Theoretical Atto-Nano Physics

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NORDITA, Stockholm, 02.06.15

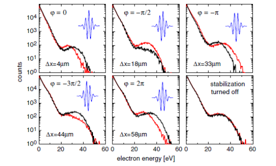
Outline

- *Conventional* Strong Field phenomena (brief description)
  - The physical picture: the three-step model
  - Typical numbers: the laser-ionized electron does not feel the spatial variation of the field
- Laser-matter processes driven by temporal and spatial synthesized laser fields
  - The (recent) past: the experiments, motivation
  - The (recent) past and present: the models (quantal, semiclassical and classical) & selected results
  - The present: conclusions
- The (near) future: outlook, perspectives & work in progress

Conventional Strong Field phenomena
Brief description

Above threshold ionization (ATI)

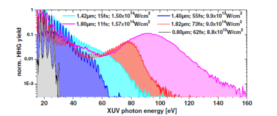
- Atomic or molecular bound electrons absorb many more photons than the minimum required to reach the continuum
- Excess energy converted to kinetic energy
- Production of direct and rescattered electrons
- Use as a pulse characterization



Typical photoelectron spectra (experiment)

High-order harmonic generation (HHG)

- Emission of the electron excess energy in the form of high-order harmonics of the fundamental laser field (HHG) (typically XUV photons)
- HHG spectra features: decay, plateau and cutoff
- Utilization as a source of attosecond pulses and molecular imaging tool

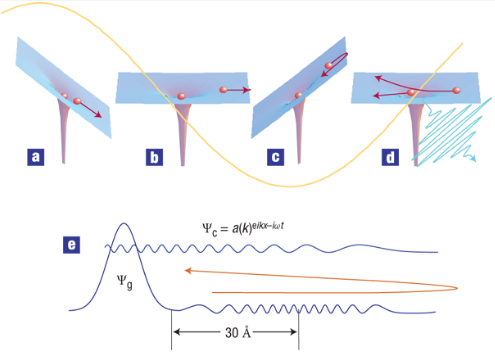


Typical HHG spectra in atoms (experiment)

Main theoretical assumption: the laser electric field is spatially homogeneous where the electron dynamics takes place

G. G. Paulus, et al., Phys. Rev. Lett. **91**, 253004 (2003); B. E. Schmidt, et al., J. Phys. B. **45**, 074008 (2012)

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



(a) tunnel ionization; (b) the electron is pulled away from the atom; (c) the electron is driven back; (d) it can 'recollide' during a small fraction of time (sub-fs) and convert its kinetic energy in a high energy and 'ultrashort' photon (or it can rescatters, double ionize, etc.); (e) the quantum mechanical picture (split of the wavefunction in a bound and a continuum part, overlap after).

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)

Conventional Strong Field phenomena
Typical numbers

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

@ 800 nm

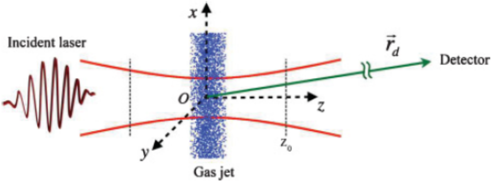
- $I \approx 10^{13}$ W/cm^2 $\Rightarrow \alpha \approx \pm 0.28$ nm
- $I \approx 10^{14}$ W/cm^2 $\Rightarrow \alpha \approx \pm 0.87$ nm

@ 1800 nm

- $I \approx 10^{13}$ W/cm^2 $\Rightarrow \alpha \approx \pm 1.5$ nm
- $I \approx 10^{14}$ W/cm^2 $\Rightarrow \alpha \approx \pm 4.5$ nm

@ 3.1 μm

- $I \approx 10^{13}$ W/cm^2 $\Rightarrow \alpha \approx \pm 4.1$ nm
- $I \approx 10^{14}$ W/cm^2 $\Rightarrow \alpha \approx \pm 13$ nm



Scheme of a typical high-order harmonic experiment using gas jets. The waist of the gaussian laser pulse is several tenths of μms (for 800 nm).

C. Hernández García, et al. Phys. Rev. A **82**, 033432 (2010)

The (recent) past

Motivation, the experiments I

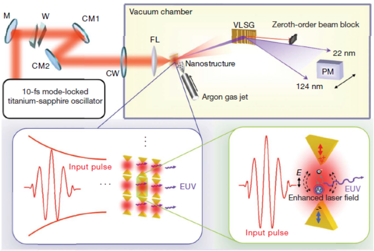
HHG in gases driven by plasmonic fields I (2008)

Vol 453|5 June 2008|doi:10.1038/nature07012 nature

LETTERS

High-harmonic generation by resonant plasmon field enhancement

Seungchul Kim[1]*, Jonghan Jin[1]*, Young-Jin Kim[1], In-Yong Park[1], Yunseok Kim[1] & Seung-Woo Kim[1]

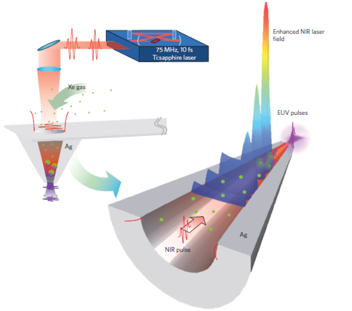


HHG in gases driven by plasmonic fields II (2011)

nature photonics LETTERS
PUBLISHED ONLINE: 16 OCTOBER 2011 | DOI: 10.1038/NPHOTON.2011.258

Plasmonic generation of ultrashort extreme-ultraviolet light pulses

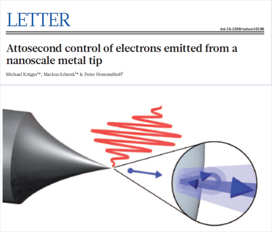
In-Yong Park[1†], Seungchul Kim[1†], Joonhee Choi[1†], Dong-Hyub Lee[1], Young-Jin Kim[1], Matthias F. Kling[2], Mark I. Stockman[3] and Seung-Woo Kim[1]*



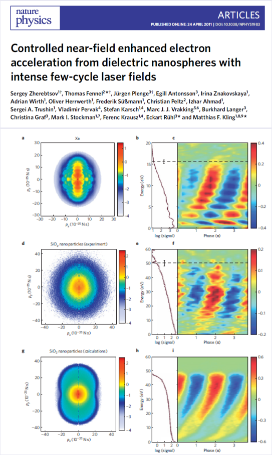
S. Kim, et al., Nature **453**, 757 (2008); I-Y. Park, et al., Nat. Phot. **5**, 677 (2011)

The (recent) past
Motivation, the experiments II

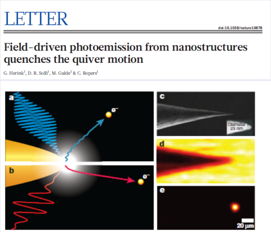
Electron emission in nanotips I (2011)



Electron emission in nanospheres (2011)



Electron emission in nanotips II (2012)



M. Krüger, et al., Nature **453**, 78 (2011); S. Zherebstov, et al., Nat. Phys. **7**, 656 (2011); G. Herink, et al., Nature **483**, 190 (2012)

The (recent) past and present
Theoretical models

We treat spatially inhomogeneous field by modifying the laser-electron coupling in the *conventional* theoretical models

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
- The laser field does not affect the bound electron
- The residual Coulomb field does not affect the *continuum* electron
- Interpretation of ATI and HHG in terms of *quantum orbits*

Classical

- Newton-Lorentz equation (1D)
- Simple man's model prescriptions
- *Extraction* of the ionization and recombination times
- Electron kinetic energy calculations
- Excellent prediction of ATI yield (energy) and HHG spectra (harmonic order) limits

The three approaches give supplementary information

M. F. Ciappina, et al. Rep. Prog. Phys (in progress) (2015)

Quantum models I

TDSE-1D

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

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

where $V_{\mathrm{atom}}(x) = -1/\sqrt{x^2 + a^2}$ is the *soft core* potential (the ionization potential is adjustable varying a) and $V_{\mathrm{laser}}(x,t) = -E(x,t)x$ with $E(x,t) = E_0 f_0(t)(1 + \varepsilon g(x)) \sin \omega t$ the potential (in the dipole approximation) due to the laser electric field including a general function $g(x)$ for the spatial variation and a (small) parameter ε that controls the inhomogeneity strength.

- Numerical solution in a grid using the Crank-Nicolson algorithm with absorbing boundary conditions (masks)
- HHG spectra calculated using the time propagated electron wavefunction via the dipole acceleration
- ATI yield calculated using the time propagated electron wavefunction via energy window techniques
- Advantages: low computational cost, allow general functional forms for $g(x)$, excellent agreement with the classical predicted limits
- Drawbacks: finer details of HHG of ATI are missing, only allow energy analysis (ATI)

Quantum models II
TDSE-3D

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

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

where $V_{\mathrm{atom}}(r)$ is the atomic potential (chosen to match the ionization potential and excited states of the atom under study) and $V_{\mathrm{laser}}(\vec{r},t) = -E(\vec{r},t)\cdot\vec{r}$ the potential due to the laser electric in the dipole approximation. We have only implemented the case for linear polarization and spatially linear nonhomogeneous fields.

- The electron wave function $\Psi(\vec{r},t)$ is written in terms of angular momenta l
- For each l (typically several hundreds) we have a set of coupled partial differential equations for the radial variable r
- Numerical solution in the radial grid using the Crank-Nicolson algorithm with absorbing boundary conditions (masks)
- HHG spectra calculated using the time propagated electron wavefunction via the dipole acceleration & ATI yield calculated using the time propagated electron wavefunction via energy window techniques
- Advantages: excellent resolution for both the HHG spectra and ATI yield; it allows angular distributions calculations (ATI); real nonhomogeneous fields can be incorporated; modeling of complex atoms with high precision
- Drawbacks: high computational cost (in particular for longer laser pulses and longer wavelengths), up to date only spatially linear nonhomogeneous fields have been studied

Strong Field Approximation (SFA) prescriptions

- The laser field does not affect the bound electron
- The residual Coulomb field does not affect the *continuum* electron

SFA for HHG

- Key ingredient: dipole transition matrix (the HHG is the Fourier transform)
- Three step: ionization, classical motion & recombination
- Saddle points treatment: Quantum Orbits interpretation
- Non-homogeneous fields modifications

  (linear case)
  - Classical action
  - Saddle points electron momentum
  - Saddle points equations

SFA for ATI

- Transition amplitudes for both the direct and rescattered processes
- ATI yield calculated as a coherent sum of direct and rescattered contributions
- Saddle points treatment: Quantum Orbits interpretation
- Non-homogeneous fields modifications

  (linear case)
  - Classical action
  - Saddle points electron momentum
  - Saddle points equations

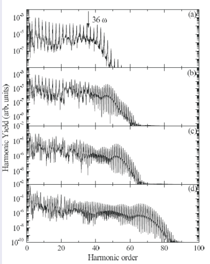
Newton-Lorentz equation (1D)

$$\ddot{x}(t) = -\nabla V_{\rm laser}(x,t)$$

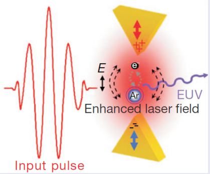
with $V_{\rm laser}(x,t) = xE(x,t)$, $E(x,t)$ being the laser electric field. Note that $E(x,t)$ is now function of both time and space

- Simple man's model prescriptions
  - Initial conditions: $x(t_i) = 0$ & $\dot{x}(t_i) = 0$ ($t_i \rightarrow$ ionization time)
  - Recollision condition $x(t_r) = 0$ ($t_r \rightarrow$ recollision time)
  - The excess of energy is *converted* at recollision
  - If the electron recombines and emits radiation $\rightarrow$ HHG
  - HHG cutoff prediction $n_c = (3.17U_p + I_p)/\omega$ (experimentally confirmed)
  - If the electron never return, $2U_p$ cutoff in ATI yield (direct electron, experimentally confirmed)
  - If the electron elastically rescatters, $10U_p$ cutoff in ATI yield (rescattered electron, experimentally confirmed)

Spatially nonhomogeneous (linear) field

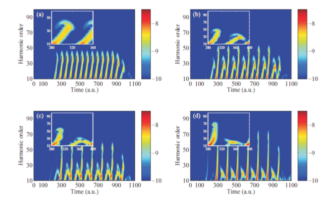


HHG for a model atom (TDSE-1D)



limited region

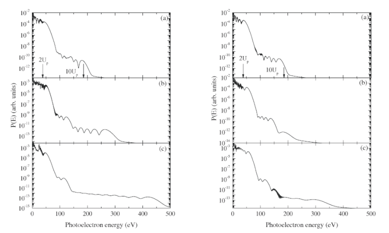
Sketch of the HHG driven by plasmonic fields



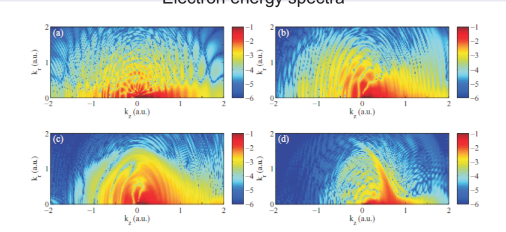
Time-frequency analysis

Selected Results II

ATI driven by linear nonhomogeneous fields

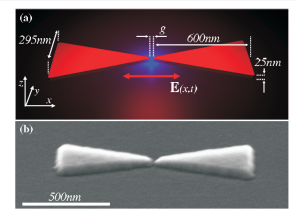


ATI for a model atom (TDSE-1D)
Electron energy spectra

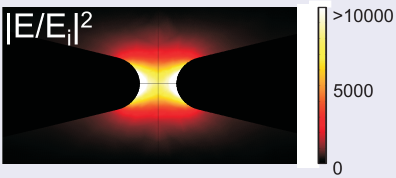


ATI for H (TDSE-3D)
Two-dimensional photoelectron spectra

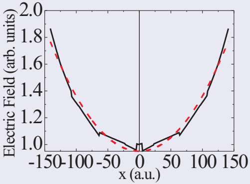
Selected Results III
HHG driven by plasmonic 'real' fields



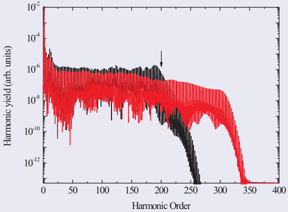
Sketch and TEM image
of the bow-tie nanostructure



FDTD simulation of the plasmonic
field enhancement

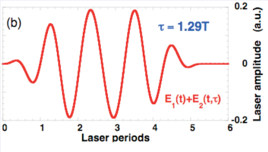


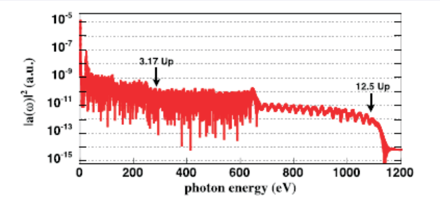
Cut along the polarization axis



HHG spectra for a model atom
(TDSE-1D)

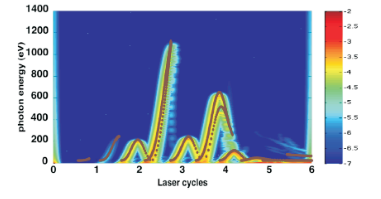
Temporal synthesized field





HHG in He (TDSE-3D)

Spatially nonhomogeneous (linear) field

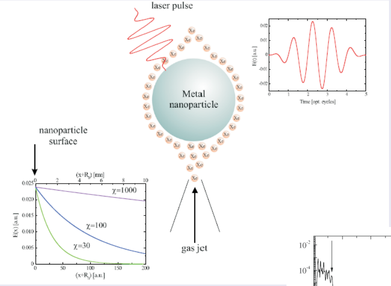


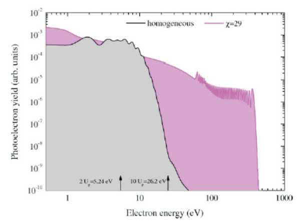
Time-frequency analysis+ classical simulations

Selected Results V

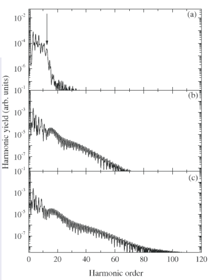
HHG and ATI driven by plasmonic near-fields

Sketch of the setup





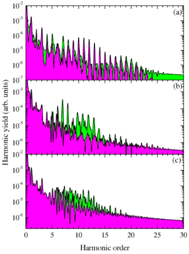
ATI for a model atom (TDSE-1D)



HHG for a model atom (TDSE-1D)

Selected Results VI
HHG driven by metal nanotip photoemission

- **TDSE-1D model (used for ATI)**
- **Potential barrier (metal surface)**
- **Image charge**
- **Actual metal tip parameters**
- **Inclusion of a DC field**



HHG spectra for a metal Au nanotip

Summary of results I
Chronological

- M. F. Ciappina, J. Biegert, R. Quidant and M. Lewenstein, *High-order harmonic generation from inhomogeneous fields*, Phys. Rev. A **85**, 033828 (2012).
- T. Shaaran, M. F. Ciappina and M. Lewenstein, *Quantum-orbit analysis of high-order harmonic generation by resonant plasmon field enhancement*, Phys. Rev. A **86**, 023408 (2012).
- M. F. Ciappina, J. A. Pérez-Hernández, T. Shaaran, J. Biegert, R. Quidant and M. Lewenstein, *Above threshold ionization by few-cycle spatially inhomogeneous fields*, Phys. Rev. A **86**, 023413 (2012).
- M. F. Ciappina, Srdjan S. Acimovic, T. Shaaran, J. Biegert, R. Quidant and M. Lewenstein, *Enhancement of high harmonic generation by confining electron motion in plasmonic nanostructures*, Opt. Exp. **20**, 26261-26274 (2012).
- T. Shaaran, M. F. Ciappina and M. Lewenstein, *Estimating the plasmonic field enhancement using high-order harmonic generation: the role of the field inhomogeneity*, J. of Mod. Opt. **59**, 1634-1639 (2012).
- M. F. Ciappina, T. Shaaran and M. Lewenstein, *High order harmonic generation in noble gases using plasmonic field enhancement*, Ann. der Phys. (Berlin) (Special Issue) **525**, 97-106 (2013).
- J. A. Pérez-Hernández, M. F. Ciappina, M. Lewenstein, L. Roso and A. Zaïr, *Beyond Carbon K-edge harmonic emission using spatial and temporal synthesized laser fields*, Phys. Rev. Lett. **110**, 053001 (2013).

Summary of results II
Chronological

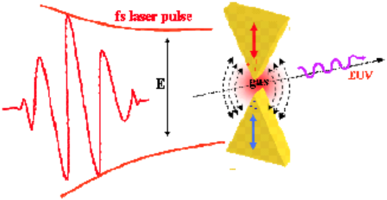
- T. Shaaran, M. F. Ciappina, R. Guichard, J. A. Pérez-Hernández, L. Roso, M. Arnold, T. Siegel, A. Zaïr and M. Lewenstein, *High-order harmonic generation by enhanced plasmonic near-fields in metal nanoparticles*, Phys. Rev. A **87**, 041402(R) (2013).
- T. Shaaran, M. F. Ciappina and M. Lewenstein, *Quantum-orbit analysis of above-threshold ionization driven by an intense spatially inhomogeneous field*, Phys. Rev. A **87**, 053415 (2013).
- M. F. Ciappina, J. A. Pérez-Hernández, T. Shaaran, L. Roso and M. Lewenstein, *Electron-momentum distributions and photoelectron spectra of atoms driven by an intense spatially inhomogeneous field*, Phys. Rev. A **87**, 063833 (2013).
- M. F. Ciappina, T. Shaaran, R. Guichard, J. A. Pérez-Hernández, L. Roso, M. Arnold, T. Siegel, A. Zaïr and M. Lewenstein, *High energy photoelectron emission from gases using plasmonics enhanced near-fields*, Las. Phys. Lett. **10**, 105302 (2013).
- M. F. Ciappina, J. A. Pérez-Hernández and M. Lewenstein, *ClassSTRONG: Classical simulations in Strong Field Physics*, Comp. Phys. Comm. **185**, 398-406 (2014).
- M. F. Ciappina, J. A. Pérez-Hernández, T. Shaaran, M. Lewenstein, M. Krüger and P. Hommelhoff, *High-order harmonic generation driven by metal nanotip photoemission: Theory and simulations*, Phys. Rev. A **89**, 013409 (2014).
- M. F. Ciappina, J. A. Pérez-Hernández, T. Shaaran and M. Lewenstein, *Coherent XUV generation driven by sharp metal tips photoemission*, EJPD (Special Issue) **68** 172 (2014).

- Theoretical modeling of novel laser-matter processes: quantal, semiclassical & classical
- Reliability of the theoretical approaches and their predictions
- Prospect of table-top high repetition rates and strong laser sources using plasmonic fields: looking for experimental alternatives

- Extension of the theoretical approaches to more complex nonhomogeneous fields & macroscopic effects modeling (problem of incoherent radiation)
- Exploring (theoretical & experimentally) strong field related phenomena (above threshold photoemission, HHG in solids, electron emission and radiation from thin films, etc.)
- Attosecond physics at nanometric scale (attosecond streaking of plasmonic fields, tailoring electron trajectories, molecular dynamics)

The (near) future
Perspectives & challenges II

- Application of Quantum Optimal Control Theory (QOCT) tools to plasmonic fields. Is it possible? Yes (theoretically)
- Experimental implementation: tailoring nanostructures, studies of damage thresholds & new materials. **Goal**: production of high energy XUV/EUV photons without amplification stage

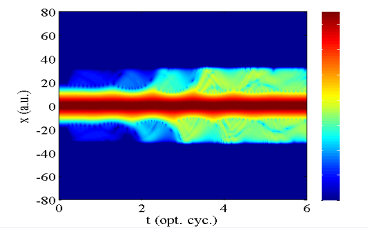


Sketch of HHG enhancement using a plasmonic structure

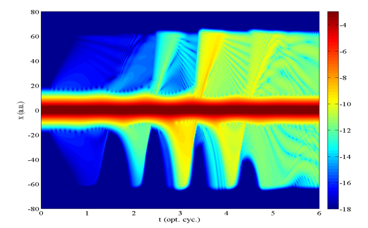
The (near) future

Work in progress: understanding the electron trajectories

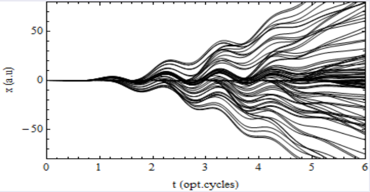
$|\Psi(x,t)|^2$ 1D-TDSE Homogeneous



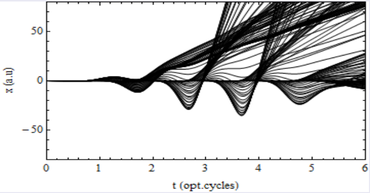
$|\Psi(x,t)|^2$ 1D-TDSE Exp decay



Electron trajectories (homog.)



Electron trajectories (exp)



Thank You

Thank you
for your attention !

Questions & Answers