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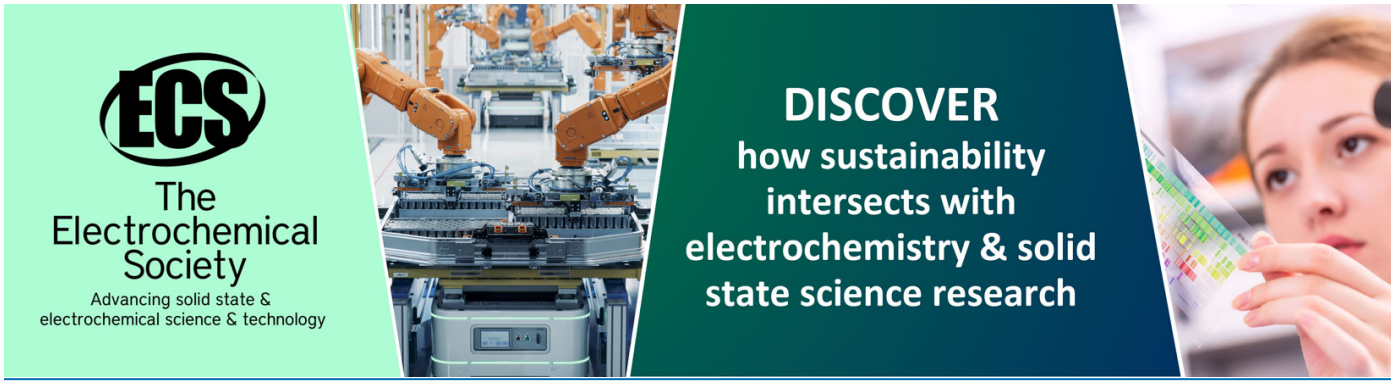

XXIX International Conference on Photonic, Electronic, and Atomic Collisions (ICPEAC2015) IOP Publishing
Journal of Physics: Conference Series **635** (2015) 112101 doi:10.1088/1742-6596/635/11/112101

Mapping the dissociative ionization dynamics of molecular nitrogen with attosecond resolution

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Synopsis We wish to understand the processes underlying the ionization dynamics of N_2 as experimentally induced and studied by recording the kinetic energy release (KER) in a XUV-pump/IR-probe setup. To this end a theoretical model was developed describing the ionization process using Dyson Orbitals and, subsequently, the dissociation process using a large set of diabatic potential energy surfaces (PES) on which to propagate. From said set of PES, a small subset is extracted allowing for the identification of one and two photon processes chiefly responsible for the experimentally observed features.

N_2 is the most abundant element is the earth's atmosphere and as such is of integral importance to processes induced by solar radiation, in particular the dissociation of N_2 induced by solar XUV light in the upper atmosphere. Recent advances in atto second technology have made it possible to gain new insights into ultra fast electron dynamics following sudden removal of an electron [1, 2], providing the perfect tools to study the photo induced dissociation of N_2.

We present the experimental and theoretical investigation of the ionization and dissociation dynamics of N_2. Isolated attosecond pulses (energy range: 16-50 eV, duration: 300 as) were used to ionize N_2 molecules, through a single photon transition. After a variable delay, 4-fs NIR/VIS CEP controlled pulses (peak intensity: $8 \cdot 10^{12}\mathrm{W/cm^2}$) were used to probe the subsequent dissociation dynamics. The angularly resolved momentum distribution of the produced N^+ fragments was measured as a function of the pump-probe delay using a velocity map imaging (VMI) spectrometer. We observed a depletion of a quasi-bound state of N_2^+, 8 fs after zero time delay, together with sub-cycle modulation of the N^+ yield.

To understand the origin of the dynamics a model was developed: The ionization process was simulated assuming an instantaneous transition of the nuclear ground state wave function of N_2 to a set of diabatic PESs of the N_2^+ ion, obtained using *ab initio* multiconfigurational methodology. The relative initial populations of said set of ionic PESs were approximated using Dyson orbitals in first order perturbation theory [3, 4] and modelling the leaving electron by a Coulomb wave. The dissociating ion subjected to the IR pulse was modelled by solving the TDSE using Split Operator [5] and Fast Fourier [6] techniques.

Careful investigation of the data produced by aforementioned techniques yielded the $F^2\Sigma_g$, $3^2\Sigma_g$, $C^2\Sigma_u$ and $5^2\Sigma_u$ states of N_2^+ as being capable of describing most features observed experimentally; most importantly a two photon transition from $F^2\Sigma_g$ to $3^2\Sigma_g$ via $5^2\Sigma_u$ was found to be responsible for the sub-femtosecond dynamics, as it causes the wave packets initially in $F^2\Sigma_g$ and $3^2\Sigma_g$ to interfere.

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