Mapping ultrafast dynamics of highly excited D_2^+ by ultrashort XUV pump - IR probe radiation

You may also be interested in:

Ultrafast Dynamics And Electronic State -- Lifetime Interferences In Chlorine-Containing Molecules
G Goldsztejn, T Marchenko, R Guillemin et al.

Ultrafast dynamics in the bifunctional PENNA neutral and cation molecule induced by ultrashort photoexcitation
Shoutian Sun, Benoît Mignolet and Françoise Remacle

Nonlinear Optical Properties of Indium Phthalocyanine AxiallyGrafted Polystyrene Thin Film
Zhu Rong-Yi, Qiu Xue-Qiong, Chen Yu et al.

Ultrafast Dynamics of Exciton–Exciton and Exciton–Longitudinal Optical-Phonon Scattering Processes in ZnO Epitaxial Thin Films
Jun Takeda, Nobuyuki Arai, Yuki Toshine et al.

Theory of strong-field attosecond transient absorption
Mengxi Wu, Shaohao Chen, Seth Camp et al.

Ultrafast dynamics of excitons in semiconductor quantum dots on a plasmonically active nano-structured silver film
Subrata Batabyal, Abhinandan Makhal, K Das et al.

Ultrafast correlated-electron dynamics and atomic structure
Steven Hutchinson, Michael A Lysaght and Hugo W van der Hart

Mapping ultrafast dynamics of highly excited D_2^+ by ultrashort XUV pump - IR probe radiation

R. Bello[*][1], **L. S. Martin**[†], **C. W. Hogle**[†], **A. Palacios**[*], **J. L. Sanz-Vicario**[§], **X. M. Tong**[‡], **F. Martín**[*◇], **M. Murnane**[†], **H. C. Kapteyn**[†] and **P. Ranitovic**[†][2]

[*] Departamento de Química, M-13, Universidad Autónoma de Madrid, Cantoblanco 28049, Madrid, Spain

[†] JILA and Department of Physics, University of Colorado and NIST, Boulder, CO 80309-0440, USA

[§] Grupo de Física Atómica y Molecular, Instituto de Física, Universidad de Antioquia, Medellín, Colombia

[◇] Instituto Madrileño de Estudios Avanzados (IMDEA) en Nanociencia, Cantoblanco, 28049 Madrid, Spain

[‡] Institute of Materials Science, University of Tsukuba, 1-1-1 Tennodai, Tsukuba, Ibaraki 305-8573, Japan

Synopsis An ultrashort XUV laser pulse ionizes the D_2 molecule creating an electronic and nuclear wave packet, with the dominant contributions from the $2s\sigma_g$ and $2p\pi_u$ ionic states. A delayed interaction with a 780 nm IR field ejects the second electron, leading to the Coulomb explosion of the molecule, whose nuclear fragments, recorded in coincidence, map the dynamics associated to those two ionic excited states. By varying the orientation of the light polarization, one can control the molecular dynamics by modifying the ratio between the ionic states. Experimental and ab initio theoretical data are jointly reported.

The ultimate goal of attosecond science is to achieve control of electron dynamics in matter. Our aim is to trace and control nuclear and electronic wave packets in excited [1] or ionized molecules [2]. In spite of its simplicity, the D_2 molecule is an ideal benchmark for coherent control mechanisms combining accurate experimental and theoretical methodologies. In our experiment, a single XUV pulse (energy 42.7 eV, 7 fs pulse duration) ionizes the D_2 molecule, creating a wave packet involving the four lowest states of D_2^+. After some delay, a 780 nm IR field ejects the second electron, leading to Coulomb explosion. All fragments are measured in coincidence using the well-known COLTRIMS technique, which allows for a complete reconstruction of the fragment momenta, and thus for identification of the molecular orientation. The nuclear kinetic energy (NKE) distributions map the evolution in the more highly excited ionic states, $2s\sigma_g$ and $2p\pi_u$, which is usually hard to obtain. The coherent superposition of nuclear wave packets in the two electronic states $2s\sigma_g$ and $2p\pi_u$ can be manipulated by varying the XUV pulse polarization with respect to the molecular axis. Moreover, the nuclear wave packet associated to each ionic state accumulates a different momentum since they follow different potential energy curves, therefore leading to nuclear fragments with different energies in the Coulomb explosion channel. Additionally, we observe interference between these two channels when identical final states are reached after the multiphoton IR absorption. This is shown in figure 1, where we plot the theoretical data for the double ionization probabilities as a function of the NKE and the time delay between the pulses

for perpendicular (left panel) and parallel (right panel) orientations. In both cases, we observe the coherent superposition of the $2s\sigma_g$ and $2p\pi_u$ states. The time-dependent simulations are carried out using a $\mathcal{L}^2$ close-coupling method [3, 4], including nuclear motion and electron-electron interactions. The wave packets created by the XUV pulse are obtained from the ab initio calculation, while the Coulomb explosion induced by the IR probe pulse is described as a sudden transition in a two-step model. A very good agreement is bound between experiment and theory.
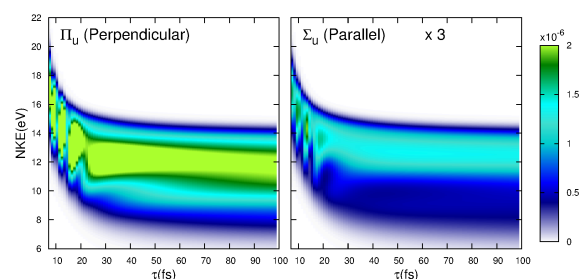

Figure 1. Calculated nuclear kinetic energy (NKE) distributions associated to the Coulomb explosion of D_2 as a function of the pump-probe time delay τ, for perpendicular ($^1\Pi_u$) and parallel ($^1\Sigma_u^+$) molecular orientations with respect to the polarization axis.

References

[1] P. Ranitovic *et al.* 2014 *PNAS* **111** 912

[2] F. Calegari *et al.* 2014 *Science* **346** 336

[3] A. Palacios *et al.* 2007 *PRA* **75** 013408

[4] J. L. Sanz-Vicario *et al.* 2006 *PRA* **73** 033410

[1]E-mail: roger.bello@uam.es
[2]E-mail: pranitovic@lbl.gov