

Novel light from high-order harmonic generation manipulated by XUV light

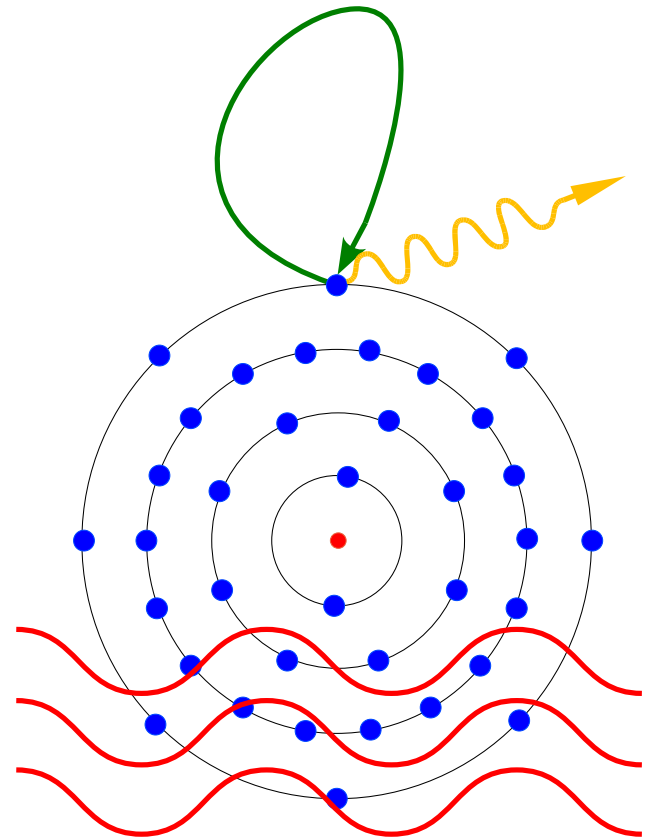
Christian Buth, Markus C. Kohler, Joachim Ullrich, Christoph H. Keitel

14 March 2011

Max-Planck-Institut für Kernphysik, Heidelberg, Germany

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

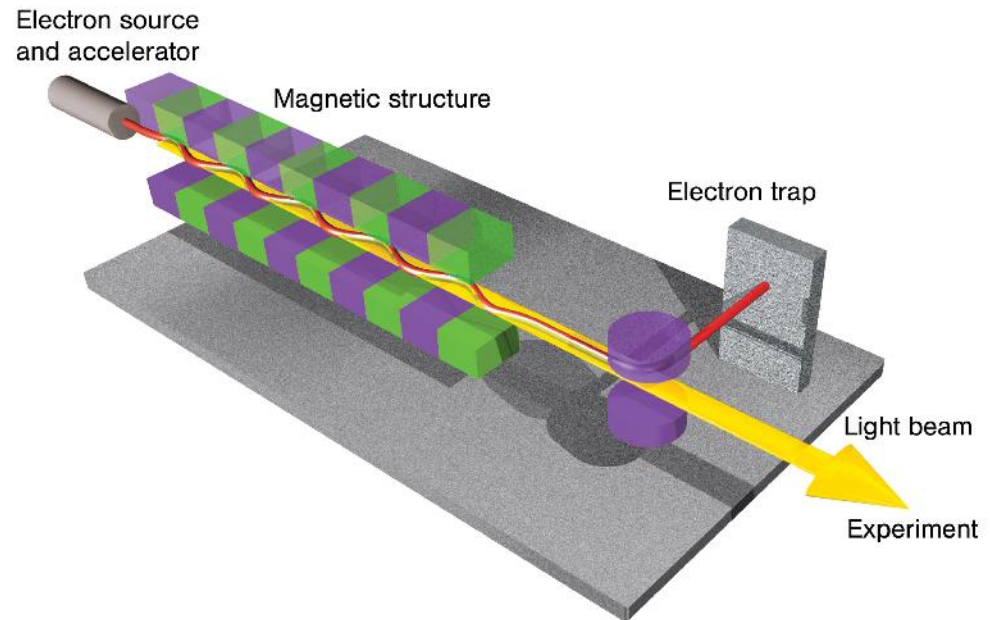


P. Kova, F. Wilken, D. Bauer, C. H. Keitel, Phys. Rev. Lett. 98, 043904 (2007)

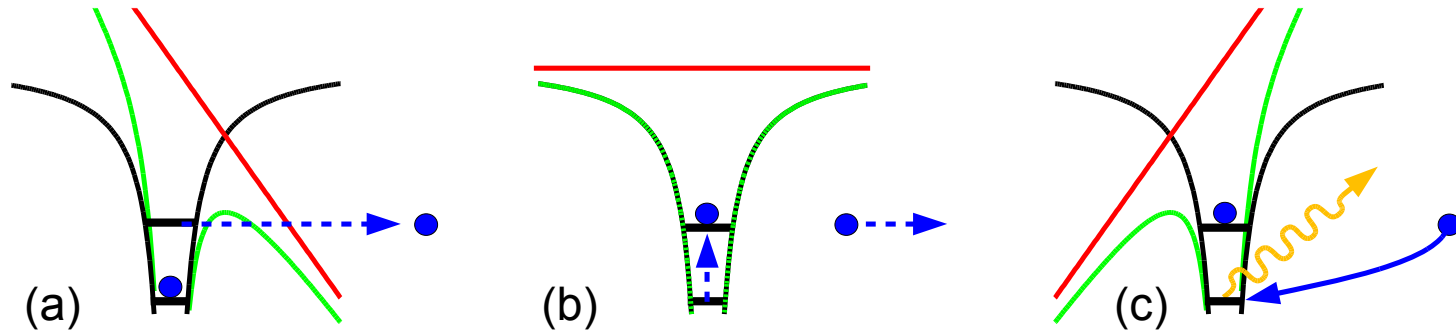
2 A. Fleischer, Phys. Rev. A 78, 053413 (2008)

XUV/x-ray free electron lasers

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



HHG with XUV in terms of the three-step model



- (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



Theory of resonant XUV excitation and HHG

■ Two-electron basis states $|a\rangle \otimes |c\rangle, |\vec{k}\rangle \otimes |c\rangle, |\vec{k}\rangle \otimes |a\rangle$

■ Hamiltonian $\hat{H} = \hat{H}_A + \hat{H}_L + \hat{H}_X$

■ Wavepacket

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

■ Equations of motion, Rabi matrix

■ Dipole matrix element

$$\begin{aligned} \tilde{D}(\Omega) = & -i \sum_{\substack{i \in \{a, c\} \\ j \in \{+, -\}}} U_{ij} w_j \int_0^\infty \sqrt{\frac{(-2\pi i)^3}{\tau^3}} e^{-iF_{0,j}(\tau)} \sum_{N=-\infty}^\infty i^N J_N \left(\frac{U_P}{\omega_L} C(\tau) \right) e^{-iN\omega_L \tau} \\ & \times \sum_{M=-\infty}^\infty \check{b}_{M-N,i}(\tau) h_{M,0,i}(\Omega, \tau) d\tau \end{aligned}$$

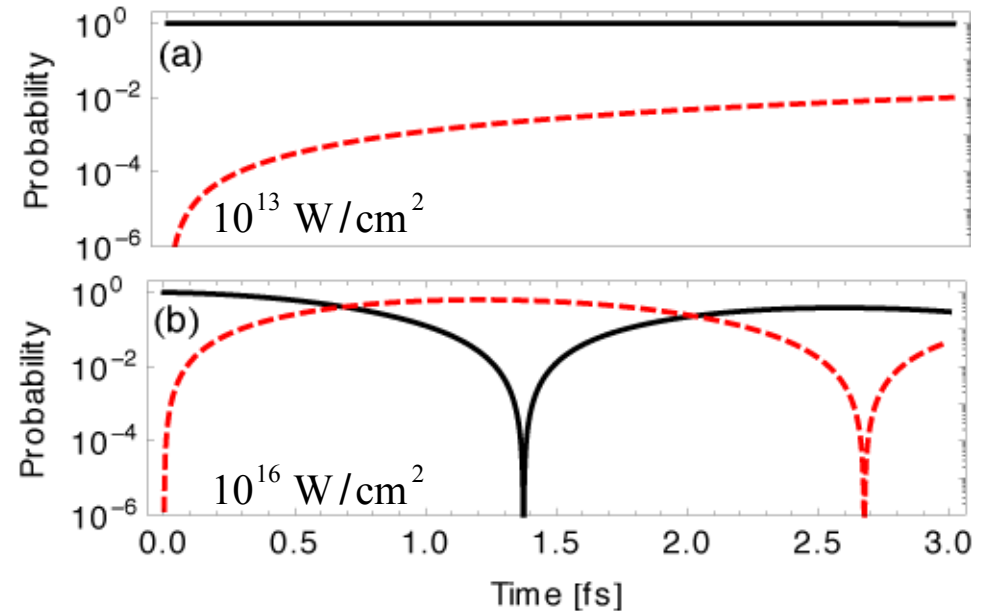
■ Harmonic photon number spectrum (HPNS) along x axis

$$\frac{d^2 P(\Omega)}{d\Omega d\Omega_s} = 4\pi \Omega \rho(\Omega) |\tilde{D}(\Omega)|^2$$



Rabi flopping in a two-level system

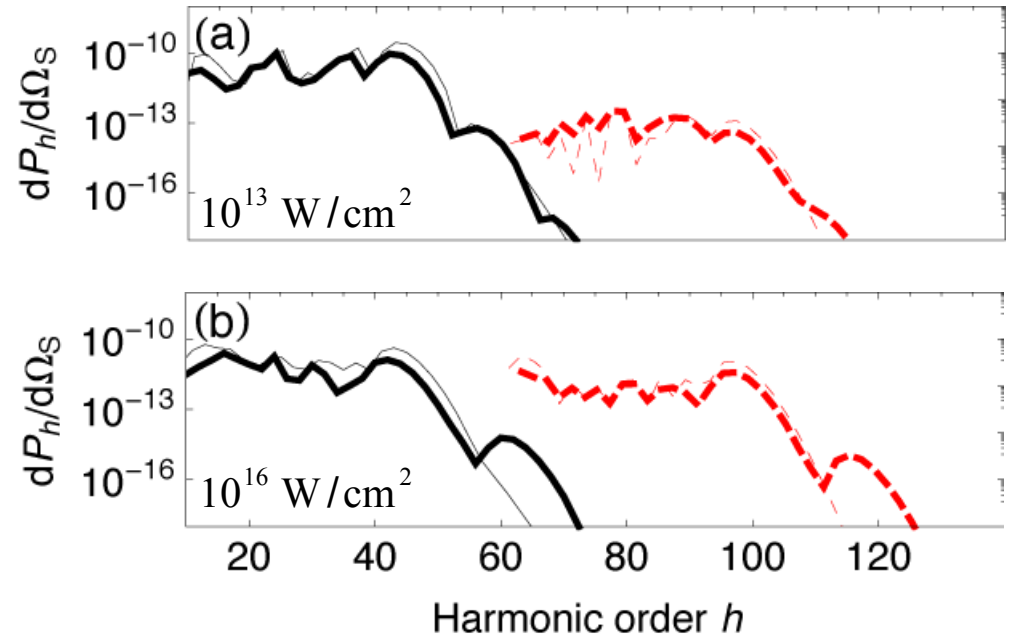
- Two-level system in **cation** $|\vec{k}\rangle \otimes |c\rangle$ and $|\vec{k}\rangle \otimes |a\rangle$
- Probabilities to find the electron in the valence or the core state
- Excursion time of electron for an 800 nm laser ≈ 1 fs
- **Rabi oscillations** on the time scale of HHG





HHG spectra for semi-CW laser and XUV light

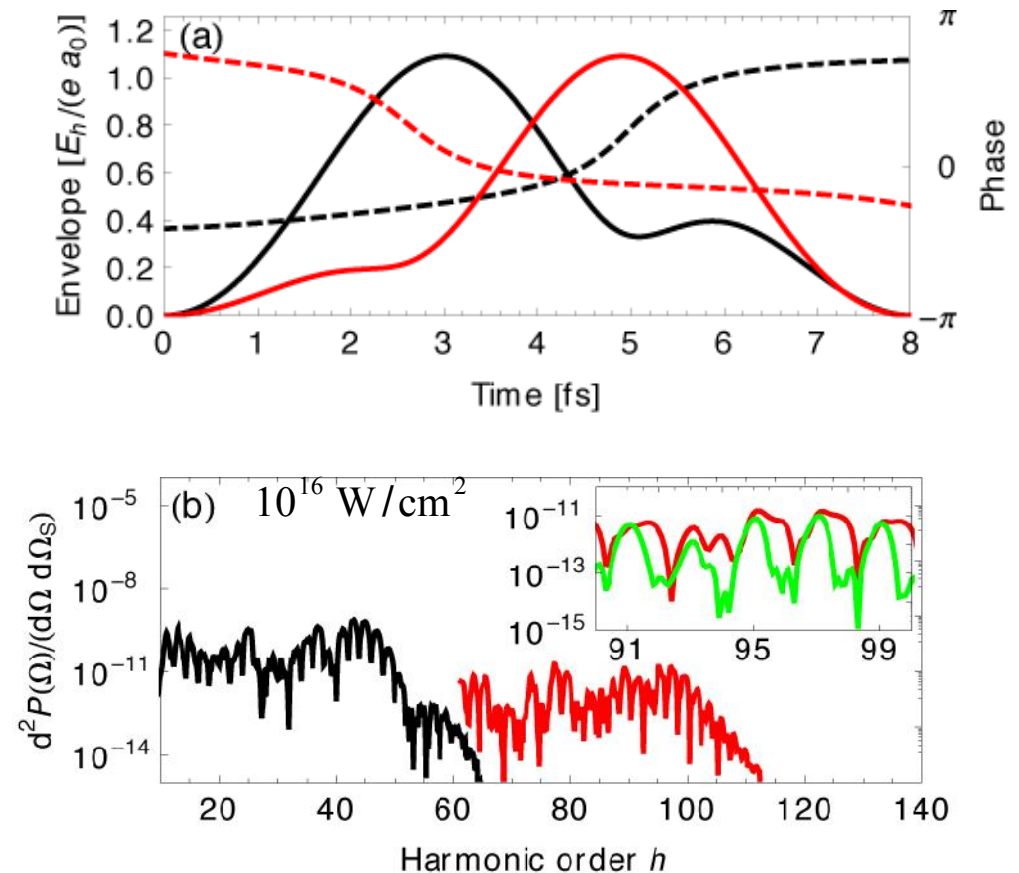
- HHG spectra of **krypton**
- 800 nm laser and XUV light with **constant amplitude** starting at $t = 0$ s
- Valence $4p$, core $3d$
- Transition $|c\rangle \rightarrow |a\rangle$ by XUV
- Core recombination is determined by probability to find electron in the valence hole





HHG with a SASE FEL pulse

- SASE pulse of FELs is only **partially coherent**
- Sensitive dependence of HHG on XUV pulse shape
- Upconversion of HHG by XUV excitation leads to **novel light**



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

8 T. Pfeifer, Y. Jiang, S. Düsterer, R. Moshhammer, J. Ullrich, Opt. Lett. 35, 3441 (2010)





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)



Conclusion

- **Two-color physics**: XUV free electron lasers and optical lasers
- Valence HHG induced by optical laser
- XUV light leads to **resonant excitation** of intermediate ion
- Recombination of continuum electron with core hole
- **Nonlinear upconversion** of HHG light
- Potential for exciting applications

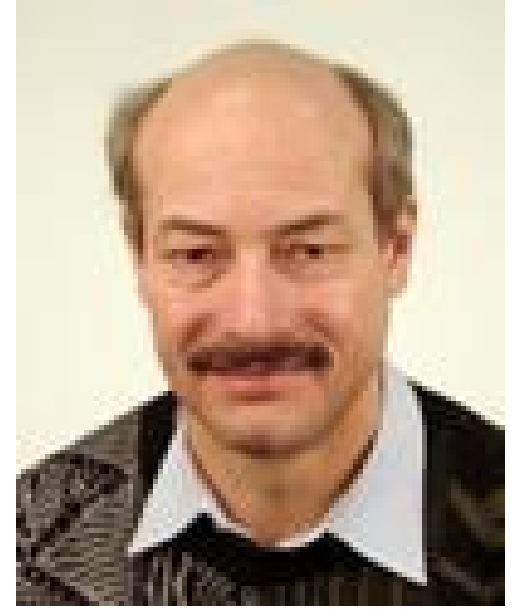
Acknowledgment



Markus C. Kohler



Christoph H. Keitel



Joachim Ullrich