Novel approaches in HHG spectroscopy

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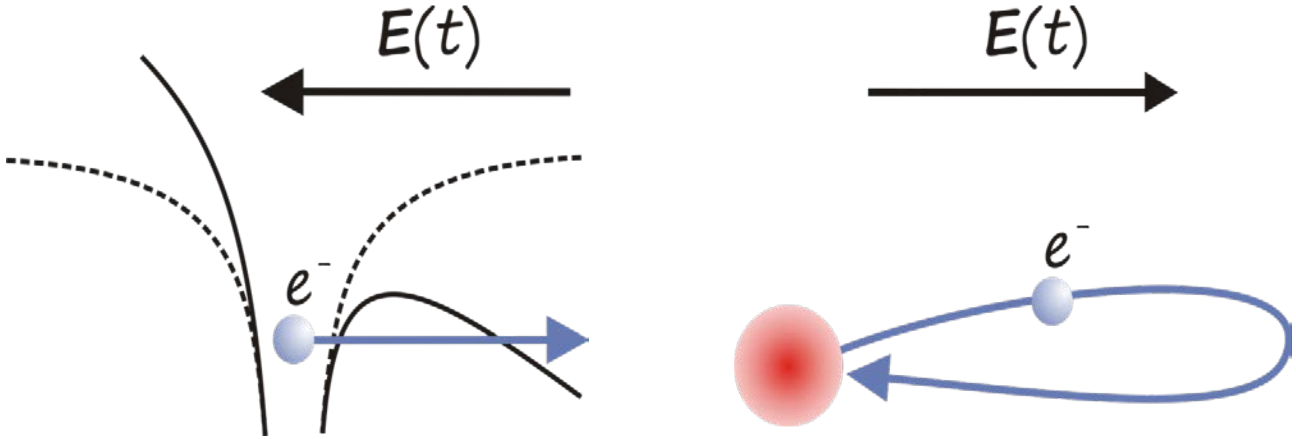
Outline

- A few reminders of HHG spectroscopy
- the study of electron correlation in xenon: experiments and interpretation
- perspective:
  development of the molecular imaging lab @ IFN-CNR

High order harmonic generation



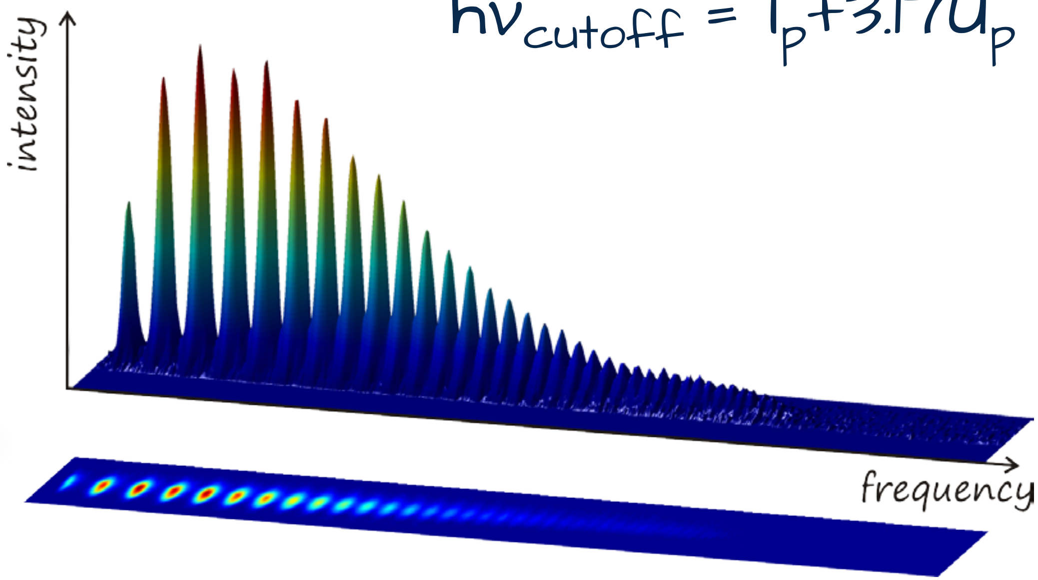
I. Ionization: the laser field detaches an electron from the valence shell via tunnel ionization

II. Propagation: the freed electron is accelerated by the laser field

III. Recombination: the energy gained by the electron is released through the emission of a XUV photon

HHG driven by longer wavelength

$h\nu_{cutoff} = I_p + 3.17U_p$ where $U_p \sim E^2\lambda^2$



- well-developed plateau in fragile molecules
- negligible contribution of multielectron effects

but...  generation yield $\sim\lambda^{-\alpha}$ with $\alpha \sim 5\text{-}6$

Lewenstein quantum model

- Strong Field Approximation
- Single Active Electron from the outermost valence shell

$$\vec{\mathcal{E}}_{XUV}[\Omega] \quad \propto \quad -(m\omega)^2 \int_{\mathbb{R}} \int_{\mathbb{R}+} \int_{\mathbb{R}^3} \underbrace{\boxed{\frac{e}{\hbar^4} \vec{E}(t-\tau) \cdot \vec{d}\left[\vec{k}(\vec{p}, t-\tau)\right]}}_{\text{I. ionization}} \underbrace{\boxed{\vec{d}\left[\vec{k}(\vec{p}, t)\right]^*}}_{\text{III. recombination}} \underbrace{\boxed{e^{-\frac{i}{\hbar} S(\vec{p}, t, \tau) + i\Omega t}}}_{\text{II. propagation}} \mathrm{d}\vec{p}\mathrm{d}\tau'\mathrm{d}t'$$

$$S(\vec{p}, t, \tau) = \int_{t-\tau}^{t} \frac{\left[\vec{p} + e\vec{A}(t')\right]^2}{2\mu} \mathrm{d}t' \qquad \vec{d}(\vec{k}) = \langle e^{i\vec{k}\cdot\vec{r}} | \vec{r} | \Gamma_0(\vec{r}) \rangle$$

orbital structure and symmetry are encoded in the harmonic spectrum!

Saddle point approximation & quantum trajectories

I. ionization II. propagation

$$\vec{\mathcal{E}}_{XUV}[\Omega] \propto -(\Omega)^2 \int_{\mathbb{R}} \int_{\mathbb{R}+} \int_{\mathbb{R}^3} \boxed{\frac{e}{\hbar^4}\vec{E}(t-\tau)\cdot\vec{d}\left[\vec{k}(\vec{p},t-\tau)\right]} \boxed{\vec{d}\left[\vec{k}(\vec{p},t)\right]^*} \boxed{e^{-\frac{i}{\hbar}S(\vec{p},t,\tau)+i\Omega t}} \mathrm{d}\vec{p}\mathrm{d}\tau'\mathrm{d}t'$$

III. recombination

$$\vec{\mathcal{E}}_{XUV}[\Omega] \propto -(\Omega)^2\vec{E}(t_s-\tau_s)\cdot\frac{e}{\hbar^4}\vec{d}\left[\vec{k}(\vec{p_s},t_s-\tau_s)\right]\ \vec{d}\left[\vec{k}(\vec{p_s},t_s)\right]^* \int_{\mathbb{R}} \int_{\mathbb{R}+} \int_{\mathbb{R}^3} \boxed{e^{-\frac{i}{\hbar}S(\vec{p},t,\tau)+i\Omega t}} \mathrm{d}\vec{p}\mathrm{d}\tau'\mathrm{d}t'$$

through the saddle point approximation

- we find a coupling between:

ionization time $t_s - \tau_s$

$$\frac{\partial S(p_s,t_s,\tau_s)}{\partial(-\tau)} = 0 = \frac{\left[\vec{p_s}+e\vec{A}(t_s-\tau_s)\right]^2}{2\mu} + I_p$$

recombination time t_s

$$\frac{\partial S(p_s,t_s,\tau_s)}{\partial t} = 0 = \frac{\left[\vec{p_s}+e\vec{A}(t_s)\right]^2}{2\mu} + I_p - \hbar\Omega$$

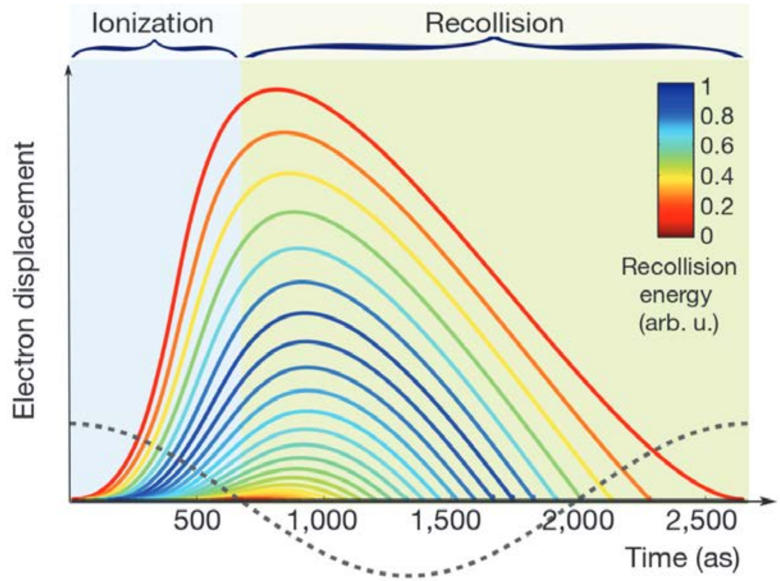
photon energy $\hbar\Omega$

$$\vec{\nabla}_{\vec{p}}S(p_s,t_s,\tau_s) = 0 = \int_{t_s-\tau_s}^{t_s} \frac{\left[\vec{p_s}+e\vec{A}(t')\right]}{\mu}dt'$$

UDynI

Attosecond dynamics probed by HHG

Each saddle point solution defines a quantum trajectory



Shafir et al. Nature 485, 343 (2012)

The attosecond nature of the process is mapped into the HHG spectrum!

Why xenon?

it is not a complex system...but:

- it shows a high harmonic yield
- it can be modeled "easily"
- it shows electronic correlation

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Strong-Field Many-Body Physics and the Giant Enhancement in the High-Harmonic Spectrum of Xenon

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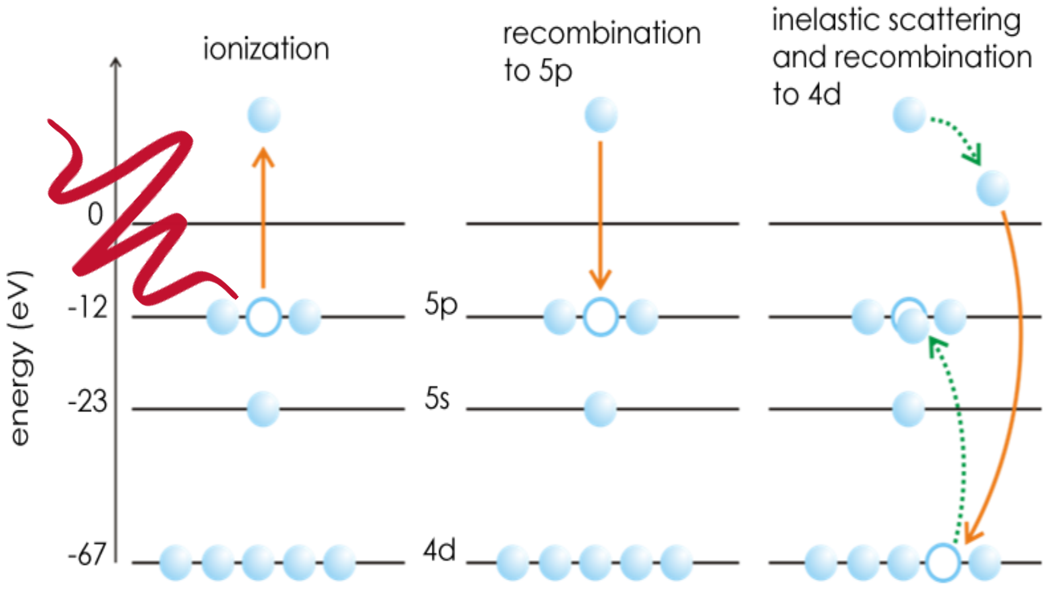
Electron correlation in xenon



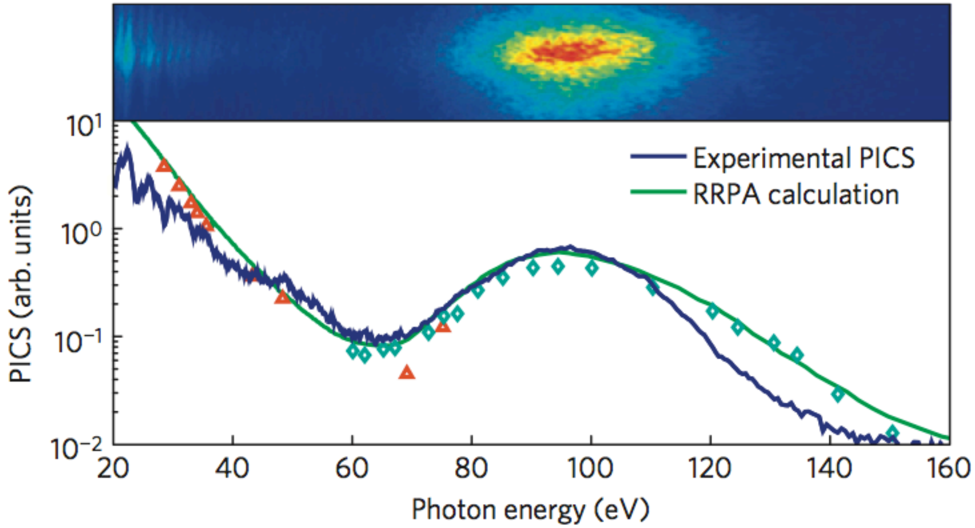
first channel: the returning electron recombines directly with the 5p electron vacancy

second channel: the returning electron promotes an electron of the 4d inner shell to the 5p valence shell via inelastic scattering and then it recombines with the 4d electron vacancy

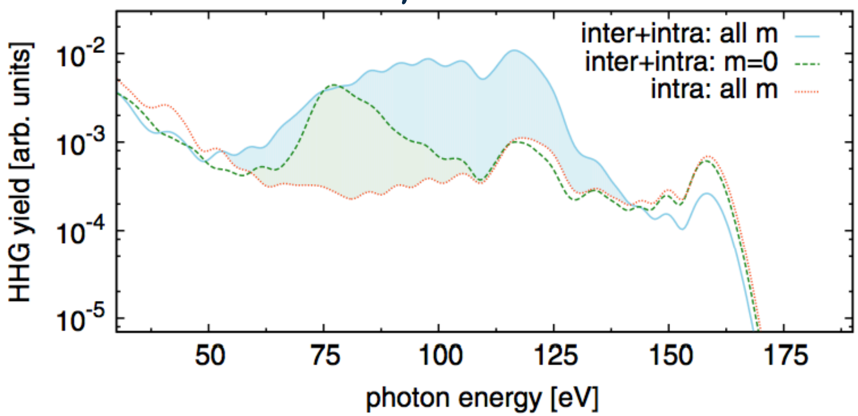
giant resonance around 100 eV

Xenon giant resonance in HHG

A. Shiner et al., Nat. Physics 7, 464 (2011)



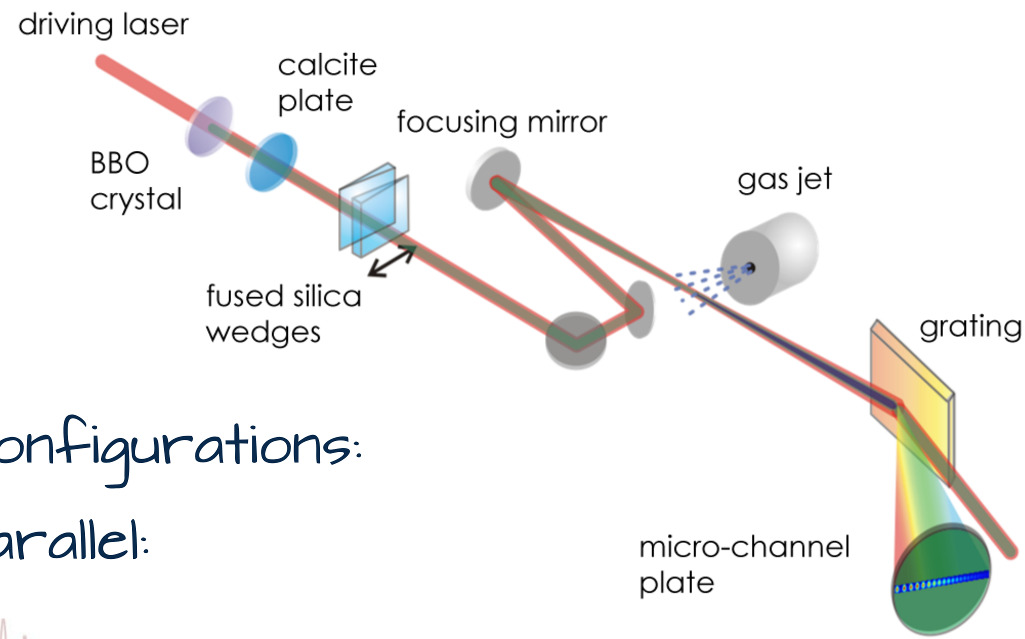
S. Pabst & R. Santra, Phys. Rev. Lett. 111, 233005 (2013)



how can we extract information about electron correlation from HHG measurement?

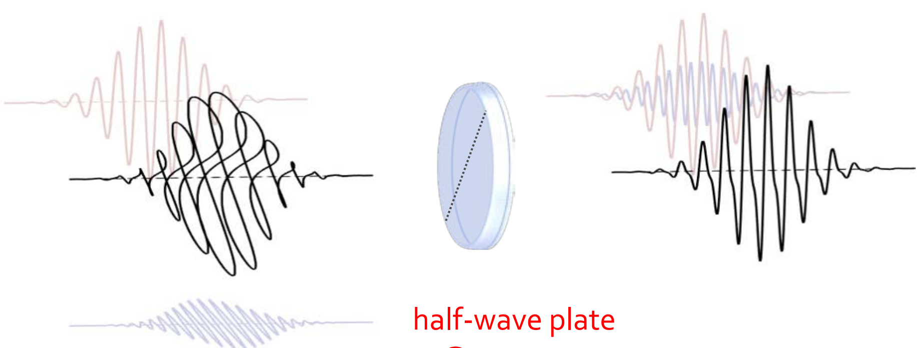
by two-color HHG!

Two-color HHG spectroscopy



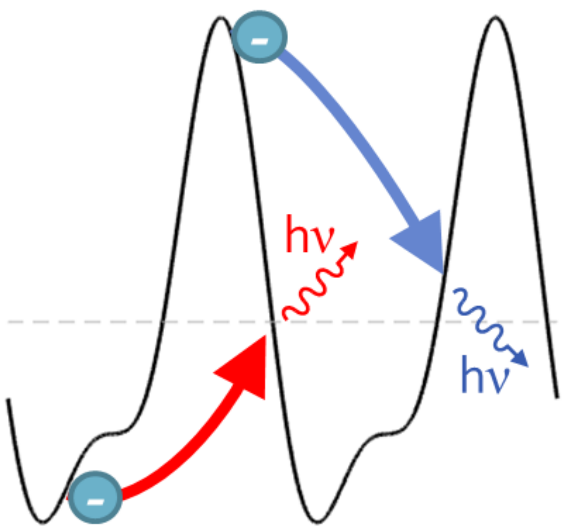
Two possible polarization configurations:

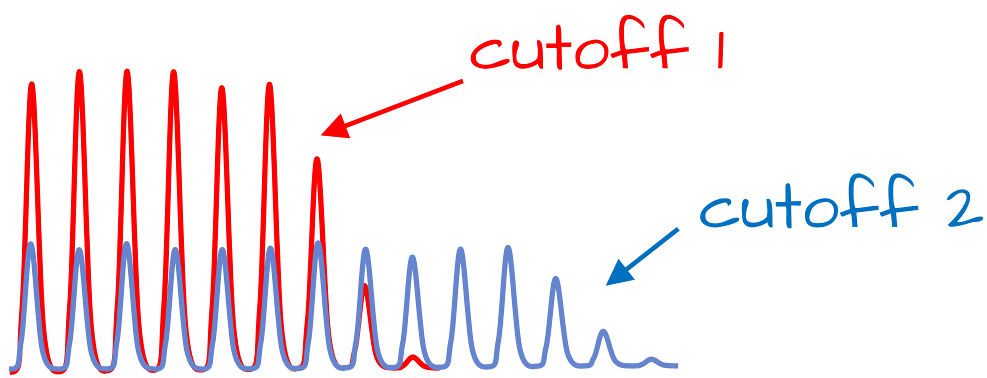
Perpendicular: Parallel:



Parallel configuration: method

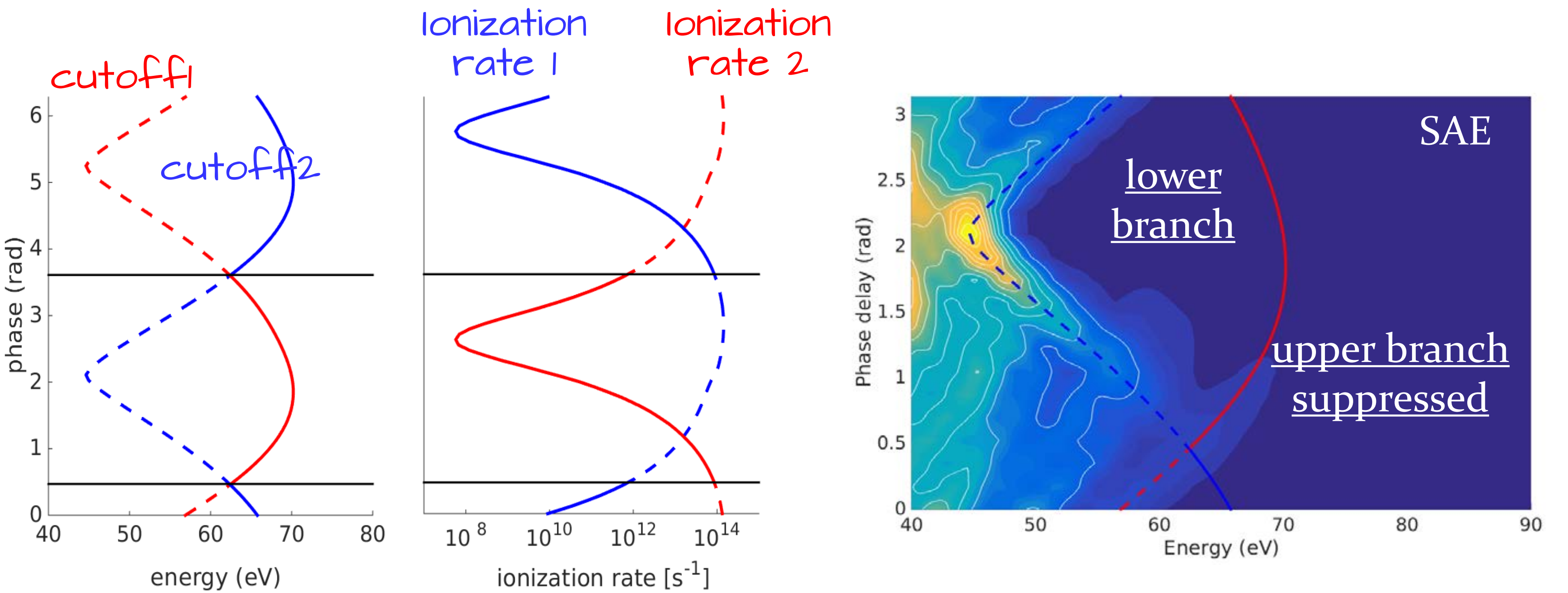
the symmetry between the two half cycles is broken

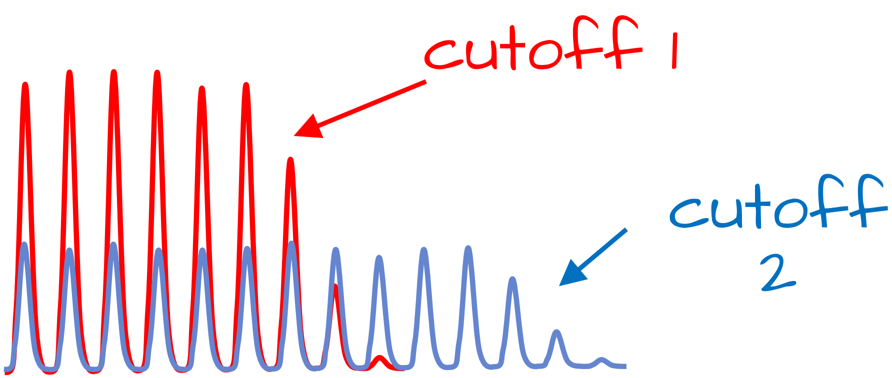




this results in two classes of trajectories with two different cutoffs and ionization probabilities

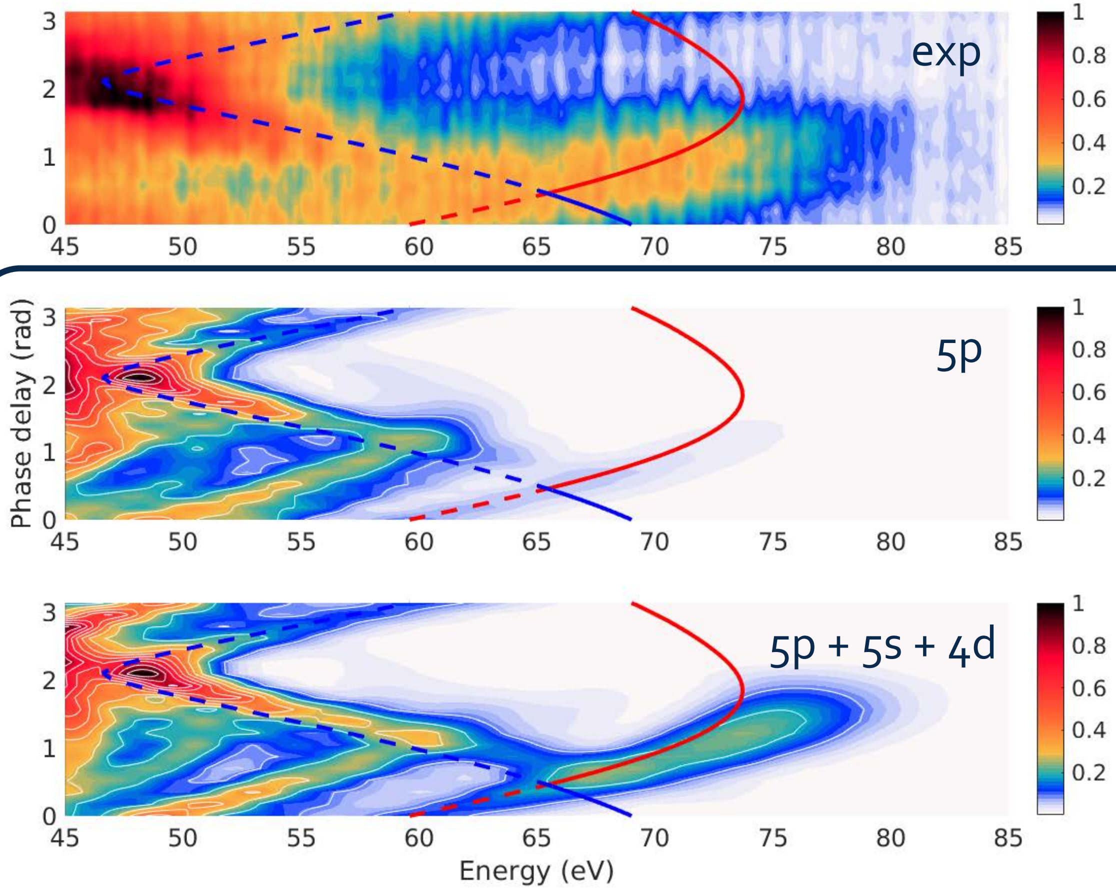
Parallel configuration: method





Frolov et al., PRA 81, 063407 (2010)

Parallel configuration: results



time-dependent configuration interaction singles (TDCIS) calculations by Stefan Pabst

Qualitative and quantitative agreement with TDCIS calculations when all 5p, 5s and 4d orbitals are interacting

- D. Faccialà et al., Phys. Rev. Lett. 117, 093902 (2016).

Parallel configuration: interpretation

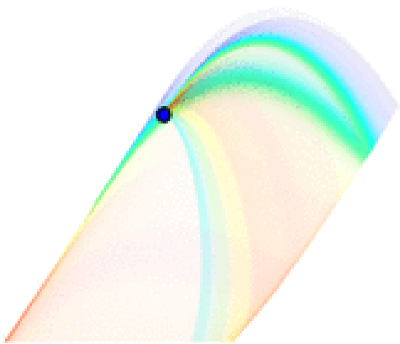
the upper branch becomes visible in the HHG spectrum only when all 5p, 5s and 4d orbitals are interacting

the GIANT RESONANCE counteracts the reduced ionization probability of the upper branch

...Can we extract information on the electron-electron correlation from HHG spectra?

In parallel polarization we cannot easily disentangle τs, ts, Ω

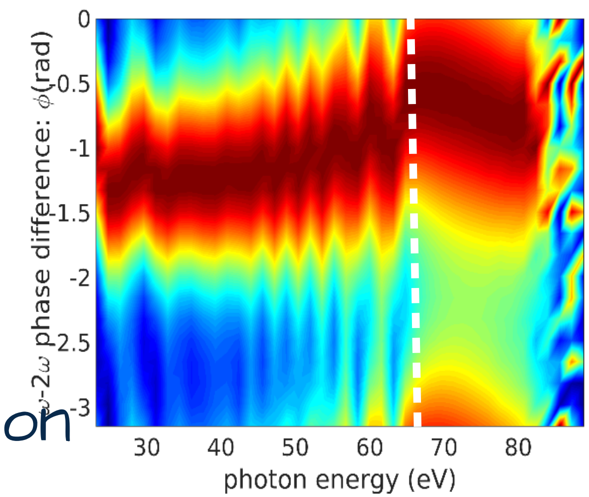
Perpendicular configuration: method



The second harmonic displaces the electron in the orthogonal direction, it acts as a gating which selects specific trajectories with a given energy

There is an optimal phase delay φ between the two fields that maximizes the probability of recombination for each harmonic photon energy.

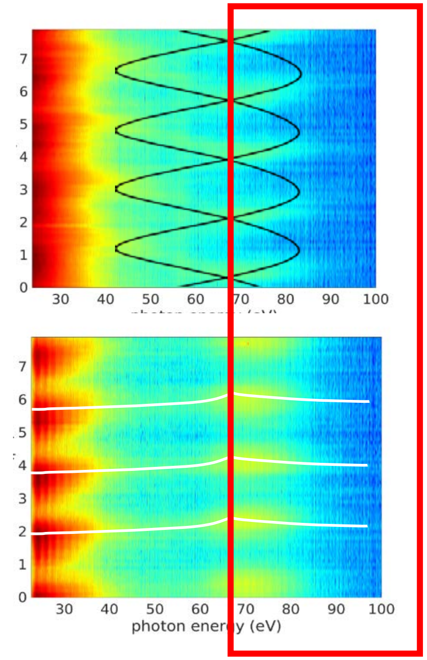
$\varphi(\Omega)$ is a measure of the recombination time of the electron t_r



...it is a clock to probe dynamics!

Conclusions

(1) The xenon giant resonance gives access to the observation of unrevealed features:



- In parallel polarization it counteracts the reduced ionization probability of the upper branch
- In perpendicular polarization it enhances the contribution of trajectories that are classically forbidden

(2) We measured the delay induced by the process in the energy region of the cutoff trajectories

- the electron is accelerated during the recombination due to the coulomb-exchange process.

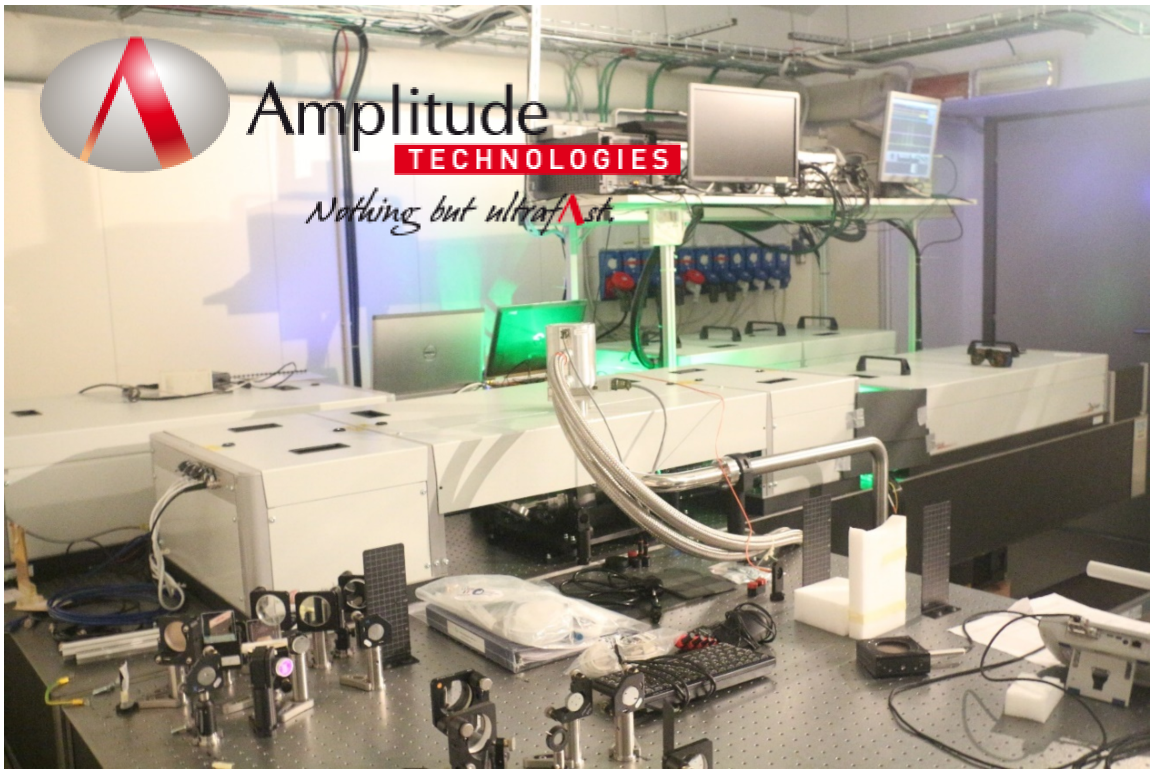
(3) Two-color HHG spectroscopy is sensitive to the phase of the dipole.

Perspectives: the new lab

A new lab @ CNR-IFN

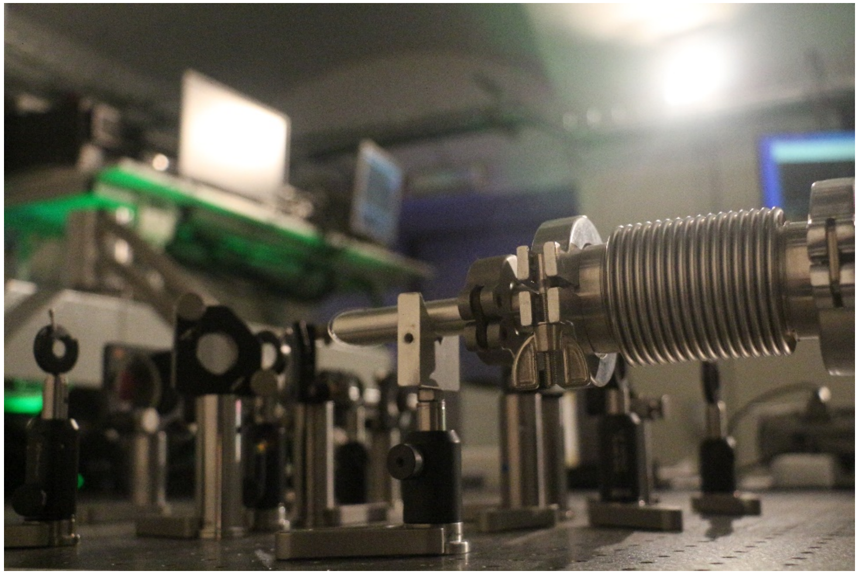
the laser source

European Research Council
Established by the European Commission

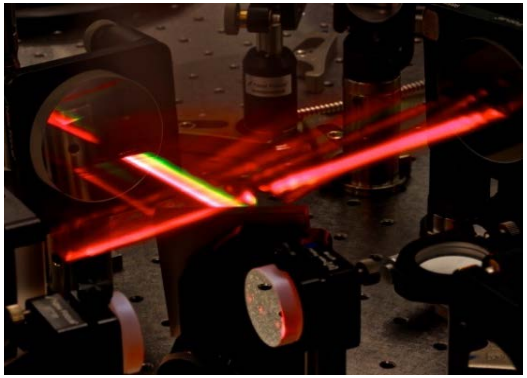


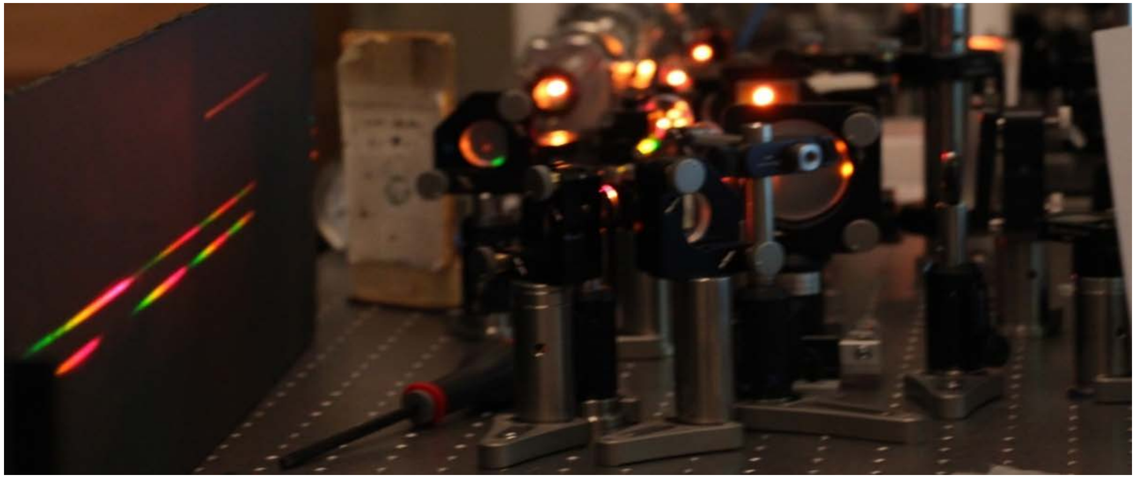
Driving laser source:
<22 fs pulses
15 mJ energy
1 kHz repetition rate

The Udyni lab @ CNR-IFN



manipulating the light



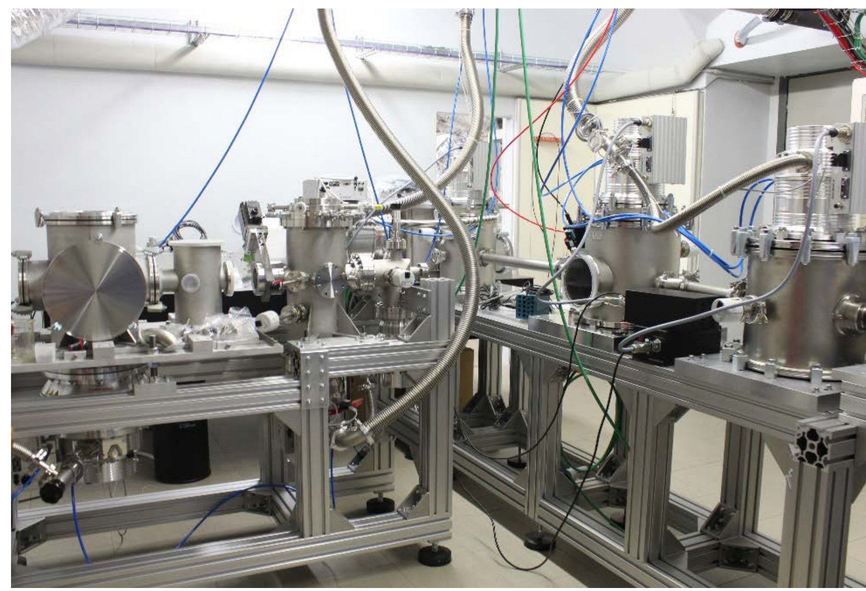


High energy OPA
- 1.2 - 3 μm
- <20 fs pulses
- 2 mJ energy
- CEP stable
- + hollow fiber

The Udyni lab @ CNR-IFN

XUV spectrometer
- gratings @ 10 nm and 1 nm
- stigmatic/astigmatic
- harmonic polarization detection

VMI spectrometer
for electrons up to 200 eV



Aknowledgments

Stefan Pabst

DESY

ITAMP

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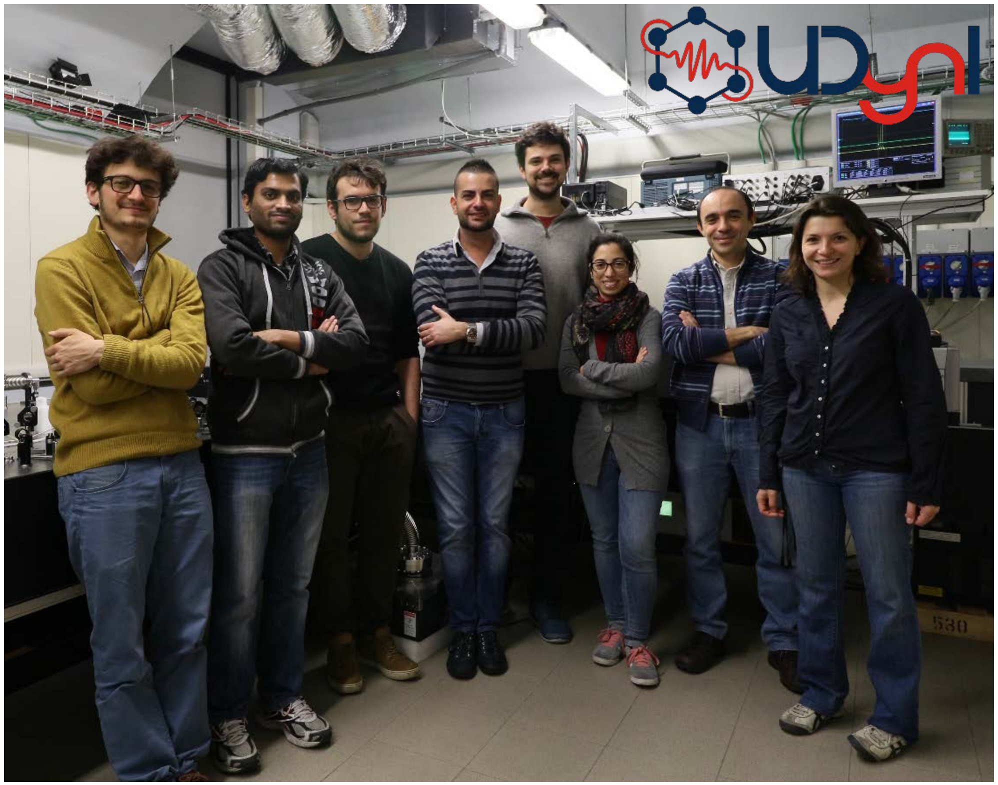

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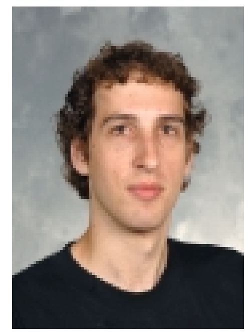

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