

Attosecond Physics gets Nano

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XXX ICPEAC, Cairns, Australia, 01/08/17

Citius, Altius, Fortius

(Olympic motto, Pierre de Coubertin, 1894) Faster, Higher, Stronger

Memorial dedicated to the City of Cairns as Host City for the beginning of the 1956 Olympic Torch relav

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Citissimus, Maximus, Brevissimus, Minimus

(Atto-nano-physics motto) Fastest, Highest, Shortest, Smallest

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 Above-threshold ionization (ATI) and high-order harmonic generation (HHG)



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- The physical picture: the three-step model



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 - The present: conclusions

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- The (near) future: outlook & perspectives

What is strong/intense? Electric field due to the attraction of an electron and a proton: |E_H| = 5.14 × 10¹¹ V/m (51.4 V/Å). Intensity equivalent: I = 3.51 × 10¹⁶ W/cm². Laser pulses in an intensity regime between 10¹⁴ - 10¹⁶ W/cm² are routinely generated worldwide
 What is short? Classical period of an electron around a proton: τ = 152 as, period of a light wave for a typical Titanium-Sapphire (Ti:Sa) laser (λ = 800 nm): 2.7 fs (1 fs = 10⁻¹⁵ s). Few-cycle laser pulses (less than 2-3 total optical cycles) are generated in several laboratories around the world

What is small? Bohr radius: 0.0529 nm (0.529 Å), typical classical excursion of an electron in an oscillating electron field: 1-10s nm (10 Å). Engineering metal/dielectrics nanostructures down to a few ten nanometers is nowadays possible

engineering at a nanometric scale: the **Atto-nano-ph** has just started!!!

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4 / 28

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For the first time we could combine attosecond time resolution with engineering at a nanometric scale: the Atto-nano-physics stage has just started!!!

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Atomic or molecular bound electrons

Excess energy converted to kinetic energy
 Production of direct and rescattered

Use as a few-cvcle pulses characterization

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5 / 28

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 HHG spectra features: decay, plateau cutoff

 Utilization as a source of coherent XUV radiation & attosecond pulses (key tools for molecular imaging)

Typical HHG spectra in atoms (experiment)

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5 / 28

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5 / 28

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1.42µm; 15fs; 1.50x10¹⁴W/cm² - 1.40µm; 55fs; 9.9x10¹ 1.80µm: 11fs: 1.57x10¹⁴W/cm² _____ 1.82µm; 73fs; 9.0x10¹⁴W/cm² HHG vield XUV photon energy [eV]

5 / 28

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5 / 28

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Conventional Strong Field phenomena The physical picture

The three-step model (it is simple and works!!!!)

a) an atomic or molecular electron is laser-ionized via tunneling; (b) i way from the atom; (c) it is driven back -when the laser electric field ts direction; (d) it can 'recollide' during a small fraction of time (sub convert its kinetic energy into a high energy and 'ultrashort' photon -

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. A 49, 2117 (1994); 6 / 28

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6 / 28

Conventional Strong Field phenomena

The physical picture

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P. B. Corkum, Phys. Rev. Lett. 71, 1994 (1993); M. Lewenstein, et al., Phys. Rev. A 49, 2117 (1994); P. B. Corkum & F. Krausz, Nat. Phys. 3, 381 (2007)

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Classical electron excursion in an oscillating electric field (quiver radius) $\alpha=E_0^2/\omega^2=I/\omega^2$



M.F. Ciappina, et al. Attosecond physics at the nanoscale, Rep. Prog. Phys. 80, 054401 (2017)

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Scheme of conventional strong field processes

Scheme of plasmonic-enhanced strong field processes

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The (recent) past Motivation, the experiments I

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HHG in gases driven by plasmonic fields I (2008)	HHG in gases driven by plasmonic fields II (2011)	

Motivation, the experiments I



Motivation, the experiments I



Motivation, the experiments I



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Motivation, the experiments II

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Electron emission in nanotips I (2011)

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Attosecond control of electrons emitted from a nanoscale metal tip

Michael Kriger¹⁴, Markus Schenk¹⁴ & Peter Hommelhoff

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ARTICLES physics Controlled near-field enhanced electron acceleration from dielectric nanospheres with intense few-cycle laser fields Screey Zherebtsoy¹¹, Thomas Fernel²⁺⁷, Jürgen Plenge³¹, Epill Antonson², Irina Znakovskawa¹, Adrian Wirth¹, Oliver Hernwerth¹, Frederik Süßenern¹, Christian Peltz², Idnar Ahmed¹, Sorgei A. Trushin¹, Vladimir Pervak⁴, Stefan Karsch¹⁴, Marc J. J. Wakking⁵⁰, Barkhard Langer², Christina Graff, Mark I, Stockman¹², Ferenc Krausa¹⁴, Eckart Rühlf* and Matthias F, Käras^{18,0}*

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Quantal

- The Time Dependent Schrödinger Equation (TDSE) 1D & 3D
- Numerical solution in a grid with absorbing boundaries
- ATI yield and HHG spectra obtained postprocessing the time propagated wavefunction
- Advantages & Drawbacks of each flavor

Semiclassical

- The Strong Field Approximation (SFA) or Lewenstein model
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Quantum models I The Time Dependent Schrödinger Equation in reduced dimensions (TDSE-1D)



- atomic potential
- laser-atom coupling
- electron wf (1D)
- Numerical solution in a grid using the Crank-Nicolson algorithm with absorbing boundary conditions (masks)
- Sector calculated Fourier transforming the time-dependent dipole extension
- ATLyield calculated using the time propagated electron wavefunction via energy window techniques
- Advantages: low computational cost, allow general functional forms for the nonhorno field, excellent agreement with the classical predicted limits of SEI are compared with the classical predicted limits.

The Time Dependent Schrödinger Equation in reduced dimensions (TDSE-1D)

- time evolution of electron wf (1D)
- kinetic energy

$$i\frac{\partial\Psi(x,t)}{\partial t} = \left(-\frac{1}{2}\frac{d^2}{dx^2} + V_{\text{atom}}(x) + V_{\text{laser}}(x,t)\right)\Psi(x,t)$$

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M.F. Ciappina, et al. Attosecond physics at the nanoscale, Rep. Prog. Phys. 80, 054401 (2017)

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The Time Dependent Schrödinger Equation (TDSE-3D) in the Single Active Electron (SAE) approximation

$$i\frac{\partial\Psi(\mathbf{r},t)}{\partial t} = \left(-\frac{1}{2}\nabla^2 + V_{atom}(r) + V_{laser}(\mathbf{r},t)\right)\Psi(\mathbf{r},t)$$

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- time-dependent dipole moment
- recombination transition matrix

$$\mathbf{D}(t) = -\mathrm{i} \int_{0}^{t} \mathrm{d}t' \int \mathrm{d}\mathbf{k} \langle \psi_{0} | - \mathbf{r} | \mathbf{k} + \mathbf{A}(\mathbf{r}, t) \rangle$$
$$\times \langle \mathbf{k} + \mathbf{A}(\mathbf{r}, t') | \mathbf{E}(\mathbf{r}, t') \cdot \mathbf{r} | \psi_{0} \rangle \exp[-\mathrm{i}S(\mathbf{k}, \mathbf{r}, t, t')]$$

- ionization transition matrix
- classical action

M. Lawrenstein, et al., Phys. Rev. A 40, 2117 (1994); M.F. Cheppins, et al., Attaneousl. physics and the neuroscient Rep. Phys. 80, 084401 (2017).

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Semiclassical model II The Strong Field Approximation (SFA) for ATI

total probability amplitude.

$$M_{\mathbf{p}}^{\mathrm{SFA}} = M_{\mathbf{p}}^{\mathrm{(d)}} + M_{\mathbf{p}}^{\mathrm{(r)}}$$

- direct electrons probability amplitude
- rescattered electrons probability amplitude

 B. Millohold, et al., J. Phys. B 96, R203 (2006); M.F. Gappins, et al., Accorecord physics at the monotohy floar Phys. Phys. 80, 164401 (2017).

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Classical approaches Newton-Lorentz equation (1D) & simple man's model

electron acceleration (1D)



- force
- Initial conditions: $z(t_i) = 0$ & $\dot{x}(t_i) = 0$ $(t_i \rightarrow \text{ionization time})$
- π Recollision condition $x(t_r) = 0$ $(t_r
 ightarrow recollision time)$
- The excess of energy is converted at recollision
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- experimentally confirmed)

Classical approaches Newton-Lorentz equation (1D) & simple man's model

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$$\ddot{x}(t) = -\nabla V_{\text{laser}}(x,t) = -\nabla [xE(x,t)]$$

force

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- \blacksquare If the electron recombines and emits radiation \rightarrow HHG
- HHG cutoff prediction $n_c = (3.17U_p + I_p)/\omega$ (experimentally confirmed)
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Newton-Lorentz equation (1D) & simple man's model

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- HHG cutoff prediction $n_c = (3.17U_p + I_p)/\omega$ (experimentally confirmed)
- If the electron never return, 2Up cutoff in ATI yield (direct electrons, experimentally confirmed)
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Newton-Lorentz equation (1D) & simple man's model





- Initial conditions: $x(t_i) = 0 \& \dot{x}(t_i) = 0$ ($t_i \rightarrow \text{ionization time}$)
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The electron confinement menace

Rationale

- caricature of the plasmonic-enhanced field (linear dependence)
- electron confinement (limited grid)
- tailoring trajectories (only short)
- HHG cutoff extension
- symmetry breaking

1D HHG spectra

Setup example

Fime-analysis

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1D HHG spectra Time-analysis

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M.F. Ciappina, et al., Phys. Rev. A 85, 033828 (2012); W.F. Ciappina, et al. Attosecond physics at the nanoscale, Rep. Prog. Phys. 80, 054401 (2017).

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0 100 300

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Time (a u)

300 500

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1D HHG spectra 10 Harmonic order

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Attacking the CEP determination



- caricature of the plasmonic-enhanced field (linear dependence)
- strong CEP dependence (energy)
- high-energy electron production
- tailoring electron trajectories

1D ATI for CEP #1

Classical mode

LD ATI for CEP #2

M.F. Ciappina, et al., Phys. Rev. A 86, 023413 (2012); M.F. Ciappina, et al. Attosecond physics at the nanoscale, Rep. Prog. Phys. 80, 054401 (2017).

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The revenge of the CEP

Rationale

- caricature of the plasmonic-enhanced field (linear dependence)
- strong CEP dependence (angular)
- tailoring electron trajectories
- directional electron bunches

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3D ATI for CEP #1	3D ATI for CEP #2

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A new and improved model for the plasmonic field

real plasmonic-enhanced field (FDTD)

Marcelo Ciappina

A new and improved model for the plasmonic field

Rationale

- real plasmonic-enhanced field (FDTD)
- tailoring trajectories
- HHG cutoff extension
- symmetry breaking
- long wavelengths

HHG (1D) & FDTD & real field #1

Sketch & TEM image

HHG (1D) & FDTD & real field #2

M.F. Ciappina, et al., Opt. Exp. 20, 26261 (2012); M.F. Ciappina, et al. Attosecond physics at the nanoscale, Rep. Prog. Phys. 80, 054401 (2017).

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Marcelo Ciappina

The high energetic electron strikes back

Rationale

- temporal synthesized field (experimentally tested)
- linear inhomogeneous field
- HHG cutoff extension
- interpretation via quantum vs. classical simulations
- supercontinuum generation (atto pulses)

HHG in 3D (He)

Temporal synthesized pulse

Fime analysis & classical

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HHG in 3D (He)

Temporal synthesized pulse

Fime analysis & classical

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The return of ATI and HHG driven by plasmonic near-fields

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Proposed setup & near-field

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Electrons 10 homogeneous 7=29 1073 hotoelectron vield (arb, units) 10* 10" 105 107 10⁻⁸ 105 10U=362eV 10¹⁰ 100 1000 Electron energy (eV)

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The high-energy structure (HES) awakens

Rationale

- linear inhomogeneous field
- high-energy structure (HES)
- independent of the atom
- validated via CTMC simulations
- highly (temporally) localized electron bunches

3D TDSE (H and He)

Ionization phase CTMC

CTMC simulations

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The last recombination site

Rationale

- HHG in solids
- Wannier picture for the valence band (sites)
- Bloch picture for the conduction band (delocalized)
- atomic-like recombination picture (different sites contribution)
- other models comparison: advantages

Comparison with Bloch-Bloch

Picture

Localization study

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- Prospect of table-top high repetition rates and strong laser sources using plasmonic fields: looking for experimental alternatives

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- Exploring (theoretical & experimentally) strong field related phenomena (above threshold photoemission, HHG in solids, electron emission and radiation from thin films, etc.)
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Thank you for your attention !

Attosecond Physics at the Nanoscale International Workshop

Scientific Coordinators:

Dr. Marcelo Ciappina (ELI-Beamlines, Czech Republic)

Prof. Seungchul Kim (POSTECH and Busan University, South Korea)

Prof. Young-Jin Kim (Nanyang Technological University, Singapore)

When: October 22-26, 2018 (Save the date!!!) Where: Center for Theoretical Physics of Complex Systems (PCS), Daejeon, South Korea Interested? Please write me: marcelo.ciappina@eli-beams.eu Partial or total support provided

