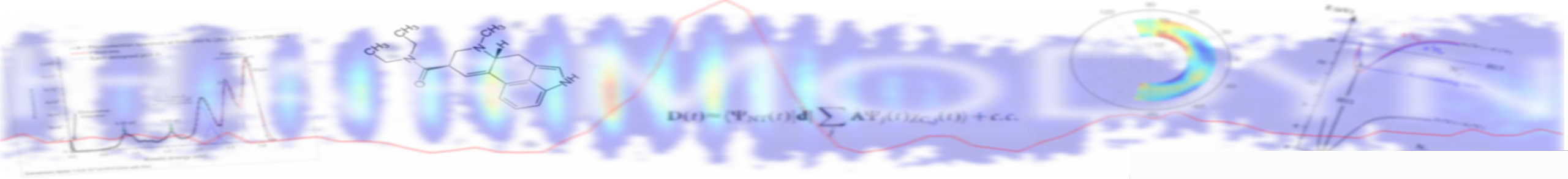


High-order Harmonic Generation and XUV Free Induction Decay From Electronic Wavepackets

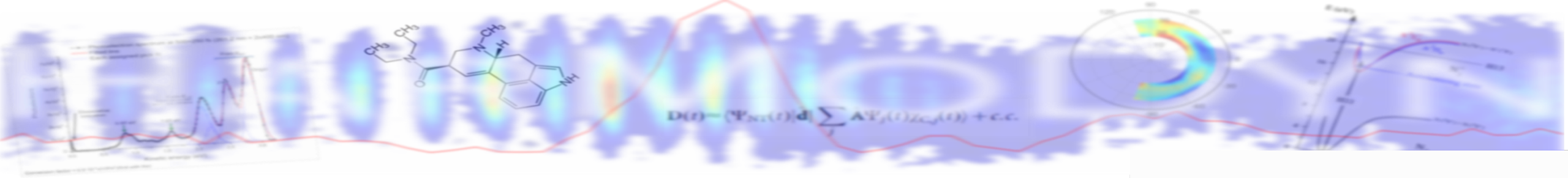
Samuel Beaulieu

INRS / Bordeaux University

INRS UNIVERSITÉ DE RECHERCHE

cnrs

CELIA



Outline

1. Genesis of HHG and Attosecond science ;

2. How we discovered a new HHG mechanism ;

3. XUV spectroscopy without using XUV pulses ;

4. Conclusion.

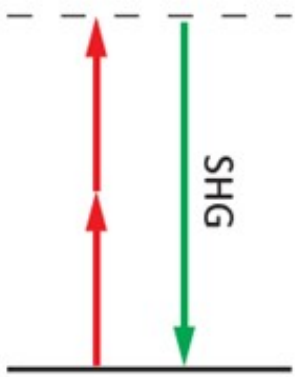
1961 : Perturbative Nonlinear Optics

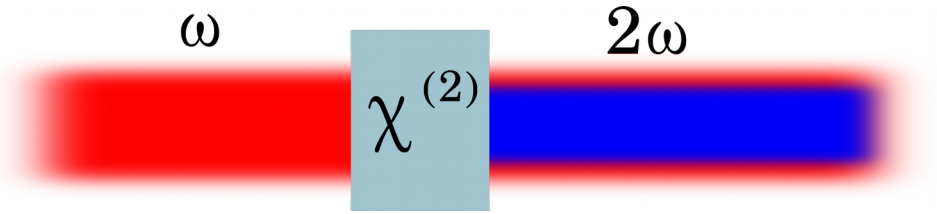
Featured in Physics | PRL Milestone | Free to Read

Generation of Optical Harmonics

P. A. Franken, A. E. Hill, C. W. Peters, and G. Weinreich
Phys. Rev. Lett. **7**, 118 – Published 15 August 1961

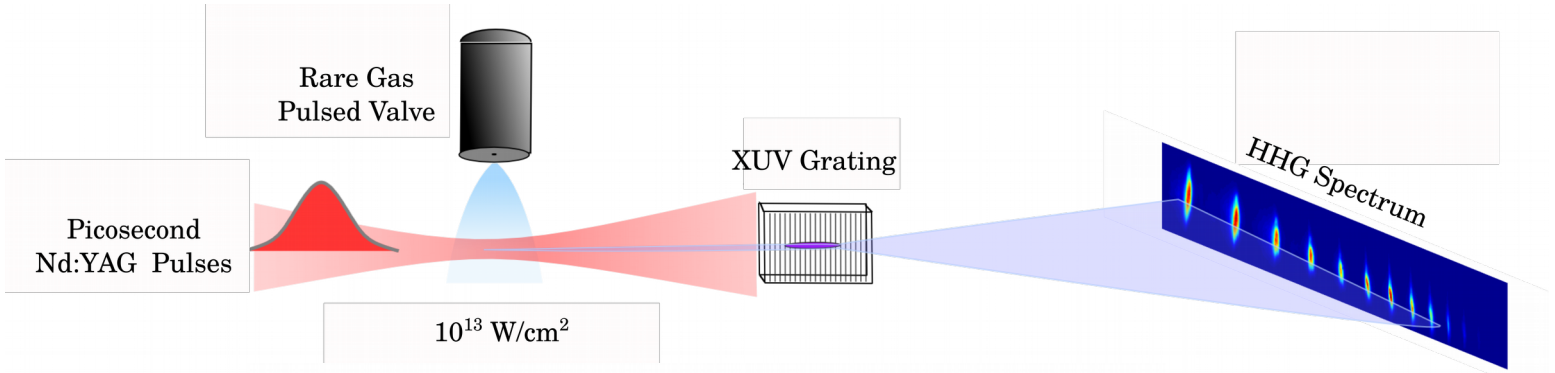
Physics See Focus story: *Landmarks*—Ruby Red Laser Light Becomes Ultraviolet

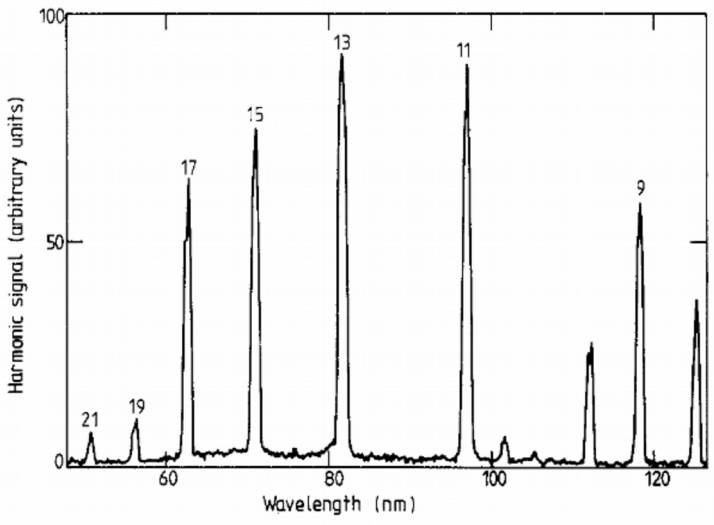




$\sim 10^9$ W/cm^2

1988 : First Observation of High-order Harmonic

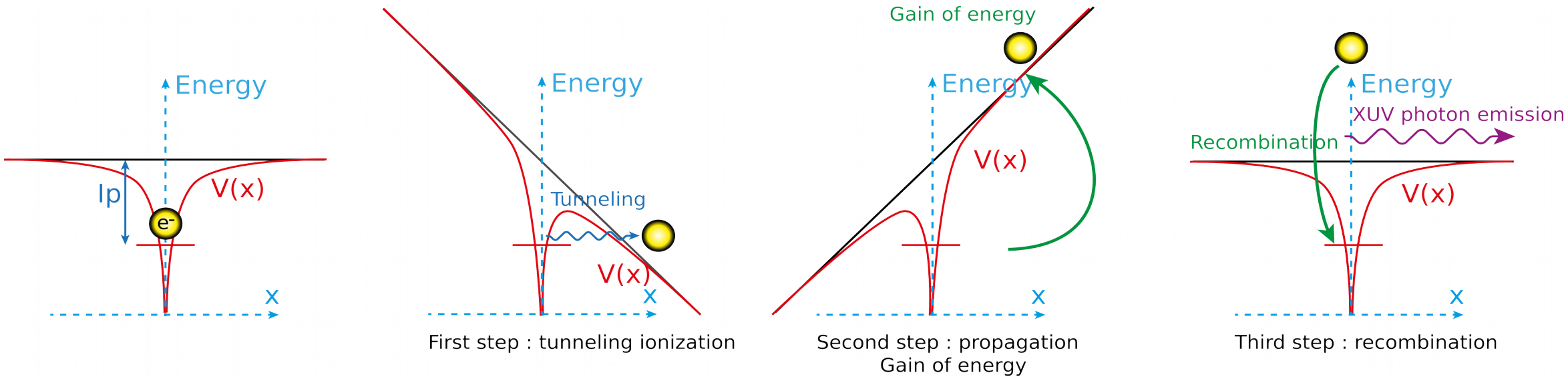




Nonperturbative NLO :
up to the 21th harmonics

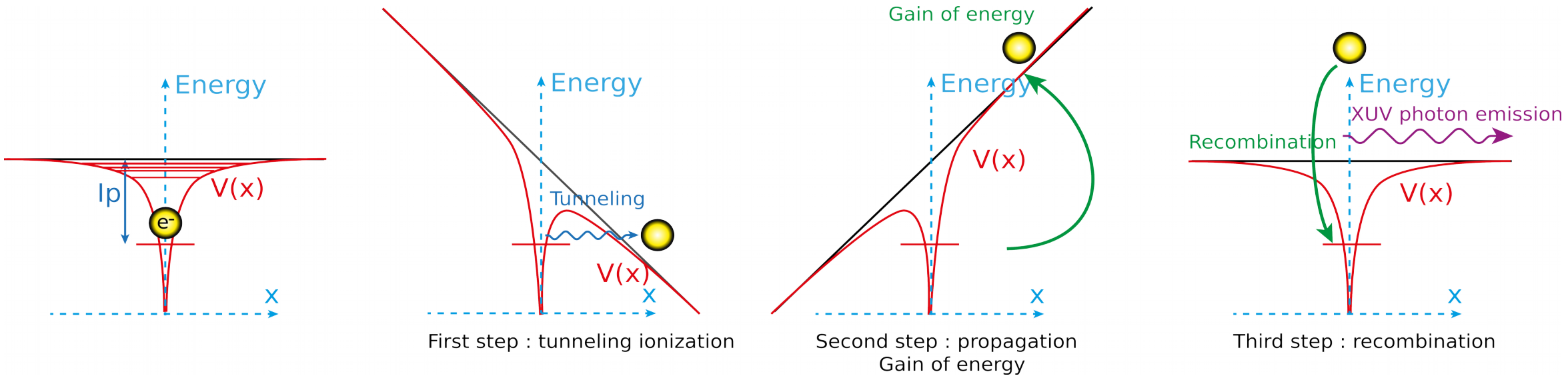
Lompray *et al.*, J. Phys. B : At. Mol. Opt. Phys. **21**, L31-L35 (1998)

Early 90s : The Three Steps Model (Paul Corkum, NRC Ottawa)



M. Yu. Kuchiev, JETP Lett. **45**, 404 (1987) ; P.B. Corkum PRL **71**, 1994 (1993) ; K.J. Schafer et al., PRL **70**, 1599 (1993)

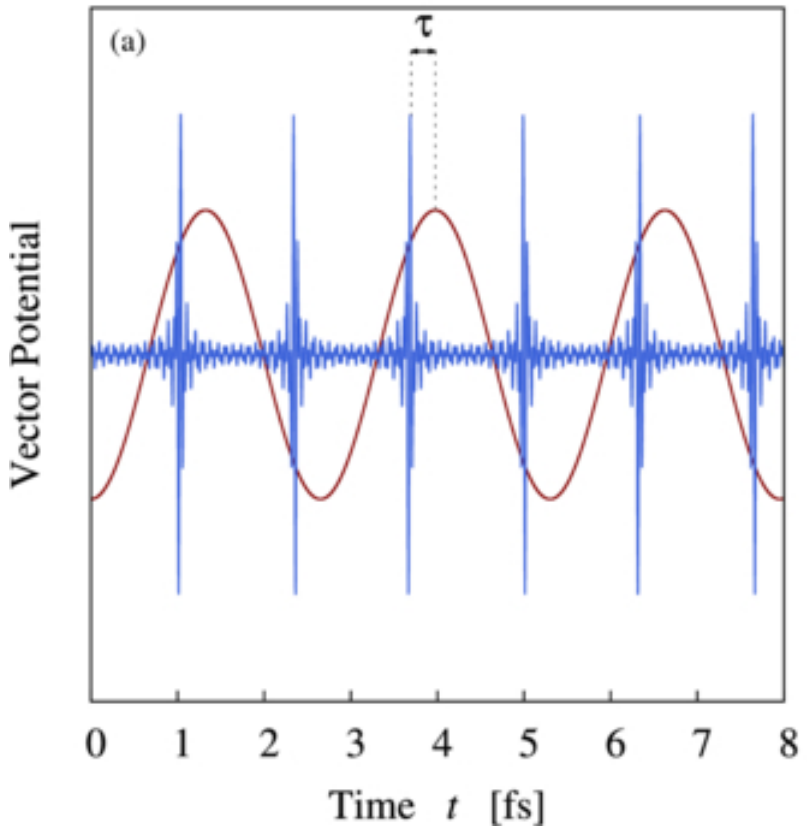
Early 90s : The Three Steps Model (Paul Corkum, NRC Ottawa)

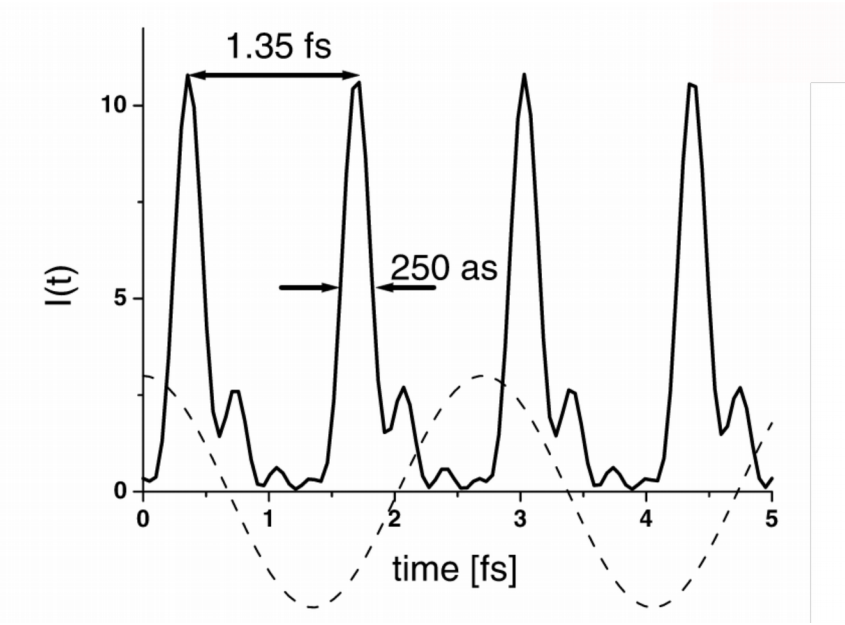


These three steps occur within a fraction of the driving laser cycle :
Attosecond (10^{-18} s) confinement of the emission

M. Yu. Kuchiev, JETP Lett. **45**, 404 (1987) ; P.B. Corkum PRL **71**, 1994 (1993) ; K.J. Schafer et al., PRL **70**, 1599 (1993)

2001 : Attosecond duration of HHG





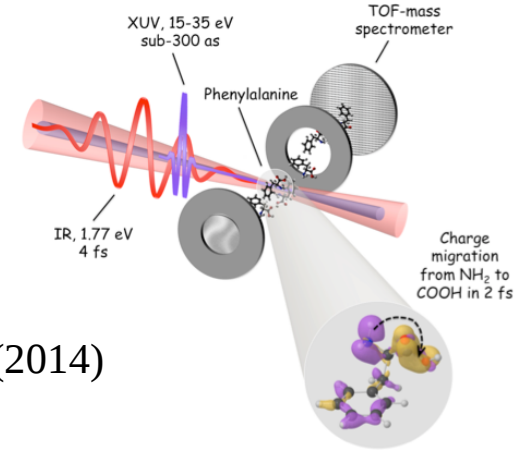
RABBIT : Reconstruction of Attosecond Beating By Interference of Two-Photon Transitions

Paul *et al.*, Science **292**, 1689-1692 (2001) ; Mairesse *et al.*, Science **302**, 1540-1543 (2003)

Trends in Attosecond Science

HHG as a light source

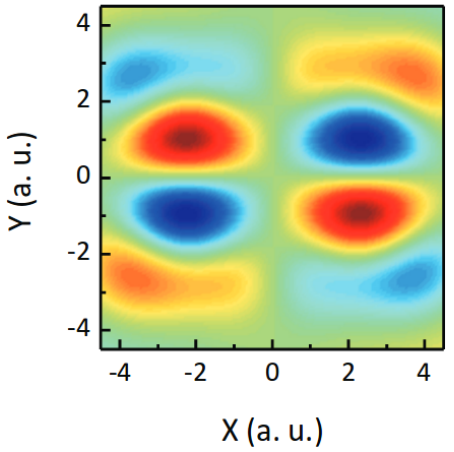
Sub-5 fs charge migration in amino acids



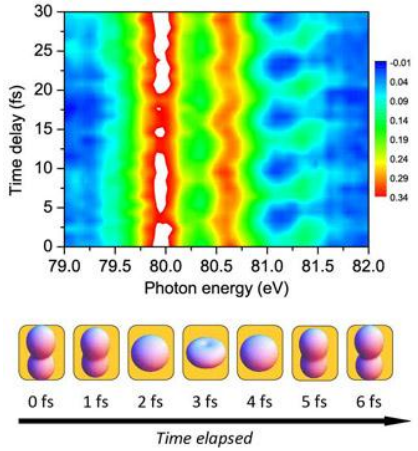
Calegari *et al.*, Science **346,** 336-339 (2014)

High Harmonic Spectroscopy

Tomographic reconstuction of molecular orbital



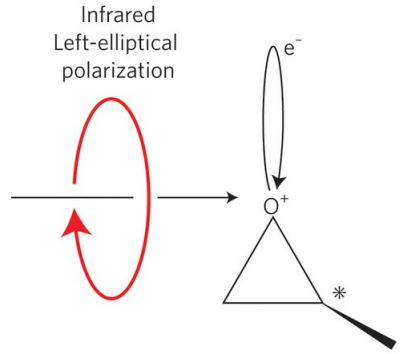
Vozzi *et al.*, Nature Physics **7**, 822–826 (2011)



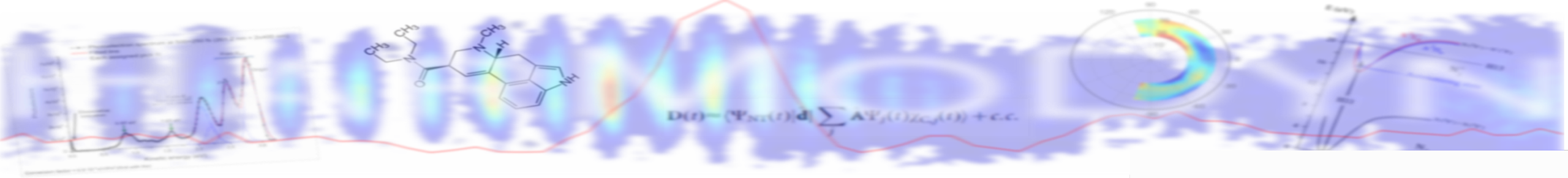
Coherent Hole dynamics using Attosecond Transient Absorption

Goulielmakis *et al.*, Nature **466**, 739–743 (2010)

Probing chirality on sub-fs time scale



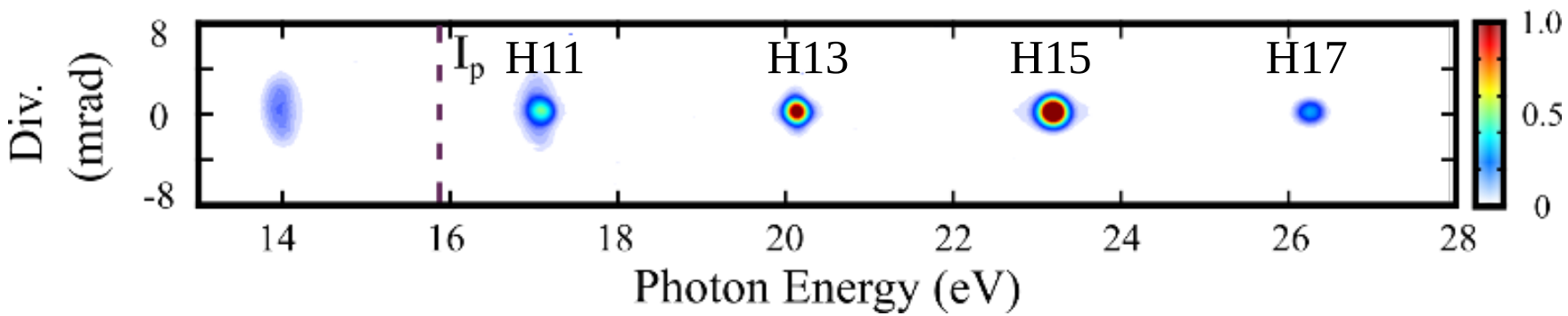
Cireasa *et al.*, Nature Phys. **11**, 654 (2015)

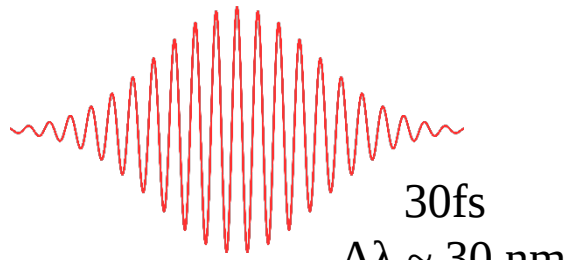


Trends in Attosecond Science

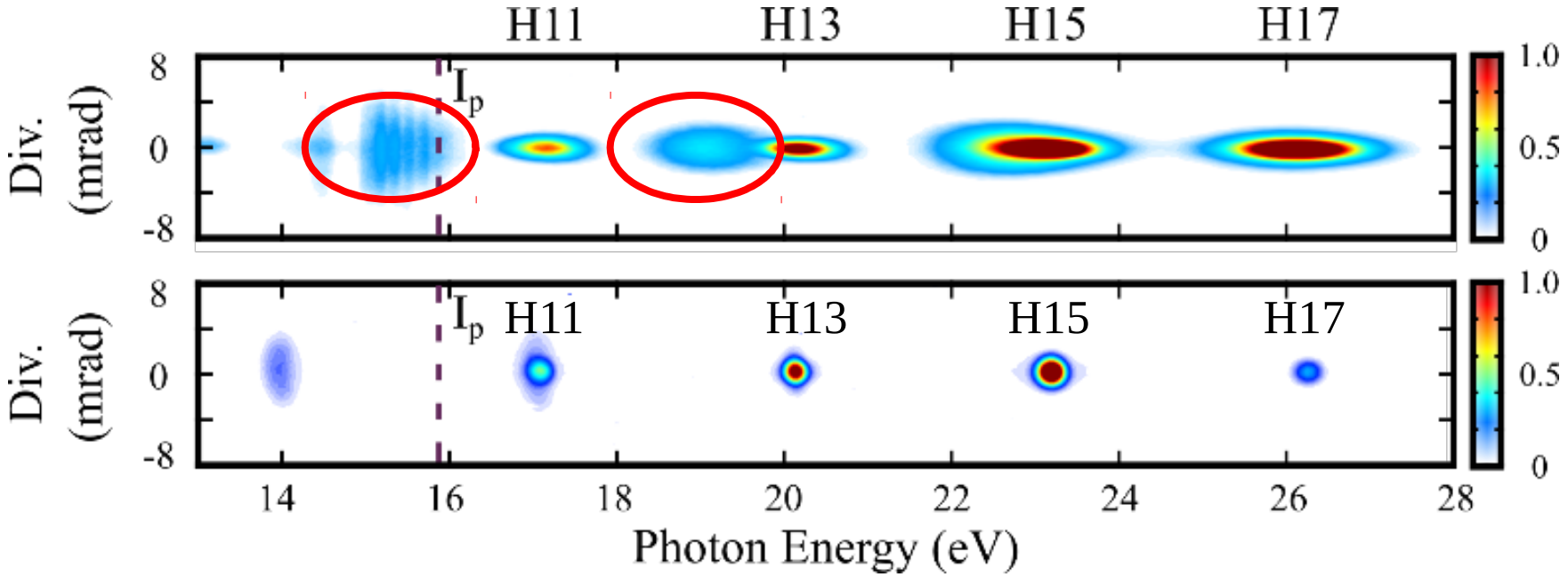
Our contribution : *(accidental)* discovery of a new HHG mecanism !

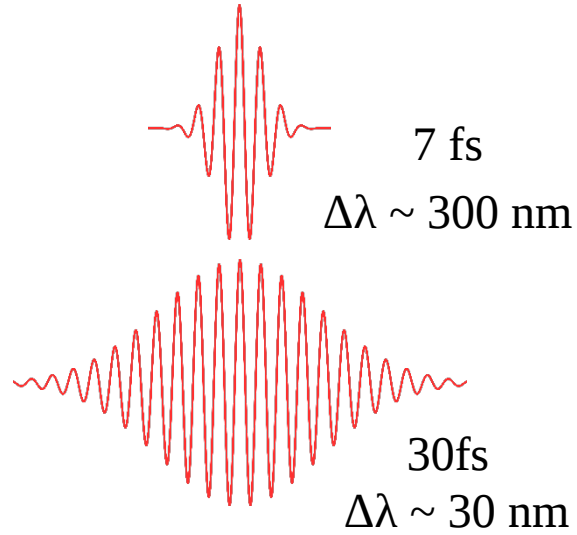
High Harmonic Generation in Ar atoms





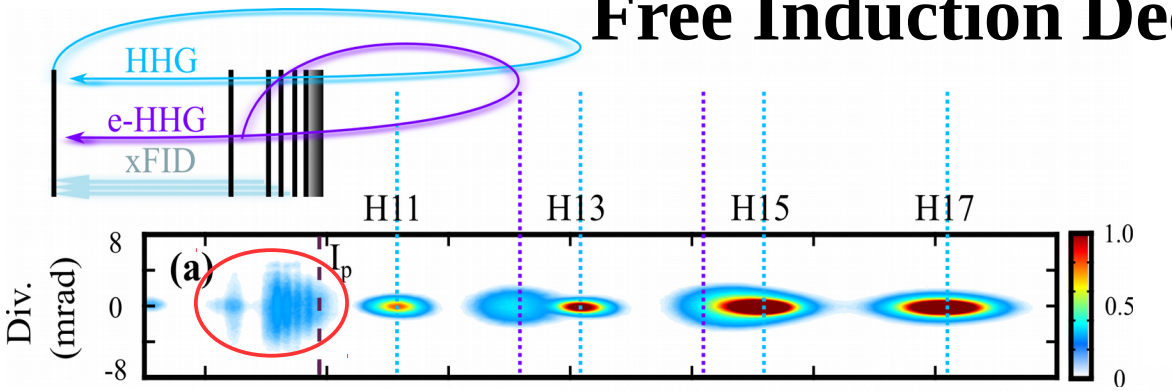
High Harmonic Generation in Ar atoms

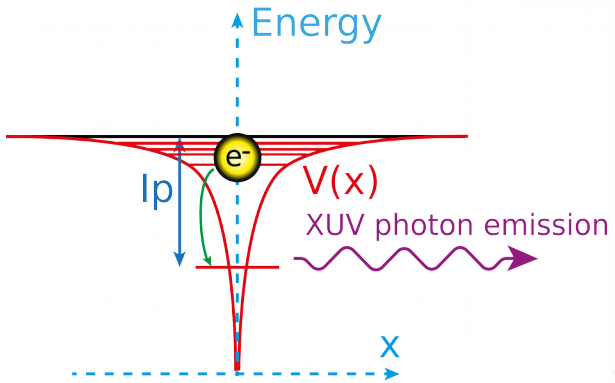




Beaulieu *et al.*, Physical Review Letters **117**, 203001 (2016)

Free Induction Decay from Rydberg States

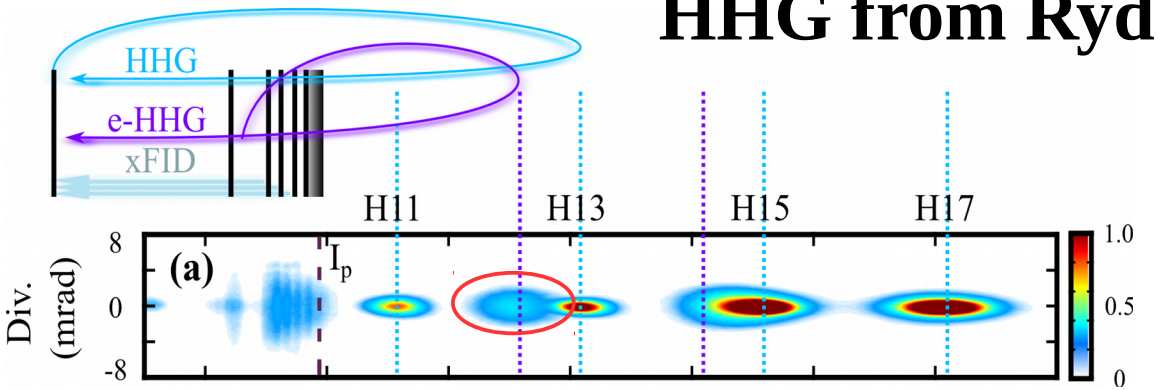




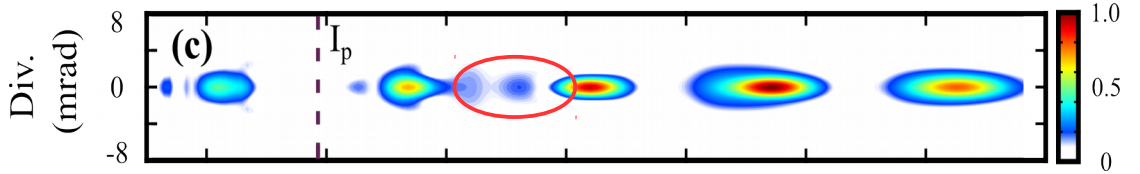
Coherent emission of XUV light by the electronic wavepacket, after the laser pulse.

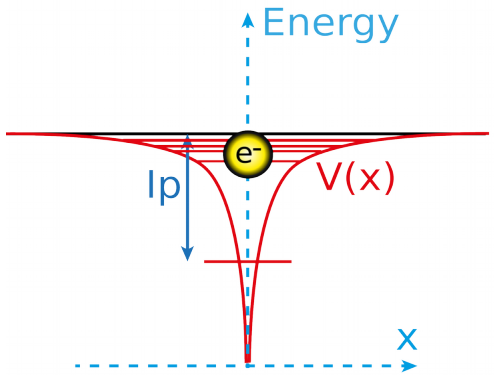
Beaulieu *et al.*, Physical Review Letters **117**, 203001 (2016)

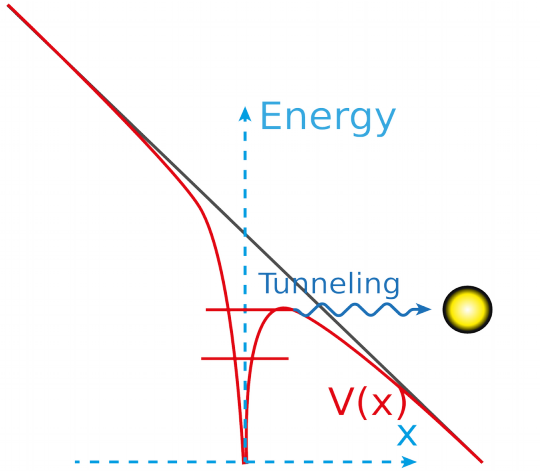
HHG from Rydberg States (e-HHG)



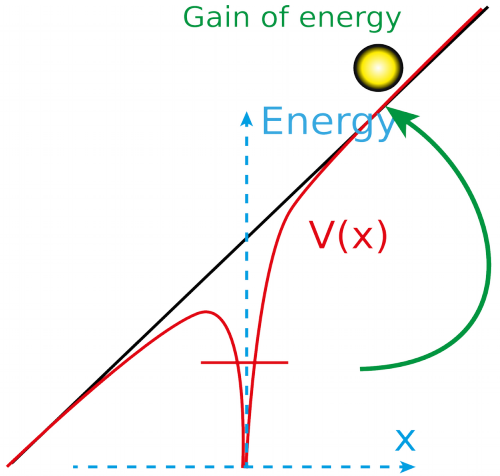
TDSE



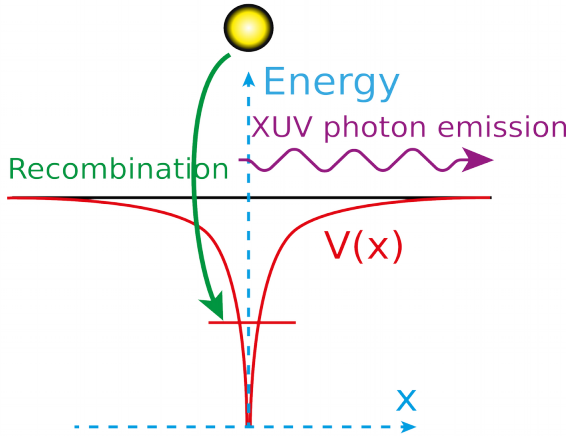




First step : tunneling ionization



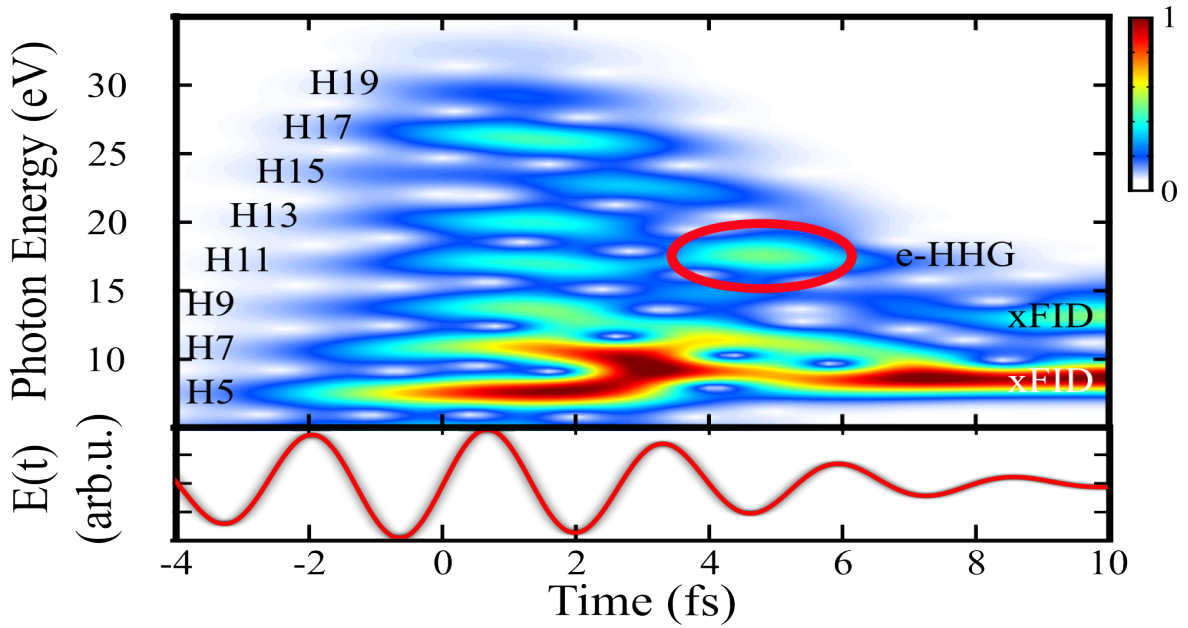
Second step : propagation
Gain of energy



Third step : recombination

Beaulieu *et al.*, Physical Review Letters **117**, 203001 (2016)

HHG from Rydberg States

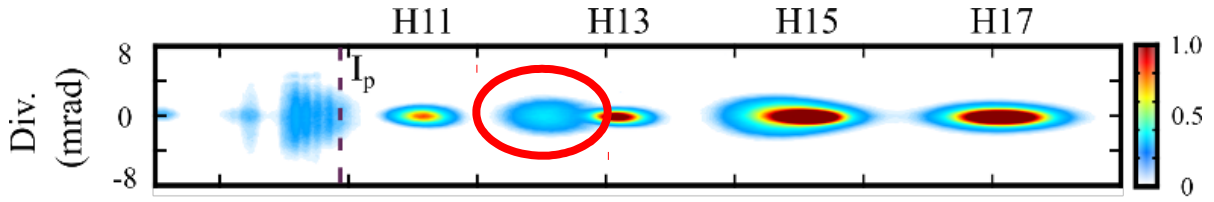


Gabor Analysis of the TDSE : e-HHG is delayed by 4 fs compared to the non-resonant HHG.

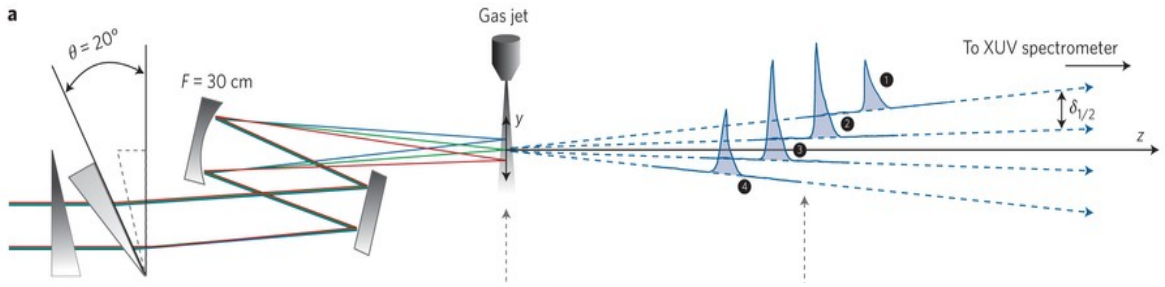
Beaulieu *et al.*, Physical Review Letters **117**, 203001 (2016)

Temporal resolution of the emission of e-HHG

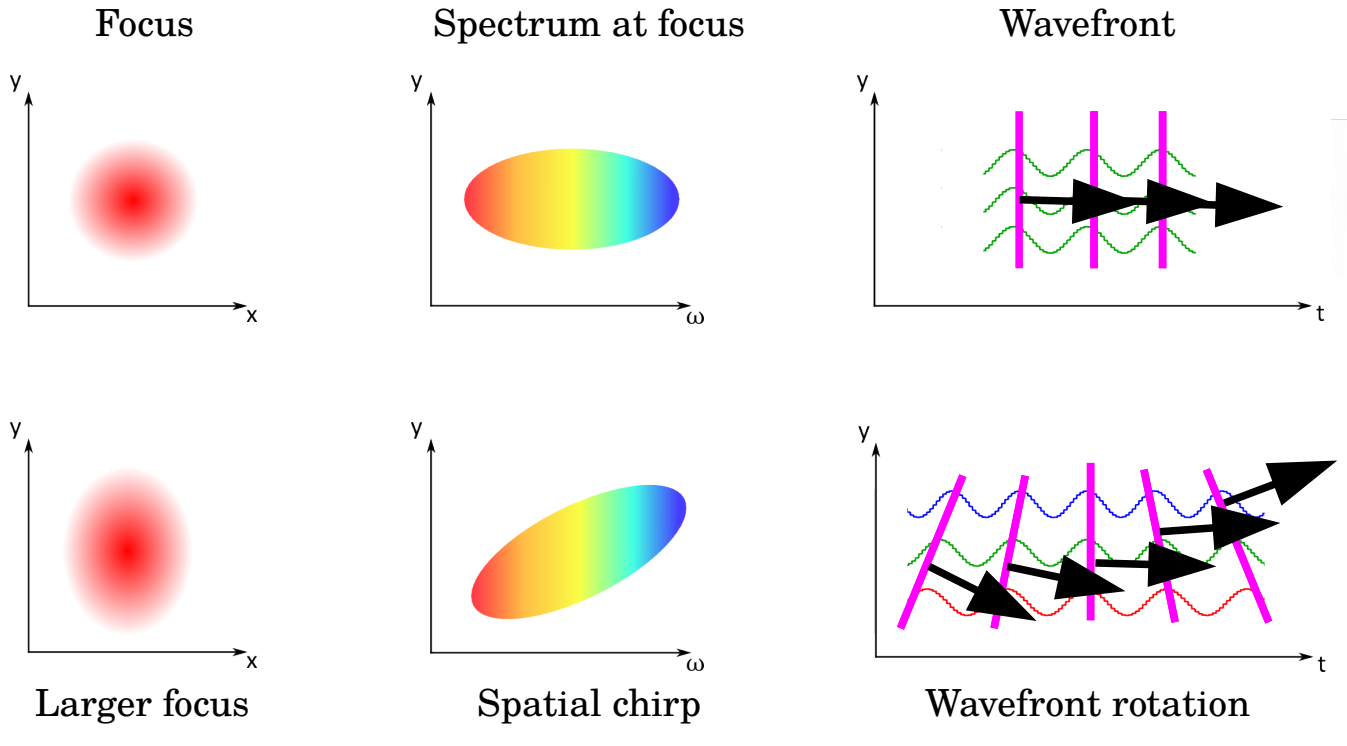
e-HHG (sub-5 fs)

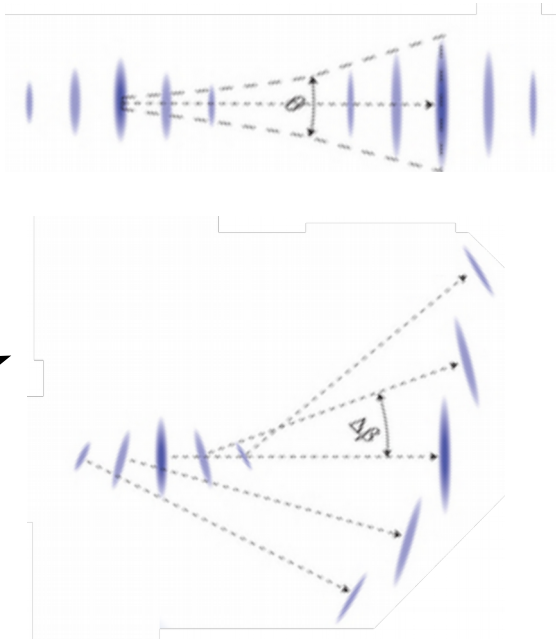


Attosecond Lighthouse



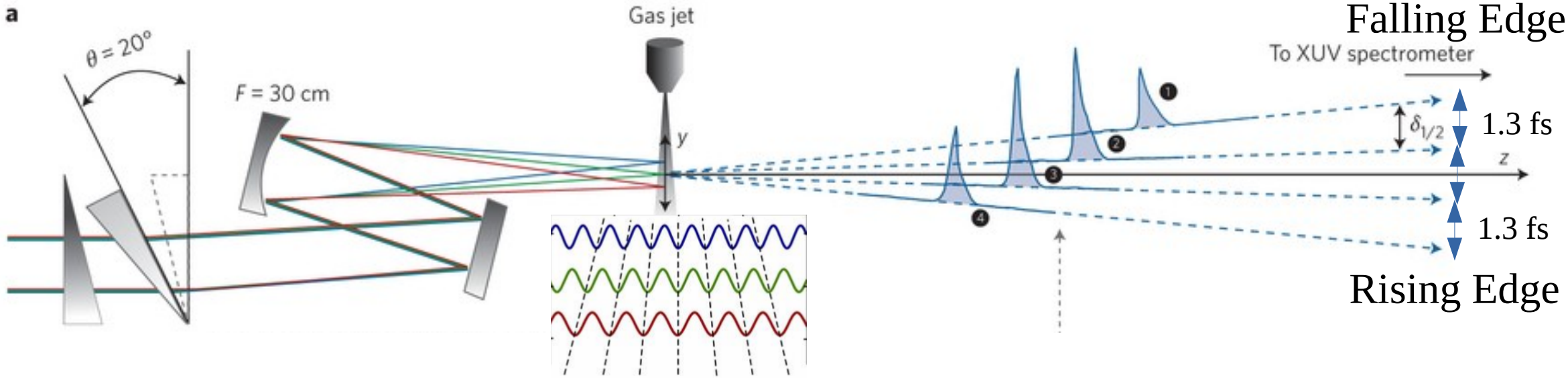
The Attosecond Lighthouse : Ultrafast wavefront rotation by focusing a spatially chirp pulse





Vincenti *et al.*, Phys. Rev. Lett. **108,** 113904 (2012) ; Kim *et al.*, Nature photonics **7**, 651-656 (2013)

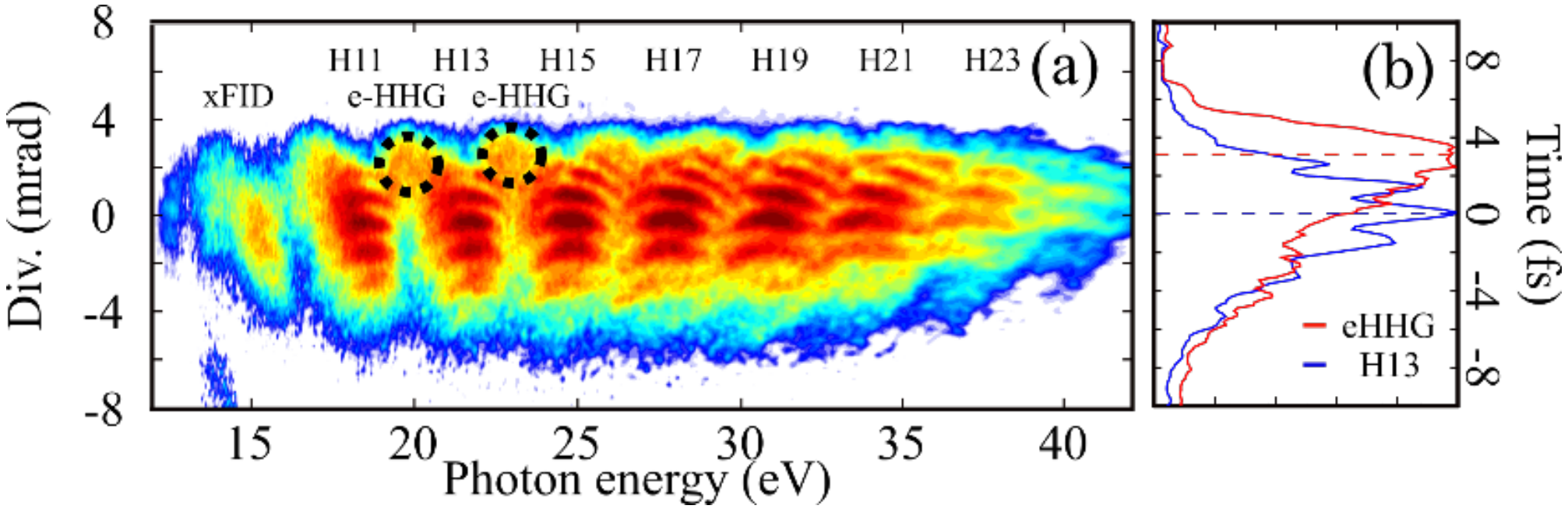
The Attosecond Lighthouse :
Ultrafast wavefront rotation by focusing a spatially chirp pulse



A simple way to isole an attosecond pulse from a pulse train

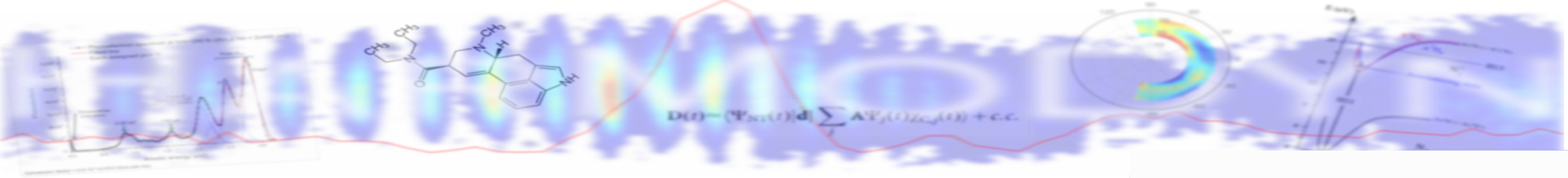
Vincenti *et al.*, Phys. Rev. Lett. **108,** 113904 (2012) ; Kim *et al.*, Nature photonics **7**, 651-656 (2013)

Attosecond Lighthouse



Single-shot measurement of the spectrum (at 1 kHz) – circumvent the needs of CEP stability

Beaulieu *et al.*, Physical Review Letters **117**, 203001 (2016)

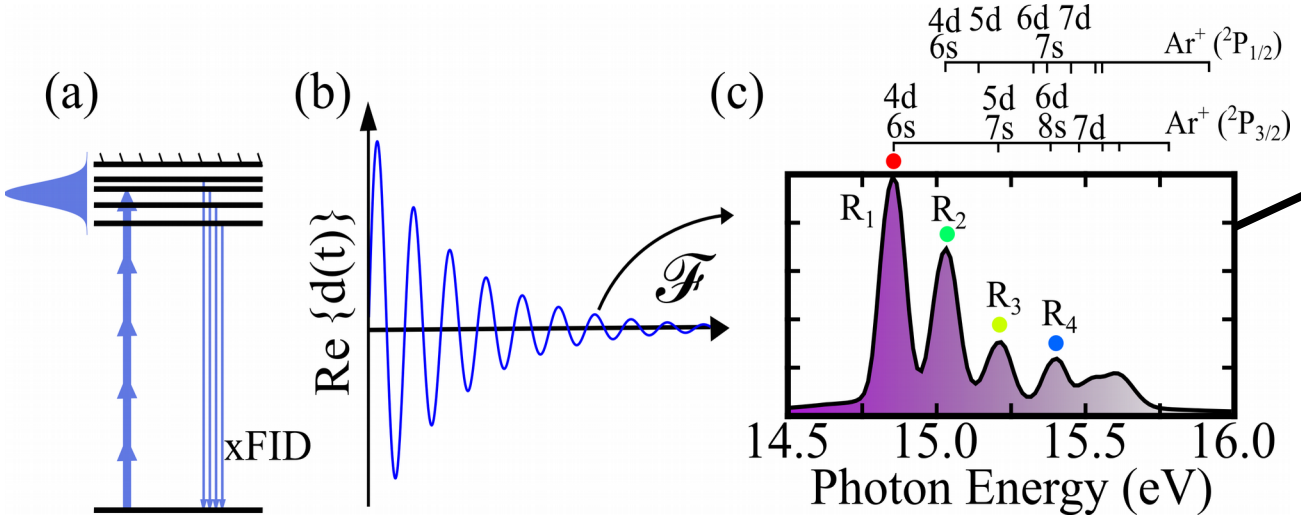


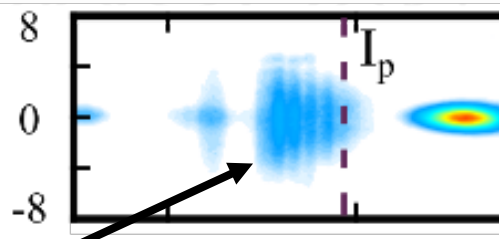
Partial Conclusion

New HHG mechanism from excited states !

Delayed in time by few femtoseconds.

XUV-Free Induction Decay (xFID)





Beaulieu *et al.*, Phys. Rev. A 95, 041401(R) (2017)

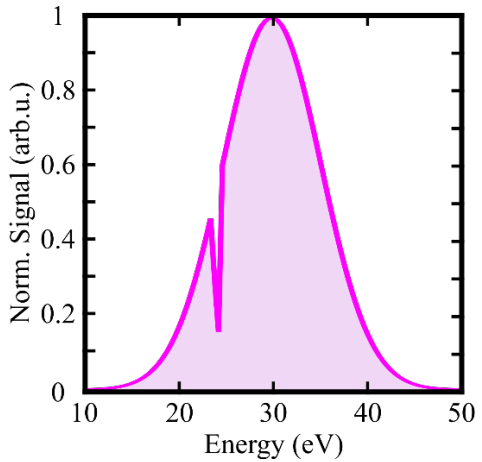
xFID vs Absorption Spectroscopy

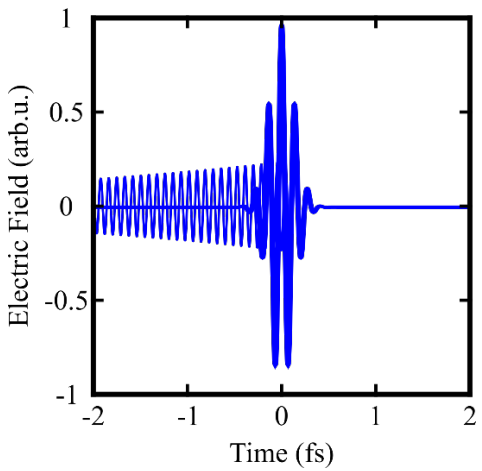
TAS

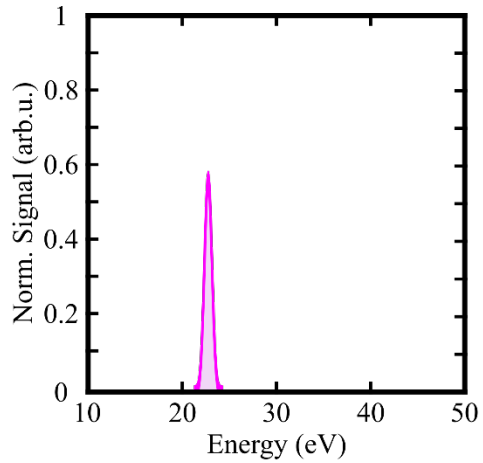
- Interference between dipole emission and incoming light

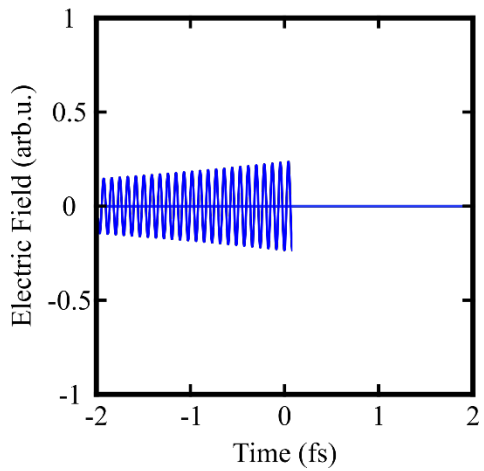
- Poor S/N ratio

- Phase encoded in the (Lorentz-Fano) Lineshape.







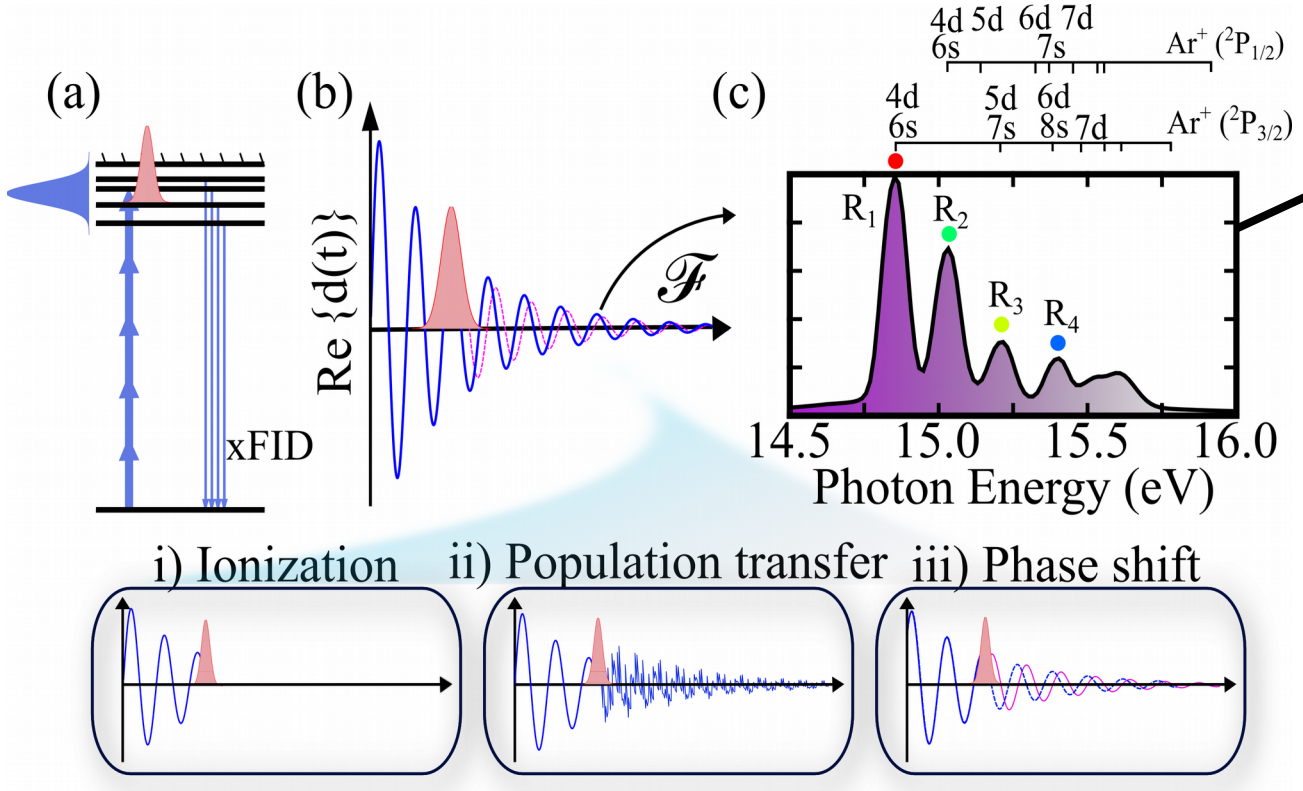


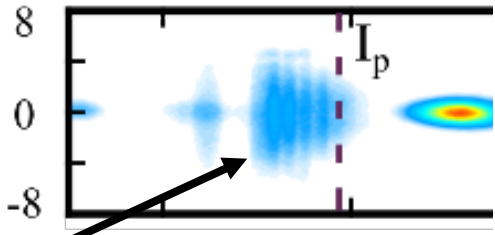
xFID

- Zero background

- Did we lost the phase information ?

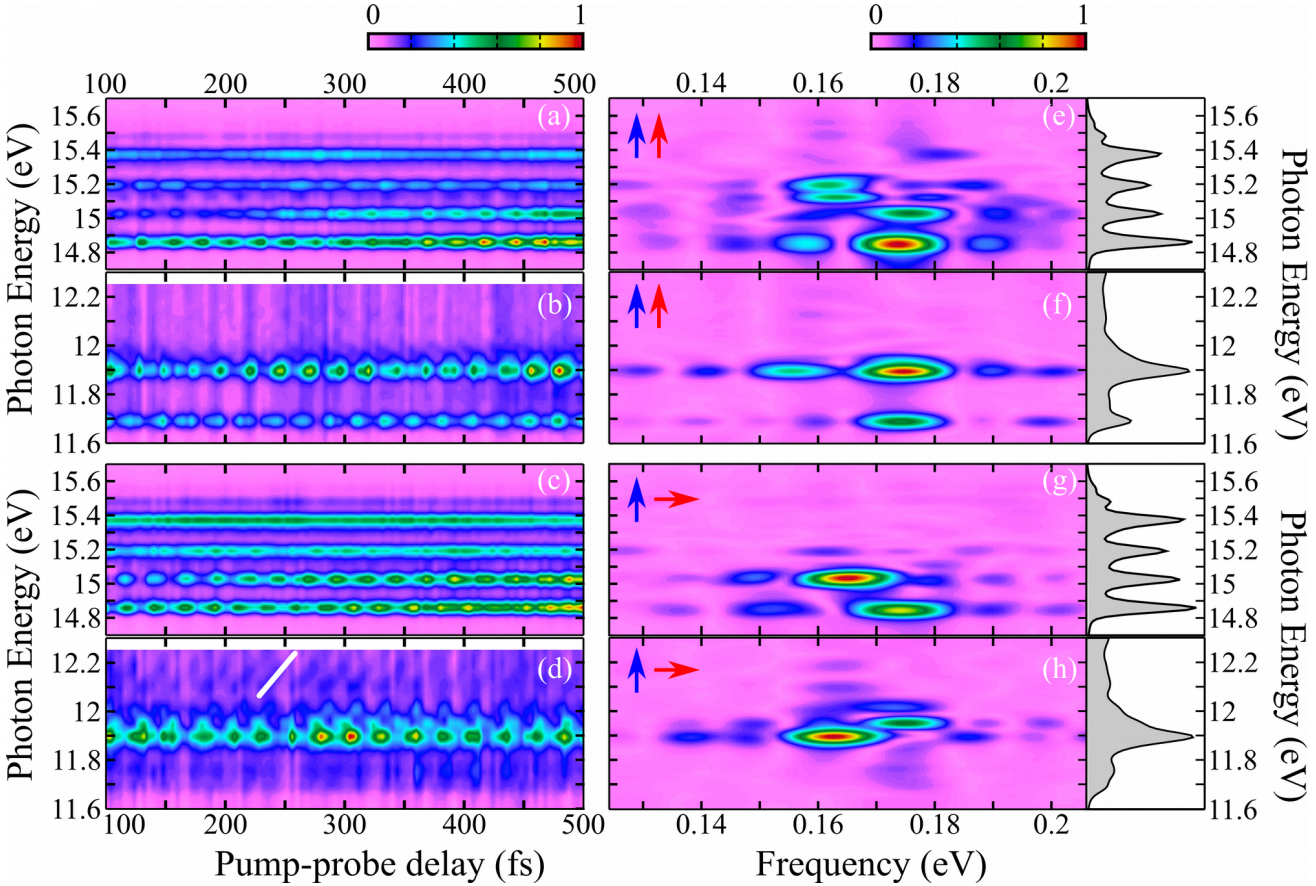
Time-Resolved xFID

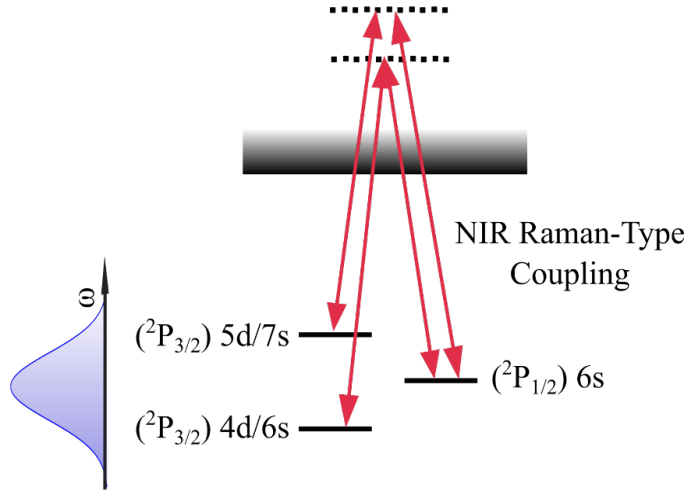




Beaulieu *et al.*, Phys. Rev. A 95, 041401(R) (2017)

Ramsey-type Spectroscopy of the xFID

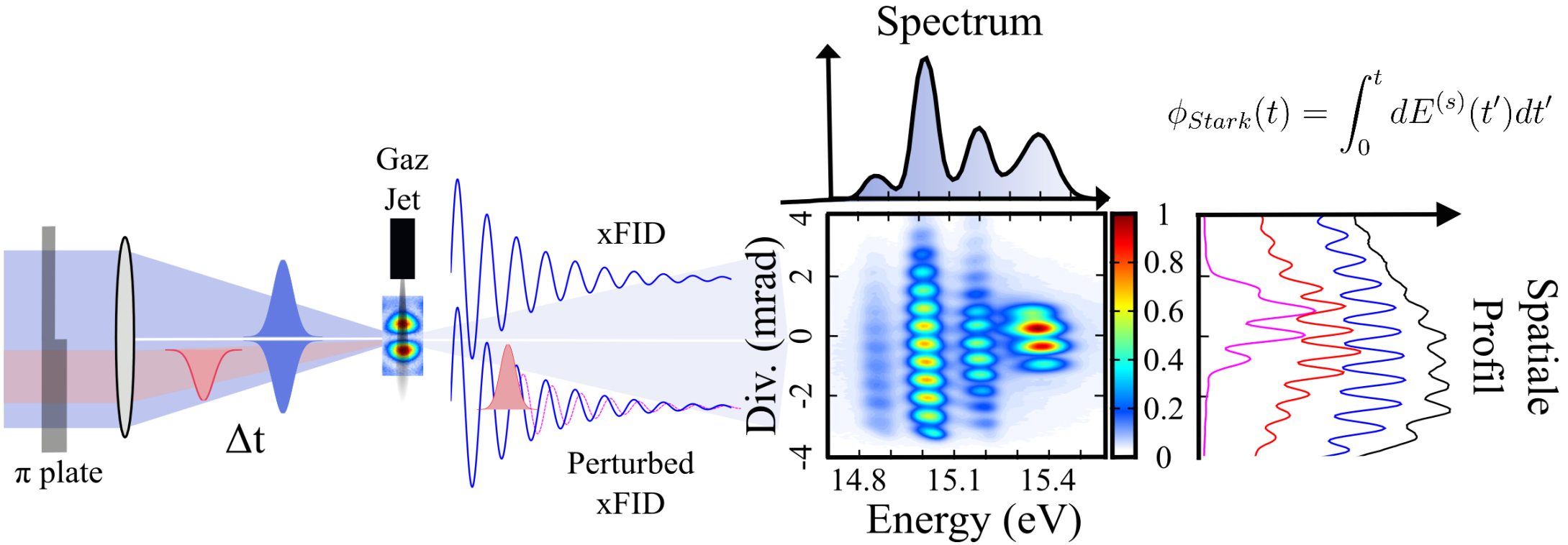




XUV spectrometer resolution
< 70 meV

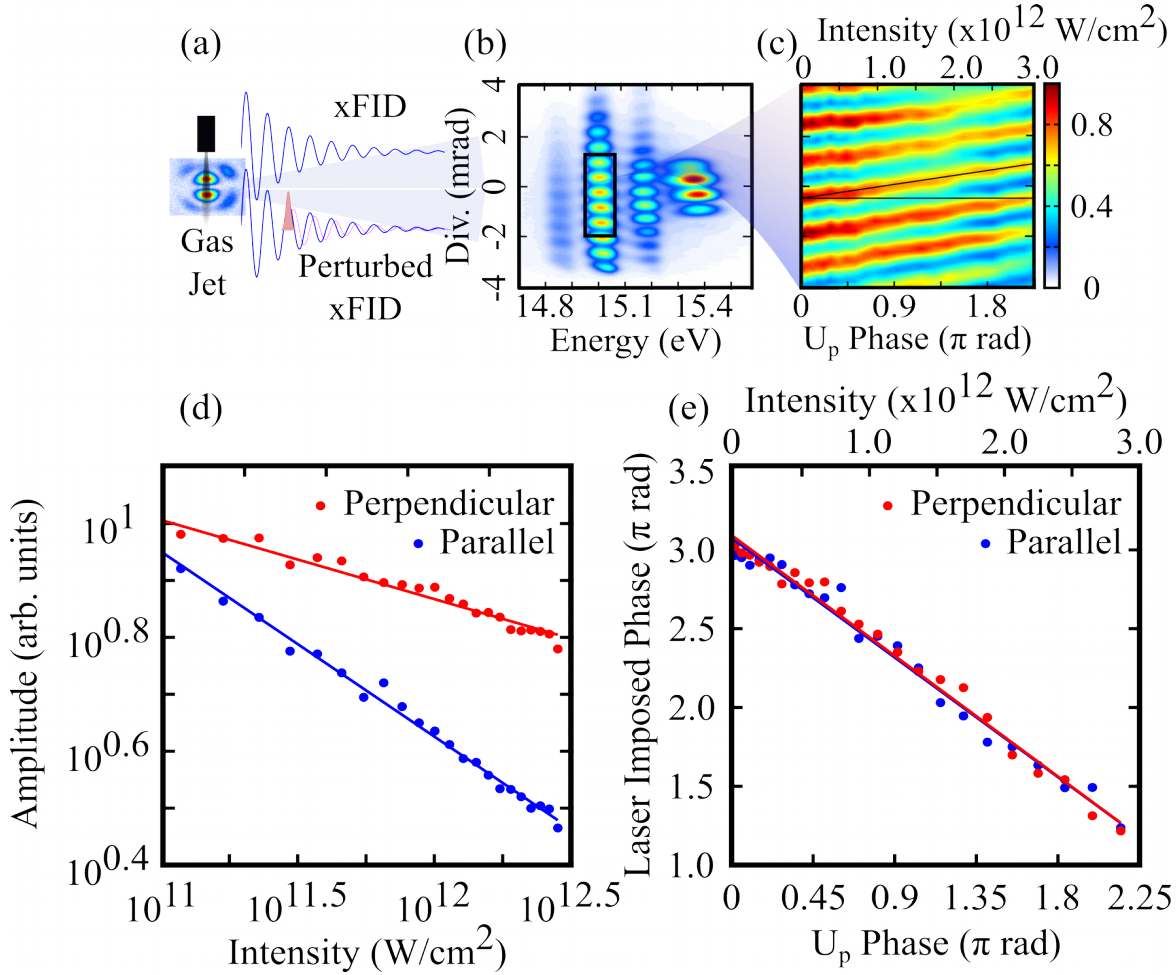
Achieved spectral resolution
> 1 meV

Laser-Imposed Phase Measurement



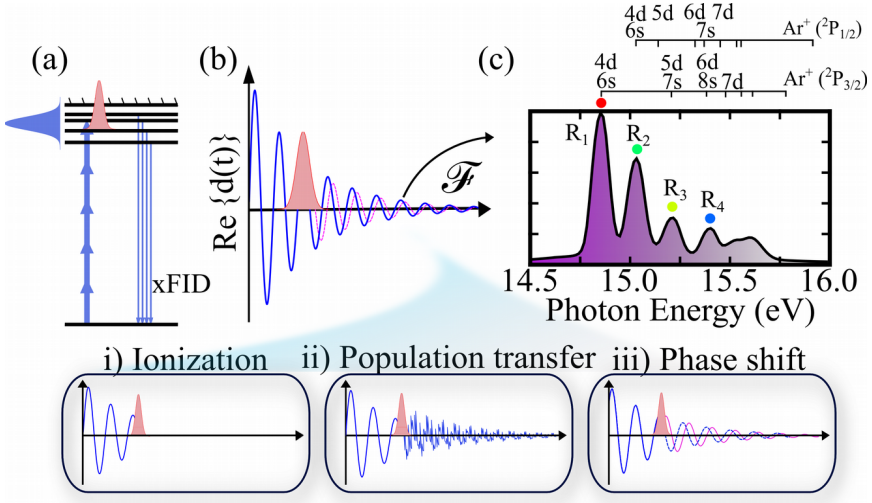
Beaulieu *et al.*, Phys. Rev. A 95, 041401(R) (2017)

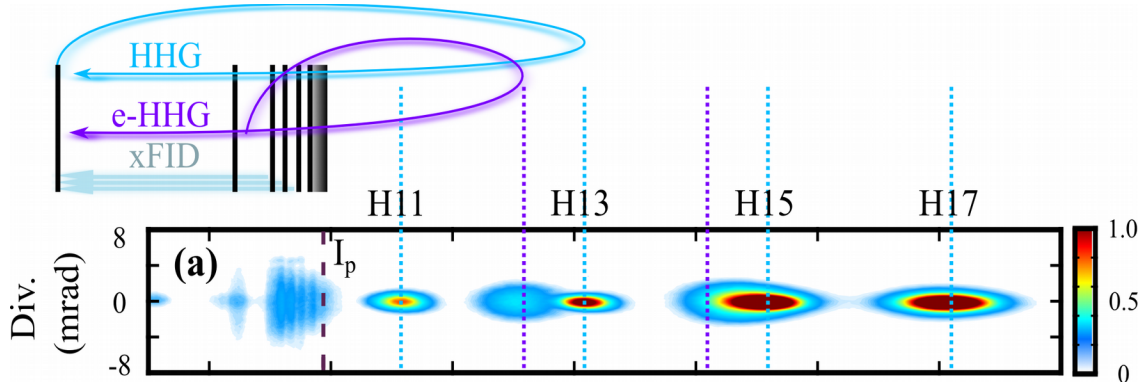
Laser-Imposed Phase Measurement



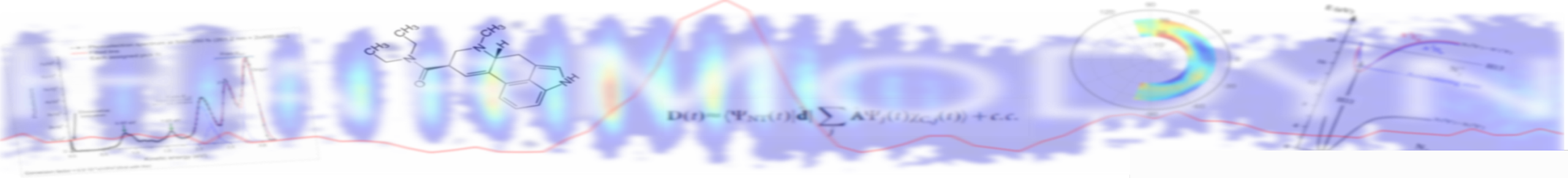
Conclusion

1) New HHG mechanism : HHG from Rydberg states





2) Novel time- and phase-resolved xFID spectroscopy of electronic wavepackets



Bordeaux

Yann Mairesse
Etienne Bloch
Fabrice Catoire
Dominique Descamps
Antoine Comby

INRS

François Légaré

CEA Saclay

Romain Géneaux
Lou Barreau

Louisiana State University

Seth Camp
Mette Gaarde
Ken Schafer

CELIA

cnrs dépasser les frontières

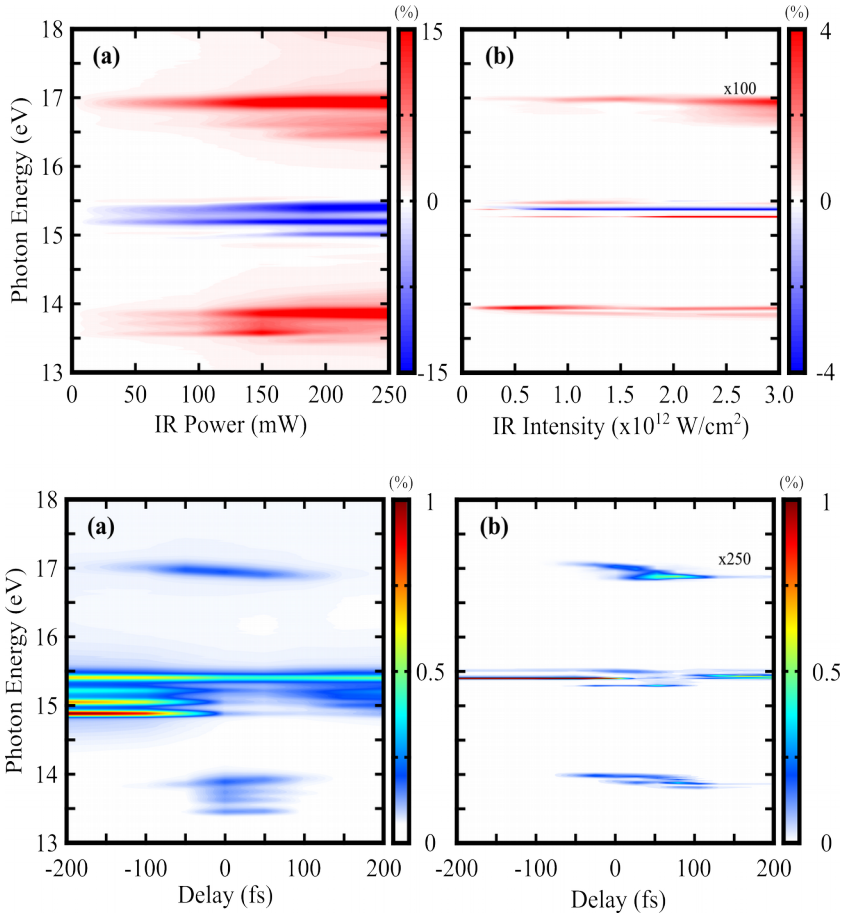
Bourses d'études supérieures du Canada Vanier Canada Graduate Scholarships

INRS Université d'avant-garde

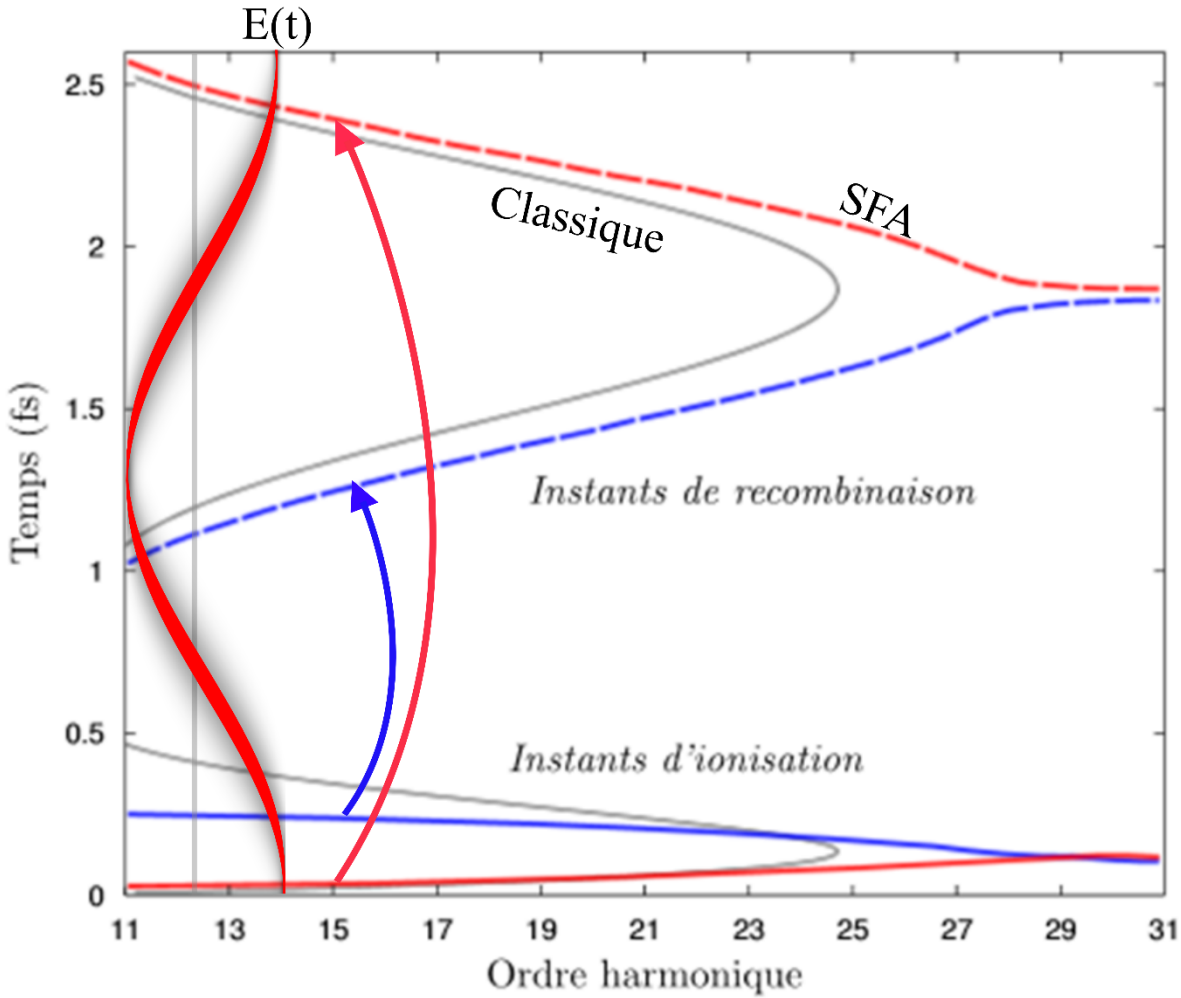
Fonds de recherche Nature et technologies Québec

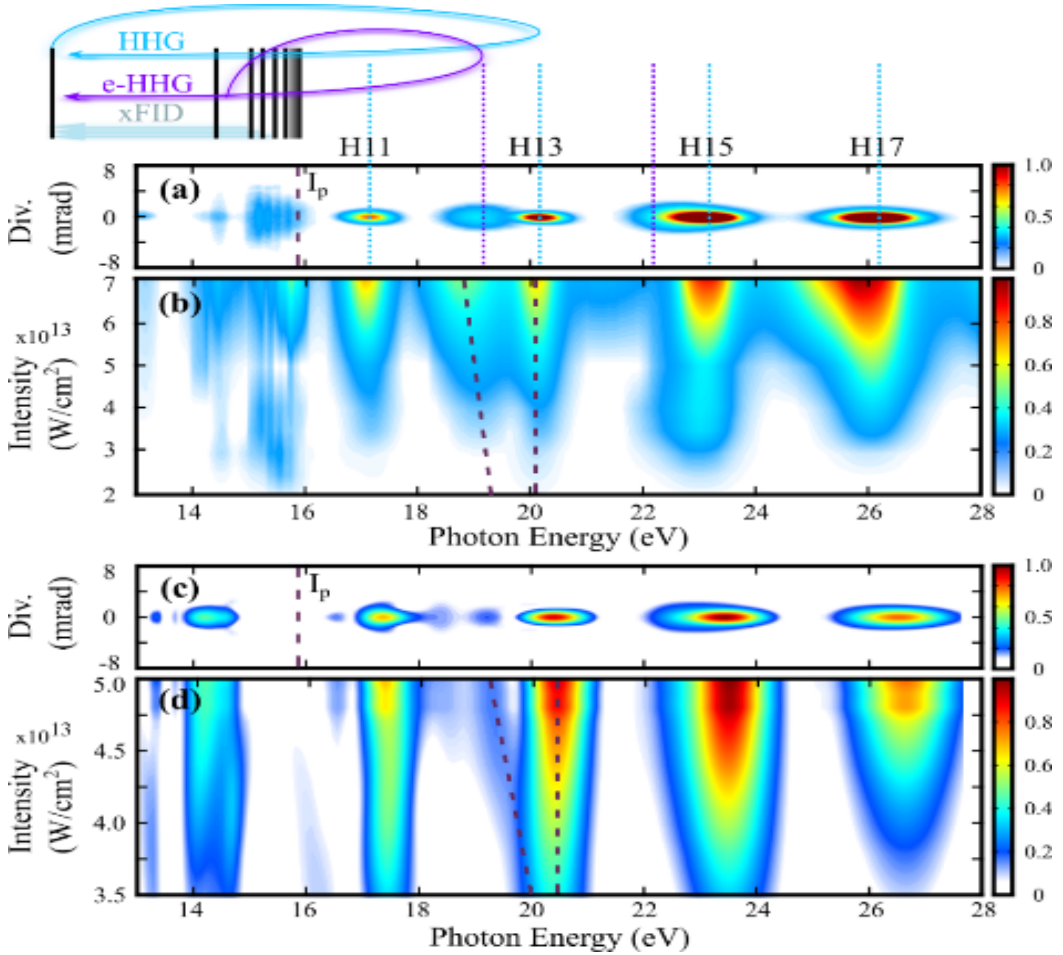
Perspectives

Hyper-Raman Effect in HHG



TDSE calculation by V. Strelkov

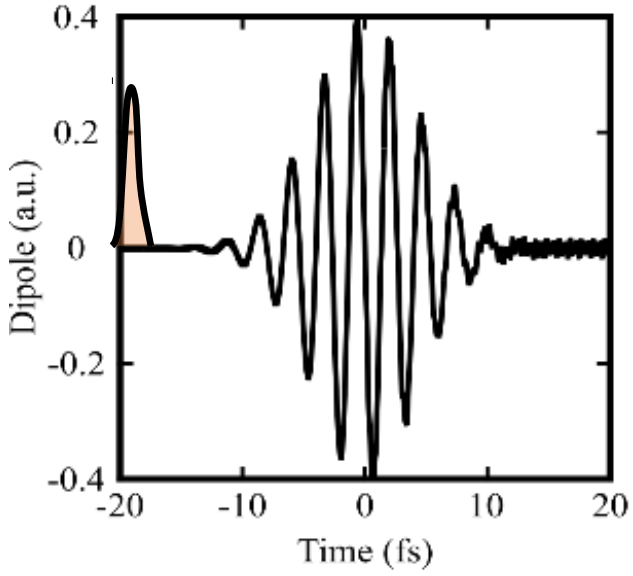

E(t)
Classique
SFA
Instants de recombinaison
Instants d'ionisation
Temps (fs)
Ordre harmonique
0
0.5
1
1.5
2
2.5
11
13
15
17
19
21
23
25
27
29
31

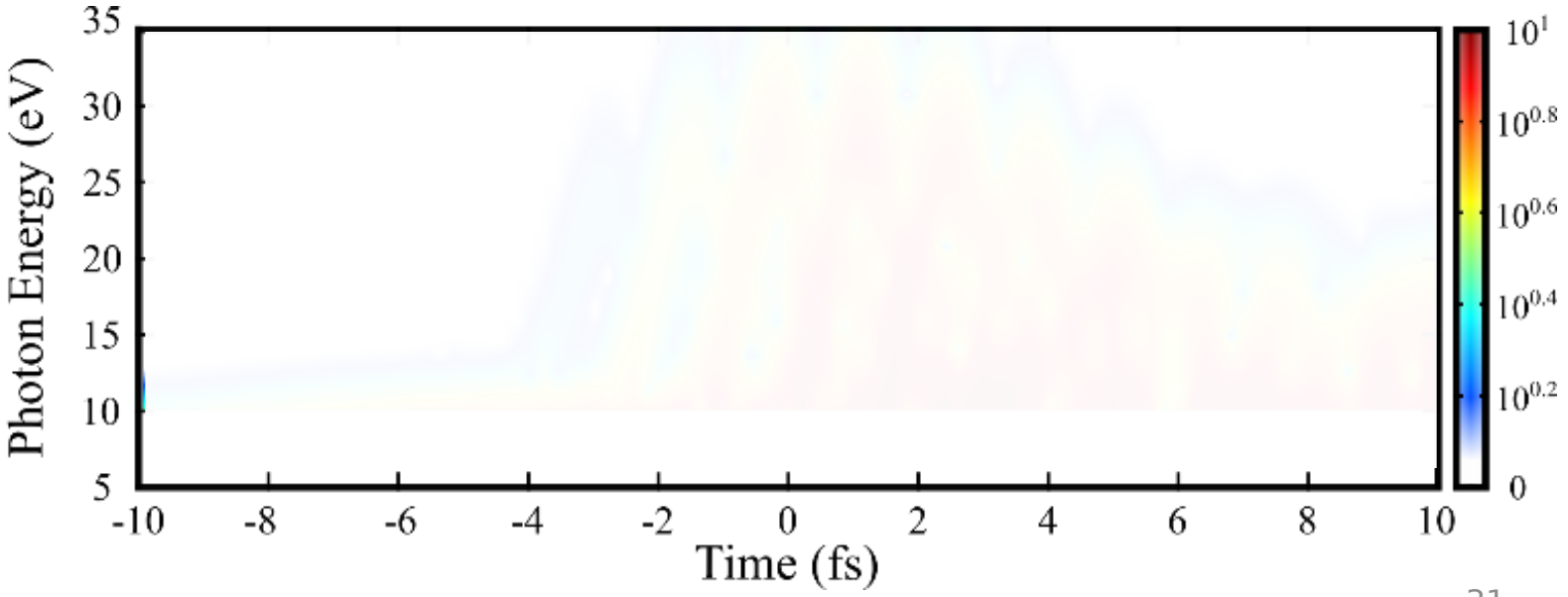

HHG
e-HHG
xFID
H11
H13
H15
H17
(a)
(b)
(c)
(d)
I_p
Div. (mrad)
Intensity (W/cm^2)
x10^{13}
Photon Energy (eV)

Nouveau mécanisme en HHG

Comment extraire de l'information du TDSE ?

Analyse de Gabor.

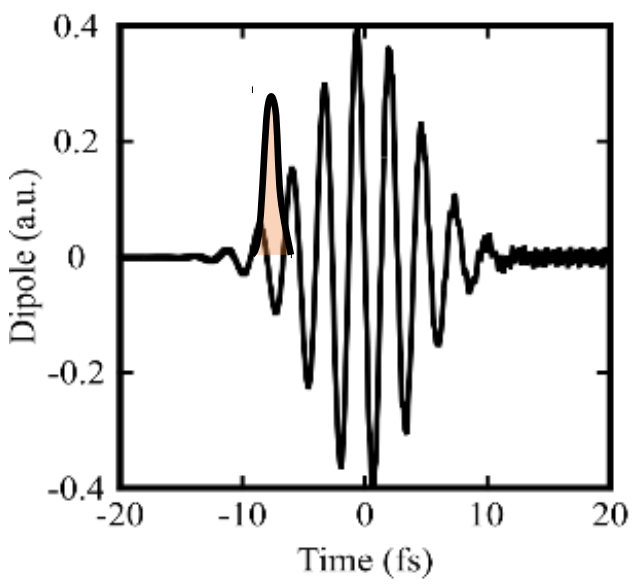


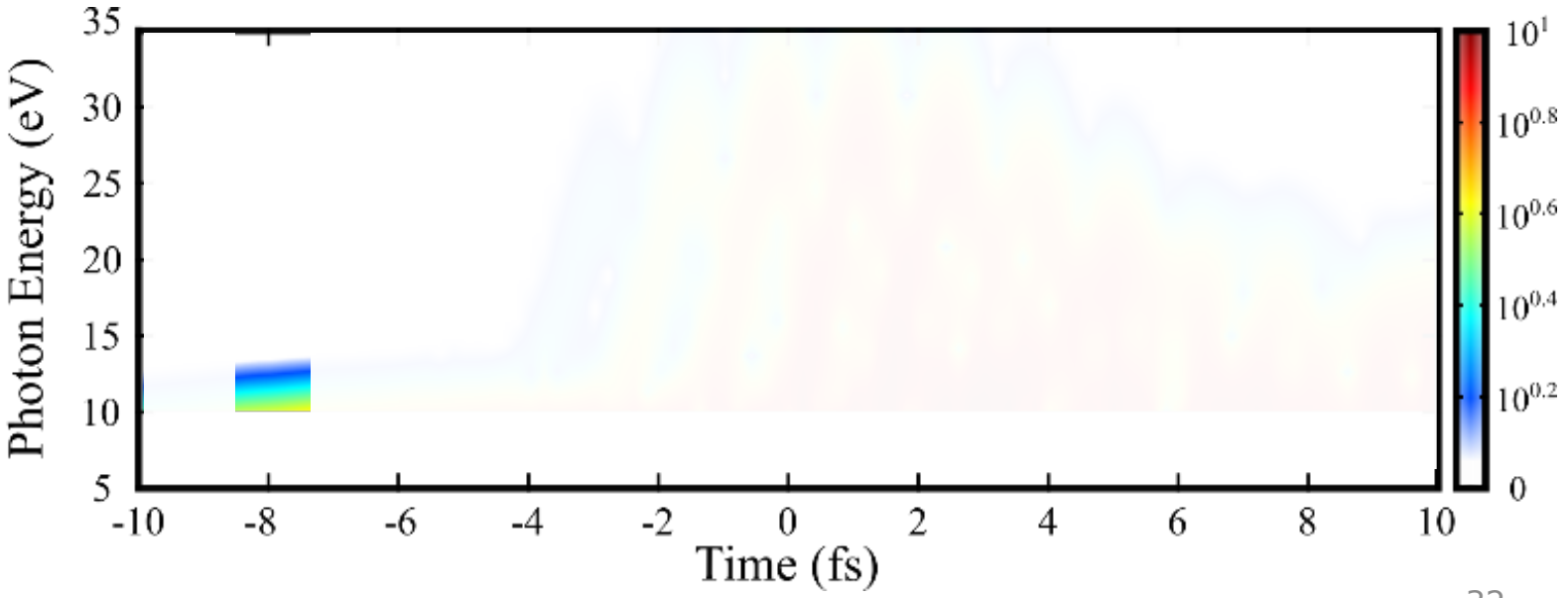


Nouveau mécanisme en HHG

Comment extraire de l'information du TDSE ?

Analyse de Gabor.

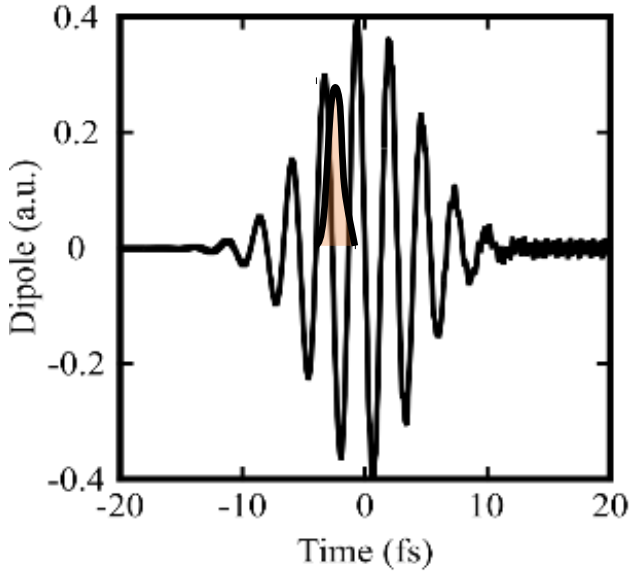


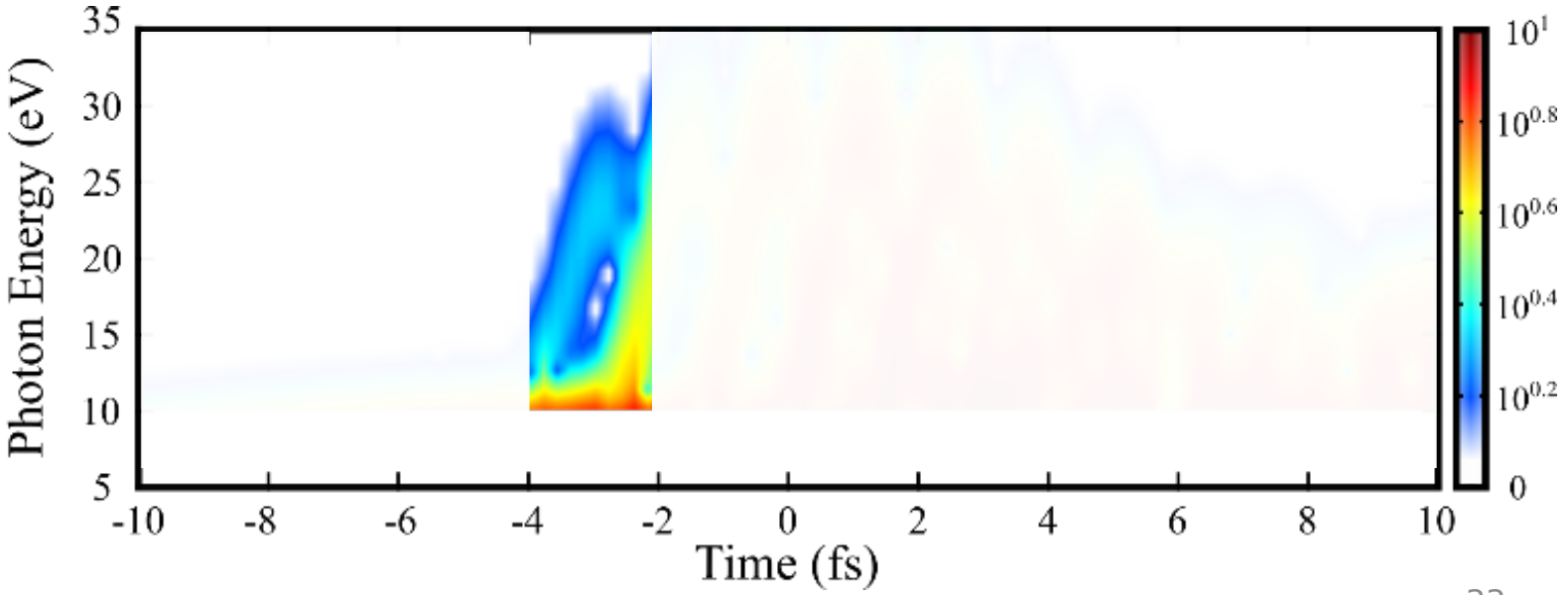


Nouveau mécanisme en HHG

Comment extraire de l'information du TDSE ?

Analyse de Gabor.

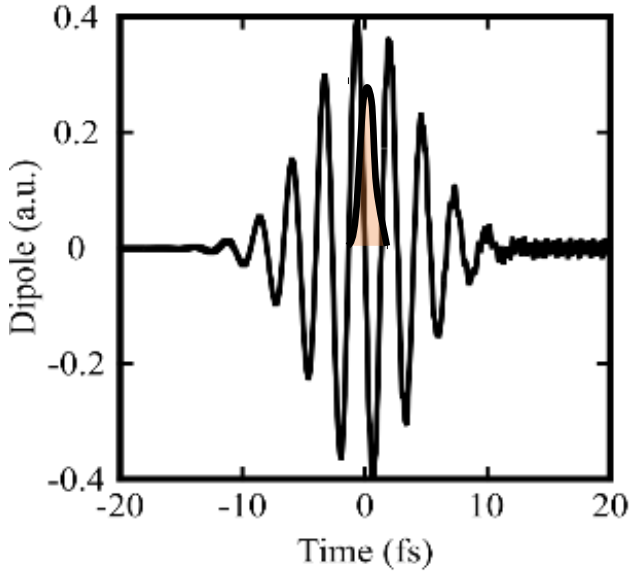


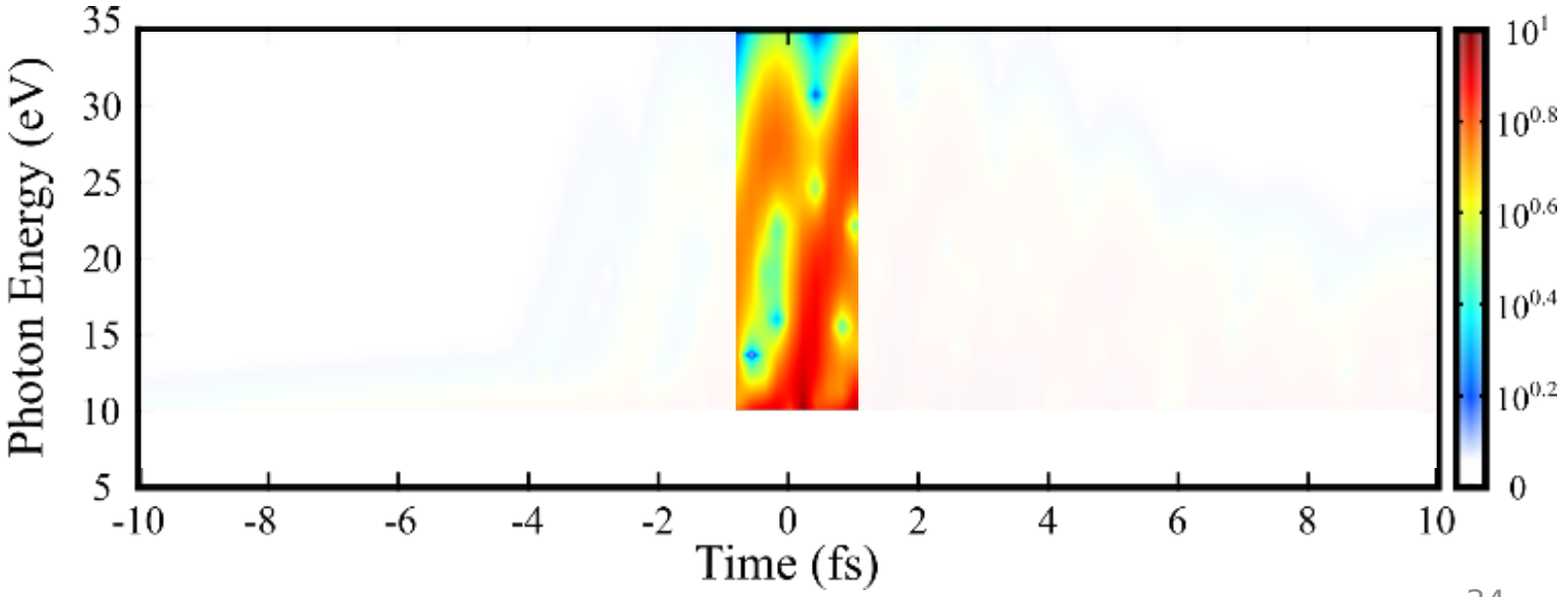


Nouveau mécanisme en HHG

Comment extraire de l'information du TDSE ?

Analyse de Gabor.

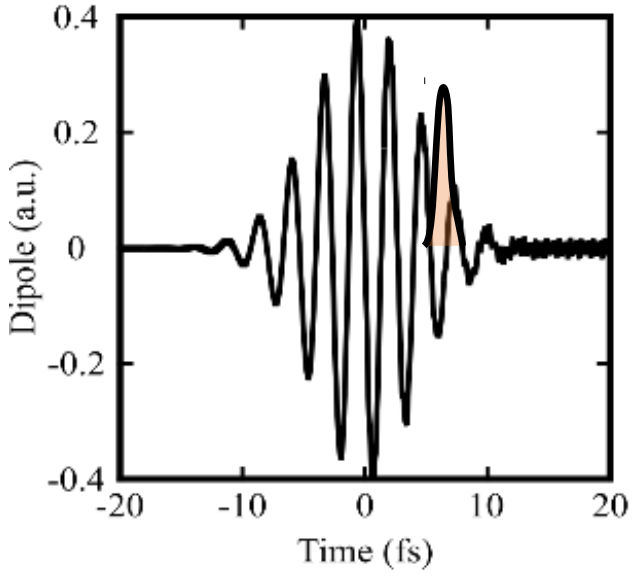


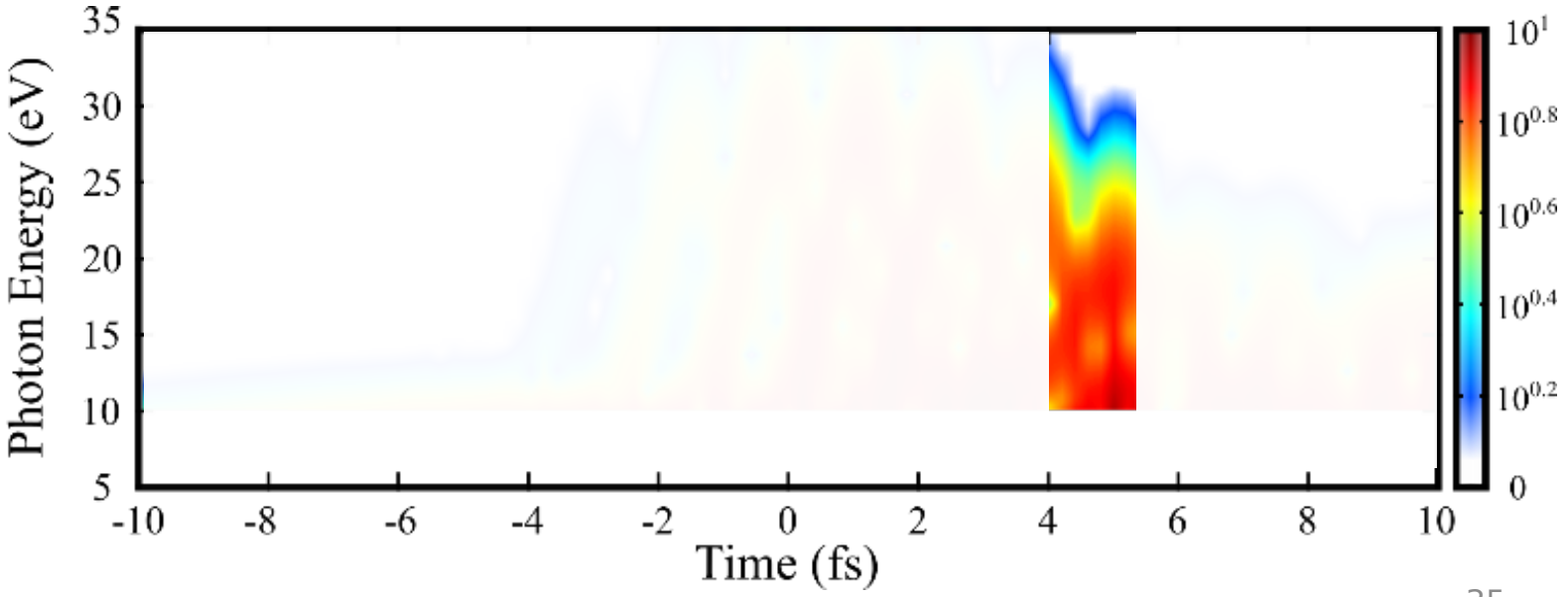


Nouveau mécanisme en HHG

Comment extraire de l'information du TDSE ?

Analyse de Gabor.

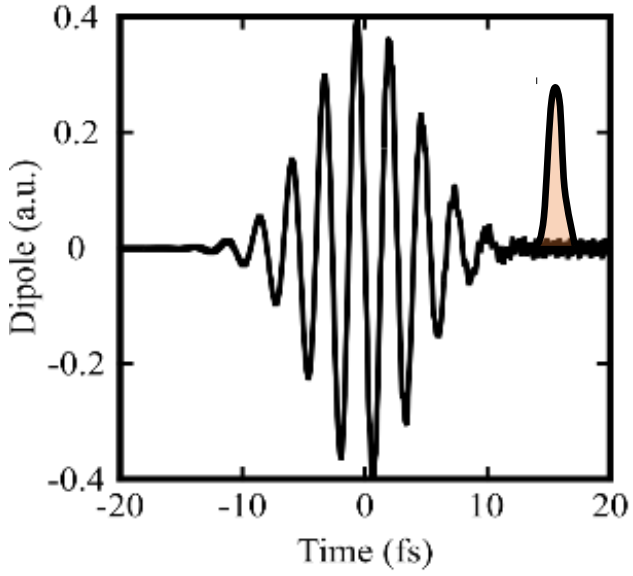


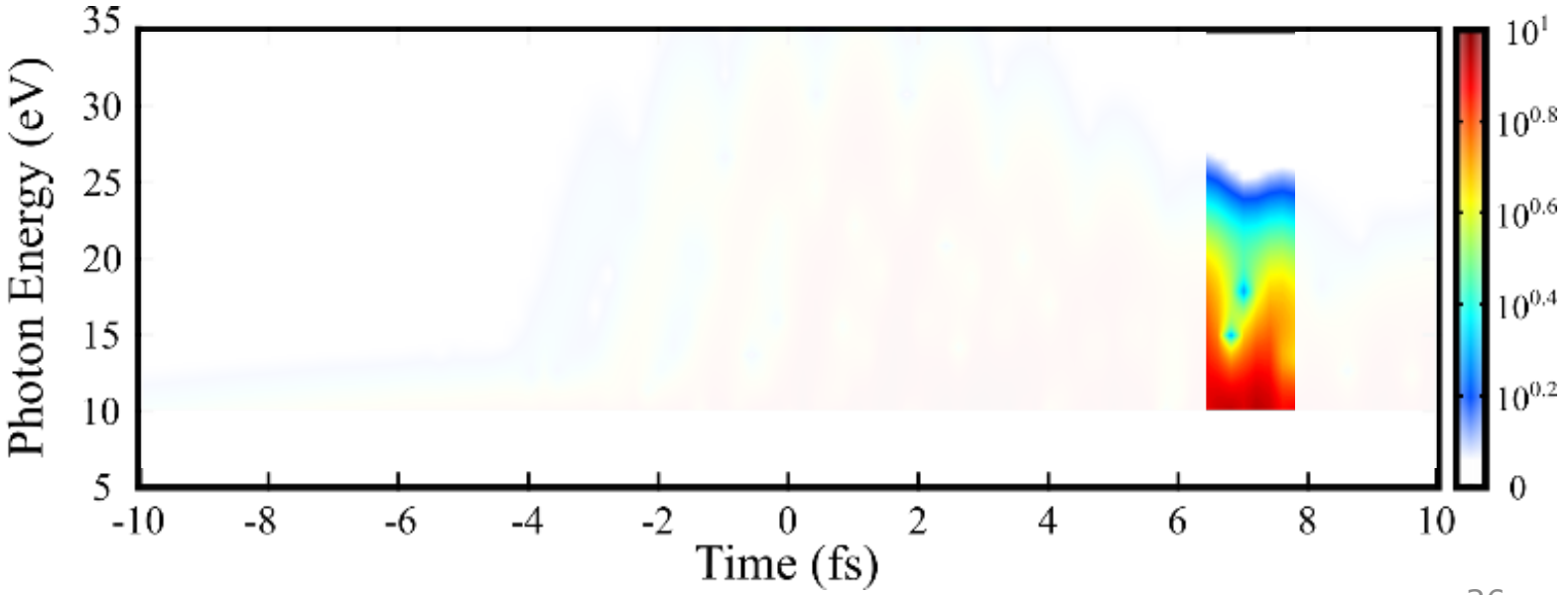


Nouveau mécanisme en HHG

Comment extraire de l'information du TDSE ?

Analyse de Gabor.





Nouveau mécanisme en HHG

Comment extraire de l'information du TDSE ?

Analyse de Gabor.