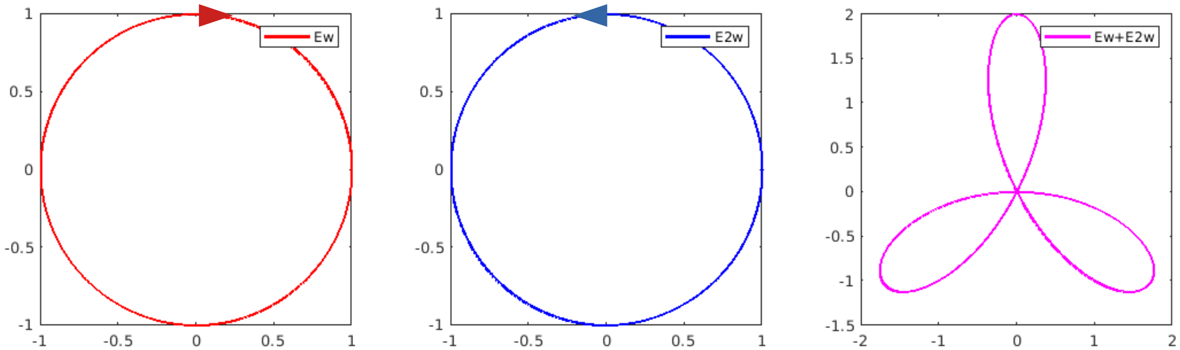
Bi-circular bichromatic fields

Bicircular fields :

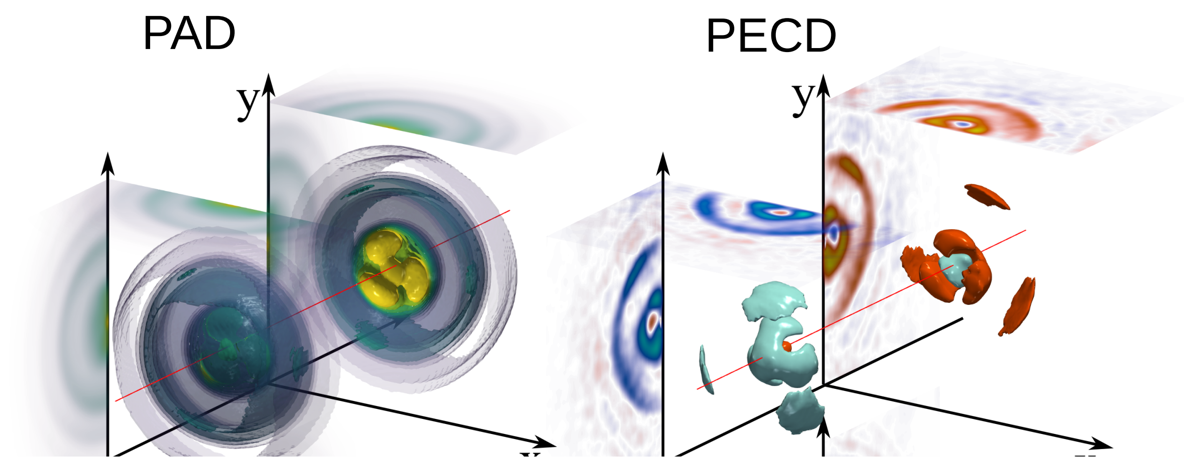
Superposition of fundamental and second harmonic fields
Intensity ratio r
Circularly polarized (helicity σ)

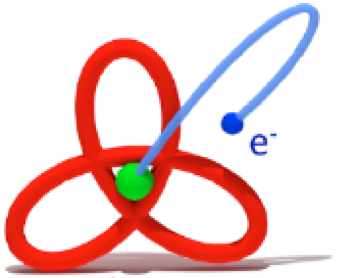
$$E_x = \sqrt{I_\omega}\sigma\left[\sin(\omega t) + \sqrt{r}\sin(2\omega t)\right]$$

$$E_y = \sqrt{I_\omega}\left[\cos(\omega t) + \sqrt{r}\cos(2\omega t)\right]$$



Chiral photoionization with clover field :



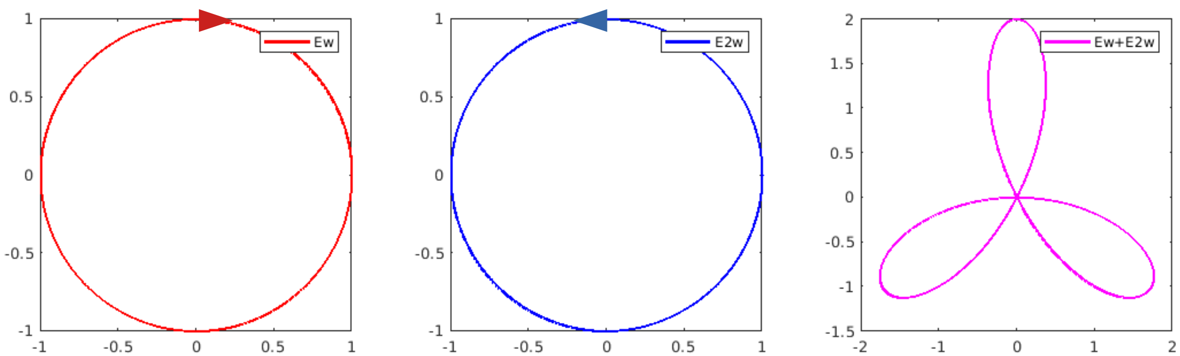


Bi-circular bichromatic fields

Bicircular fields :

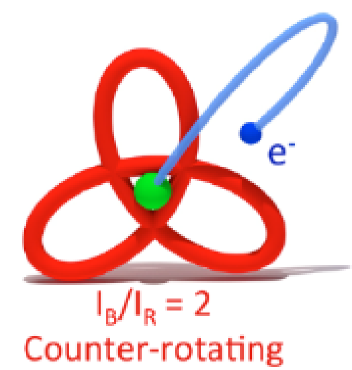
Superposition of fundamental and second harmonic fields
Intensity ratio r
Circularly polarized (helicity σ)

$$E_x = \sqrt{I_\omega}\sigma\left[\sin(\omega t) + \sqrt{r}\sin(2\omega t)\right]$$
$$E_y = \sqrt{I_\omega}\left[\cos(\omega t) + \sqrt{r}\cos(2\omega t)\right]$$



Chiral high-harmonic generation with clover field :

Combine circular polarization and highly probable re-collision
→ Great tool for chiral high-harmonic generation



IOP Publishing Journal of Physics B: Atomic, Molecular and Optical Physics
J. Phys. B: At. Mol. Opt. Phys. 48 (2015) 234005 (10pp) doi:10.1088/0953-4075/48/23/234005

Opportunities for chiral discrimination using high harmonic generation in tailored laser fields

Olga Smirnova[1], Yann Mairesse[2] and Serguei Patchkovskii[1]

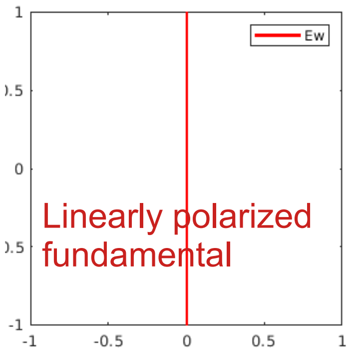
[1] Max-Born Institute for Nonlinear Optics and Short Pulse Spectroscopy, Max-Born-Strasse 2A, D-12489 Berlin, Germany
[2] CELIA. Université de Bordeaux—CNRS—CEA. F33405 Talence. France

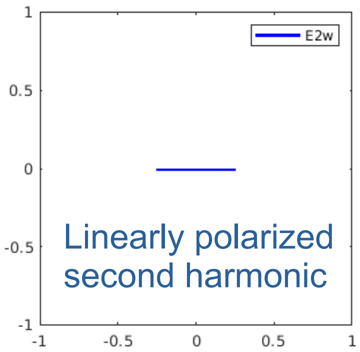
PHYSICAL REVIEW X 8, 031060 (2018)

Chiral Discrimination through Bielliptical High-Harmonic Spectroscopy

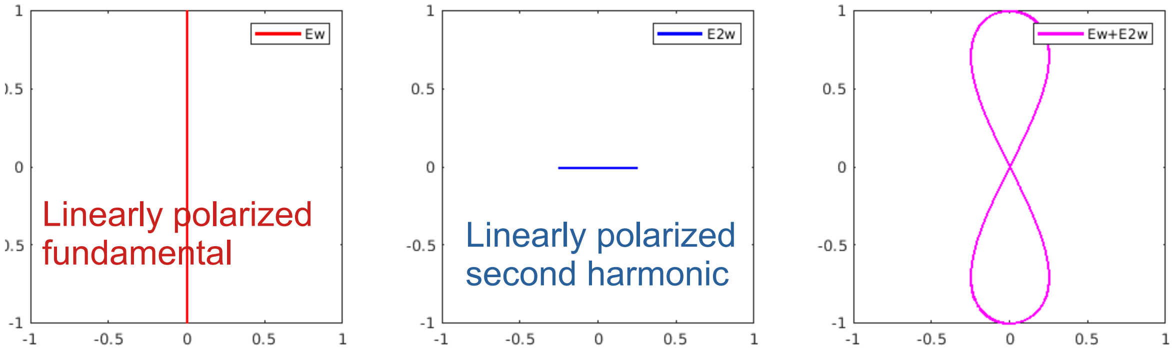
Denitsa Baykusheva and Hans Jakob Wörner*
ETH Zürich, Laboratory of Physical Chemistry, 8039 Zurich, Switzerland

Bi-linear bichromatic fields





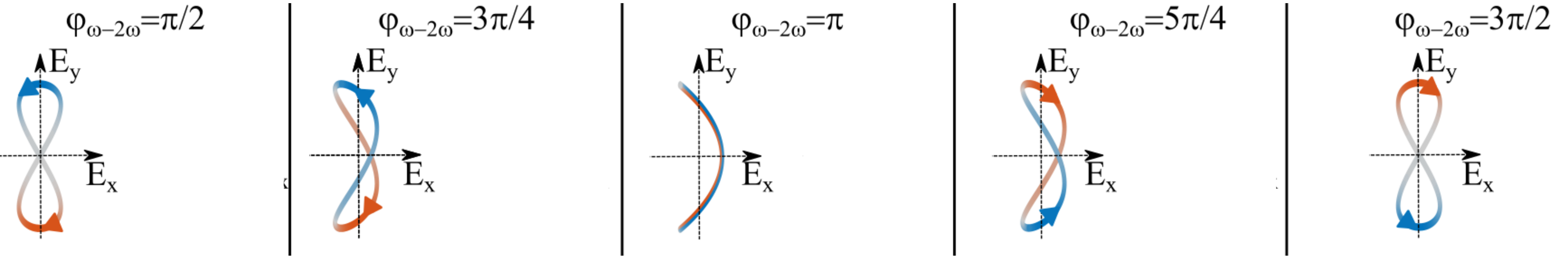
Bi-linear bichromatic fields



The field rotation direction reverses every half cycle

Shape depends on relative phase between ω and 2ω components

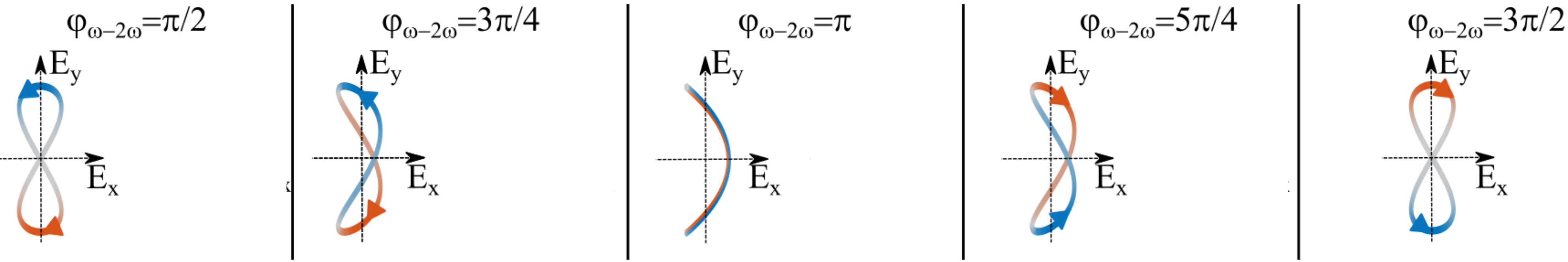
Chirality in a w-2w bilinear laser field ?



The field rotation direction reverses every half cycle

Shape depends on relative phase between ω and 2ω components

Chirality in a w-2w bilinear laser field ?



The field rotation direction reverses every half cycle
Shape depends on relative phase between ω and 2ω components

Can a field with zero net chirality produce a chiroptical signal ?

Sub-cycle chirality

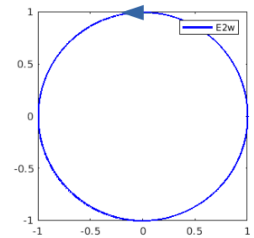
PHYSICAL REVIEW X **9,** 031004 (2019)

Featured in Physics

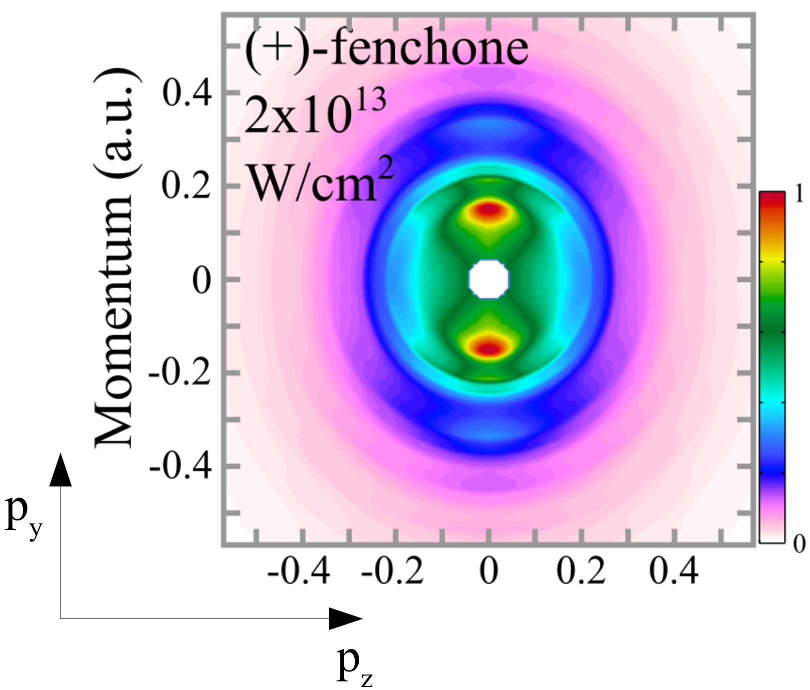
Controlling Subcycle Optical Chirality in the Photoionization of Chiral Molecules

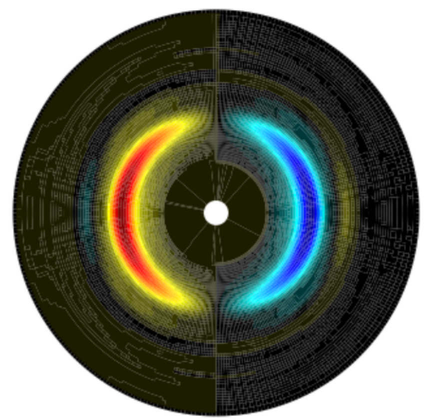
S. Rozen,[1,*] A. Comby,[2,*] E. Bloch,[2] S. Beauvarlet,[2] D. Descamps,[2] B. Fabre,[2] S. Petit,[2]
V. Blanchet,[2] B. Pons,[2] N. Dudovich,[1] and Y. Mairesse[2]

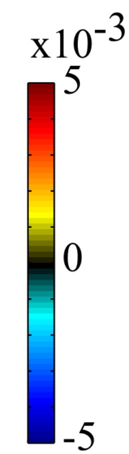
[1]*Weizmann Institute of Science, Rehovot, 76100, Israel*
[2]*Université de Bordeaux, CNRS, CEA, CELIA, UMR5107, F33405 Talence, France*



Circular polarization







Sub-cycle chirality

PHYSICAL REVIEW X 9, 031004 (2019)

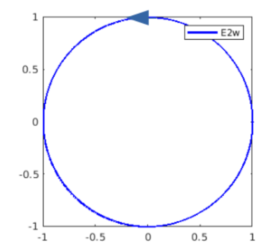
Featured in Physics

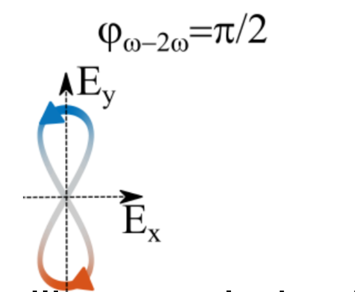
Controlling Subcycle Optical Chirality in the Photoionization of Chiral Molecules

S. Rozen,[1,*] A. Comby,[2,*] E. Bloch,[2] S. Beauvarlet,[2] D. Descamps,[2] B. Fabre,[2] S. Petit,[2] V. Blanchet,[2] B. Pons,[2] N. Dudovich,[1] and Y. Mairesse[2]

[1]*Weizmann Institute of Science, Rehovot, 76100, Israel*

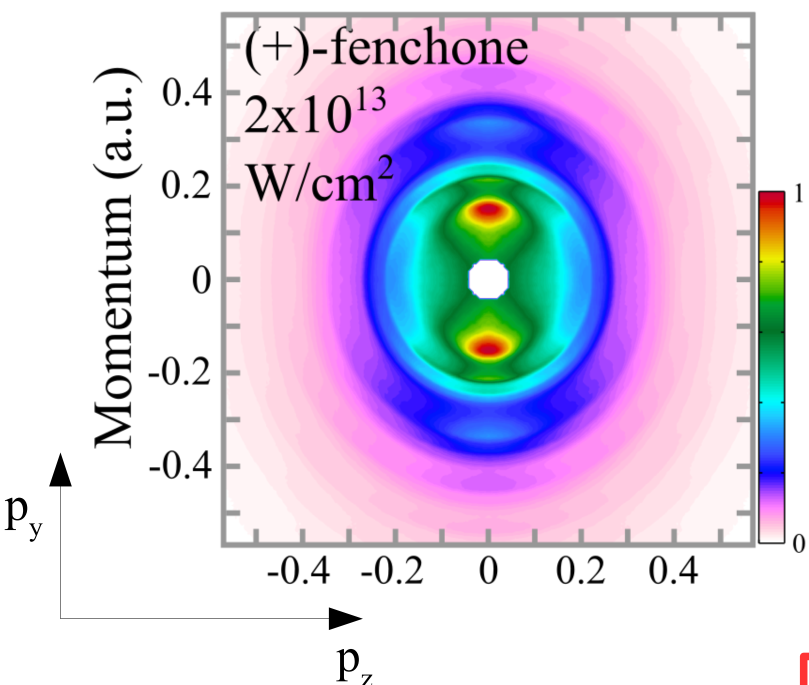
[2]*Université de Bordeaux, CNRS, CEA, CELIA, UMR5107, F33405 Talence, France*

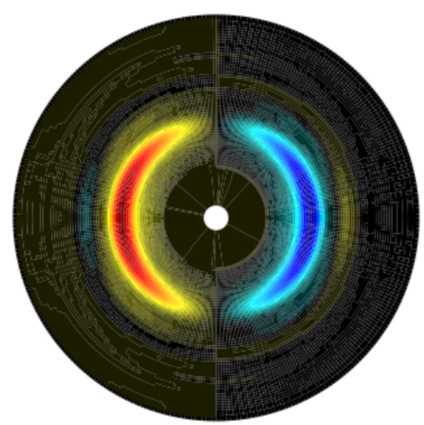


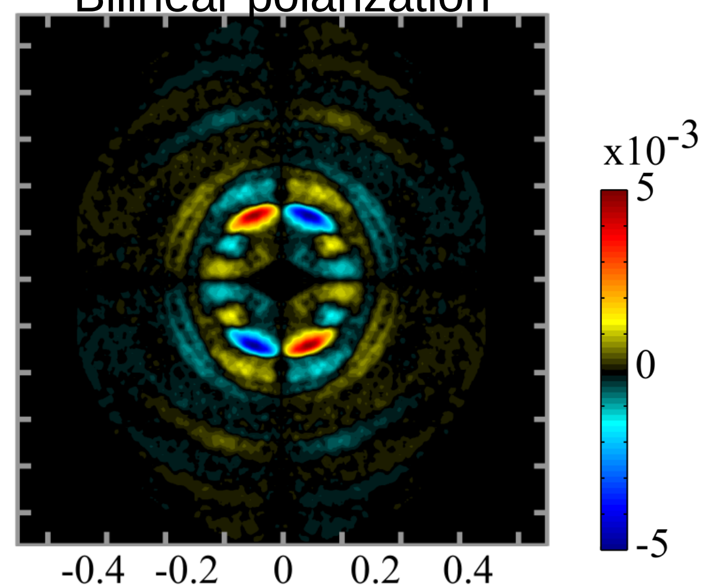


Circular polarization

Bilinear polarization

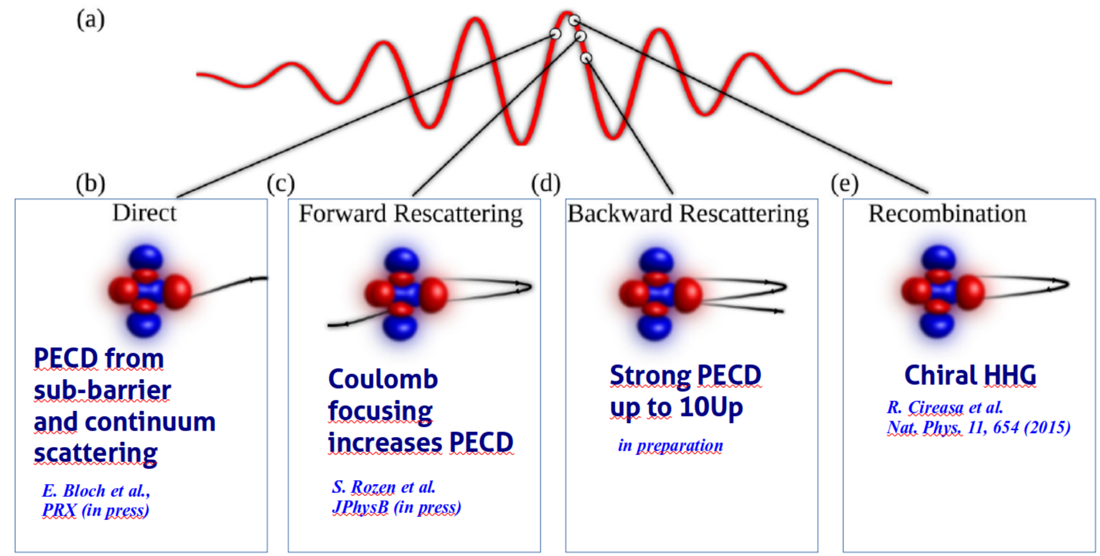






Clear up/down antisymetric chiroptical signal
Signature of sub-cycle chiral response

Conclusion : the strong-field chiral toolbox



+ nonsequential double ionization
Strong CD
J.L Bégin et al., Sci. Reports 10:14074 (2020)

Processes

attosecond electron dynamics

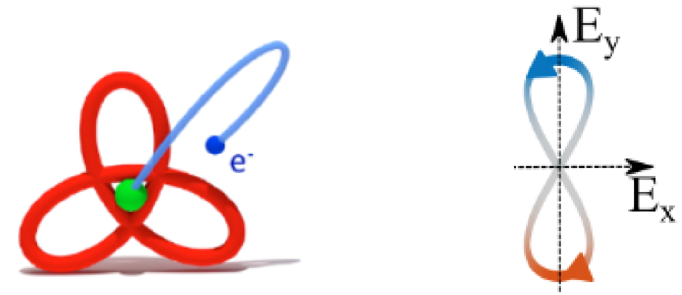
+spin-orbit interaction

molecular dynamics in pump-probe spectroscopy

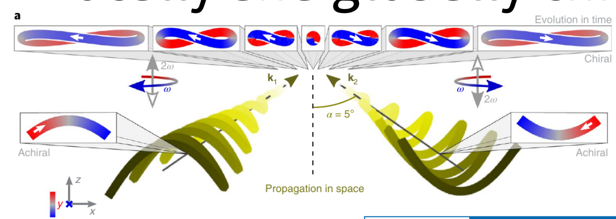
complex chiral systems
– attempts on beetles (with CEMES)
chiral Pt (with ISM)

Targets

Electron trajectory manipulation
Control of optical chirality



+Locally and globally chiral light



nature photonics
LETTERS
https://doi.org/10.1038/s41566-019-0531

Synthetic chiral light for efficient control of chiral light-matter interaction

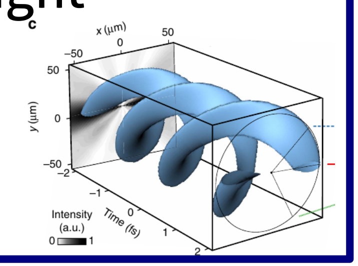
David Ayuso[1,7]*, Ofer Neufeld[2,7], Andres F. Ordonez[1,3], Piero Decleva[4], Gavriel Lerner[2], Oren Cohen[2], Misha Ivanov[1,5,6]* and Olga Smirnova[1,3]*

+Orbital angular momentum in strong field or XUV light

ARTICLE
Received 13 Apr 2016 | Accepted 13 Jul 2016 | Published 30 Aug 2016 DOI: 10.1038/ncomms12583 OPEN

Synthesis and characterization of attosecond light vortices in the extreme ultraviolet

R. Géneaux[1], A. Camper[2], T. Auguste[1], O. Gobert[1], J. Caillat[3], R. Taïeb[3] & T. Ruchon[1]



Tools

CHIMERA

Partner	Name	First name
Bordeaux University CBMN	ODA	Reiko
	POUGET	Emilie
	NLATE	Sylvain
	PRANEE	Piyanan
	To be recruited	
Bordeaux University ISM	BASSANI	Dario
	BONHOMMEAU	Sébastien
	BUFFETEAU	Thierry
	DAUGEY	Nicolas
	DEL GUERZO	André
	RAFFY	Guillaume
	TALAGA	David
Bordeaux University CELIA	BLANCHET	Valérie
	DELAGNES	Jean-Christophe
	MAIRESSE	Yann
	to be recruited	
University of Lorraine LCP-A2MC	BATTIE	Yann
	BROCH	Laurent
	EN NACIRI	Aotmane
	to be recruited	

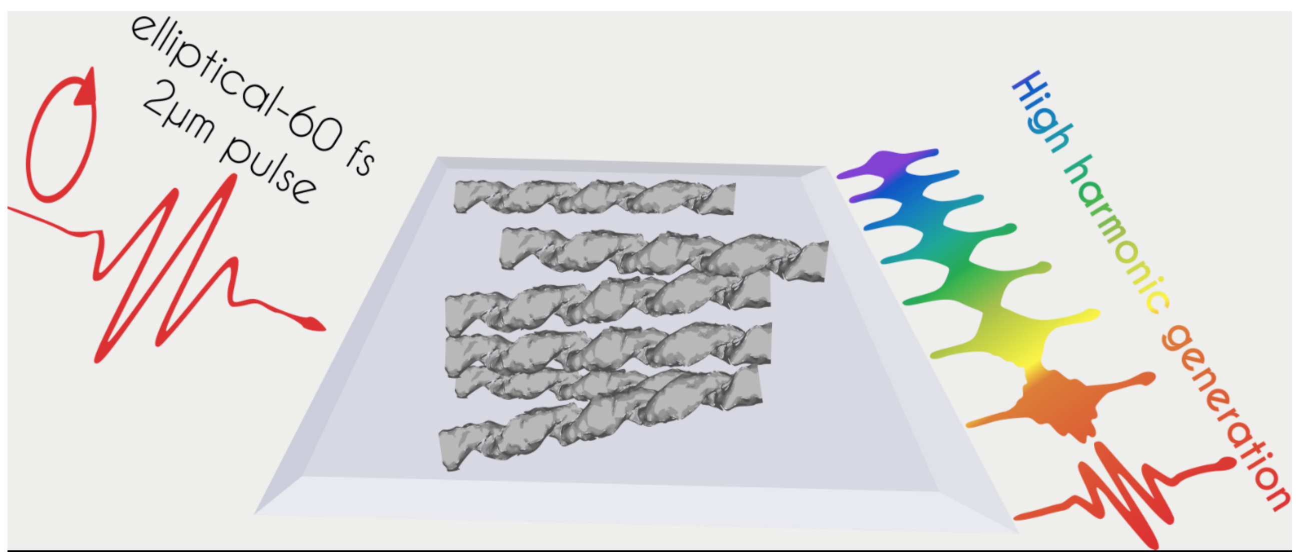
Chiral Induction from Microns to Electrons for Radiative Anisotropy ANR2021

Experiments @ CELIA:

- Coherently drive electrons with a strong field in chiral nanohelices
  - → chirosensitive high-harmonic generation
- use orbital angular momentum in the XUV range
  - → light helix with a pitch adjusted to the nanostructure

Compare to well established spectroscopies

Investigate the link from chiral nanoassembly down to the electron's response



Chirality at CELIA and Chirality in strong laser fields

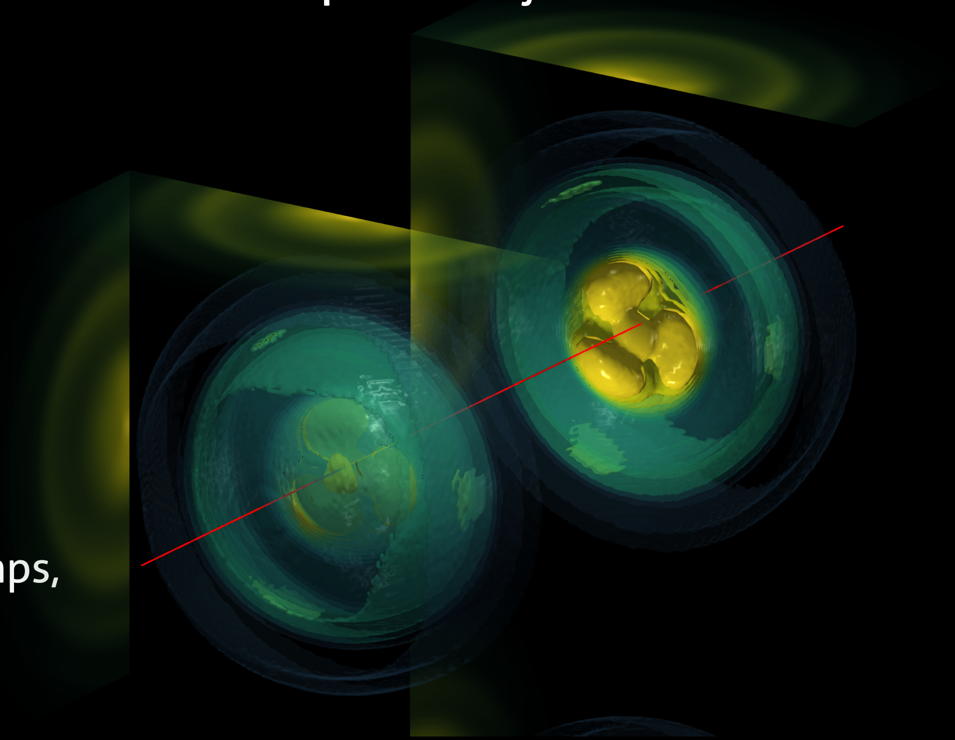
Yann Mairesse,
CELIA, Université de Bordeaux – CEA – CNRS
http://harmodyn.celia.u-bordeaux.fr

CELIA Bordeaux

Experiments :
Samuel Beaulieu,
Sandra Beauvarlet,
Etienne Bloch,
Antoine Comby,
Debobrata Rajak,
Valérie Blanchet,
Yann Mairesse

Laser Team :
Dominique Descamps,
Stéphane Petit

Theory :
Alex Clergerie,
Sylvain Larroque,
Baptiste Fabre,
Bernard Pons



Weizmann Rehovot
Shaked Rozen
Ayelet Julie Uzan
Nirit Dudovich

LCPMR Paris
Richard Taïeb

cnrs
université de BORDEAUX
cea
RÉGION Nouvelle-Aquitaine
erc
European Research Council
Established by the European Commission

This project has received funding from the European Research Council (ERC) under the European Union's Horizon 2020 research and innovation programme (grant agreement No 682978)