High-order Harmonic Generation: a spectroscopic tool on the attosecond and angstrom scale

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Outline

- Introduction on light-matter interaction in the strong-field regime
- High-order Harmonic Generation in gases (a basic discussion)
- High-order Harmonic Generation in crystals (a basic discussion)
- Applications of gas-phase HHG (some examples)
- New frontiers in HHG

Strong-optical-field phenomena

Optical phenomena are said in "strong field regime" when the electric-field component of light becomes comparable to the atomic Coulomb field

Modern laser technology provides access to this regime:

1 atomic unit of electric field **$E_a = 5.14\times10^{11}$ V/m**

Peak intensity of a laser pulse with energy of 15 mJ, duration of 25 fs, focused to a spot of 30-μm radius: **$I = 2\times10^{20}$ W/m²**

Peak electric field **$E_p = \sqrt{\dfrac{2I}{c\varepsilon_0}} = 4\times10^{11}$ V/m**

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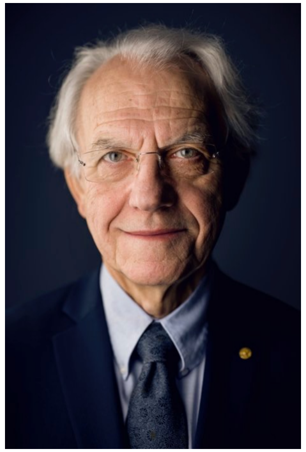
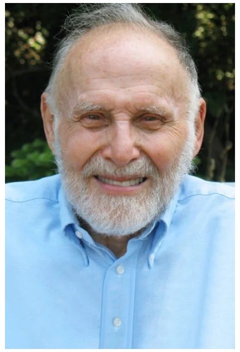


Nobel Prize in Physics 2018 to Gérard Mourou and Donna Strickland "for their method of generating high-intensity, ultra-short optical pulses…

… and to Arthur Ashkin "for the optical tweezers and their application to biological systems"



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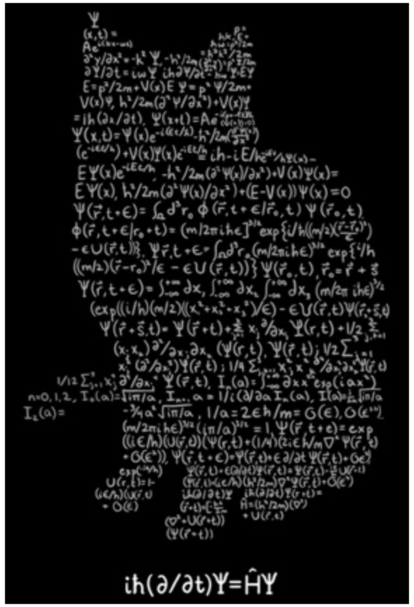
Strong-optical-field phenomena

What happens to matter exposed to intense laser pulses?

The fundamental constituents of matter are atoms and molecules and they obey quantum mechanics



Quantum description: time-dependent Schrödinger equation (TDSE)

$$\left[\hat{H} + U_{int}(t)\right]|\Psi\rangle = i\frac{\partial|\Psi\rangle}{\partial t}$$

(atomic units)

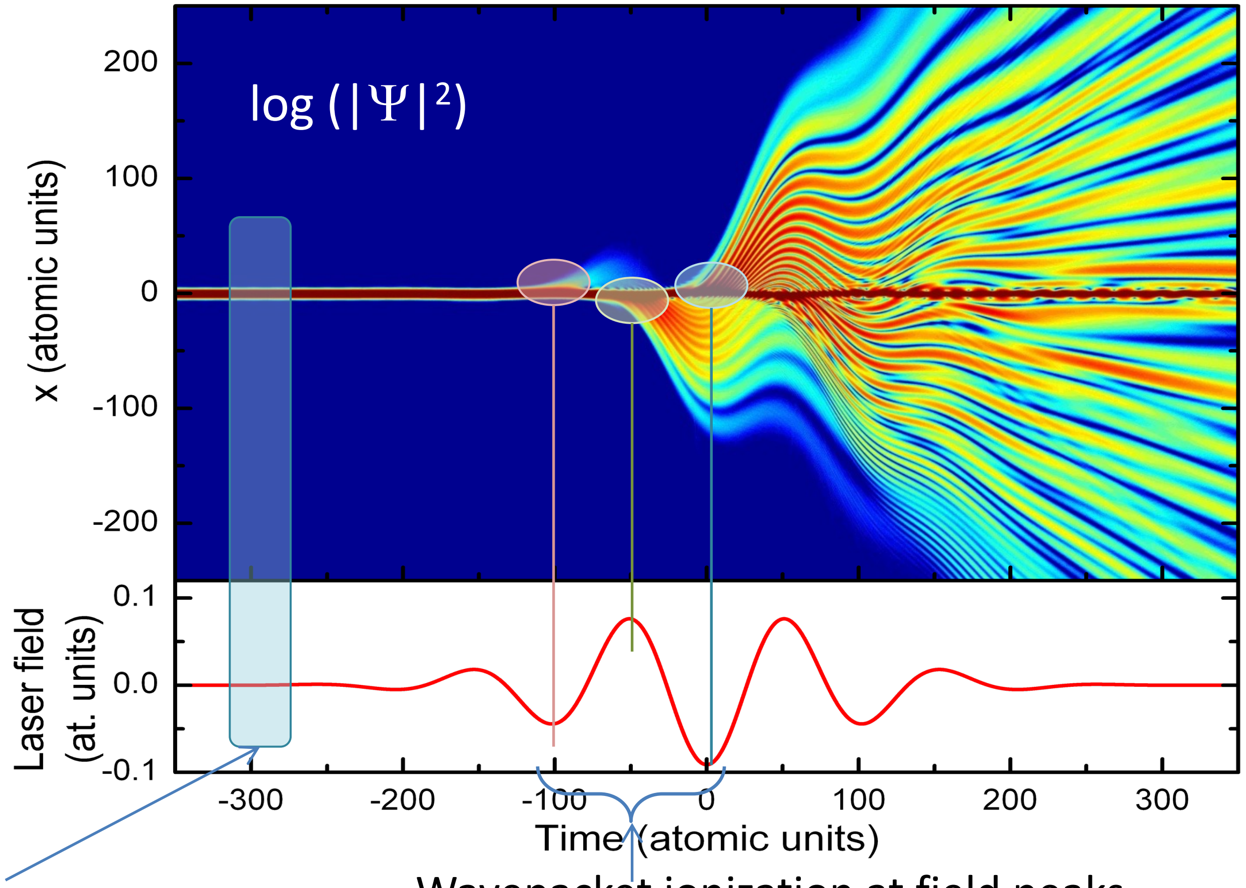
where $\hat{H} = -\frac{\nabla^2}{2} + V_{atom}(\boldsymbol{r})$ and $U_{int}(t) = \boldsymbol{r}\cdot\boldsymbol{E}(\mathrm{t})$

Simple case: 1D model atom with $V_{atom} = -\frac{1}{1+|x|}$

and ground state $\Psi_0(x) = \sqrt{\frac{2}{5}}e^{-|x|}(1+|x|)$ ⇒ I_p = 13.6 eV

(hydrogen ionization potential)

Laser-atom interaction



Unperturbed ground state

Wavepacket ionization at field peaks
Interference patterns are visible

Laser pulse features

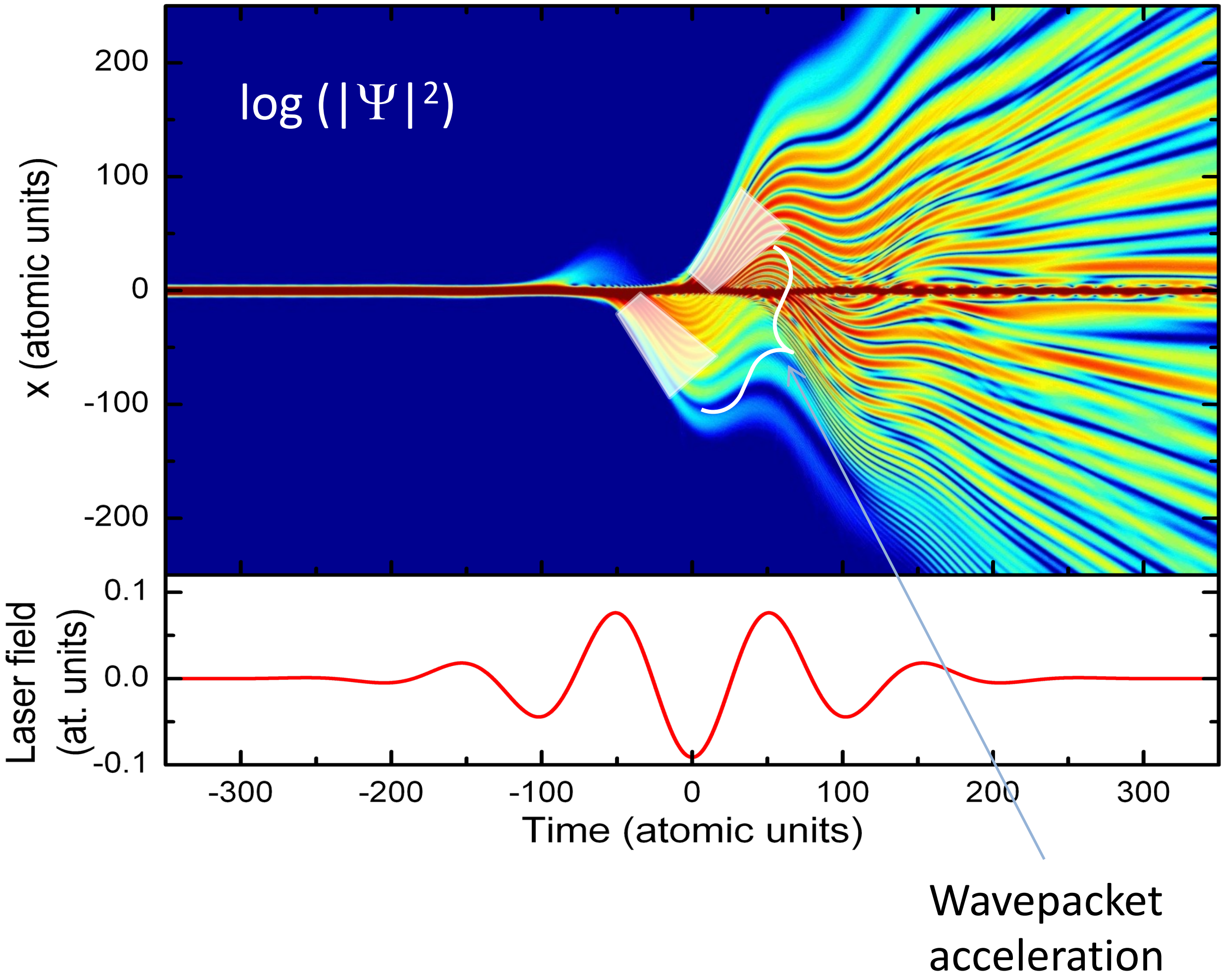
- λ = 800 nm
- duration = 3.5 fs
- peak intensity = 3.5×10^{14} W/cm^2

Atomic units

Time: 100 a.u. = 2.4 fs

Length: 100 a.u. = 5.3 nm

Laser-atom interaction



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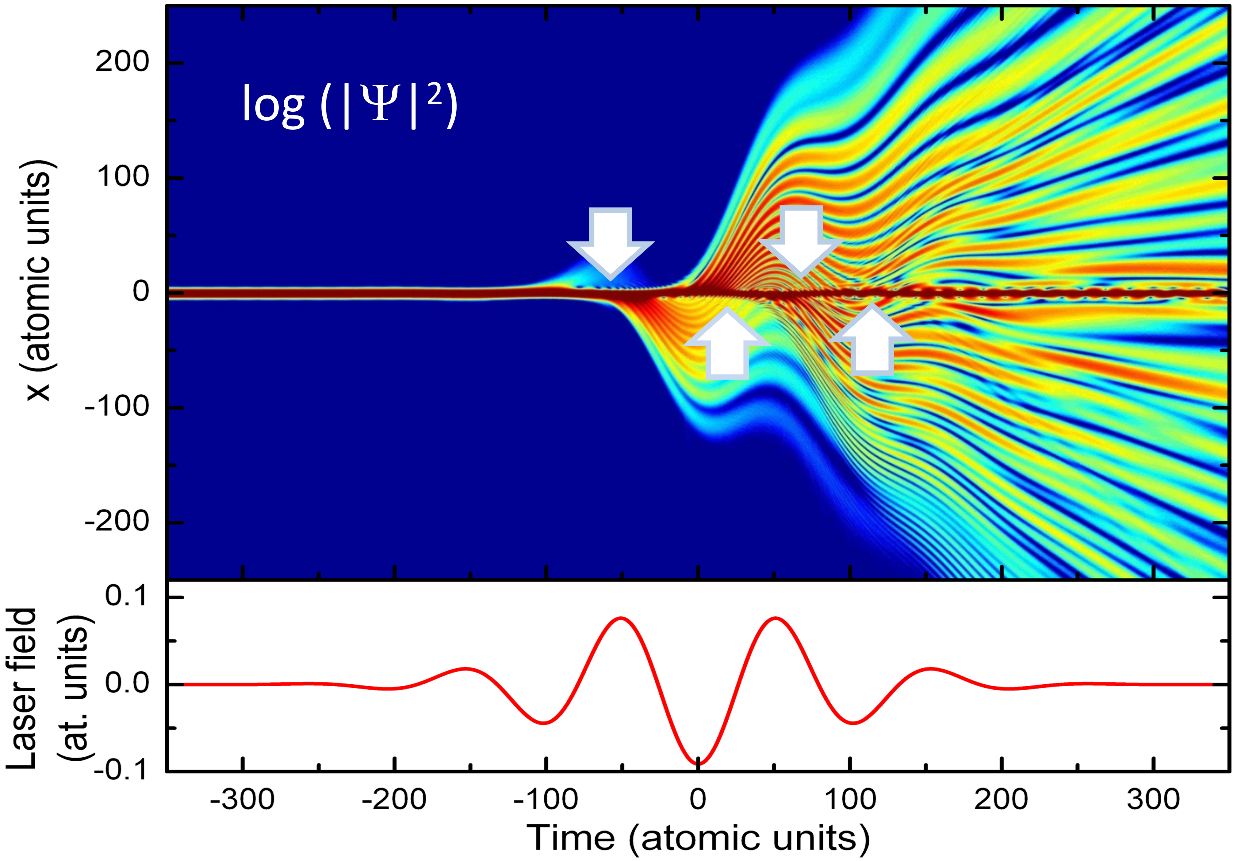

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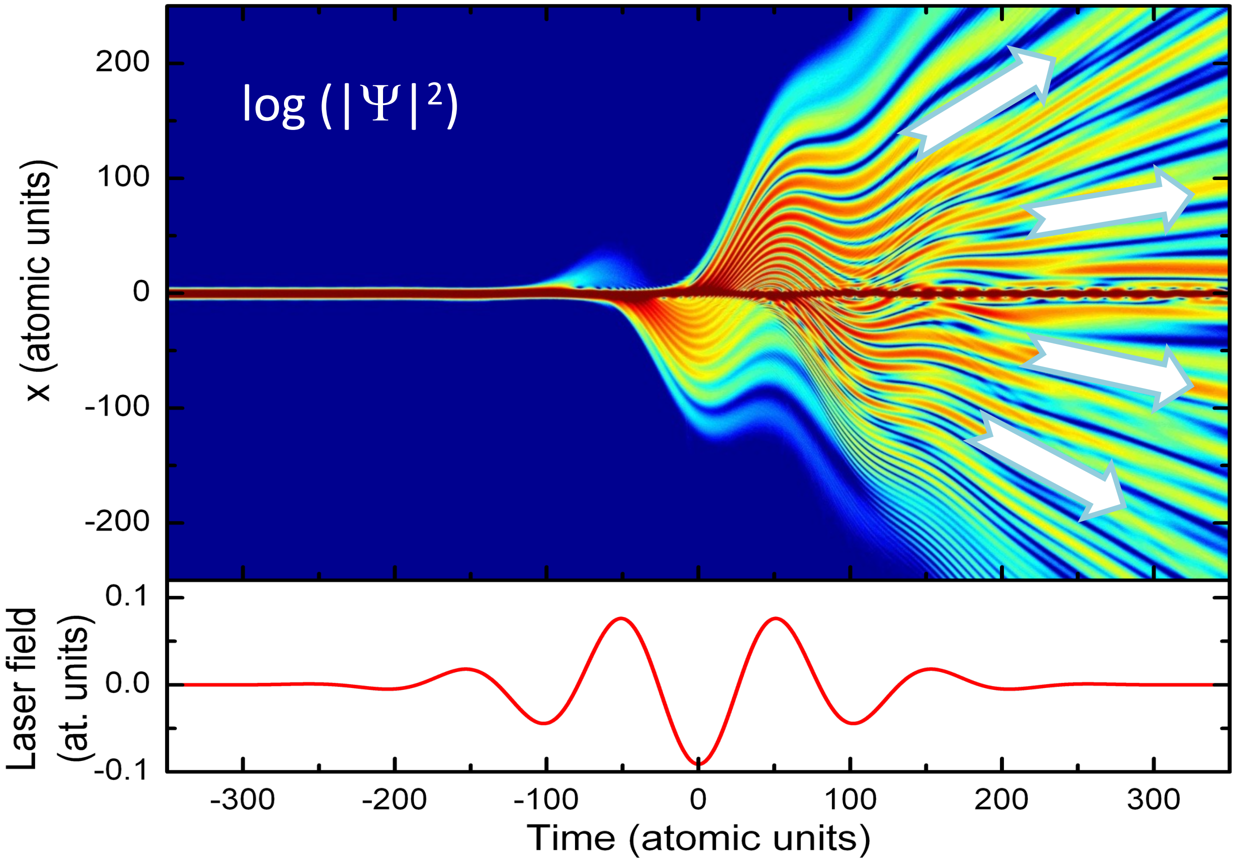
Wavepacket recollision

Laser-atom interaction



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The ionized wavepacket finally leaves its parent atom

Three-steps: the essence of strong field processes

Recollision

Ionization

Acceleration

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Strong Field processes in classical mechanics

Simple assumptions:

1) After ionization, electron motion described by Newtonian mechanics under the laser field (**Coulomb field neglected**):

$$-e\boldsymbol{E} = \frac{d\boldsymbol{k}}{dt}$$

electron charge

laser electric field

electron momentum: *m***v**

Since the vector potential ***A*** is given by:

$$\boldsymbol{E} = -\frac{d\boldsymbol{A}}{dt}$$

we get:

$$\frac{d}{dt}(\boldsymbol{k} - e\boldsymbol{A}) = 0$$

$\boldsymbol{P} = (\boldsymbol{k} - e\boldsymbol{A})$ called *canonical momentum,* is constant along the electron trajectory

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Strong Field processes in classical mechanics

2) At ionization (time t_i) the electron is in $\boldsymbol{r}(t_i) = 0$ with initial velocity $\boldsymbol{v}(t_i) = 0$

$$\boldsymbol{k}(t_i) = \boldsymbol{P} + e\boldsymbol{A}(t_i) = 0 \Longrightarrow \boldsymbol{P} = -e\boldsymbol{A}(t_i)$$

hence $\boldsymbol{v}(t) = \frac{\boldsymbol{k}(t)}{m} = \frac{\boldsymbol{P}+e\boldsymbol{A}(t)}{m} = \frac{e[\boldsymbol{A}(t)-\boldsymbol{A}(t_i)]}{m}$

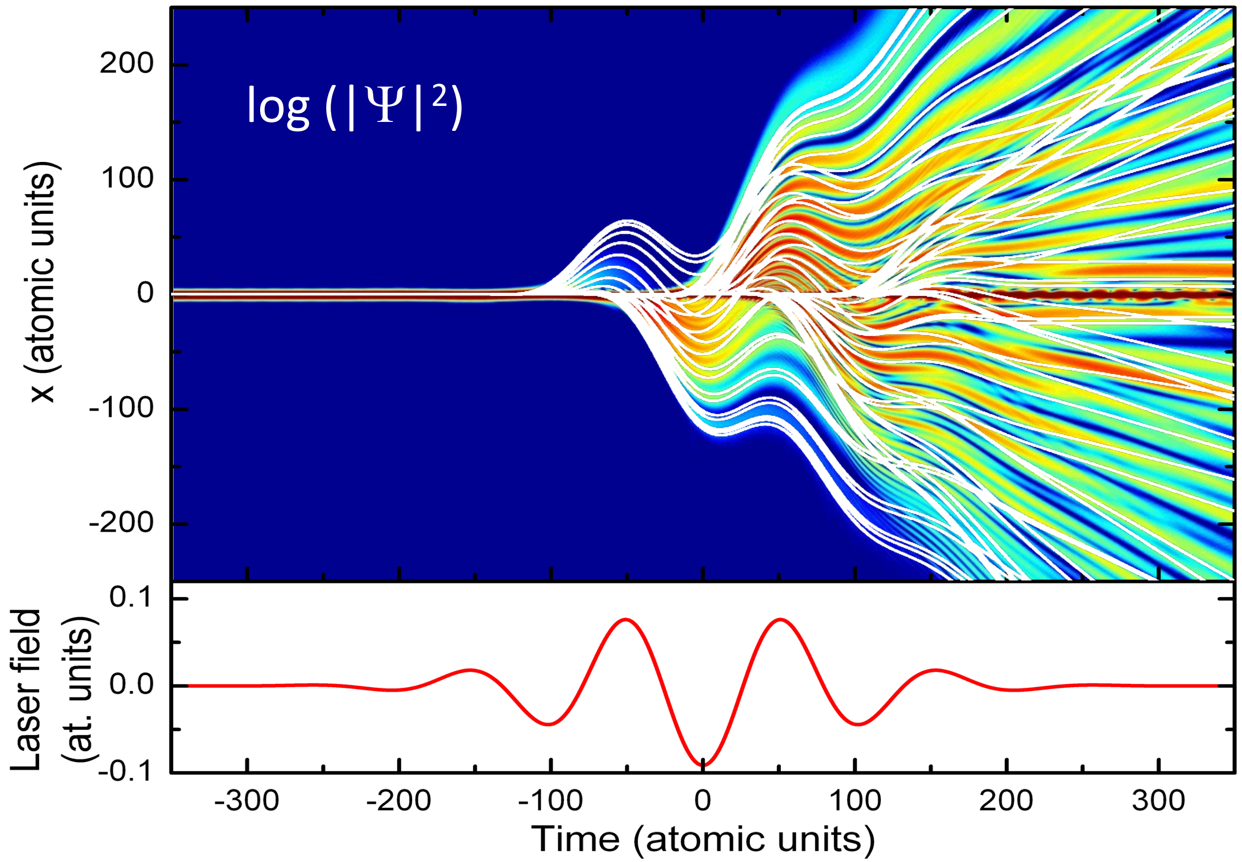
and then $\boldsymbol{r}(t_i, t) = \int_{t_i}^{t} \boldsymbol{v}\, dt = \int_{t_i}^{t} \frac{e[\boldsymbol{A}(t)-\boldsymbol{A}(t_i)]}{m} dt$

Electron trajectory for a given ionization time t_i

Ok… but I said that only quantum models are fine!

Why I'm bothering you with Newtonian mechanics?

Classical mechanics is not so bad…



- Classical electron trajectories follow qualitatively the wavefunction evolution
- Thus we can still keep a qualitative "particle" description of the processes
- However only 3D TDSE gives a quantitative picture (and it is computationally demanding)

Laser-atom interaction in SFR

Several phenomena take place during the interaction:

- ❖ Direct Photoionization (tunneling regime)
- ❖ Recombination of the freed electron after some excursion in the continuum (emission of radiation)
- ❖ Elastic scattering of the ionized electron in the ionic potential (Above Threshold Ionization etc.)
- ❖ Inelastic scattering of the ionized electron (Nonsequential Double Ionization etc.)

High order Harmonic Generation (HHG)

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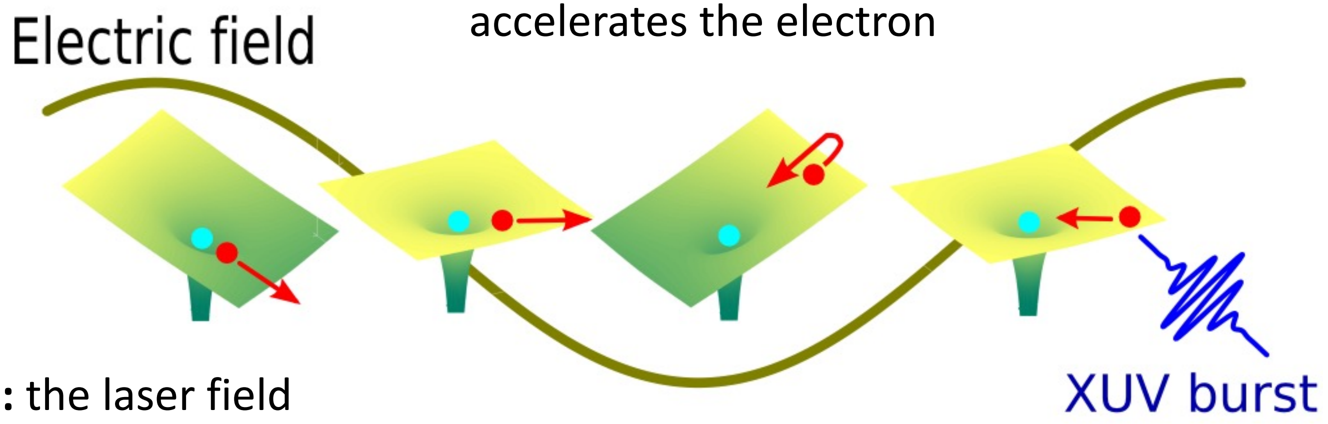
HHG: three-step model

Proposed by Corkum and Kulander et al. in 1993

2: the laser field
accelerates the electron



1: the laser field
ionizes the electron

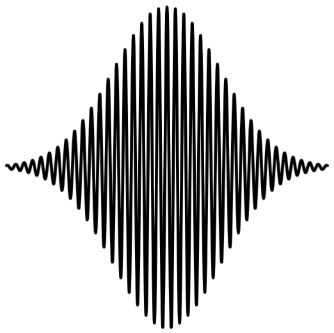
3: the electron collides with the parent
ion emitting a burst of XUV-soft X light

- XUV (eXtreme Ultra Violet) : 10-300 eV
- soft X Rays: 300-3000 eV

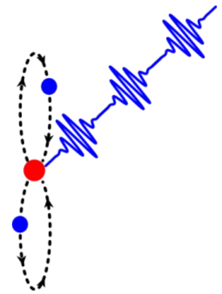
HHG: three-step model

Proposed by Corkum and Kulander et al. in 1993

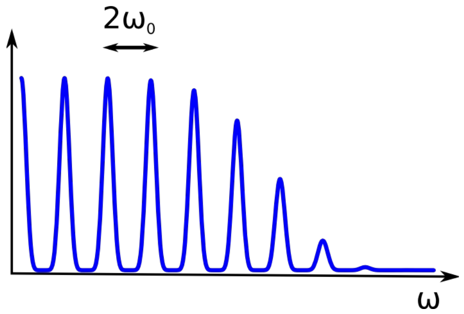
"Long" laser pulse



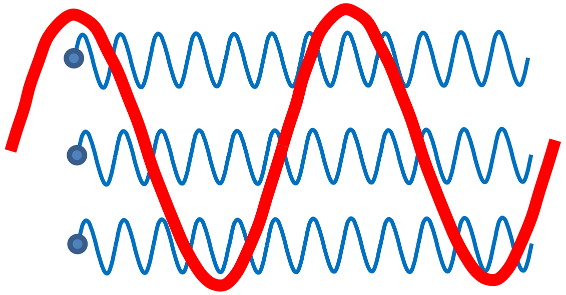
Multiple XUV bursts



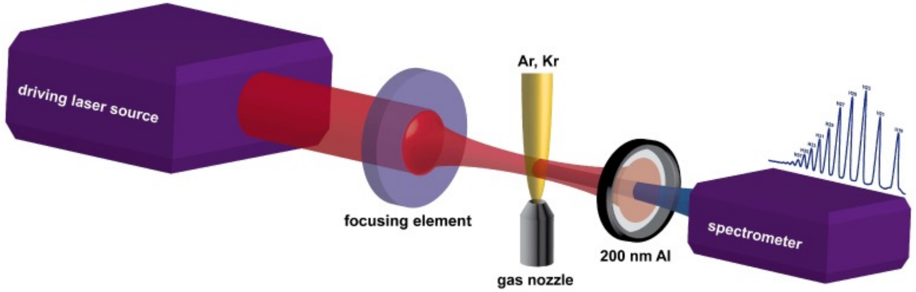
Odd harmonics
in spectral domain (HHG)



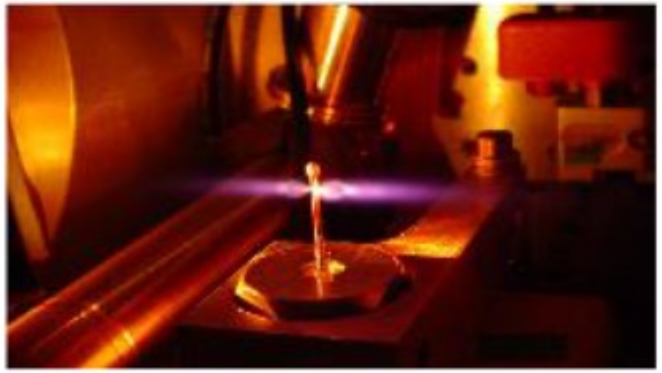
HHG is a coherent process:
all the atoms in the macroscopic medium are driven by the same laser field



Typical HHG experimental spectra

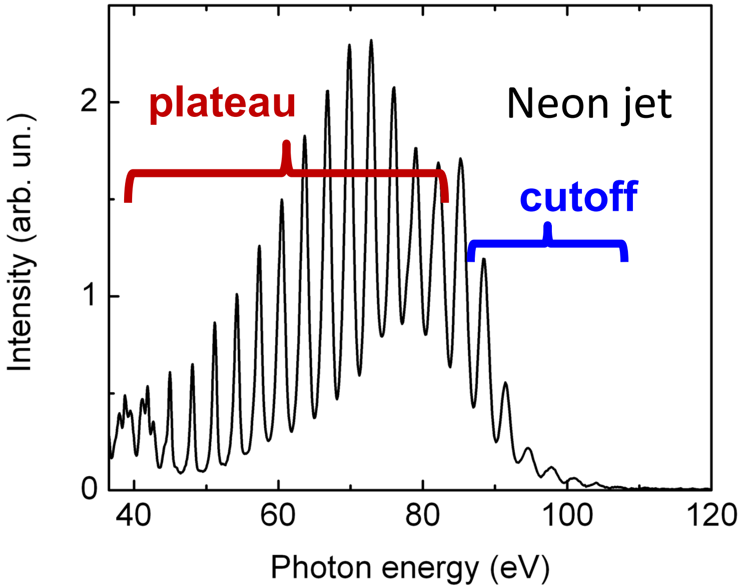


credits: S. Hädrich et al., *Opt. Express* **18,** 20242 (2010)



credits: J. Seres et al., *Photonics* **2**, 104 (2015)

Acquired in our laboratory (2016)



- HHG spectra are typically produced in gas jets/cells inside vacuum chambers
- Spectral analysis performed with grazing-incidence XUV spectrometers
- The HHG spectra present a plateau and a cutoff region

Typical features of HHG in gases

Spectral region: from 80 nm to about 1 nm

Generation yield: 10^{-4} to 10^{-7} according to laser parameters and driven target

Energy per shot: few μJ to few pJ

Harmonic beam divergence: few mrad

Duration of a single light burst: 1 fs to tens of attoseconds (1 as = 10^{-18} s)

Polarization: **linear** (more about this afterwards)

HHG in classical mechanics

Simple assumptions:

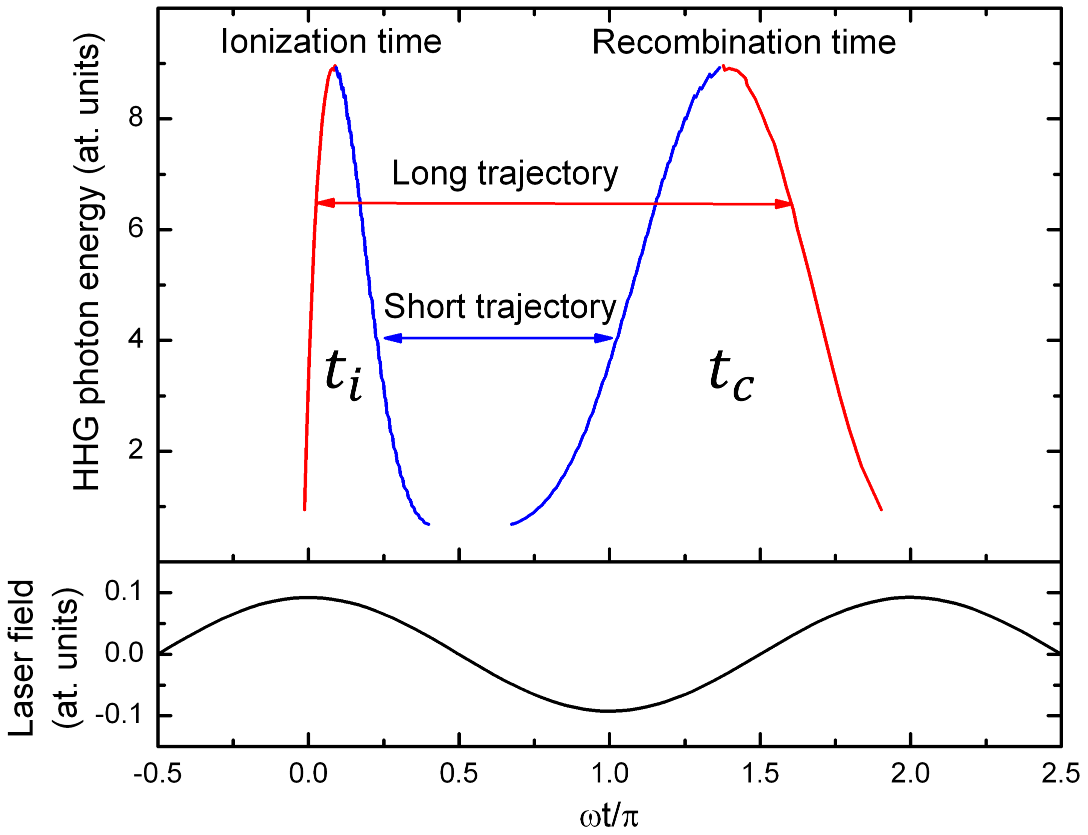
2) $$\boldsymbol{r}(t_i,t) = \int_{t_i}^{t} \boldsymbol{v}\, dt = \int_{t_i}^{t} \frac{\boldsymbol{P} + e\boldsymbol{A}(t)}{m} dt = \int_{t_i}^{t} \frac{e[\boldsymbol{A}(t) - \boldsymbol{A}(t_i)]}{m} dt$$

3) At recombination (time t_c) the electron collides with the ion in $\boldsymbol{r}(t_i, t_c) = 0$ and its kinetic energy (plus ionization energy) is released as a photon of frequency ω:

$$\frac{k^2(t_c)}{2m} + I_p = \frac{|\boldsymbol{P} + e\boldsymbol{A}(t_c)|^2}{2m} + I_p = \frac{e^2|\boldsymbol{A}(t_c) - \boldsymbol{A}(t_i)|^2}{2m} + I_p = \hbar\omega$$

Only few electron trajectories contribute to HHG

HHG: classical predictions



Two electron trajectories contribute to high order harmonic generation:

- **Short trajectory**: electron flight time shorter than half laser optical cycle
- **Long trajectory**: electron flight time comparable to an optical cycle

Different photon energies ħω are emitted at different times t_c ! (Attochirp)

Quantum modelling of HHG

Modelling of HHG by the TDSE is a computationally intensive task

⇩

An approximated (semiclassical) model is required

Lewenstein model:

- Single active electron
- Role of excited bound states neglected
- Ionized electron wavepacket: ***plane waves*** driven by the laser field (atomic field neglected)
- Plug these assumptions into the TDSE and find an approximated solution

M. Lewenstein et al., *Phys. Rev. A* **49**, 2117 (1994).

Lewenstein model

Define:

- the *quasi-classical action* $S(\boldsymbol{P}, t_i, t_c) = \int_{t_i}^{t_c} \left\{ \frac{[\boldsymbol{P}+e\boldsymbol{A}(t\prime)]^2}{2m} + I_p \right\} \frac{dt'}{\hbar}$, with I_p ionization potential of the atom

recollision times

ionization times

- the *bound-free transition dipole moment* $\boldsymbol{D}(\boldsymbol{k}) = \langle \Psi_0 | -e\boldsymbol{r} | \boldsymbol{e}^{i\boldsymbol{k}\cdot\boldsymbol{r}/\hbar} \rangle$

then the HHG spectral intensity is given by:

step 2: motion in the continuum

$$I(\omega) \propto \left| \omega^2 \int_{-\infty}^{\infty} e^{i\omega t_c}\, dt_c \int_{-\infty}^{t_c} dt_i \int d^3\boldsymbol{P}\ \boldsymbol{D}^*[\boldsymbol{P} + e\boldsymbol{A}(t_c)]\ e^{-iS(\boldsymbol{P},t_i,t_c)}\ \boldsymbol{E}(t_i) \cdot \boldsymbol{D}[\boldsymbol{P} + e\boldsymbol{A}(t_i)] \right|^2$$

step 3: recollision

step 1: ionization

M. Lewenstein et al., *Phys. Rev. A* **49**, 2117 (1994).

Lewenstein model: luckily just few quantum paths are dominant

The Lewenstein model requires integration over infinite possible paths…

However, few paths are likely to be taken by the system, thus the integral can be reduced to a discrete sum…

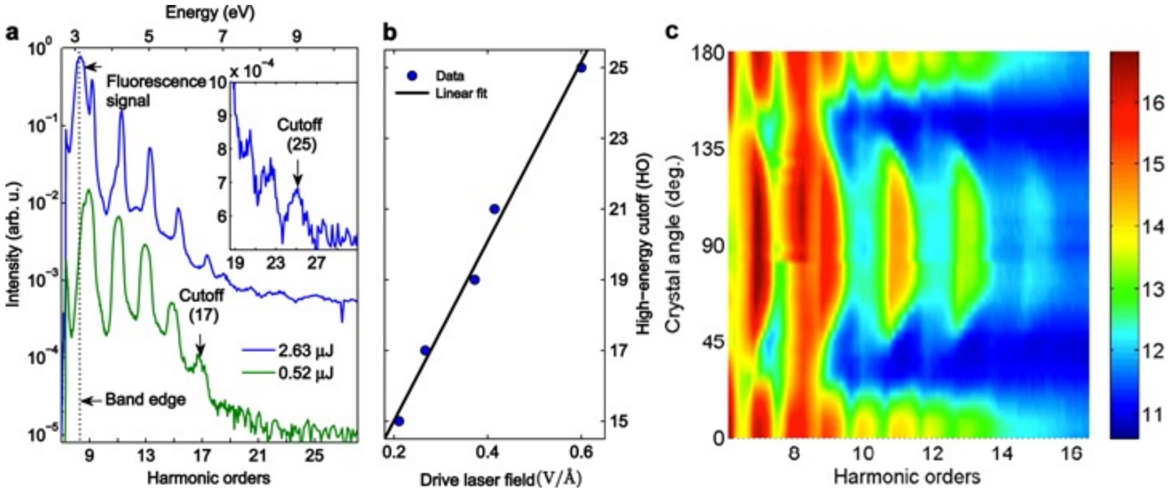
Those relevant paths can be found by suitable approaches (stationary solutions)

P. Salieres et al., *Science* **292**, 902 (2001).

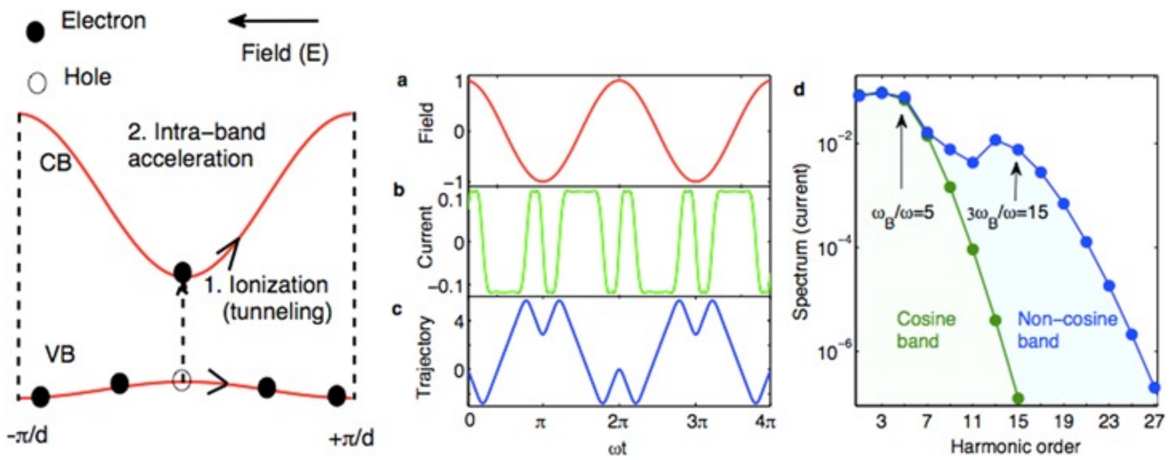
From gases to crystals

Strong-field physics in solids: an unexpected outcome

UNEXPECTED: High harmonic generation in crystals

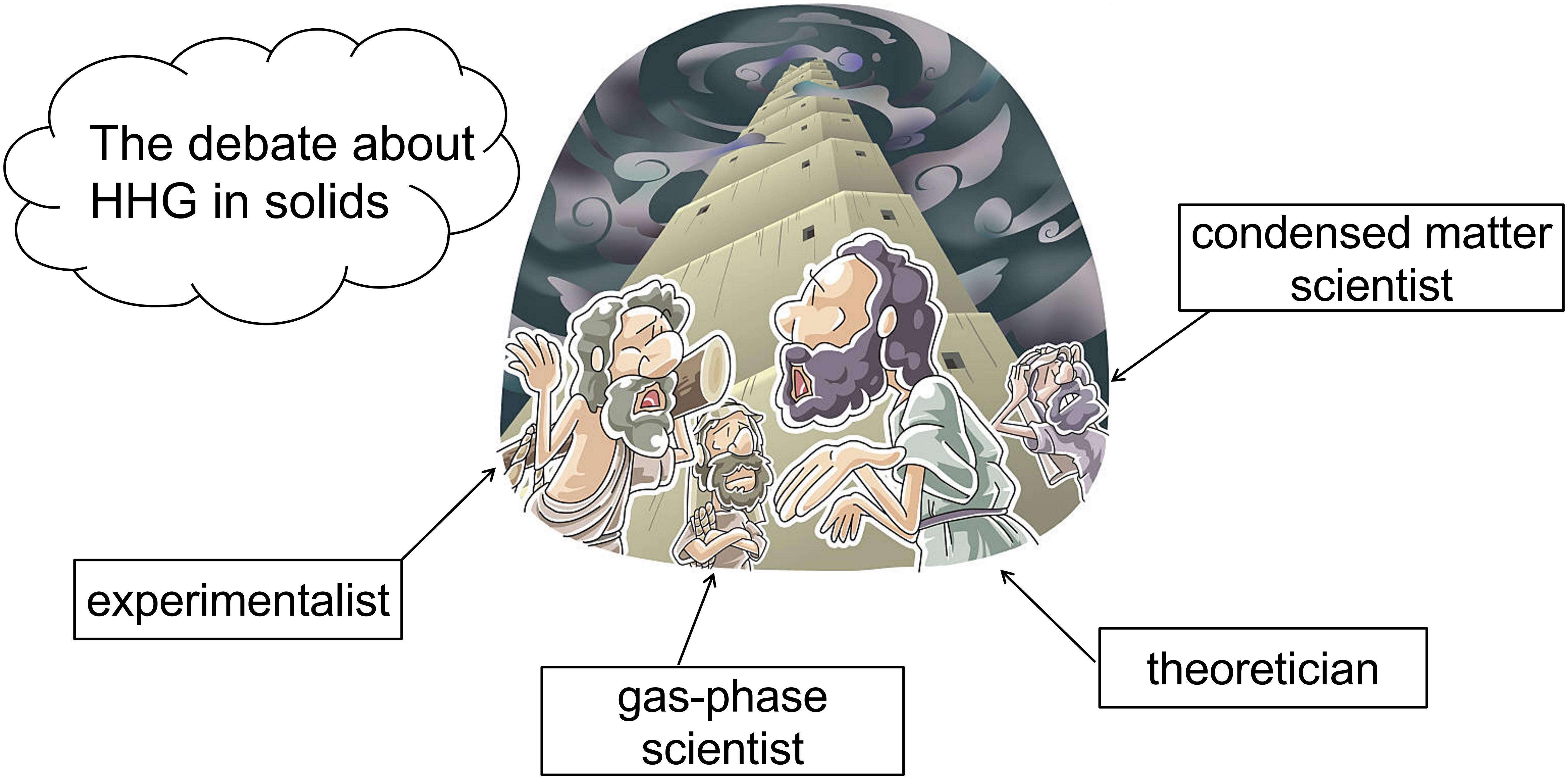


S. Ghimire et al., *Nat. Phys.* **7** 138 (2011)



HHG nowadays observed in several crystals:

ZnSe, ZnTe, GaAs, SiO_2, ZnO, MgO, GaSe, graphene


The debate about HHG in solids
condensed matter scientist
experimentalist
gas-phase scientist
theoretician

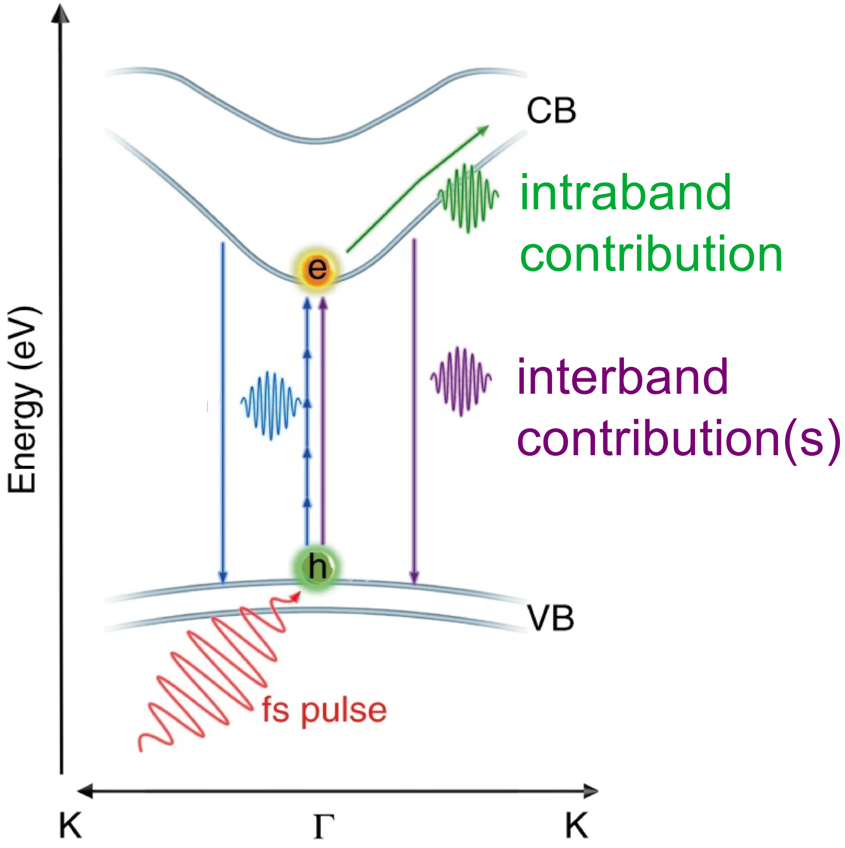
HHG in solids: the three-step model is there

Three step model in the momentum k space

1. Electron-hole creation
2. Propagation of electron and hole in the respective band:
   a. intraband contribution to HHG
3. Electron-hole recombination:
   b. interband contributions to HHG

HHG emission encodes information on:

- electronic band structure
- ultrafast dynamics triggered by fs pulse



Han, S., Ortmann, L.,
Nat. Commun ***10,*** *3272 (2019)*

A typical case: HHG in ZnTe

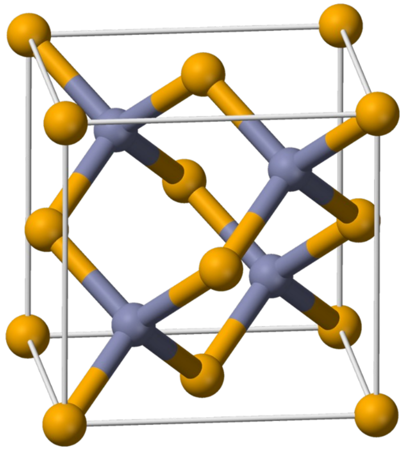
sample: ZnTe (Zinc Telluride)

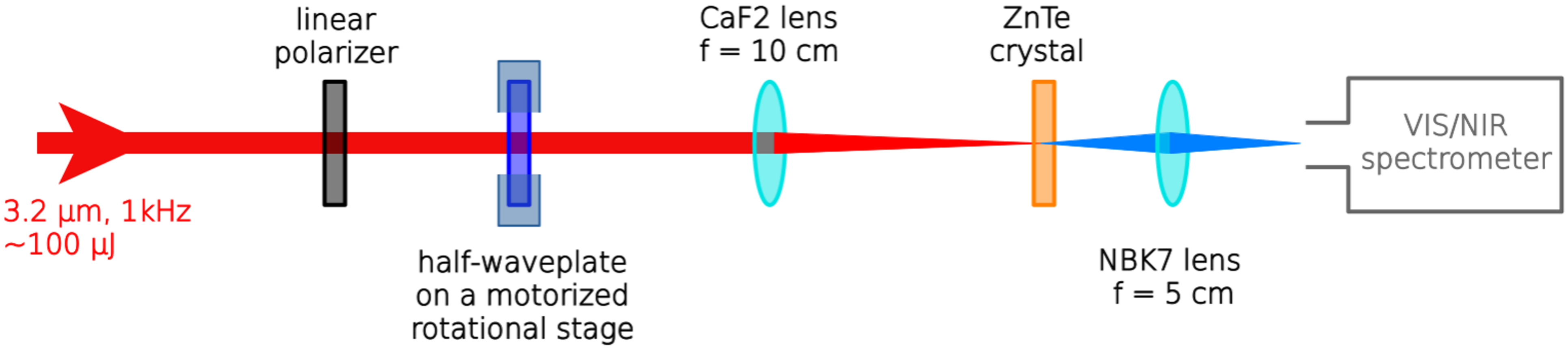
non centrosymmetric crystal (odd and even harmonics)

bandgap: 2.26 eV

transmission geometry

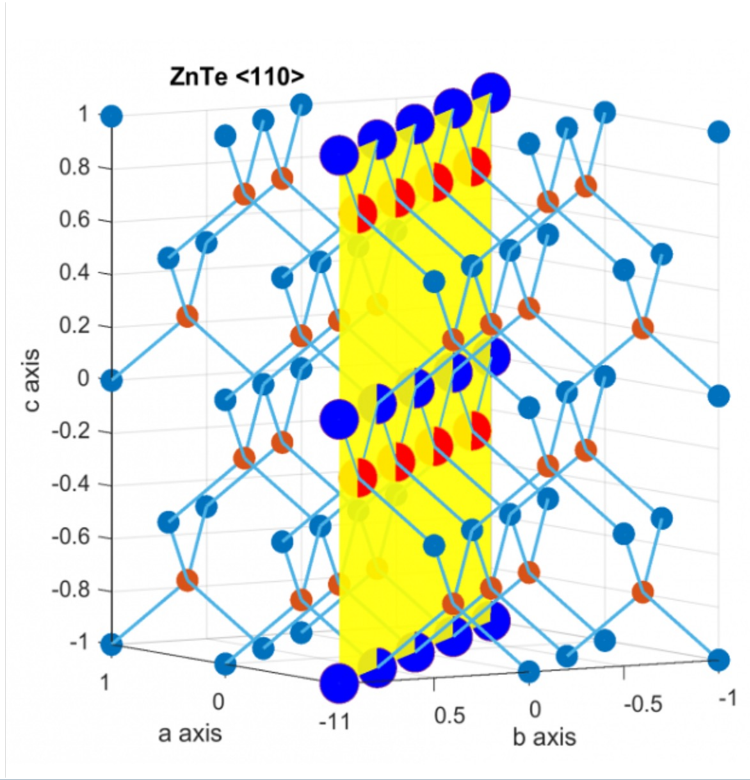
scan of polarization direction with half-waveplate on motorized stage



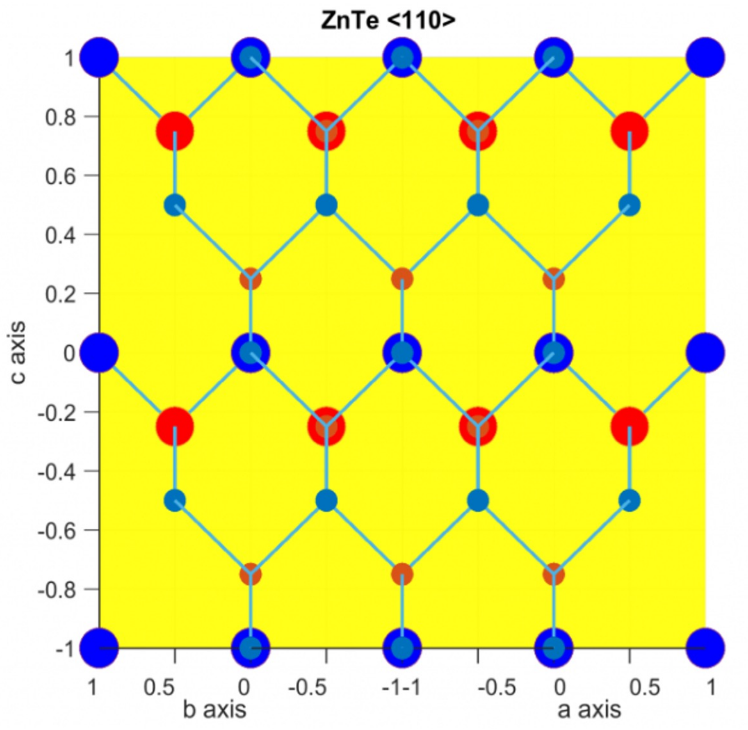


ZnTe lattice and laser polarization

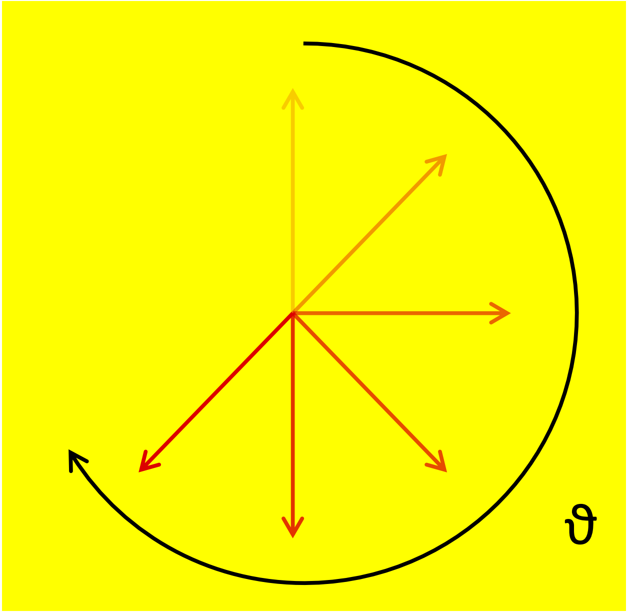
zincblende structure



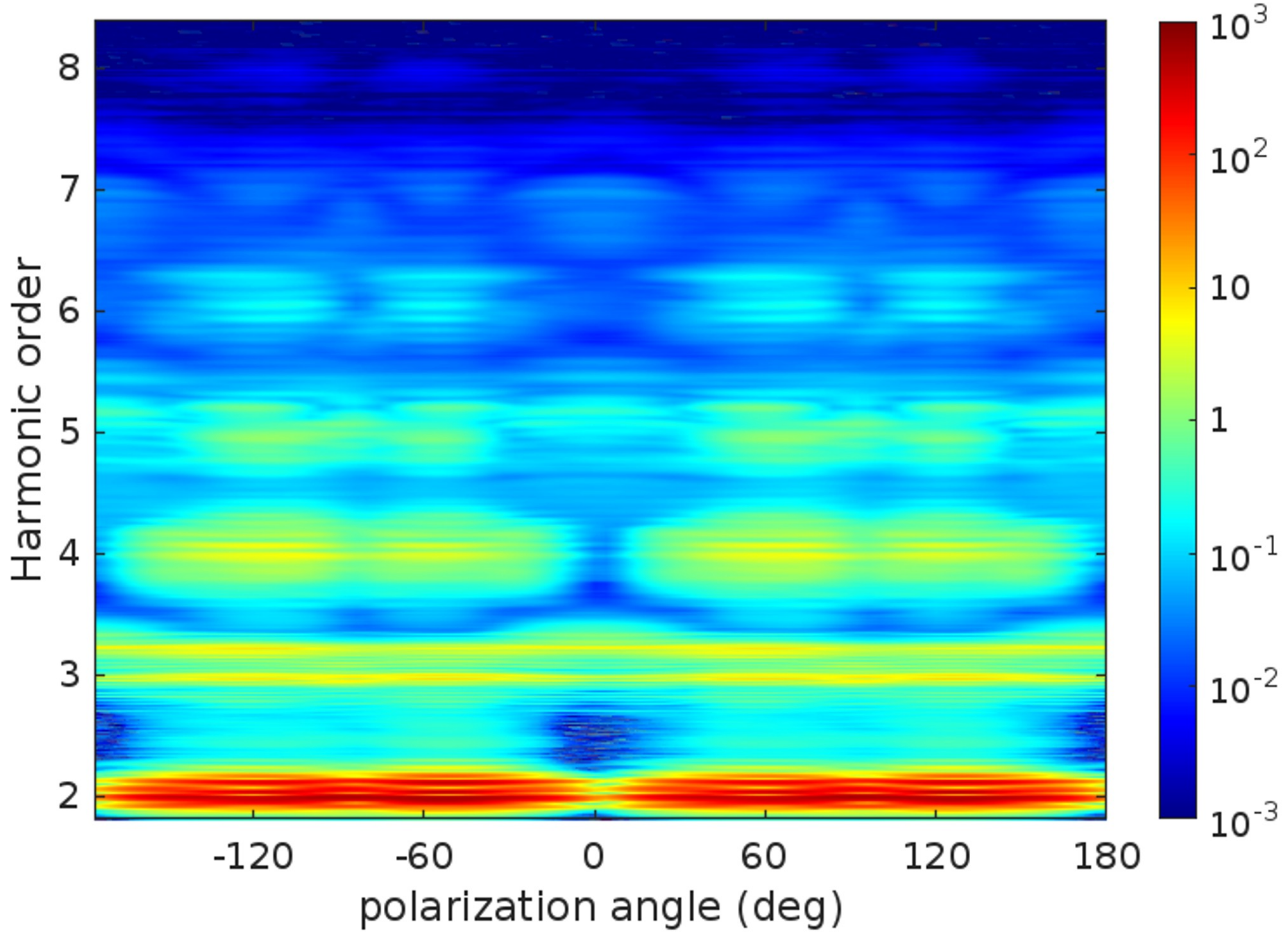
ZnTe cut along the 110 plane

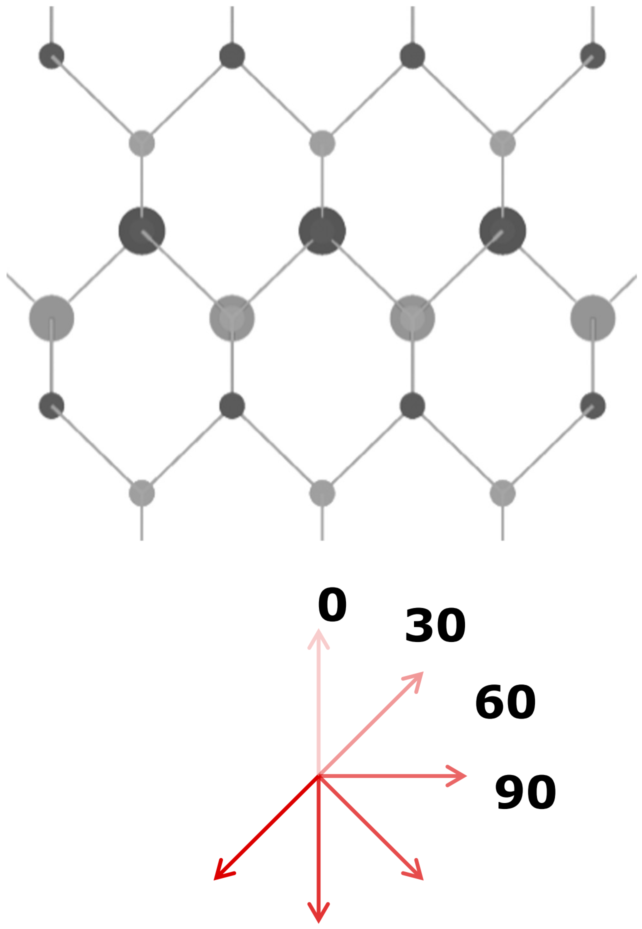


Laser field polarization



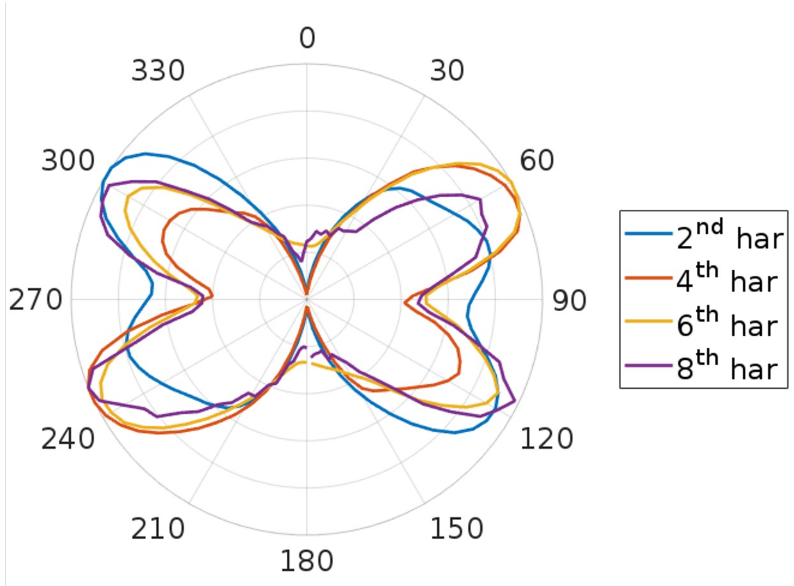
HHG spectra vs. ZnTe crystal orientation



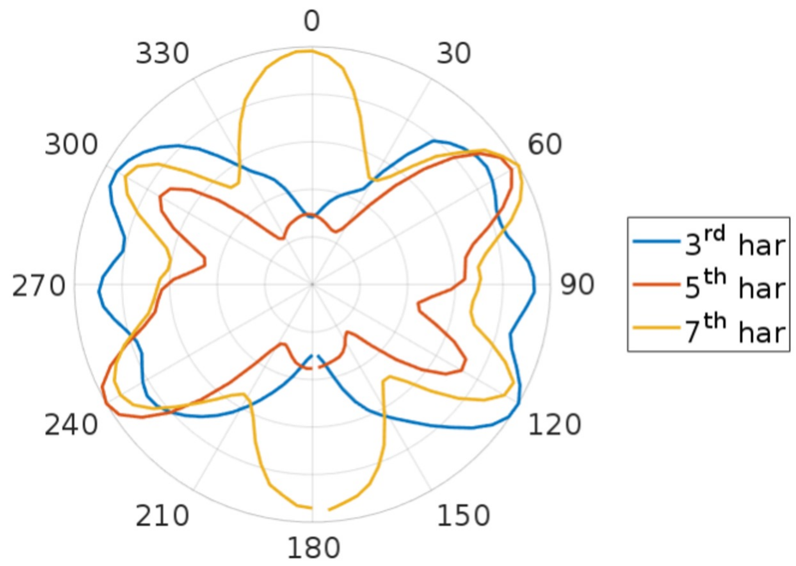


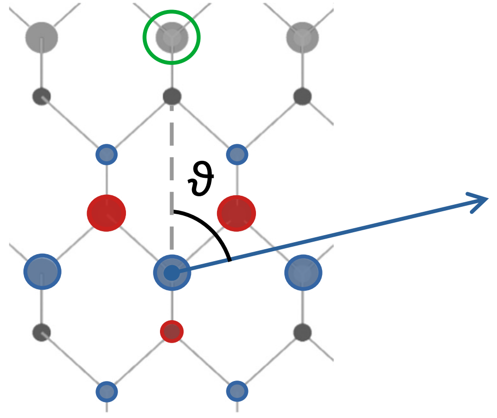
Even vs. odd harmonics in ZnTe

Even harmonics



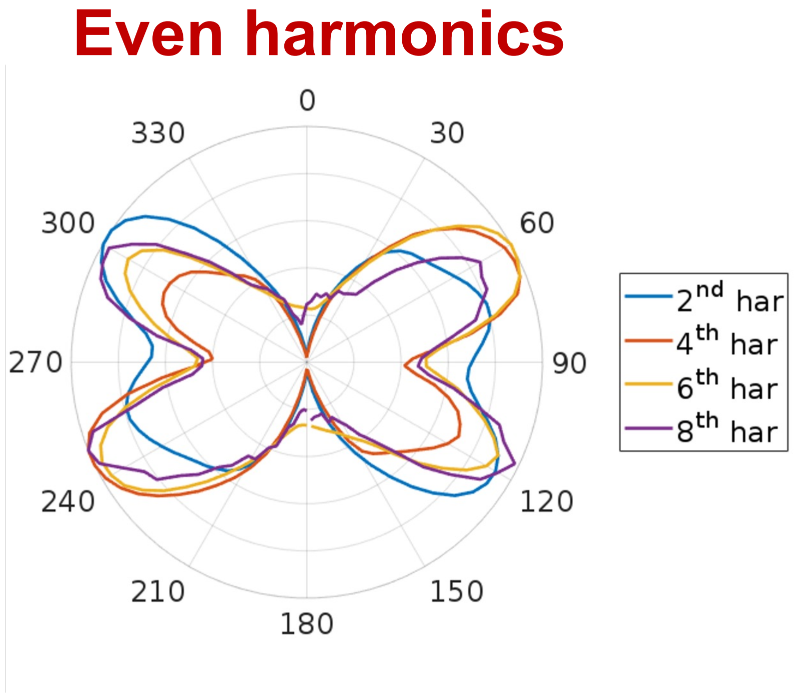
Odd harmonics

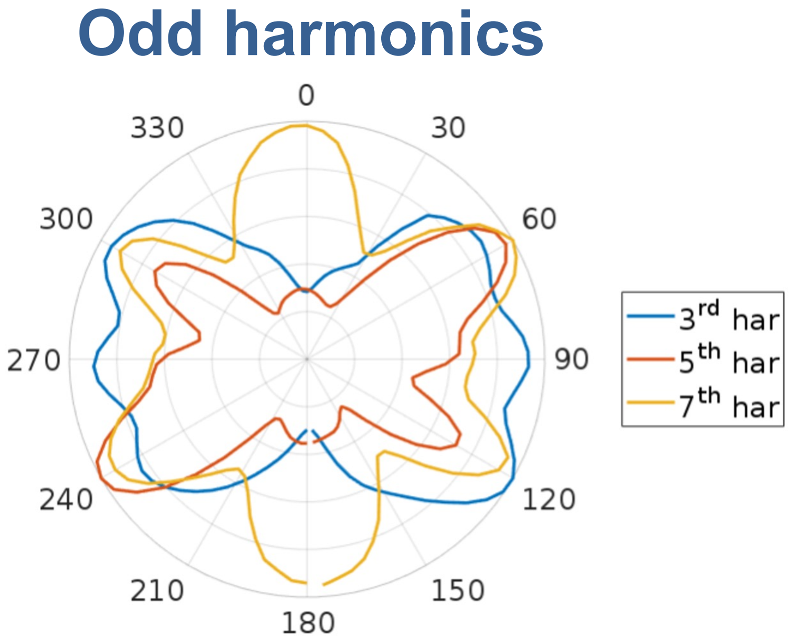


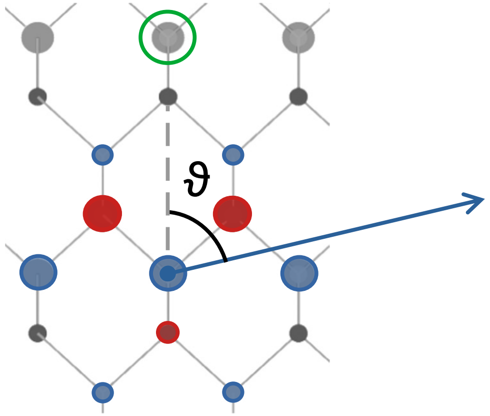


Even & odd harmonics maximize along directions connecting in-plane Zn and Te sites (no inversion symmetry along them)

Even vs. odd harmonics in ZnTe



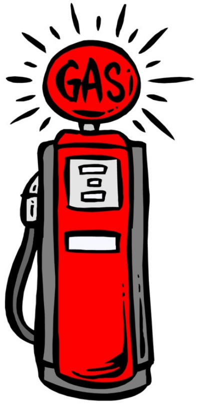




Odd harmonics also maximize along directions connecting in-plane similar atomic sites (inversion symmetry is present along them)

Peculiarities of HHG in solids

- Low damage threshold limits the use of arbitrary high intensities
- Longer wavelength and shorter duration pulses are needed, compared to gases



- Both even and odd high harmonics may be observed
- The polarization state of the harmonics can be non trivial
- Detection in transmission and in reflection are both feasible (only reflection in opaque samples)

Applications of HHG (in gases)

Structural information in HHG spectra

The spectral intensity of harmonic radiation can be approximated as:

colliding wavepacket
(plane wave with wavevector **k**)

amplitude of the
colliding wavepacket

$$I(\omega) \approx \omega^4 \left| a(\boldsymbol{k}) \int -e\boldsymbol{r}\, \Psi_0(\boldsymbol{r}) e^{-i\boldsymbol{k}(\omega)\cdot\boldsymbol{r}} d^3\boldsymbol{r} \right|^2$$

The harmonic spectrum is linked to the spatial Fourier transform of the atomic/molecular ground state wavefunction $\boldsymbol{\Psi_0}$ (times $-\boldsymbol{r}$)

The wavevector **k** of the colliding electron is related to the emitted photon energy $\hbar\omega$ and the ionization potential I_p by **energy conservation:**

$$|\hbar\, \mathbf{k}|^2/2m_e + I_p = \hbar\omega$$

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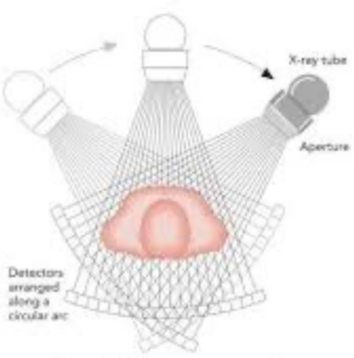
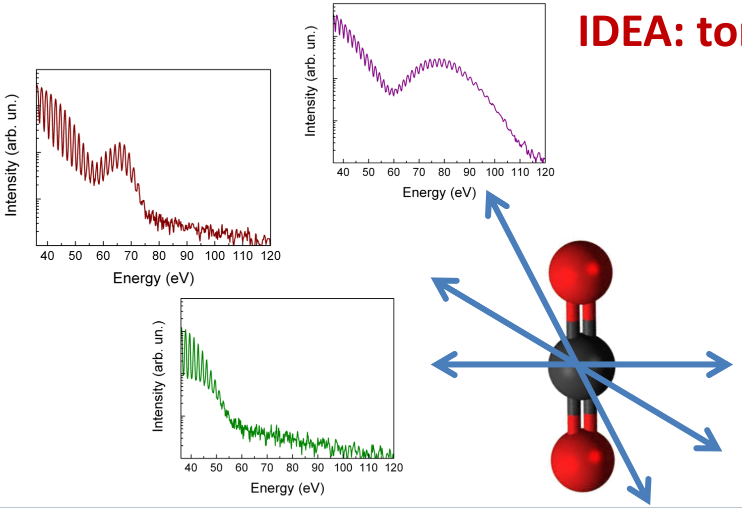
Molecular orbital tomography

Computed tomography of a human being



Some complex algorithm

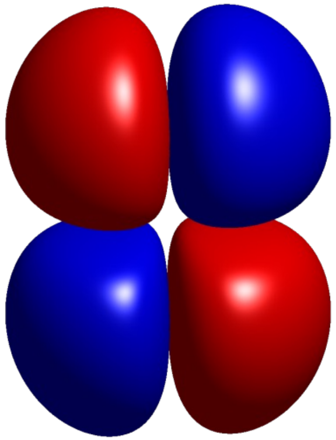
IDEA: tomography of a molecule with HHG



Some complex algorithm

Laser pulses at different polarization directions

Molecular orbital



J. Itatani et al., *Nature* **432**, 867 (2004)

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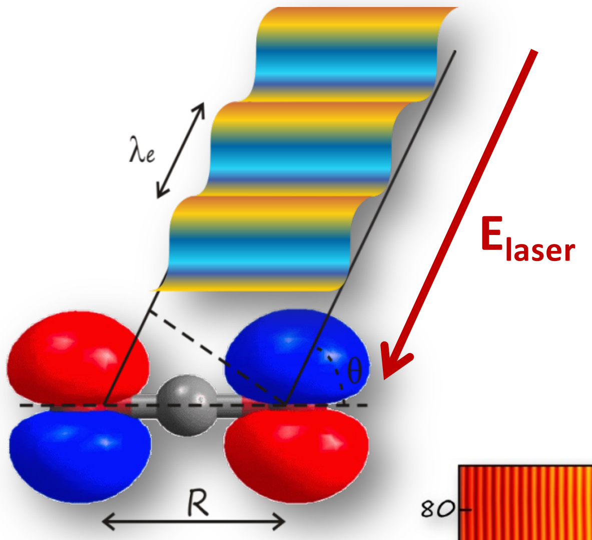
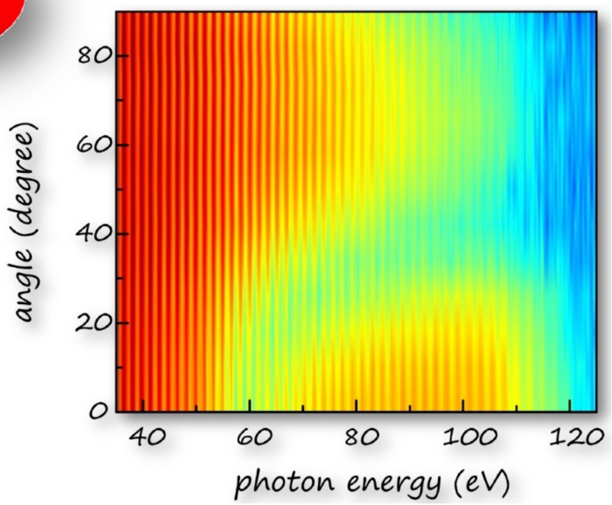
HHG tomography



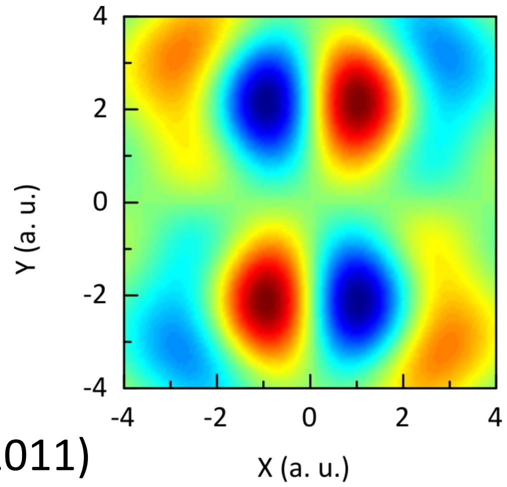


1) align the target molecules (**with a first laser pulse**)

2) collect HHG spectra I(ω,θ) at different angles θ between the molecular axis and the E field **of a second laser pulse**

3) obtain $\omega^4 \left| a(\mathbf{k}) \mathcal{F}_{\mathbf{k}(\omega)}\{\mathbf{r}\,\Psi_0(\mathbf{r})\} \right|^2$ from HHG spectra by mapping the optical frequencies ω to the spatial frequencies k

4) make some assumptions on $a(\mathbf{k})$, on the phase of $\mathcal{F}_{\mathbf{k}}\{\mathbf{r}\,\Psi_0(\mathbf{r})\}$ and on the orbital symmetry; then inverse transform.

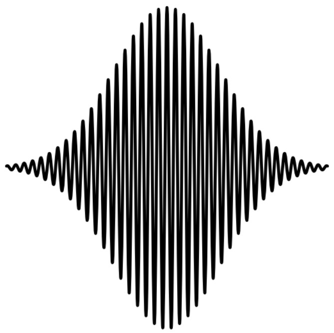


Retrieved orbital in CO_2
C. Vozzi et al., *Nature Phys.* **7**, 822 (2011)

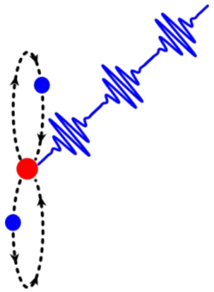
Attosecond Science

HHG in time domain

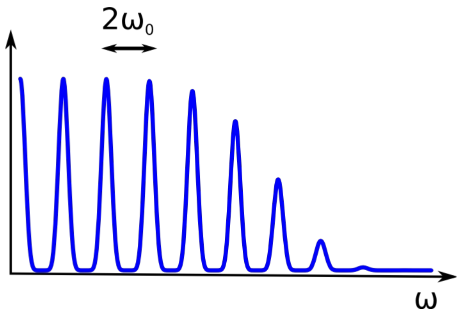
"Long" laser pulse



Multiple XUV bursts

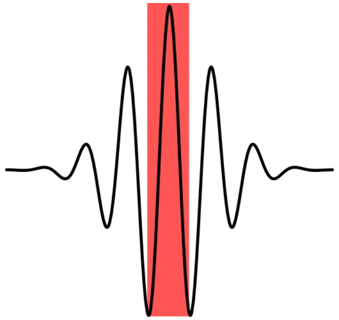


Odd harmonics
in spectral domain (HHG)

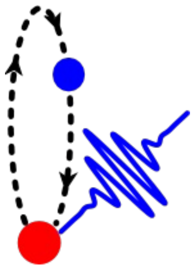


Nobel Prize in Physics 2023 to Pierre Agostini, Ferenc Krausz and Anne L'Huillier "for experimental methods that generate attosecond pulses of light for the study of electron dynamics in matter"

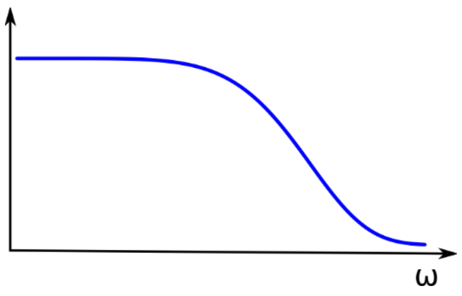
"Short" laser pulse +
gating technique



Single attosecond pulse



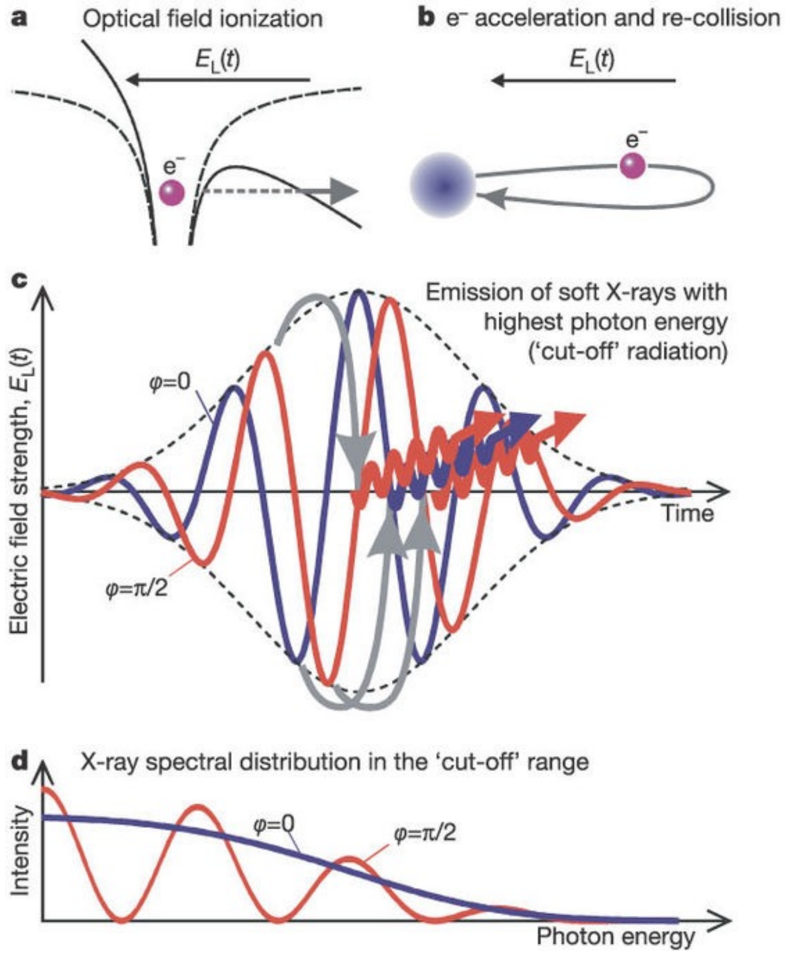
Continuous XUV spectrum



Attosecond Science and Carrier-Envelope Phase

The Carrier-Envelope Phase Offset of the driving laser pulses matters!

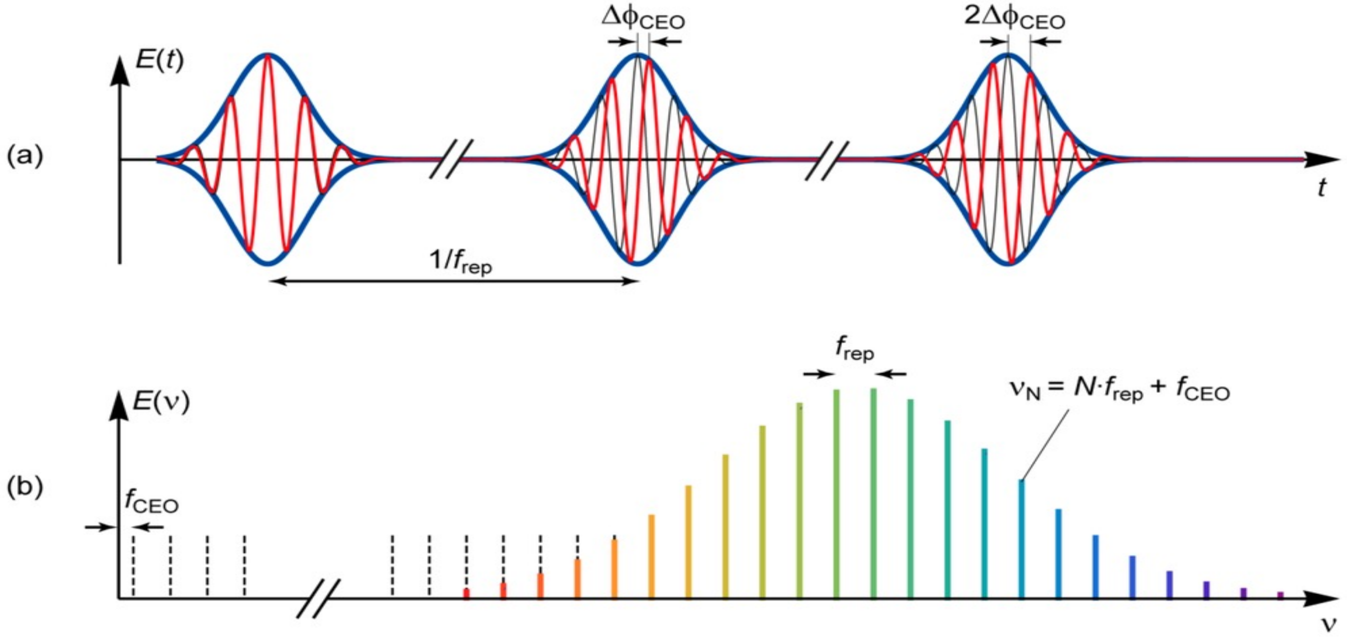
It must be **stabilized** in order to produce reliable attosecond pulses



from A. Baltuska et al, Nature 421, 611 (2003)

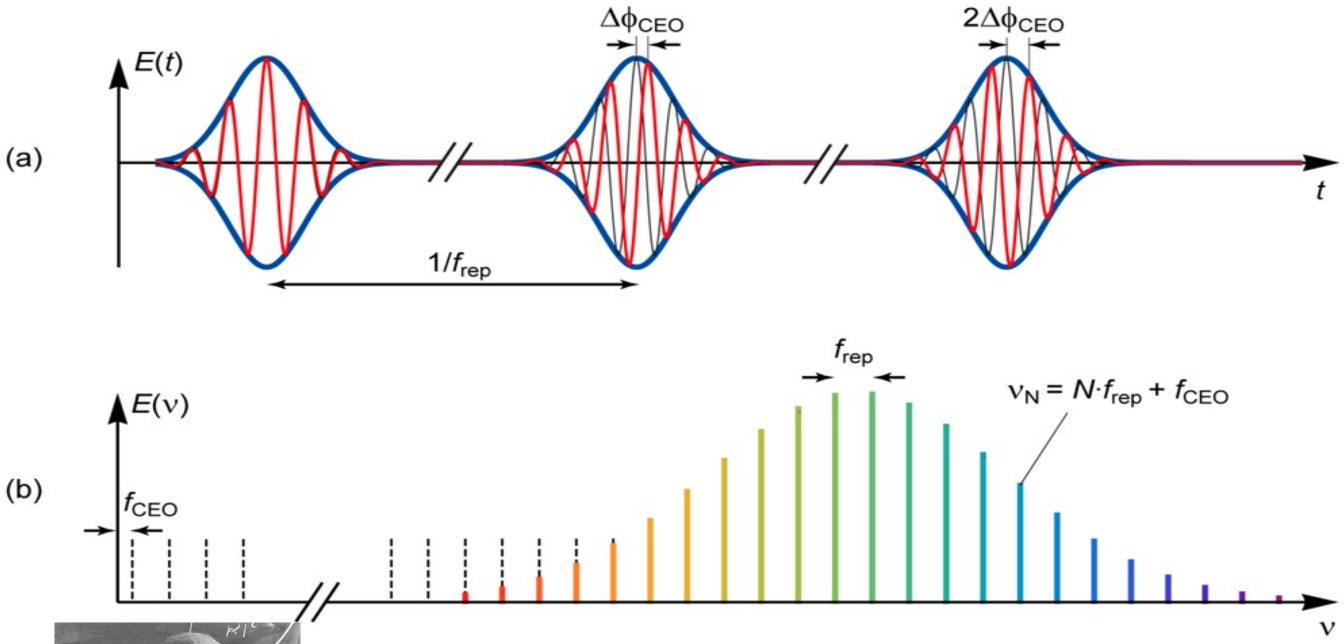
Attosecond Science and Carrier-Envelope Phase

Carrier-envelope phase (CEP) of laser pulses:
changes from pulse to pulse with rate f_{CEO} **which is not stable over time!**

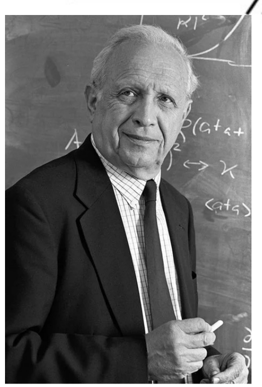


A train of laser pulses corresponds to a frequency comb in the spectral domain
with comb frequencies drifting as f_{CEO} changes over time

Attosecond Science and Carrier-Envelope Phase



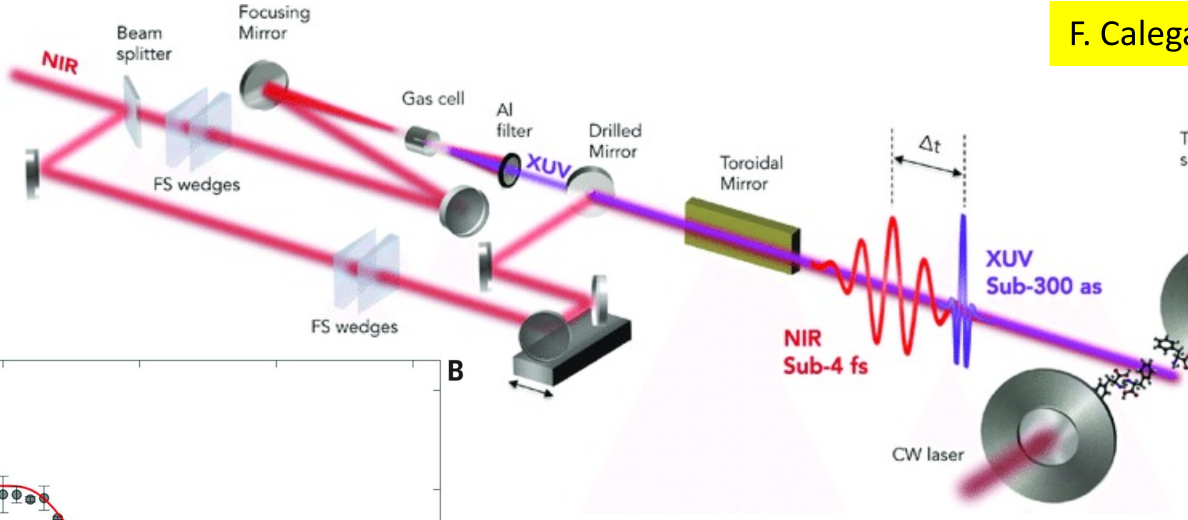
Nobel Prize in Physics 2005 to John L. Hall and Theodor W. Hänsch “for their contributions to the development of laser-based precision spectroscopy, including the **optical frequency comb technique**”…

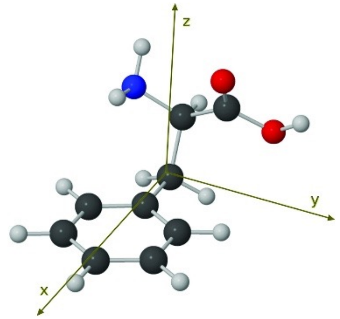


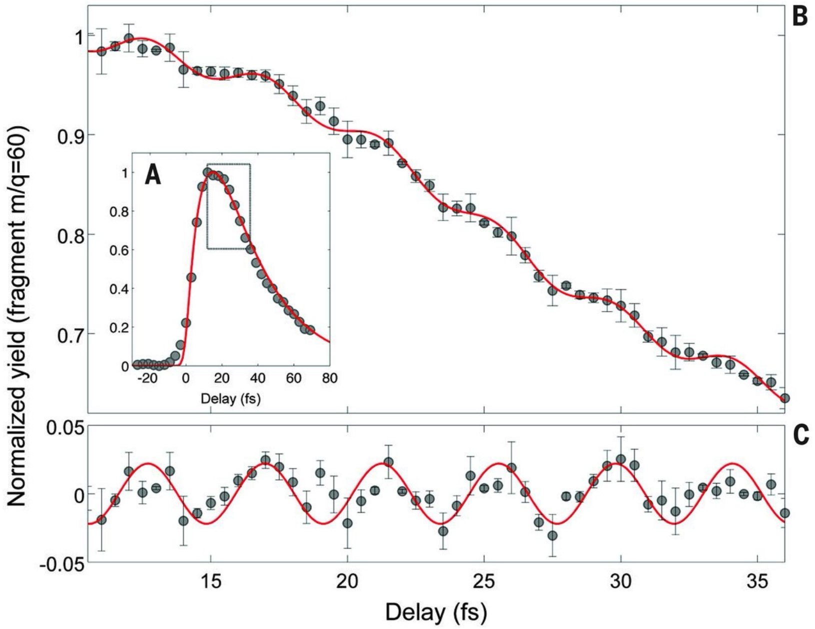
… and to Roy J. Glauber “for his contribution to the quantum theory of optical coherence”

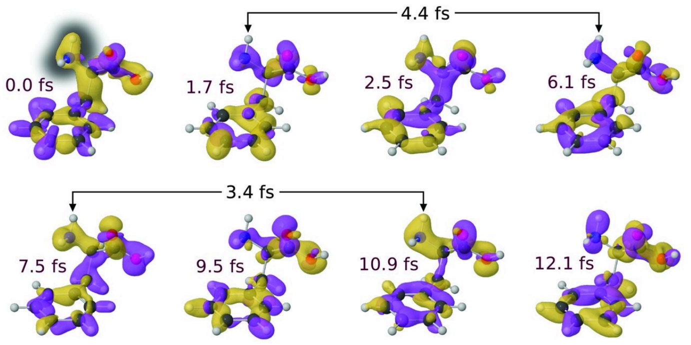
Ultrafast electron dynamics in phenylalanine initiated by attosecond pulses

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



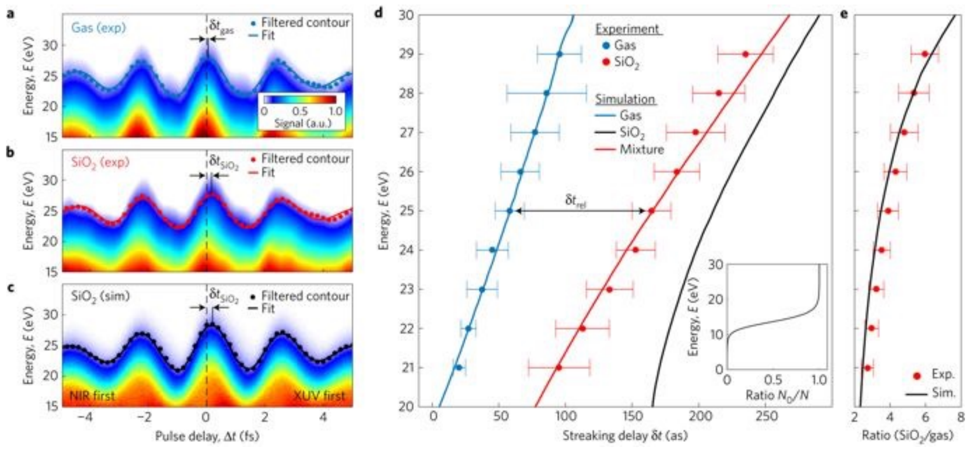


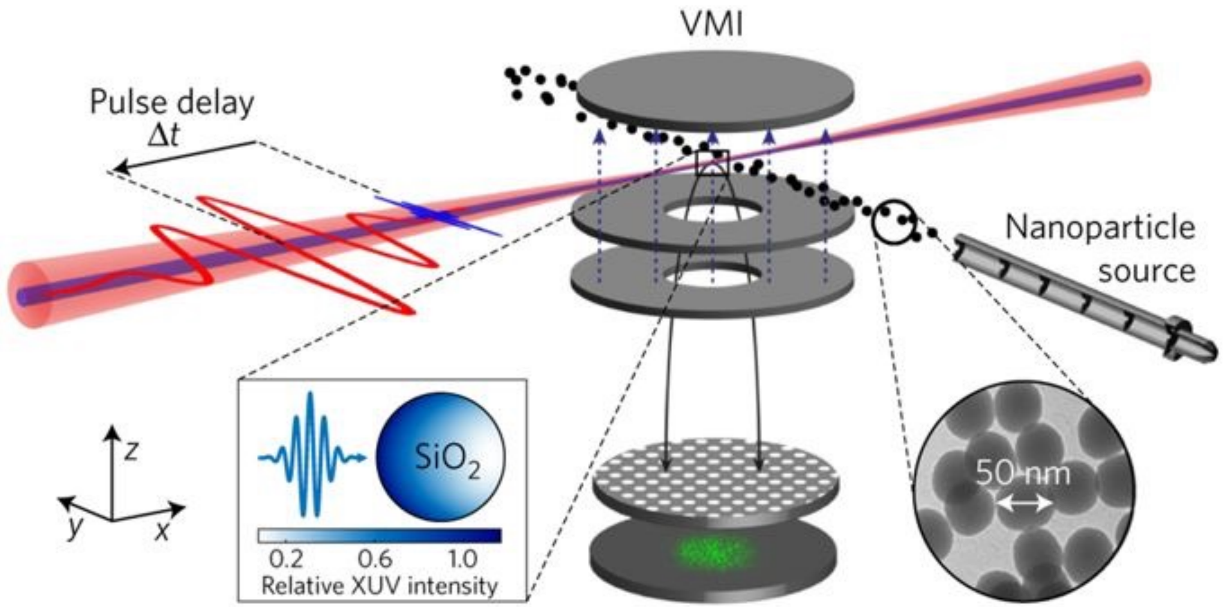




- Ionization induced by attosecond pulse
- doubly charged fragment produced by a probe pulse by ejection of a second electron
- charge migration manifests as a sub-4.5-fs oscillation in the yield of this fragment as a function of pump-probe delay.

Attosecond chronoscopy of electron scattering in dielectric nanoparticles



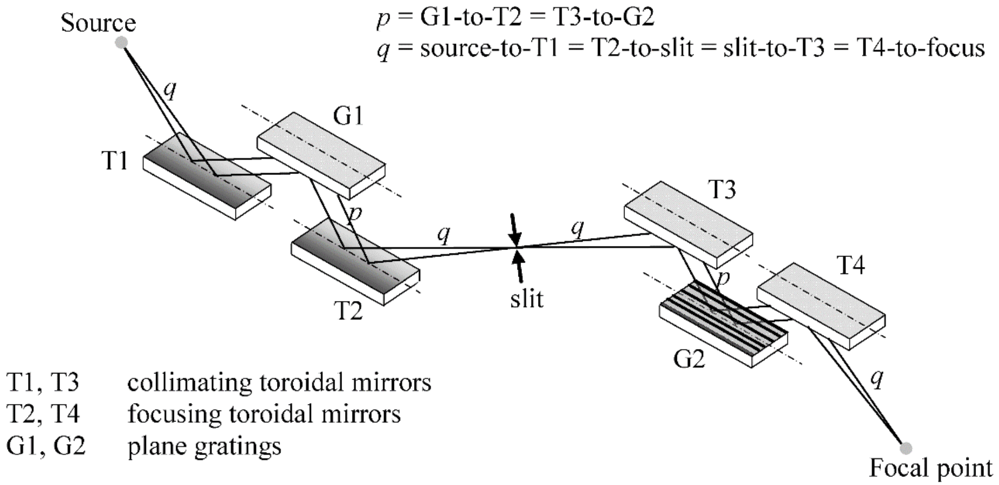


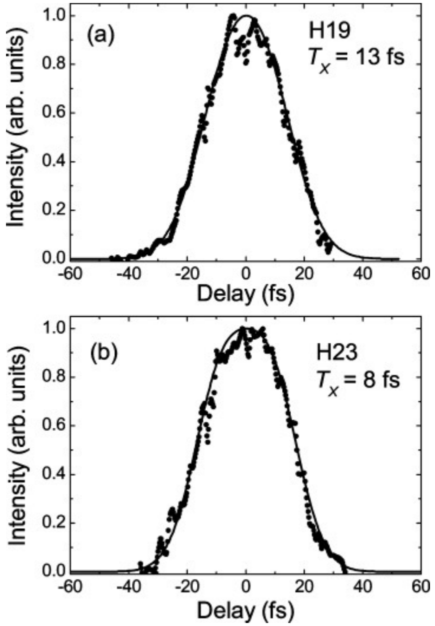
- Access to electron scattering by attosecond streaking on dielectric nanoparticles
- photoelectrons are generated inside the nanoparticles
- transport through the material and photoemission tracked on an attosecond timescale.

L. Seiffert et al.,
Nature Physics ***13***, 766 (2017)

Single harmonic selection

Time-compensated monochromator





L. Poletto, F. Frassetto, *Applied Sciences* **8**, 5 (2018)

L. Poletto, et al., *Opt. Lett.* **32**, 2897 (2007)

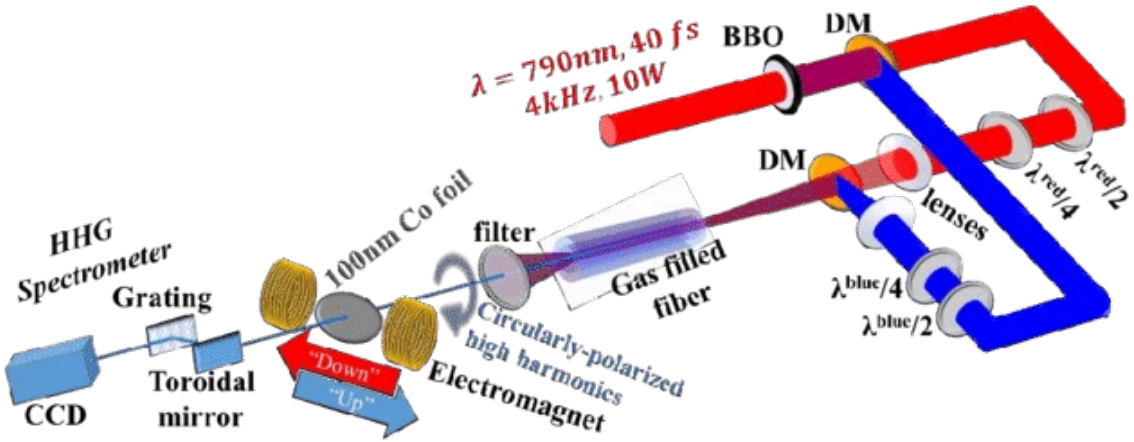
A single harmonic can be selected preserving the temporal duration

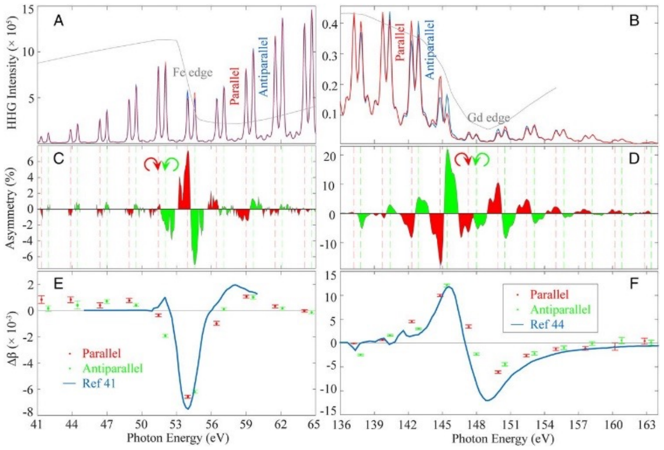
Selective excited-state time-resolved spectroscopy

Circularly-polarized harmonics

Bichromatic pulses, circularly polarized with opposite helicity, drive HHG in gas

Circularly polarized high order harmonics!!!!!





O. Kfir, et al., CLEO 2014, *paper FTu3B.1*

T. Fan et al., *PNAS* **112**, 14206 (2015)

- EUV and X-ray magnetic circular dichroism of Fe and Gd
- XMCD asymmetry of Fe and Gd
- Extracted magneto-optical absorption coeff. at the Fe $M_{2,3}$ and the Gd $N_{4,5}$ edges

New frontiers in HHG

Future directions in HHG: quantum optics

Current HHG description: quantum matter, classical e.m. field

$$[\hat{H} + U_{int}(t)]|\Psi\rangle = i\frac{\partial|\Psi\rangle}{\partial t}$$

where $\hat{H} = -\frac{\nabla^2}{2} + V_{atom}(\boldsymbol{r})$ and $U_{int}(t) = \boldsymbol{r}\cdot\boldsymbol{E}(\mathrm{t})$

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Future directions in HHG: quantum optics

A step forward: fully quantum HHG description

Open investigations in the field

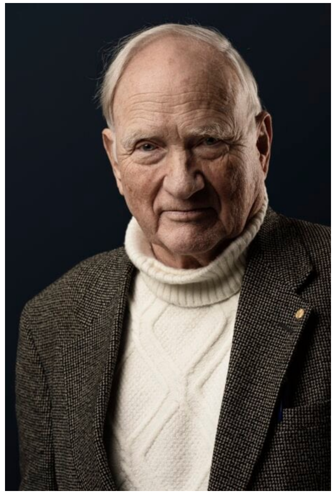
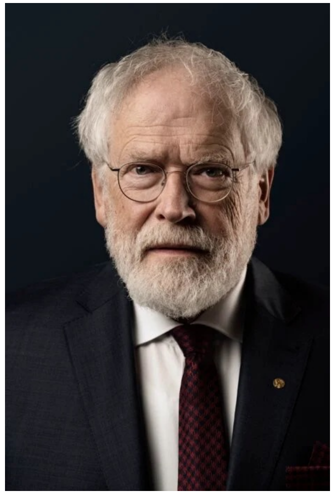
- HH radiation driven by "quantum light pulses" (e.g. Bright Squeezed Vacuum)
- Non-poissonian statistics of HH photons driven by "classical light pulses"
- Entanglement between matter and HH photons (e.g. HH photons resonant with absorption lines)

Future directions in HHG: quantum optics

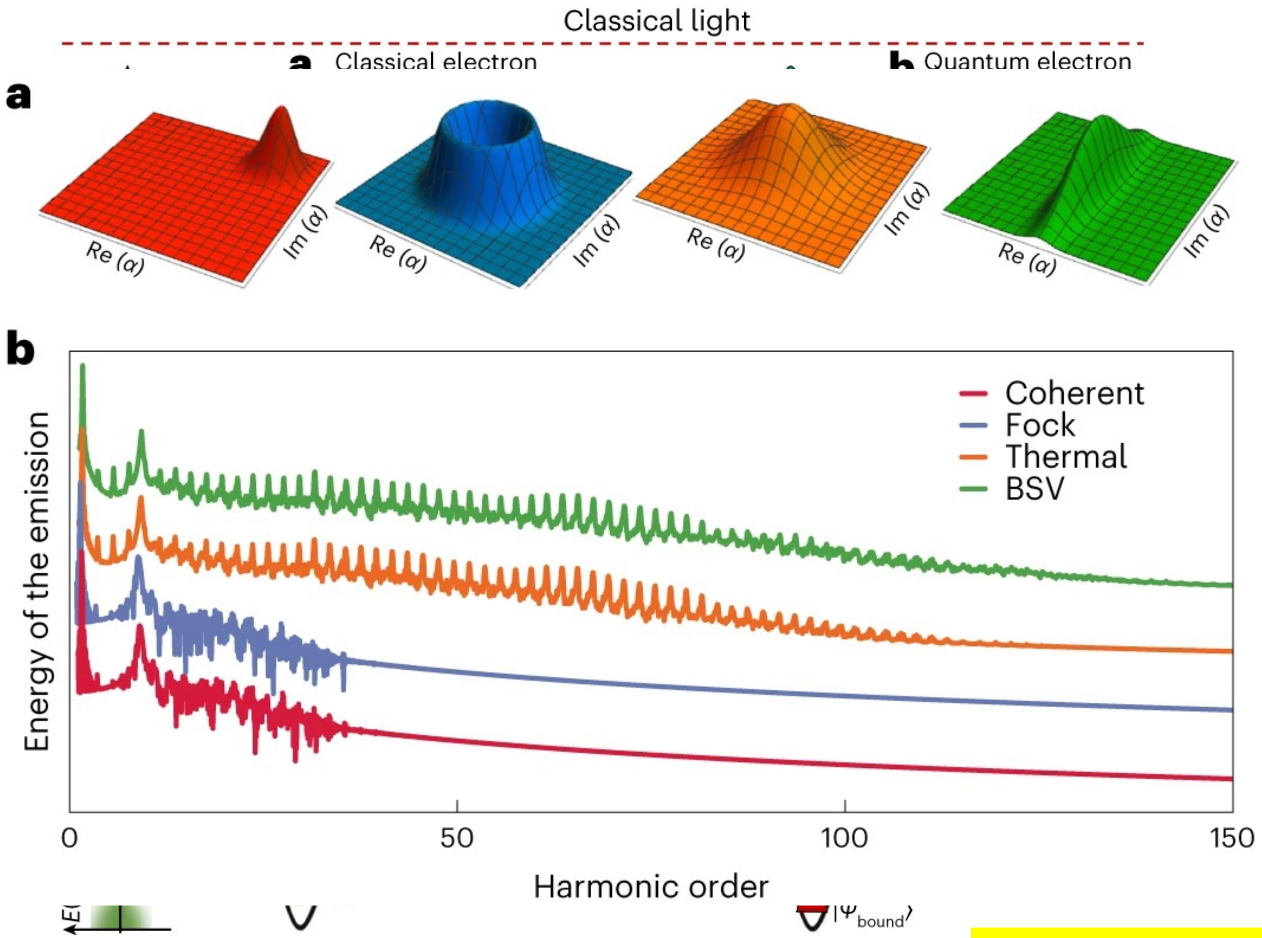
A step forward: fully quantum HHG description

Nobel Prize in Physics 2022 to Alain Aspect, John Clauser and Anton Zeilinger

“for experiments with entangled photons, establishing the violation of Bell inequalities and pioneering quantum information science”

HHG driven by non-classical light

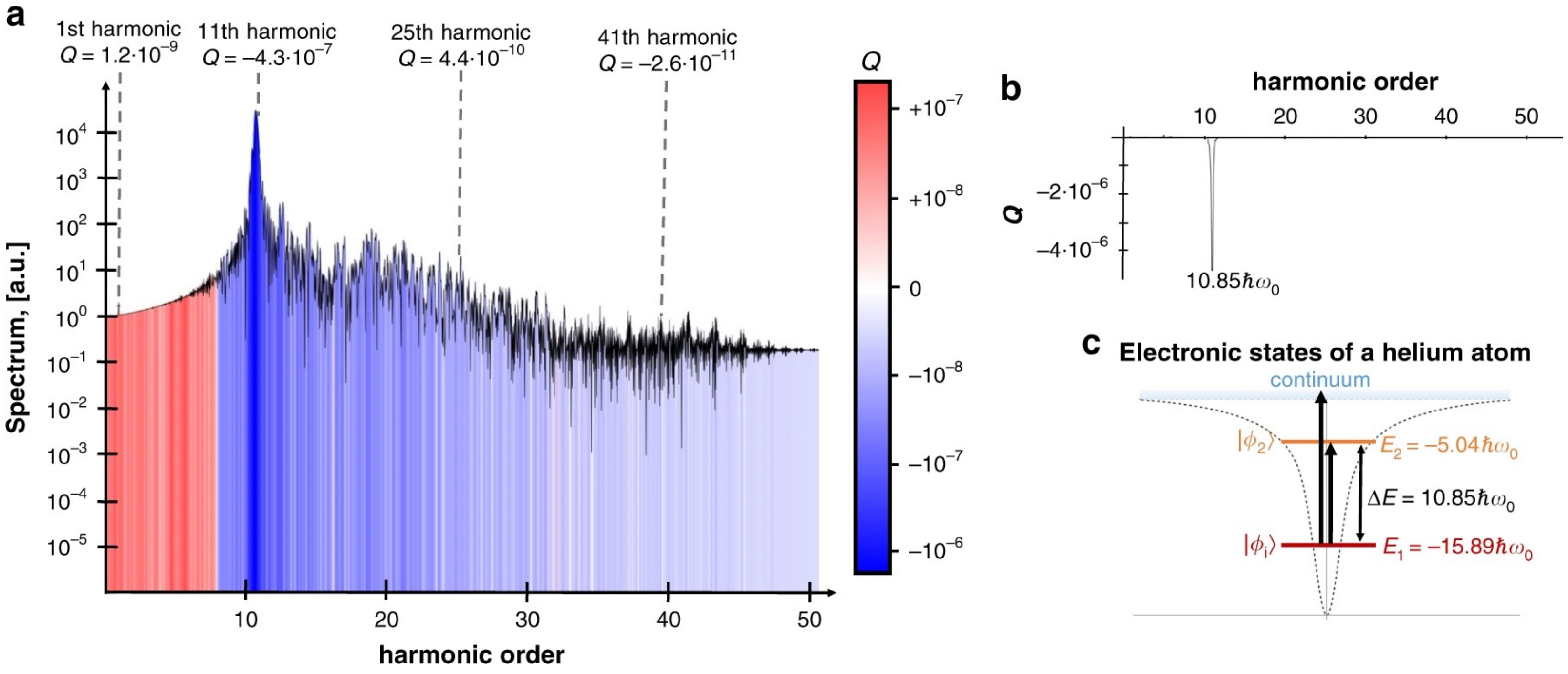


Intense squeezed light could drive the electron dynamics over trajectories not accessible by “classical light”

More extended harmonic spectra could be emitted with respect to usual experimental outcomes (using coherent states of light)

A. Gorlach et al., *Nature Physics* **19**, 1689 (2023)

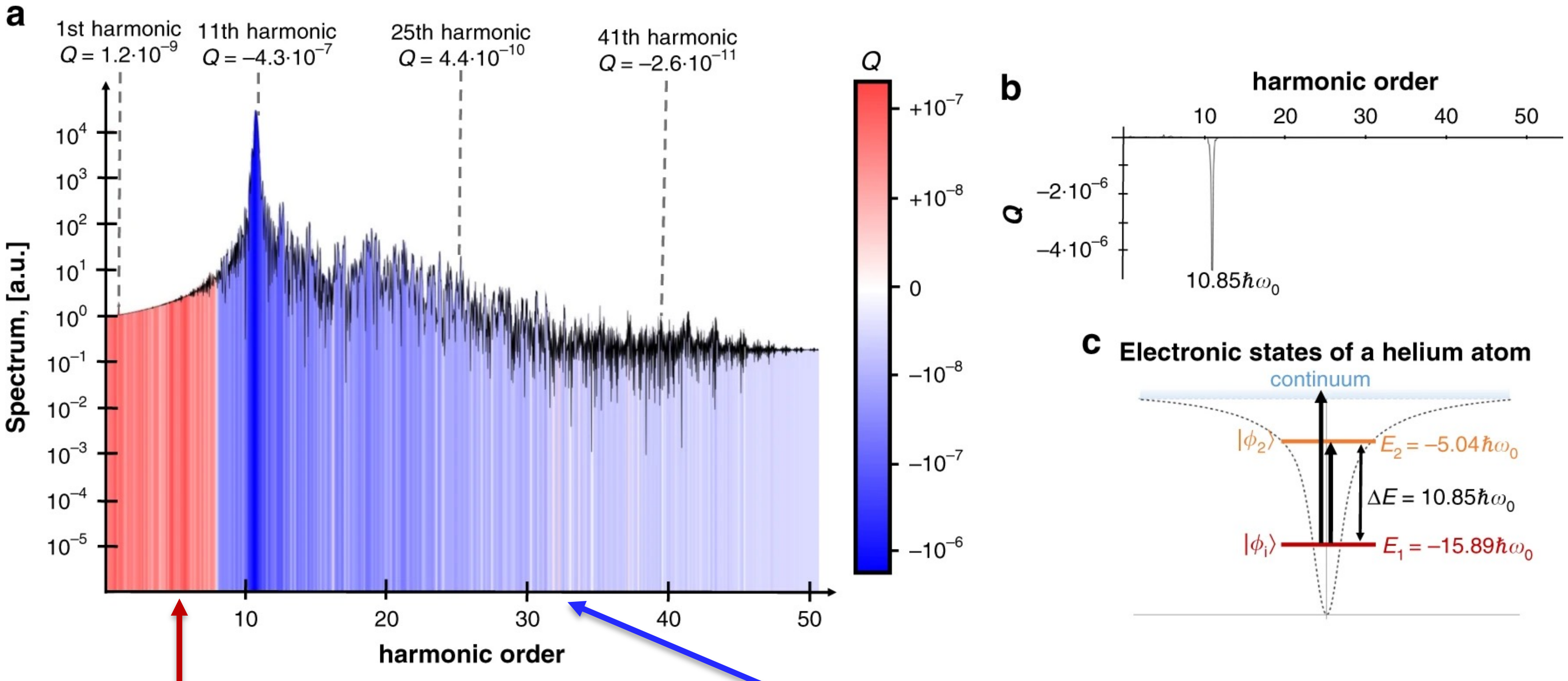
Photon statistics in single-atom HHG



Harmonics driven by "classical light pulses" could show non-classical properties

A. Gorlach et al., *Nature Communications* **11**, 4598 (2020)

Photon statistics in single-atom HHG



Q < 0 sub-Poissonian statistics

Q > 0 super-Poissonian statistics

Mandel parameter Q

$$Q = \frac{\langle n^2\rangle - \langle n\rangle^2}{\langle n\rangle} - 1$$

with n number of photons

Q = 0 for Poissonian statistics (classical coherent light)

A. Gorlach et al., *Nature Communications* **11**, 4598 (2020)

Conclusions

High-order Harmonic Generation can be used to:

- Detect properties of the driven medium (HHG spectroscopy)
- Generate coherent femtosecond pulses in the EUV-soft X region for spectroscopic applications (e.g. NEXAFS, XMCD)
- Generate coherent attosecond pulses for studying electron dynamics in molecules, liquids and solid-state systems

Novel trends (HHG in solids, quantum HHG) are emerging and will trigger applications in diverse fields of research.

Thanks for your attention!

APPENDIX

Lewenstein model: a Feynman's path integral interpretation

Feynman formulation of quantum mechanics

"The probability amplitude of any quantum mechanical process can be represented as a coherent superposition of contributions of all possible spatio-temporal paths that connect the initial and final state of the system.

The weight of each path is a complex number whose phase is equal to the classical action along the path"

$$I(\omega) \propto \left| \omega^2 \int_{-\infty}^{\infty} dt_c \, e^{i\omega t_c} \int_{-\infty}^{t_c} dt_i \int d^3\boldsymbol{P} \; \boldsymbol{D}^*[\boldsymbol{P} + e\boldsymbol{A}(t_c)] \, e^{-iS(\boldsymbol{P},t_i,t_c)} \, \boldsymbol{E}(t_i) \cdot \boldsymbol{D}[\boldsymbol{P} + e\boldsymbol{A}(t_i)] \right|^2$$

P. Salieres et al., *Science* **292**, 902 (2001).

Stationary solutions (1)

$$I(\omega) \propto \left| \omega^2 \int_{-\infty}^{\infty} e^{i\omega t_c}\, dt_c \int_{-\infty}^{t_c} dt_i \int d^3\boldsymbol{P}\ \boldsymbol{D}^*[\boldsymbol{P}+e\boldsymbol{A}(t_c)]\ e^{-iS(\boldsymbol{P},t_i,t_c)}\ \boldsymbol{E}(t_i)\cdot\boldsymbol{D}[\boldsymbol{P}+e\boldsymbol{A}(t_i)] \right|^2$$

step 2: motion in the continuum

step 3: recollision

step 1: ionization

Integration is not required: look for stationary phase conditions in the exponential terms

1. Define $\Theta = S(\boldsymbol{P}, t_i, t_c) + \omega t_c$

2. Look for stationary points $\left[\boldsymbol{P}^{(s)}, {t_i}^{(s)}, {t_c}^{(s)}\right]$ that are solutions of: $\begin{cases} \dfrac{\partial\Theta}{\partial t_i} = 0 \\ \dfrac{\partial\Theta}{\partial t_c} = 0 \\ \nabla_{\boldsymbol{P}}\Theta = 0 \end{cases}$

3. The integral reduces to a sum over a few contributions (*quantum electron trajectories*)

$$I(\omega) \propto \left| \sum_s \omega^2 A_s \boldsymbol{D}^*\left[\boldsymbol{P}^{(s)} + e\boldsymbol{A}({t_c}^{(s)})\right]\boldsymbol{E}\left({t_i}^{(s)}\right)\cdot\boldsymbol{D}\left[\boldsymbol{P}^{(s)} + e\boldsymbol{A}({t_i}^{(s)})\right]\ e^{i\left[\omega {t_c}^{(s)} - S\left(\boldsymbol{P}^{(s)}, {t_i}^{(s)}, {t_c}^{(s)}\right)\right]} \right|^2$$

Suitable function of the stationary solutions

Stationary solutions (2)

Stationary equations can be written and understood as follows:

$$\begin{cases} I_p + \dfrac{\left|\boldsymbol{P}^{(s)} + e\boldsymbol{A}[t_c{}^{(s)}]\right|^2}{2m} = \hbar\omega \\ \dfrac{\left|\boldsymbol{P}^{(s)} + e\boldsymbol{A}[t_i{}^{(s)}]\right|^2}{2m} = -I_p \\ \boldsymbol{P}^{(s)} = \dfrac{1}{t_c{}^{(s)} - t_i{}^{(s)}} \displaystyle\int_{t_i{}^{(s)}}^{t_c{}^{(s)}} e\boldsymbol{A}(t'')dt'' \end{cases}$$

Ionization potential + collision kinetic energy of the electron = emitted photon energy

Electron tunnels out at ionization with negative kinetic energy, since it is bound (pure quantum effect)

Simply states that the electron is ionized and collides at the same spatial position (its parent ion)

Quantum electron trajectories contributing to HHG show a semi-classical interpretation

Structural information in HHG spectra

$$I(\omega) \propto \left| \sum_s \omega^2 A_s \boldsymbol{D}^*\left[\boldsymbol{P}^{(s)} + e\boldsymbol{A}({t_c}^{(s)})\right] \boldsymbol{E}\left({t_i}^{(s)}\right) \cdot \boldsymbol{D}\left[\boldsymbol{P}^{(s)} + e\boldsymbol{A}({t_i}^{(s)})\right] \; e^{i\left[\omega {t_c}^{(s)} - S\left(\boldsymbol{P}^{(s)}, {t_i}^{(s)}, {t_c}^{(s)}\right)\right]} \right|^2$$

Structural information on the atom/molecule is here!
It is a dominant contribution to HHG spectrum

Structural information in HHG spectra

$$I(\omega) \propto \left| \sum_s \omega^2 A_s \boldsymbol{D}^*[\boldsymbol{P}^{(s)} + e\boldsymbol{A}(t_c{}^{(s)})]\boldsymbol{E}(t_i{}^{(s)}) \cdot \boldsymbol{D}[\boldsymbol{P}^{(s)} + e\boldsymbol{A}(t_i{}^{(s)})]\; e^{i[\omega t_c{}^{(s)} - S(\boldsymbol{P}^{(s)}, t_i{}^{(s)}, t_c{}^{(s)})]} \right|^2$$

For energy conservation the emitted photon energy is

$$\hbar\omega = I_p + \frac{\left|\boldsymbol{P}^{(s)} + e\boldsymbol{A}[t_c{}^{(s)}]\right|^2}{2m} = I_p + \frac{\left|\boldsymbol{k}(t_c{}^{(s)})\right|^2}{2m} \Rightarrow \boldsymbol{k} = \boldsymbol{k}(\omega)$$

$$I(\omega) \propto \left| \sum_s a_s \omega^2 \boldsymbol{D}^*[\boldsymbol{P}^{(s)} + e\boldsymbol{A}(t_c{}^{(s)})] \right|^2 \approx \omega^4 |a(\boldsymbol{k})\boldsymbol{D}^*[\boldsymbol{k}(\omega)]|^2 =$$

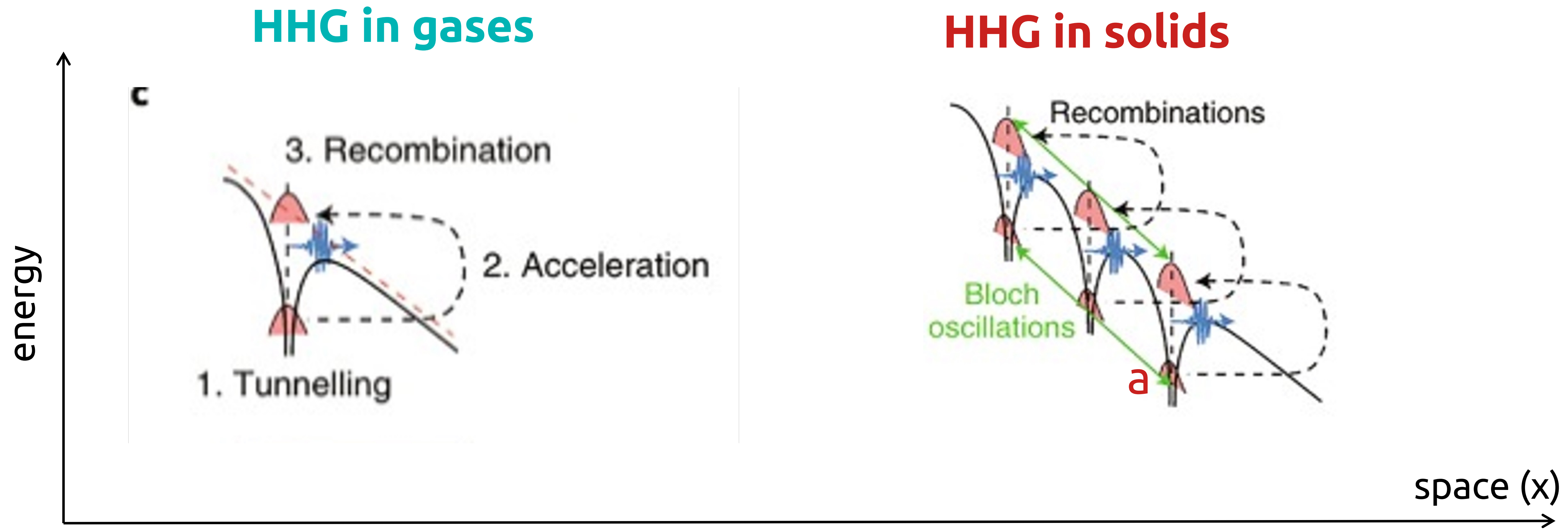
$$\omega^4 \left| a(\boldsymbol{k}) \int -e\boldsymbol{r}\, \Psi_0(\boldsymbol{r}) e^{-i\boldsymbol{k}(\omega)\cdot\boldsymbol{r}/\hbar} d^3\boldsymbol{r} \right|^2$$

*a(**k**)* is the amplitude of the colliding wavepacket

The spectrum is linked to the spatial Fourier transform of the ground state wavefunction Ψ_0 (times $-\boldsymbol{r}$)

HHG in solids vs. HHG in gas

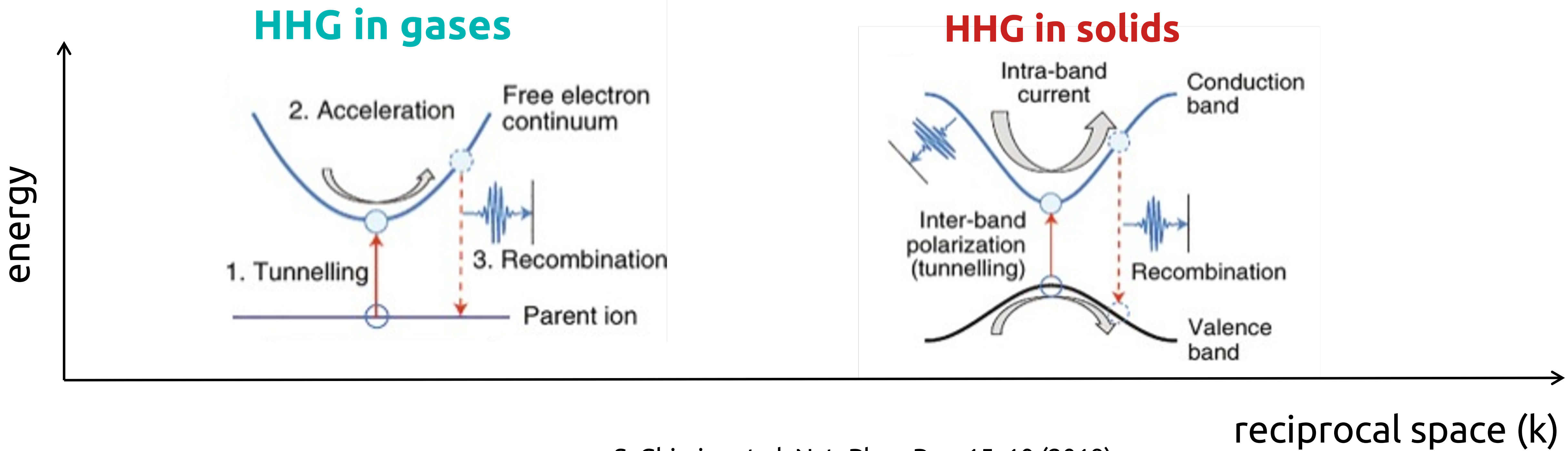
Real Space



S. Ghimire et al. Nat. Phys. Rev. 15, 10 (2019)

HHG in solids vs. HHG in gas

Momentum Space



S. Ghimire et al. Nat. Phys. Rev. 15, 10 (2019)

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Even vs. odd harmonics in ZnTe

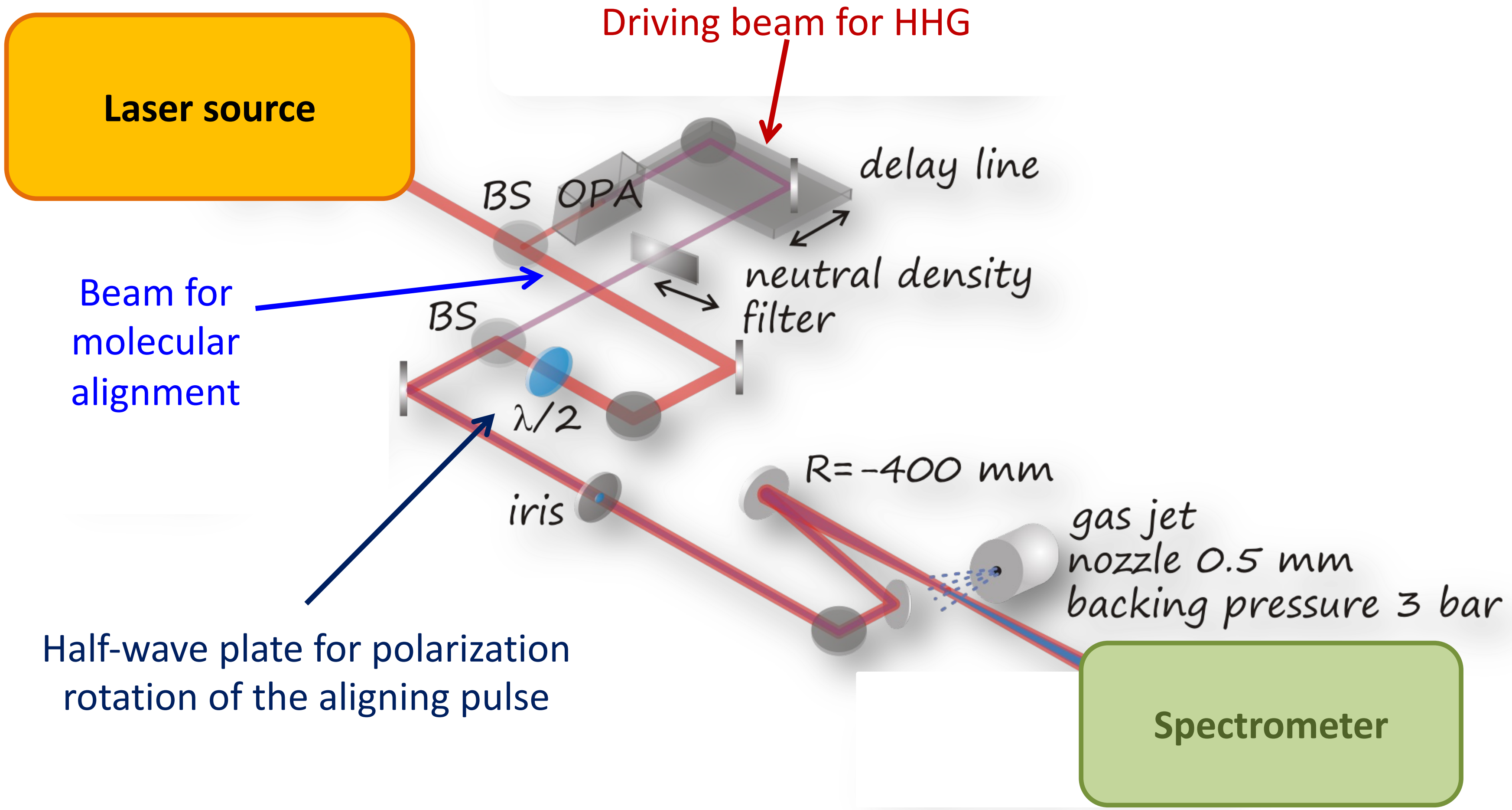
Simple-man picture

1. The laser pulse promotes electrons in the conduction band
2. The electrons accelerate along the light polarization direction
3. Harmonic emission maximizes for directions connecting atomic sites

   a) Directions without inversion symmetry (Zn-Te): one collision per optical period, even and odd harmonics are generated

   b) Directions with inversion symmetry (Zn-Zn or Te-Te): two collisions per optical period, only odd harmonics are generated

Experimental setup for HHG tomography



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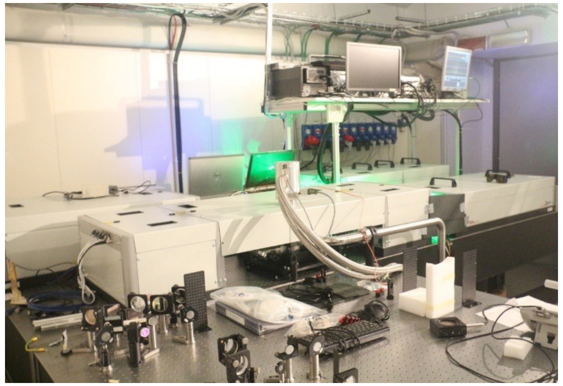
An HHG tomography lab

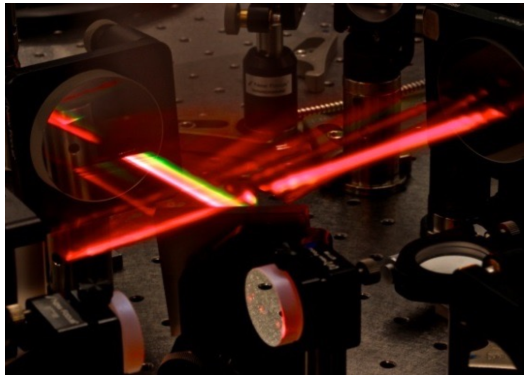


Typical driving laser source (Ti:Sa laser):

- ❑ <25 fs pulses
- ❑ 15-mJ energy
- ❑ 1-kHz repetition rate

HHG beamline:

- ❑ Vacuum chambers hosting:
  - ➢ HHG section with gas jet
  - ➢ Harmonics beam transport to diagnostics
  - ➢ Harmonics polarization analyzer
  - ➢ Grazing-incidence XUV-soft X spectrometer



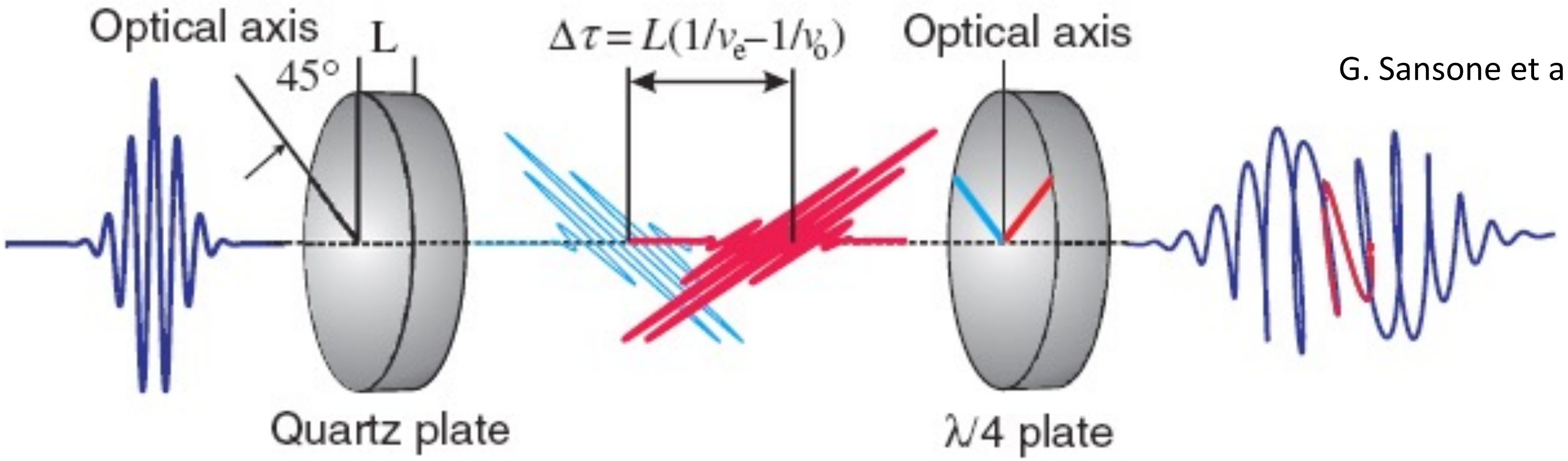
Light pulse manipulation

- ❑ Temporal compression
- ❑ Wavelength down- or up-conversion
- ❑ Multi-color laser pulse combining

How to generate Attosecond Pulses: Polarization Gating

HHG yield suppressed for circularly polarized laser pulses

Linear polarization confined to a short window selects a single attosecond pulse emission



G. Sansone et al., *Science* **314**, 443 (2006)