High-order harmonic generation enhanced by x rays

Christian Buth

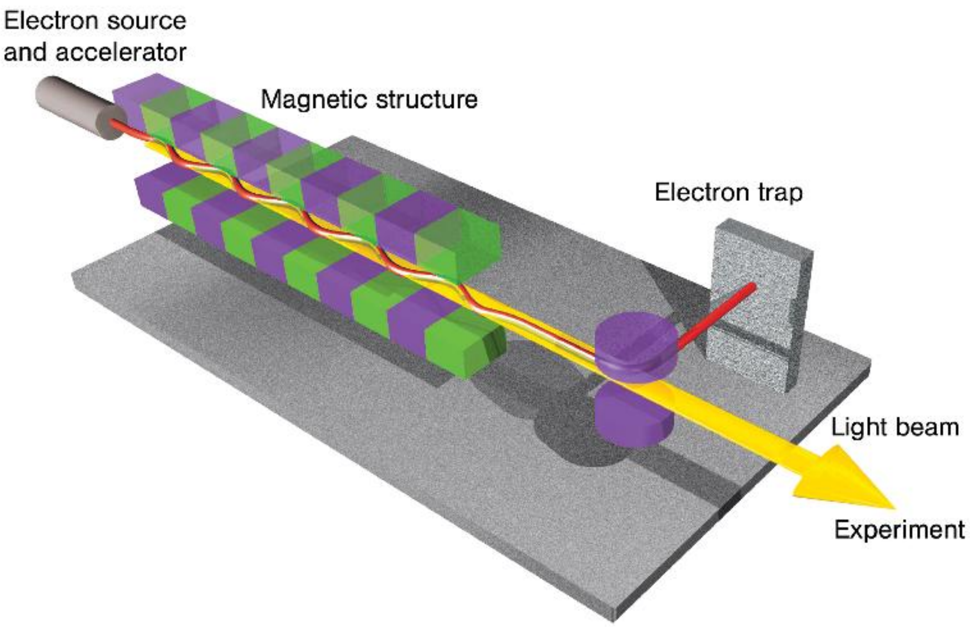
21 July 2011

Atomic, Molecular, and Optical Physics Group, X-Ray Science Division,
Advanced Photon Source, Argonne National Laboratory, Argonne, Illinois, USA

Contents

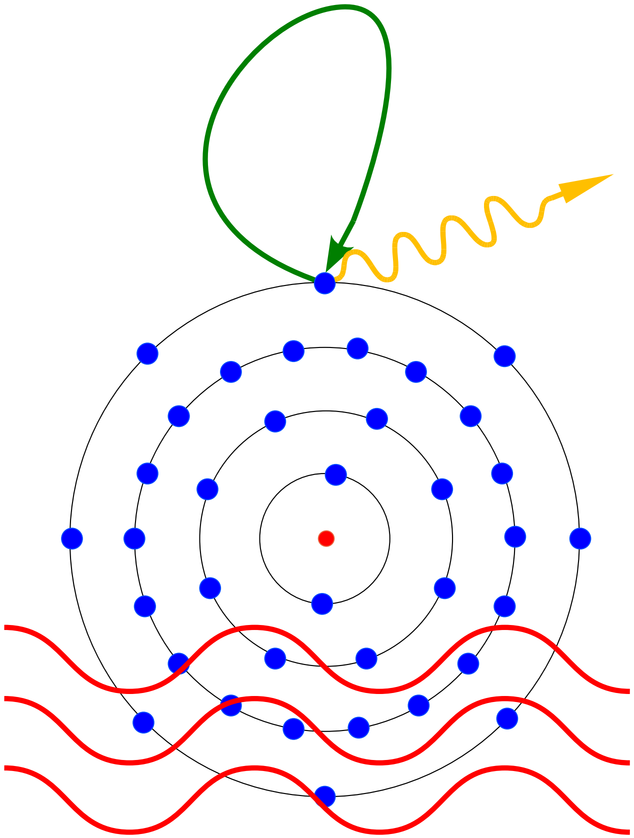
XUV / x-ray free electron lasers (FELs)

- Free-electron laser in Hamburg (FLASH)
- Linac Coherent Light Source (LCLS)
- Electrons emit x rays spontaneously
- Downstream: electrons interact with the radiation
- Self-amplified spontaneous emission (SASE)
- **Unprecedented XUV / x-ray intensities**



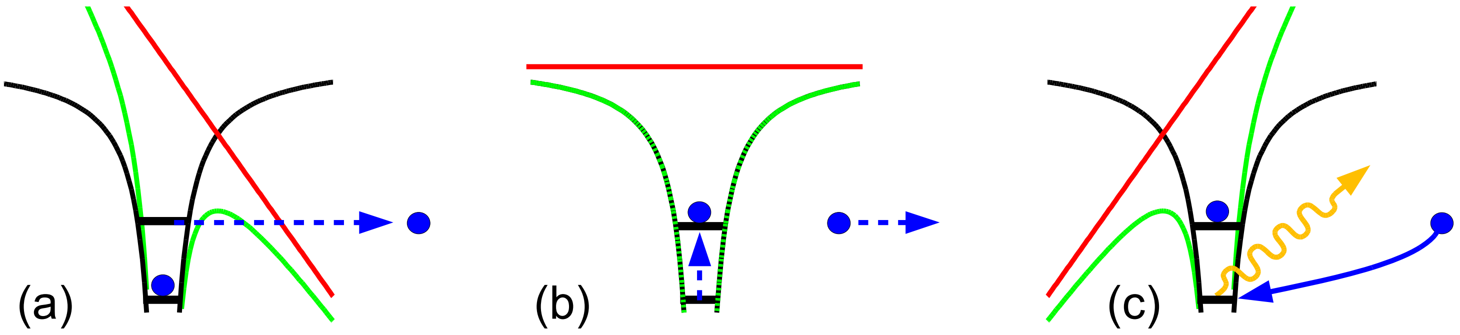
High harmonic generation (HHG)

- High harmonic generation (HHG) by atoms in intense optical laser fields
- Theory mostly based on the **single-active electron** (SAE) approximation
- Restriction to HHG from **valence electrons**
- Two-electron scheme of a nonsequential double recombination
- HHG in the presence of XUV light with many-electron effects with a frequency-dependent polarization



P. Kova, F. Wilken, D. Bauer, C. H. Keitel, Phys. Rev. Lett. 98, 043904 (2007)
A. Fleischer, Phys. Rev. A 78, 053413 (2008)

HHG with resonant excitation by x rays



(a) the atomic valence is **tunnel ionized**

(b) **free propagation** in the electric field of the optical laser

(c) the electron **recombines** with the ion emitting HHG radiation

C. Buth, M. C. Kohler, J. Ullrich, C. H. Keitel, submitted, arXiv:1012.4930

Theory of HHG with resonant excitation by x rays

- **Two-electron basis** states $|a\rangle\otimes|c\rangle, |\vec{k}\rangle\otimes|c\rangle, |\vec{k}\rangle\otimes|a\rangle$
- Hamiltonian $\hat{H}=\hat{H}_{\mathrm{A}}+\hat{H}_{\mathrm{L}}+\hat{H}_{\mathrm{X}}$
- Wavepacket

$$|\Psi,t\rangle=a(t)\,\mathrm{e}^{-\mathrm{i}\phi_1 t}|a\rangle\otimes|c\rangle+\int\limits_{\vec{k}\in R^3}\Big[b_a(\vec{k},t)\,\mathrm{e}^{-\mathrm{i}\phi_2 t}|\vec{k}\rangle\otimes|c\rangle+b_c(\vec{k},t)\,\mathrm{e}^{-\mathrm{i}\phi_3 t}|\vec{k}\rangle\otimes|a\rangle\Big]\mathrm{d}^3 k$$

- Equations of motion, Rabi matrix
- Dipole matrix element

$$\tilde{D}(\Omega)=-\mathrm{i}\sum_{\substack{i\in\{a,c\}\\ j\in\{+,-\}}} U_{ij}\,w_j\int\limits_0^\infty\sqrt{\frac{(-2\pi\mathrm{i})^3}{\tau^3}}\,\mathrm{e}^{-\mathrm{i}F_{0,j}(\tau)}\sum_{N=-\infty}^{\infty}\mathrm{i}^N J_N\!\left(\frac{U_{\mathrm{P}}}{\omega_{\mathrm{L}}}C(\tau)\right)\mathrm{e}^{-\mathrm{i}N\omega_{\mathrm{L}}\tau}$$
$$\times\sum_{M=-\infty}^{\infty}\check{b}_{M-N,i}(\tau)\,h_{M,0,i}(\Omega,\tau)\,\mathrm{d}\tau$$

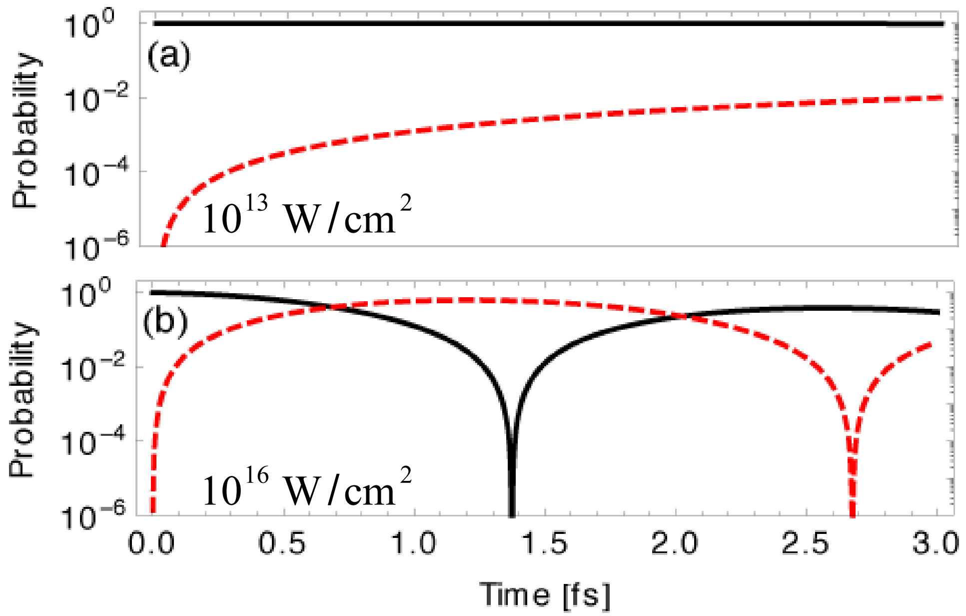
- Harmonic photon number spectrum (HPNS) along x axis

$$\frac{\mathrm{d}^2P(\Omega)}{\mathrm{d}\Omega\,\mathrm{d}\Omega_{\mathrm{S}}}=4\pi\,\Omega\,\rho(\Omega)\,|\tilde{D}(\Omega)|^2$$

C. Buth, M. C. Kohler, J. Ullrich, C. H. Keitel, submitted, arXiv:1012.4930

Rabi flopping in a two-level system

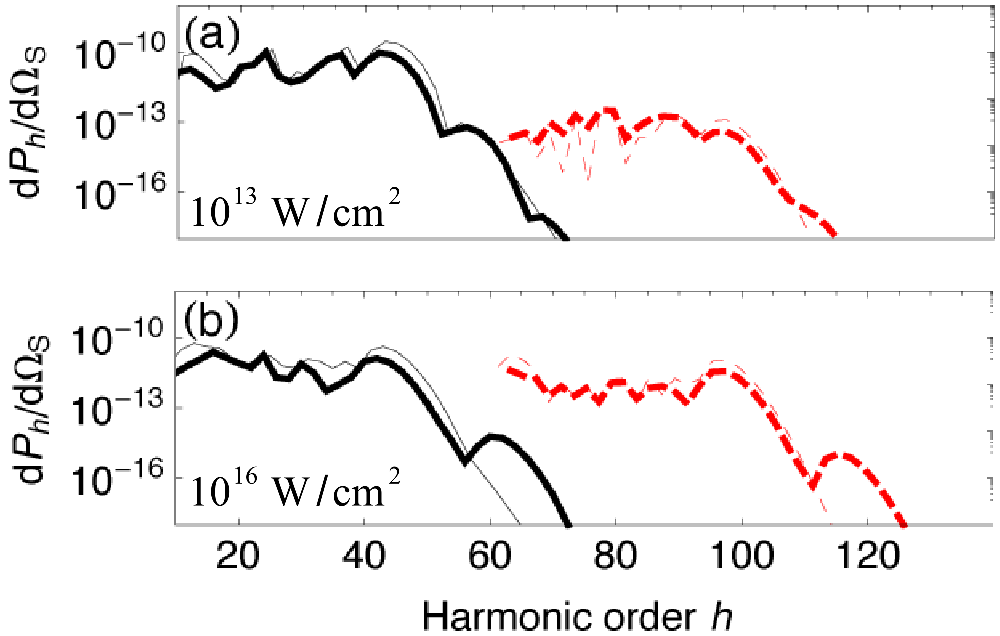
- Two-level system in **krypton cation** $|\vec{k}\rangle \otimes |c\rangle$ and $|\vec{k}\rangle \otimes |a\rangle$ with valence 4*p* and core 3*d*
- Probabilities to find the electron in the **valence** or the **core** state
- Excursion time of electron for an 800 nm laser ≈ 1 fs
- **Rabi oscillations** on the time scale of HHG
- **Krypton** $|c\rangle \rightarrow |a\rangle$ transition by XUV light with 82.6 eV photon energy



C. Buth, M. C. Kohler, J. Ullrich, C. H. Keitel, submitted, arXiv:1012.4930

HHG spectra for semi-CW laser and XUV light

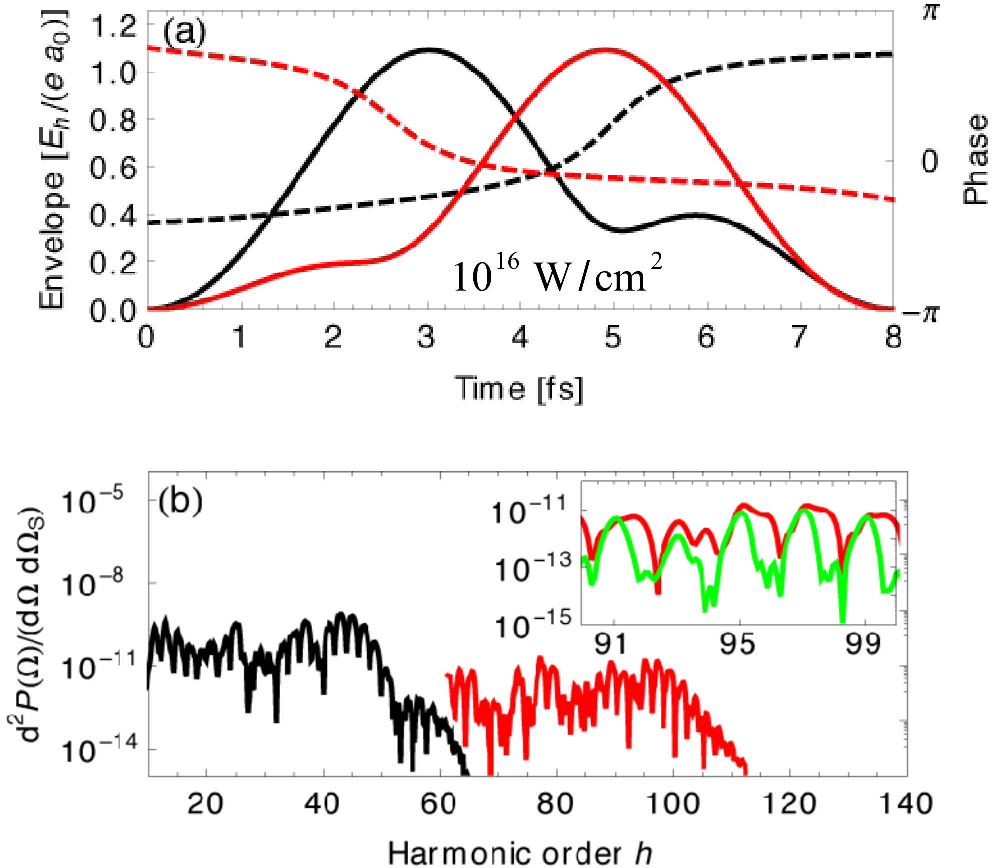
- HHG spectra of **krypton**
- 800 nm laser and XUV light with **constant amplitude** starting at $t = 0$ s with 3×10^{14} W / cm^2
- Pulse duration of both XUV and optical light is 3 optical cycles
- Valence 4*p*, core 3*d*
- Transition $|c\rangle \rightarrow |a\rangle$ by XUV light with 82.6 eV photon energy
- **Valence-hole** and **core-hole** recombination
- Core recombination is determined by probability to find electron in the valence hole



C. Buth, M. C. Kohler, J. Ullrich, C. H. Keitel, submitted, arXiv:1012.4930

HHG with a SASE FEL pulse for krypton

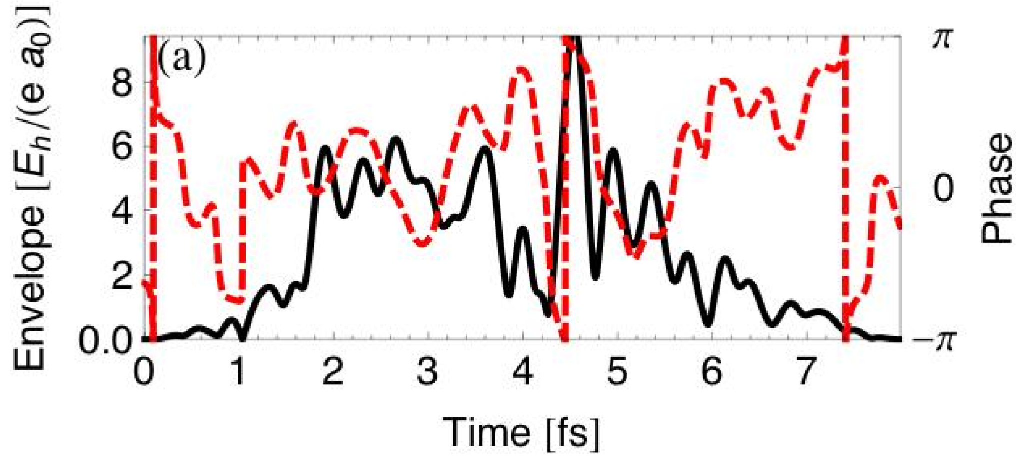
- SASE pulses of FELs are only **partially coherent**
- Sensitive dependence of HHG on XUV pulse shape (solid lines: amplitude, dashed lines: phase)
- Upconversion of HHG by XUV excitation leads to **novel light**
- **Valence-hole** and **core-hole** recombination



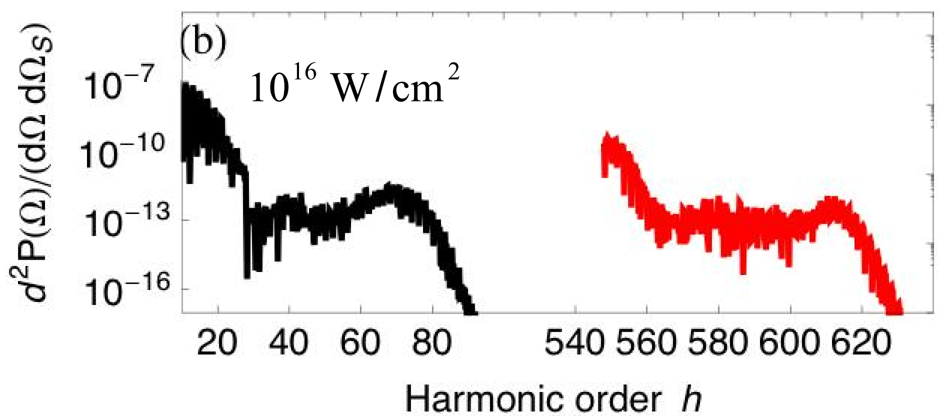
C. Buth, M. C. Kohler, J. Ullrich, C. H. Keitel, submitted, arXiv:1012.4930
T. Pfeifer, Y. Jiang, S. Düsterer, R. Moshammer, J. Ullrich, Opt. Lett. 35, 3441 (2010)

HHG with resonant x rays for neon

- SASE pulse at 848.3 eV of 7.5 fs duration, 0.2 fs coherence time
- **Amplitude** and **phase**



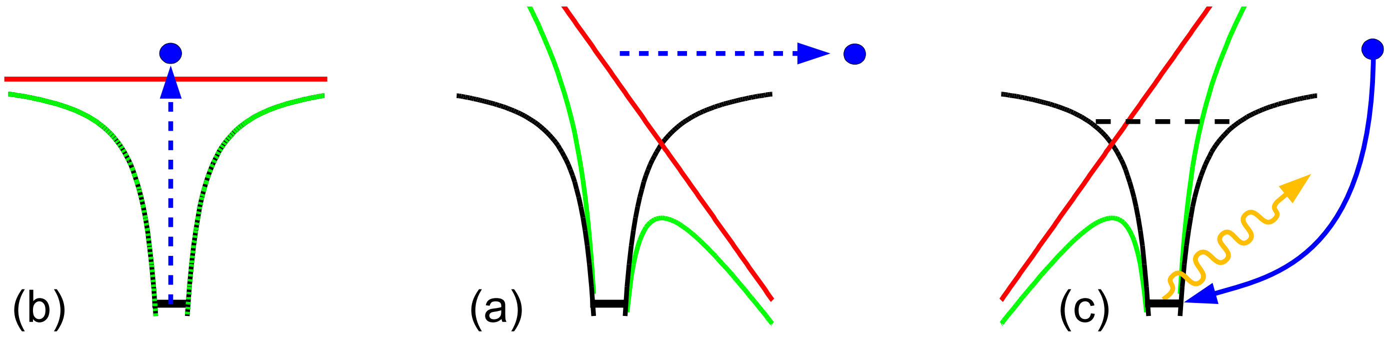
- Harmonic photon number spectrum for **neon**
- **Valence-hole** and **core-hole** recombination



C. Buth, T. Pfeifer, J. Ullrich, *et al.*, LCLS beam time proposal, 13 September 2011

Contents

HHG with direct ionization by x rays



(a) a core electron is **ionized** by x rays

(b) **free propagation** in the electric field of the optical laser

(c) the electron **recombines** with the core hole emitting HHG radiation

Theory of x-ray ionization and HHG

- For **x-ray ionization**, use one-electron basis states $|c\rangle$, $|\vec{k}\rangle$
- Hamiltonian $\hat{H} = \hat{H}_{\mathrm{A}} + \hat{H}_{\mathrm{L}} + \hat{H}_{\mathrm{X}}$
- Wavepacket

$$|\Psi, t\rangle = \mathrm{e}^{-\mathrm{i} I_{\mathrm{P}} t}\left(a(t)|c\rangle + \int_{\vec{k}\in R^3} b(\vec{k}, t)|\vec{k}\rangle \mathrm{d}^3 k \right)$$

- Equations of motion; find time-dependent dipole moment $D(t)$
- Transition dipole matrix element

$$\tilde{D}(\Omega + \omega_{\mathrm{X}}) = \frac{(\Omega + \omega_{\mathrm{X}})^4}{T} \int_0^T D(t)\, \mathrm{e}^{-\mathrm{i}\Omega t}\, \mathrm{d}t$$

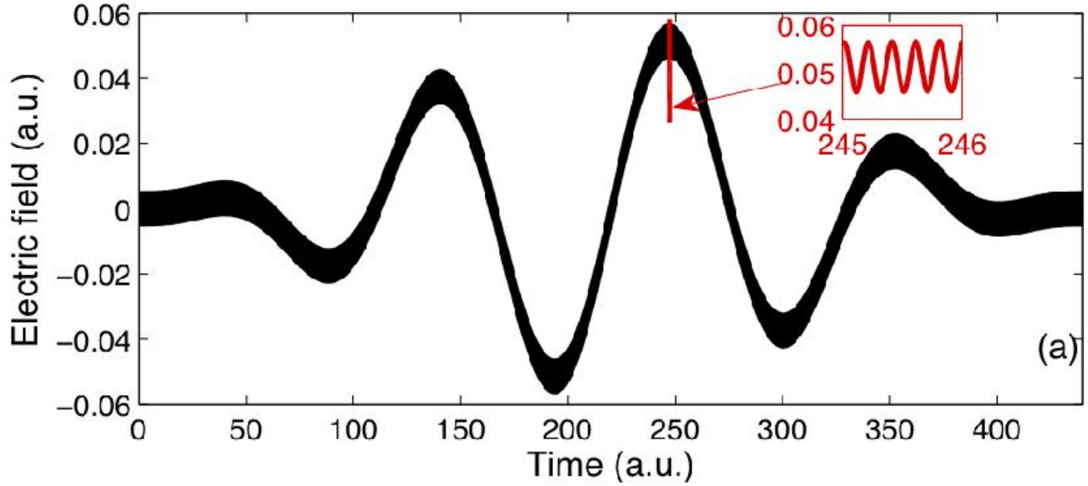
- Harmonic photon number spectrum (HPNS) along x axis

$$\frac{\mathrm{d}^2 P(\Omega)}{\mathrm{d}\Omega\, \mathrm{d}\Omega_{\mathrm{S}}} = 4\pi\Omega\rho(\Omega)|\tilde{D}(\Omega)|^2$$

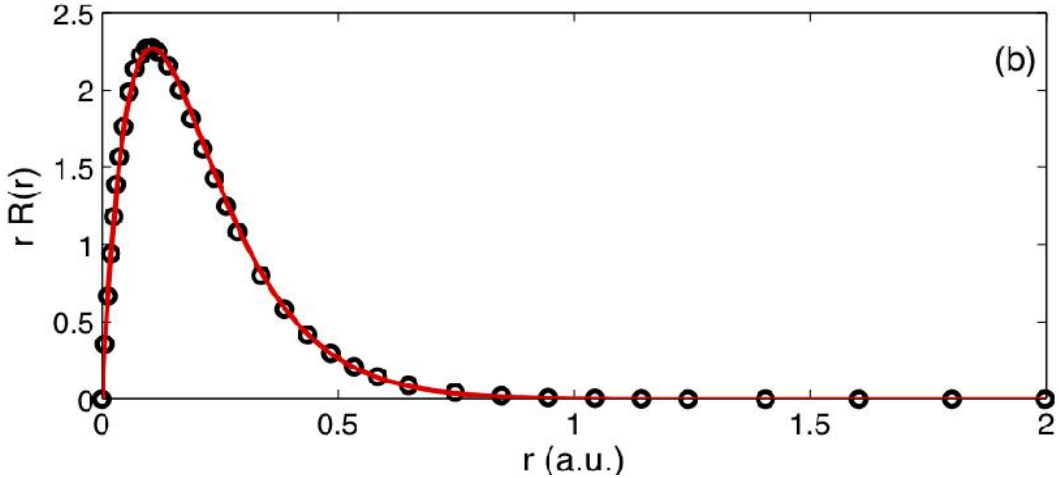
S. V. Popruzhenko, D. F. Zaretsky, W. Becker, Phys. Rev. A 81, 063417 (2010)

Electric fields and atomic orbital for neon

(a) The superimposed optical and x-ray electric fields

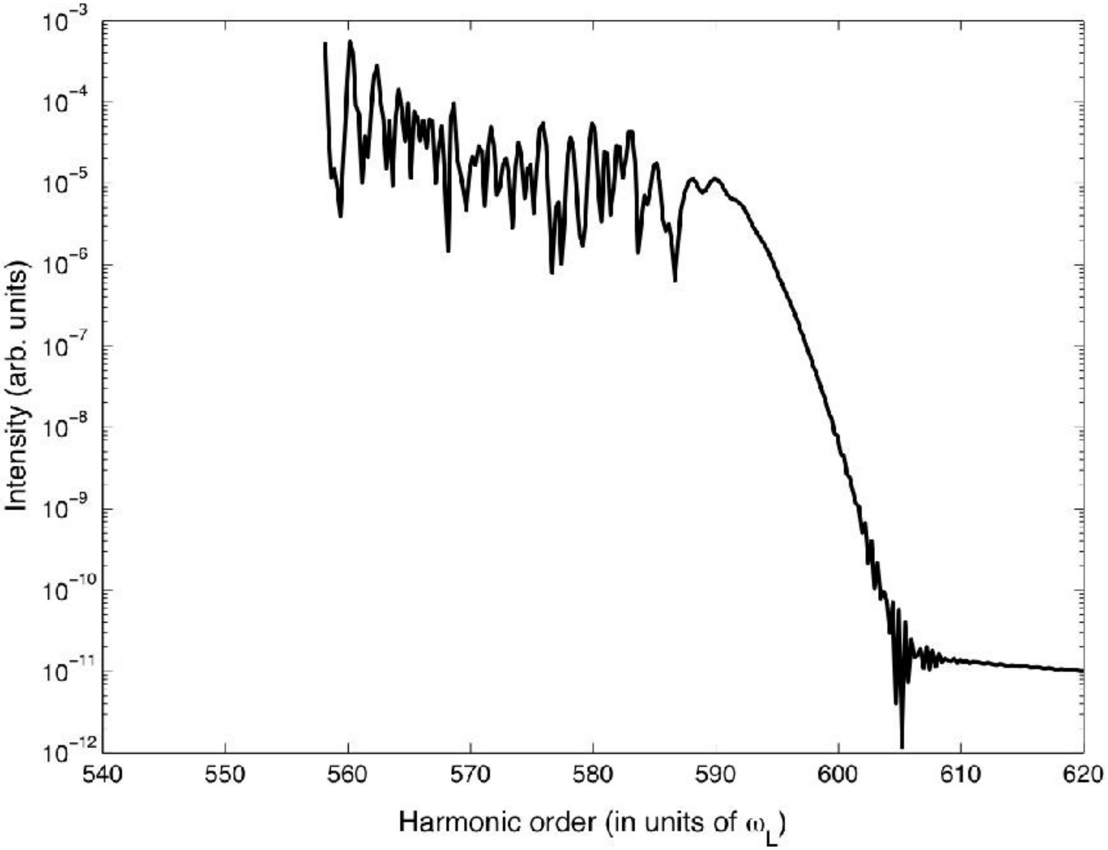


(b) The radial wavefunction of the 1*s* orbital of a **neon** atom



High harmonic spectrum with x rays for neon

- The **optical laser** has the properties:
  - Intensity: 3×10^{14} W / cm^2
  - Pulse duration: 4 optical cycles
  - Pulse shape: $\sin^2$ profile
  - Carrier envelope phase: $\vartheta = 0$
- The **1*s*-ionization potential of neon** corresponds to about 558 ω_L



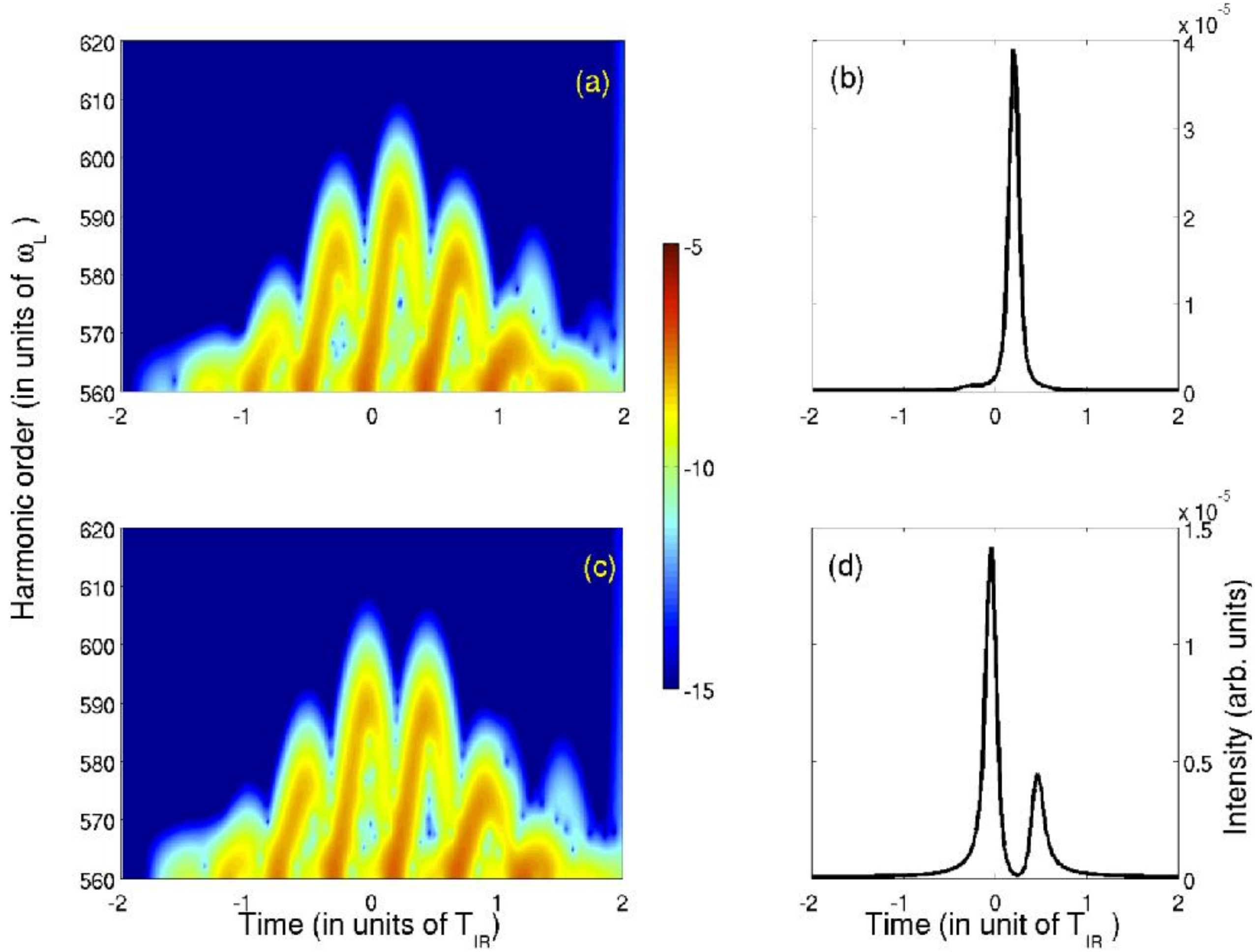
Time-frequency analysis of HHG for neon

(a) Carrier envelope phase is $\theta = 0$

(b) Attosecond pulses from harmonics with $\Omega > 590\ \omega_L$ for (a)

(c) Case (a) with $\theta = \pi / 2$

(d) Case (b) for (c)



Applications of the novel light

- Generate isolated **attosecond x-ray pulses**

  E. Goulielmakis *et al.*, Science 320, 1614 (2008)

- Reconstruction of SASE FEL pulses with **frequency resolved optical gating** (FROG)

  R. Trebino, *Frequency-resolved optical gating: the measurement of ultrashort laser pulses* (Kluwer Academic Publishers, Boston, Dordrecht, London, 2000)

- Ultrafast time-dependent **chemical imaging** of core holes

  T. Morishita *et al.*, Phys. Rev. Lett. 100, 013903 (2008)

C. Buth, M. C. Kohler, J. Ullrich, C. H. Keitel, submitted, arXiv:1012.4930

Conclusion

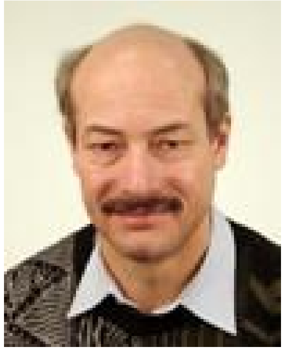
- **Two-color physics**: x-ray free electron lasers and optical lasers
- Valence HHG induced by optical laser
- X rays lead to **resonant excitation** of the intermediate ion

  OR

  x rays lead to **direct excitation / ionization** of the neutral atom
- Recombination of the continuum electron with the core hole
- **Nonlinear upconversion** of HHG light
- Potential for exciting applications

Acknowledgment

Christoph H. Keitel



Joachim Ullrich

Karen Z. Hatsagortsyan

Feng He

Markus C. Kohler