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fragmentation

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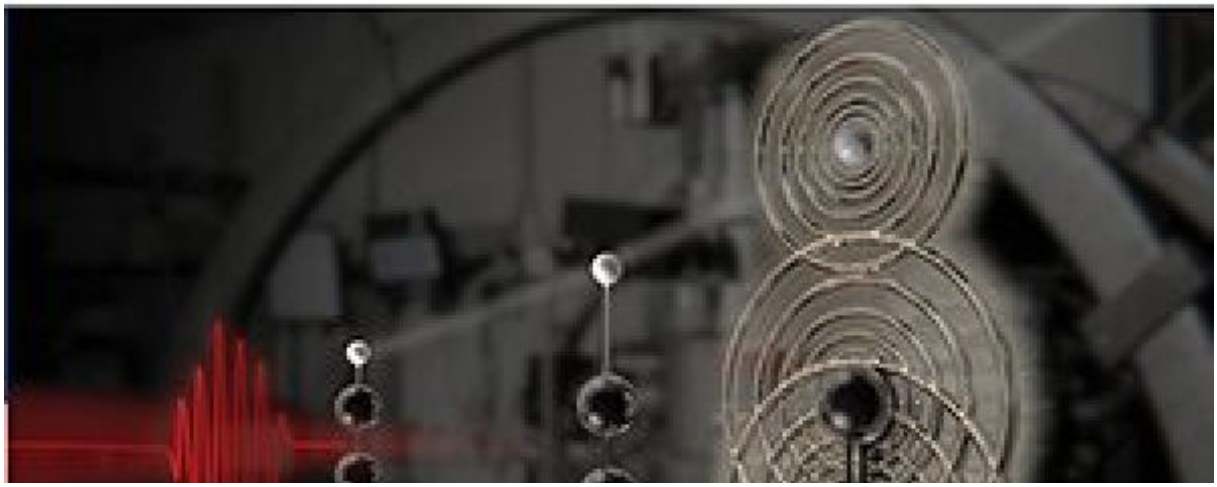

PhD Thesis Defense BENJAMIN WOLTER 'Electron Re-Collision Dynamics in Strong Mid-IR Fields for Diffraction Imaging of Molecular Structure and Its Fragmentation'

BENJAMIN WOLTER

February 26, 2018

Monday, February 26, 2018, 11:00. ICFO Auditorium

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One of the grand challenges of modern science is to image chemical reactions and biological functions while they are taking place. To realize these molecular movie, experimental

techniques need to resolve the relevant molecular motions on their natural dimensions, namely sub-atomic (? 10-10 m) spatial dimensions and in few- to hundreds of femtoseconds (10-15 s) in duration.

A developing molecular imaging technique is based on laser-induced electron diffraction (LIED). Here, an intense electric field is used to liberate an electron from the target molecule before being accelerated and driven back to its parent ion by the same laser field. Upon return, the electron wave packet re-scatters off the target nuclei and an imprint of the molecular structure is encoded in the resultant diffraction pattern. LIED follows similar principles to those used in conventional electron diffraction techniques but occurs in the presence of a strong laser field. Due to the single optical cycle nature of the strong-field induced re-collision process, LIED inherits an intrinsic temporal resolution that fundamentally lies in the sub-femtosecond temporal regime.

In this thesis, a novel experimental approach is presented that allows us to use the LIED technique for dynamic imaging of polyatomic molecular systems, a feat that had never before been realized. Our method involves combining an intense mid-IR field with 3D coincidence momentum detection. On the one hand, the mid-IR laser allows the creation of high energy re-collision electrons, and ionization within the quasi-static regime, hence permitting semi-classical treatment of the process. On the other hand, coincidence detection imaging of the entire momentum space allows for a selective assignment of the molecular reaction channel with sufficient signal-to-noise. Both points have been the main drawbacks to use LIED as a readily available and resilient method that can be utilized for structurally imaging of ultrafast processes in gas phase molecular systems.

The experiments conducted throughout this thesis are divided in two main parts:

First, the interaction of intense mid-IR waveforms with atomic and molecular targets is

investigated and tested against established theories. Here, we measured the photo electron spectrum for very low energies with high resolution and for the first time in full 3D. Thereby, we observed various orders of the recently found ionization surprise being electrons bunching for specifically low energies when crossing the ionization threshold. They predominantly exist when tunnel-ionized at longer wavelength (??1 m). Next, mid-IR induce double ionization was investigated for xenon atoms as an example of a many-electro system. By probing the transition between different ionization regimes as a function o intensity, we found divergent behavior as compared to 800 nm driving fields. These initia experiments serve as a foundation and confirmation that the main parts of strong-fiel interactions with mid-IR lasers can be accurately modeled with classical simulations. Mor importantly, this re-insures the interpretation of LIED as re-scattering electrons on classica trajectories which sets the scene for our work on molecular imaging with LIED

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