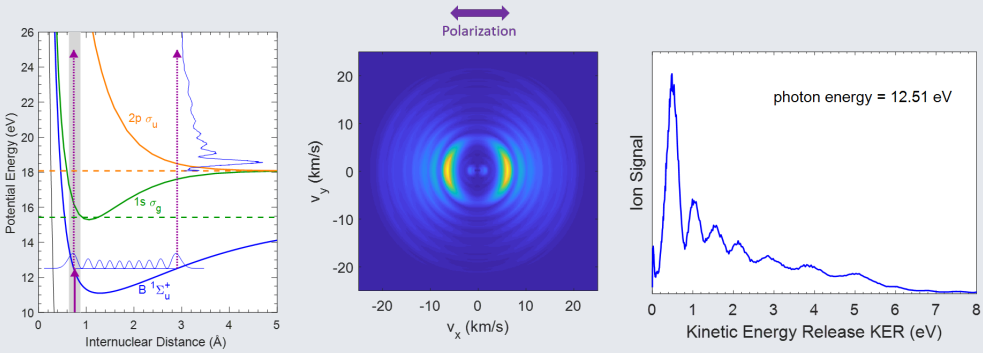
Highlight X. Imaging ultrafast electronic and nuclear dynamics in small molecules using electron-ion coincidence momentum spectroscopy

Fabian Holzmeier, Jennifer Joseph, Jean-Christophe Houver, Danielle Dowek (ISMO), David Bresteau, Thierry Ruchon, Bertrand Carré (LIDyL), John Bozek (SOLEIL)

The achievement of time-resolved images of the coupled electronic and nuclear dynamics in molecules at the atto-to-femtosecond time scale is a hot topic in molecular physics and chemistry communities. Photoionization by XUV pulses is currently involved in such studies either to initiate electronic dynamics in the continuum or to probe the relaxation of transient excited molecular states. Taking advantage of dissociative photoionization, the ImDynCo project combines the use of electron-ion coincidence momentum spectroscopy and that of high order harmonic generation on the 10 kHz beamline at ATTOLAB to access most complete observables consisting of time-resolved molecular frame photoelectron angular distributions (MFPADs) [1], with complementary spectrally resolved experiments at SOLEIL.

For extended studies on the role of electron and nuclear Degrees Of Freedom (DOFs) in photoionization in the non-linear regime, higher pulse intensities are necessary. Free electron lasers constitute such powerful sources of femtosecond XUV pulses and the FERMI facility is unique for the production of coherent VUV pulses.



We have performed first studies of non-linear two-photon dissociative (DI) and non-dissociative (NDI) photoionization of H_2 and D_2 involving resonant excitation of vibrationally resolved intermediate molecular excited states at the FERMI Low Density Matter beamline. We have demonstrated that this process leads to a remarkable enhancement of the DI/NDI ratio by about two orders of magnitude relative to the one-photon ionization at comparable excitation energy [2]. Furthermore, the DI/NDI ratio increases with increasing vibrational levels, highlighting the control of the photoionization outcome by the nuclei DOF of the hydrogenic molecule. This trend supports the striking predictions of Time Dependent Schrödinger Equation ab initio calculations [3]. Velocity Map Images (VMI) of the emitted photoelectrons and photoions were recorded, this technique being well adapted to the low 10-50 Hz repetition rate of FERMI. These energy and angularly resolved results elucidate the main reaction pathways of DI and NDI and establish unambiguously the enhanced role of repulsive excited states of the H_2^+ molecular ions in DI.

This work has been performed.

[1] K. Veryinas, V. Gruson, S.J. Weber, L. Barreau, T. Ruchon, J.-F Hergott, J.-C. Houver, R.R. Lucchese, P. Salières, and D. Dowek *"Molecular frame photoemission by a compb of elliptical high-order harmonics: a sensitive probe of both photodynamics and harmonic complete polarization state" ;Faraday Discussions,* 194, 161 - 183 (2016) (DOI: 10.1039/C6FD00137H)

[2] F. Holzmeier, R. Bello, M. Hervé, T. Baumann, A. Achner, M. Meyer, M. Di Fraia, O. Plekan, P. Finetti, E. Roussel, D. Gauthier, C. Callegari, R. Richter, K. Prince, H. Bachau, A. Palacios, F. Martin and D. Dowek *"Control of H2 dissociative ionization In the non-linear regime using EUV femtosecond pulses @ FERMI' in preparation, to be submitted in 2017*

[3] A. Palacios, H. Bachau, and F. Martin, " *Enhancement and control of H2 dissociative ionization by VUV laser pulses" Phys. Rev. Lett.* 96, 143001 (2006)

Résultats obtenus dans le cadre du projet ImDynCo financé par le thème 3 du LabEx PALM et porté par Danielle Dowek (ISMO), Thierry Ruchon et Bertrand Carré (LIDYL) in collaboration with C. Callegari et al (LDM, FERMI), K. Prince et al (ELETTRA Sincrotone), M. Meyer et al (EU-XFEL Hamburg), F. Martin et al (UAM, Madrid), H. Bachau (CELIA, Université de Bordeaux).