

DIRECT TIME RESOLVED OBSERVATION OF MOLECULAR DYNAMICS INDUCED BY EXTREME-ULTRAVIOLET PHOTOIONIZATION

Arvinder Sandhu*, Etienne Gagnon*, Ariel Paul*, Margaret Murnane*, Henry Kapteyn*
Predrag Ranitovic†, Lew Cocke†

* JILA, University of Colorado and NIST, Boulder, Colorado 80309-0441 USA

† J.R. Macdonald Lab, Physics Department, Kansas State University, Manhattan, KS 66506, USA

Laser-generated high-order harmonics provide a source of extreme-ultraviolet (EUV) radiation with unique capabilities for probing atomic and molecular dynamics. Here we present the first ultrafast molecular dynamics studies that employ high harmonics with coincidence electron ion 3D momentum imaging.

We generate femtosecond EUV pulses at ~ 42 eV photon energy by upconverting intense ($> 10^{14}$ Wcm $^{-2}$) 25 fs laser pulses in an argon-filled waveguide. We use an ultrashort EUV pulse to photoionize simple diatomic molecules: D $_2$, N $_2$ and CO. We observe the population of highly excited states of the singly charged molecule. To our knowledge there have been no reports of time resolved measurement of dynamics on these states, although numerous synchrotron studies have observed the products. We probe the states near the molecular double-ionization threshold on femtosecond time scales by employing a moderate intensity infrared field as a probe. The infrared leads to dication formation by multiphoton ionization. The kinetic energy release of the dissociative dication states evolves as a function of time delay (internuclear distance), allowing us to map excited state dynamics.

Here we present our results for the case of the N $_2$ molecule. The EUV pulse photoionizes the molecule, leaving a significant population in highly-excited states between 25 and 45 eV [1]. The dynamics of these states unfolds along different channels, which we can identify by observing correlations between electron and recoil energy in our coincidence 3D momentum imaging setup.

We identify at least three dissociation channels. The time delayed IR pulse multiphoton-ionizes the dynamically evolving N $_2^+$ system, and the kinetic energy release (KER) is obtained in the N $^+$ - N $^+$ channel as a function of time delay between EUV and IR pulses (Fig 1). Near zero time delay, we obtain a high kinetic energy release of 6-8 eV. This agrees well with previously observed KER for the dication [2]. However, with increasing time delay

we observe a significant shift of the KER to lower values. We believe that the presence of structure in the KER spectra, with a prominent 3 eV component, indicates that we are probing (1) dynamics of non-adiabatic transitions between highly excited states and/or (2) the evolution of a wave packet on a molecular Rydberg state built on a N $_2^{2+}$ core. Based on the present knowledge of potential energy surfaces, the second possibility seems more likely.

In summary, we have demonstrated a technique for time resolving highly excited states using laser based harmonics. The KER spectra as a function of time delay in N $_2$ probes the evolution of wave packets on inner-valence excited states and on molecular Rydberg states.

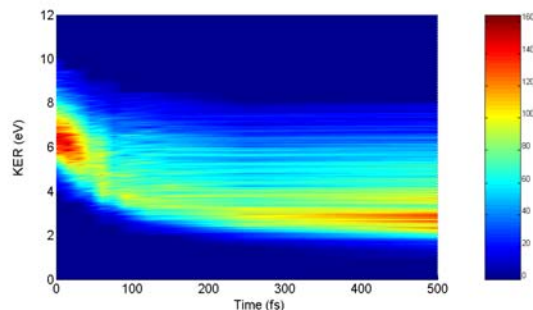


Fig. 1 Highly excited states near double ionization threshold are probed by a time delayed IR pulse which ionizes N $_2^+$. KER for the coincident N $^+$ /N $^+$ channel is obtained as a function of time delay.

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References

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