

XUV pump-XUV probe studies of 1fs scale dynamics in atoms and molecules

P. Tzallas¹, E. Skantzakis^{1,2}, P. A. Carpeggiani, A. Palacios³, D. Gray¹, F. Martín^{3,4} and D. Charalambidis^{1,2}

¹Foundation for Research and Technology - Hellas, Institute of Electronic Structure and Laser, PO Box 1527, GR-711 10 Heraklion, Crete, Greece

²Department of Physics, University of Crete, PO Box 2208, GR71003 Heraklion, Crete, Greece

³Departamento de Química, Módulo 13, Universidad Autónoma de Madrid, 28049 Madrid, Spain

⁴Instituto Madrileño de Estudios Avanzados en Nanociencia (IMDEA-Nanociencia), Cantoblanco, 28049 Madrid, Spain

Abstract: Exploiting intense coherent XUV continua, supporting attosecond pulse formation, we have performed the first ever XUV-pump-XUV-probe studies of 1fs scale dynamics in atoms and molecules. Progress towards single shot non-linear XUV autocorrelation is also reported.

Summary

Utilizing many cycle high peak power laser fields in conjunction with Interferometric Polarization Gating [1], [2], coherent 9 eV broad XUV continuum radiation confined in 1fs scale isolated pulses of sub-100nJ pulse energy have been generated. These pulses tightly focused to intensities close to 10^{14}W/cm^2 are exploited in inducing two-XUV-photon ionization of atomic Xenon and molecular Hydrogen.

In Xenon experimental conditions have been achieved at which two-XUV-photon direct double ionization (TPDDI) develops to the dominant double ionization process. Exploiting the observed TPDDI process we succeed in performing a second order autocorrelation measurement of the continuum radiation. Two-XUV-photon double ionization is further utilized in the ever first XUV-pump-XUV probe study of 1fs scale electron dynamics. A coherent manifold of doubly excited and inner-shell excited autoionizing states is pumped through single-XUV-photon absorption. The thus induced and rapidly evolving atomic coherence [3] is probed through double ionization caused by a second XUV-photon, absorbed from a delayed replica of the XUV pulse. Since the pulse duration and the decay time of the excited electron wave packet are very different, pulse duration and wave packet dynamics can be extracted from different regions of the measured trace [4].

Extending the above work to molecules, two-XUV-photon dissociative ionization of H_2 was achieved. One-photon absorption of the broad XUV continuum coherently populated all the optically allowed excited states of H_2 . Nuclear and electronic 1fs scale dynamics are subsequently investigated by means of XUV pump-XUV probe measurements, which are compared to the results of *ab initio* calculations [5]. The revealed dynamics reflects the intrinsic molecular behaviour as the XUV probe pulse hardly distorts the molecular potential. One of the main findings of the work is the measurement of the time needed in order the one photon excite molecule to stretch such that the channel to first dissociative continuum of the ion opens at the given photon energies.

In parallel to the XUV pump-XUV probe applications we are currently developing a single shot non linear XUV autocorrelation approach and device, based on spatially resolved ionization of a gas by two crossed XUV radiation beams. Observation of spatially resolved ionization is through a high spatial resolution ($1\mu\text{m}$) ion microscope. The non linear process required is a two XUV photon ionization of a gas target. The device and its optimization is modeled [6] and spatially resolved two-XUV-photon ionization has been achieved [7].

References

- [1] P. Tzallas, et al., *Nature Phys.* **3**, 846 (2007).
- [2] E. Skantzakis et al., *Opt. Lett.* **34**, 1732 (2009).
- [3] E. Skantzakis, et al., *Phys. Rev. Lett.* **105**, 043902 (2010).
- [4] P. Tzallas et al., *Nature Physics* **7**, 781 (2011).
- [5] P.A. Carpeggiani et al. *PRA*, *in press*
- [6] G. Kolliopoulos et al. (submitted)
- [7] G. Kolliopoulos et al. (submitted)