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Research Field: Atomic Molecular and Optical Physics

Expected Semester of Graduation: Spring 2018

Molecules Interacting with Intense Laser Pulses

Abstract:

Ultrashort high intensity laser pulses have allowed for the observation of ultrafast dynamics in atoms and molecules. Due to the complexity, these multielectron systems interacting with ultrashort intense laser fields are often theoretically studied using the single active electron approximation (SAE). We present here results of simulations within Time Dependent Density Functional Theory which addresses the multielectron nature of the studied systems. Results for ionization of several molecules, High Harmonic Generation (HHG) properties, dynamic localization, ionization and high order harmonics ellipticity are discussed. We show how the resonance effects can modify properties of these processes.

Prospectus:

High intensity ultrashort laser pulses allow for the observation of ultrafast dynamics in atoms, molecules, and nanostructures. These nonoperturbative field driven phenomena include processes such as High harmonic generation (HHG), Ionization, Electron localization, and Above threshold Dissociation.

High harmonic generation is a highly nonlinear process by which higher order odd harmonics at the frequency of the laser pulse are generated. Such a process is most typical at laser intensities of the order of 10^{14} W/cm^2. Typically, the intense pulse ionizes an electron wave packet into the continuum. The electron wave packet then undergoes a recombination due to the change in direction of the laser field polarization and recombination is accompanied by emission of higher order harmonics. Within the context of HHG, dynamic electron localization gives a picture into the contributions of the valence electrons of multielectron systems and offer new regime for electron dynamics.

Typically, the single active electron approximation is used to study complex multielectron systems interacting with an intense laser field. However, Time dependent density functional theory (TDDFT) is another theoretical method which allows for the explicit treatment of all or only valence electrons in molecule or atom interacting with the laser field. The systems that have been the primary focus of this project include, but are not limited to, the Hydrogen atom, H_2^+ ion, N_2^+ ion, and CO_2.

The mentioned atomic/molecular systems have been studied in the presence of a high intensity ultrashort pulse laser. Under the influence of such fields, high harmonic generation has

been observed in each of the systems. To further study HHG, influence of electron localization and ionization within the systems are analyzed. Further calculations have shown that resonant coupling between states can be observed by tuning laser frequencies to the resonant frequency of the states. In the case, HHG spectra have shown additional Mollow sidebands at intensities comparable to the main harmonics. This resonant coupling is also visible in the context electron localization, and the valence contributions to these effects can be determined. In addition, ellipticity of higher order harmonics generated by the interaction of CO_2 with an intense pulse is calculated and the partial orbital contributions are studied.

Bibliography:

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Timeline:

Spring 2017:

- Began attending group meetings and meeting with Agnieszka to discuss possibilities of projects as well as research interests.
- Began to learn theoretical methods used in our calculations

May 2017:

- Learned how to utilize JILA clusters for calculations.

- Began running first sample calculations on TDDFT software Octopus, these included tutorials located in the software website.
- Began first full TDDFT calculation of the hydrogen atom.
    - Included processing the data for High harmonic generation.
    - Calculating High Harmonic spectrum

Summer 2017:

- Began TDDFT calculations for H_2^+.
    - Included processing and visualization of High harmonic and electron Localization Data.
- Poster Presentation at Trends in Ultrafast Laser Science workshop in Boulder.

Fall 2017:

- Obtained general allocation for SUMMIT supercomputing cluster to perform TDDFT calculations.
- Began TDDFT calculations for N_2^+.
    - Included processing and visualization of High harmonic, electron Localization, and Ionization Data.
    - Multicolor laser calculations performed.
- Poster Presentation at JILA poster fest Boulder.
- Poster Presentation at APS Four Corners Meeting Ft. Collins.
- Began TDDFT calculations for CO_2.
    - Obtained ground state convergence for DFT calculation.
    - Determine most efficient way to partition the grid for TDDFT calculation, still in progress.

Spring 2018:

- Finalize calculations and result analysis for N_2^+.
    - This includes all High harmonic, two-color, as well as ionization calculations.
- Finalize calculations and result analysis for CO_2.
    - Process ellipticity data.
- Begin Writing Thesis.
    - Introduction Chapters completed no later than early February.
    - Begin Results Chapters and finish by beginning March.
    - Begin conclusions Chapters and finish by mid/late march, this includes comparison of results with experiments and other theories.
    - Have a draft of Thesis latest by end of March.
    - Finalize thesis, including edits by advisor, and peers by end of March/early April.
- 04/2018 Defend.