Project Title:

Atoms and molecules driven by relativistic laser fields

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Description of the project

Background and purpose

Recent technical developments in the area of laser physics have made it possible to produce short laser pulses with unprecedented qualities: (i) High peak intensity. Well-controlled laser pulses can be produced that have a peak intensity of $I=10^{16}$ W/cm^2 (corresponding to an electric field strength of 3×10^{11} V/m) or above. The electronic density in an atom or molecule subjected to such a laser pulse becomes strongly distorted, leading to multi-electron rearrangement and/or ionization. (ii) Short pulse duration. Laser pulses as short as 1 fs ($=10^{-15}$ s) have been demonstrated. This time scale approaches that of the motion of electrons in excited states in atoms and molecules, suggesting that electrons can be probed, or even controlled, in real time.

The above described technical advances suggest that real-time control of both electronic and nuclear motion may be feasible using well-defined laser pulses. However, an atom or molecule is a complicated object, and it is often not straightforward to interpret experimental measurements. The understanding of the correlated motion of electrons driven by intense laser fields requires the development of new methods of simulation.

This contribution summarizes two projects carried out using the RICC facility.

The first is "Creation of core-hole atoms by recolliding electrons". Here, we demonstrate that by irradiating single carbon atoms by an intense, few-cycle laser pulse, a valence electron can be ejected by field ionization and accelerated in the laser field to come back close to the atomic core with high kinetic energy. When the intensity of the laser pulse is high, the kinetic energy of the returning electron can be large enough to knock out a still bound innershell (core) electron, creating a highly charged carbon ion in the final state. In this way, the creation of core-hole ions can be achieved by near-infrared laser pulses instead of x-ray light sources.

The second project is "Control of electron localization in a laser-driven polyatomic molecule". We demonstrate theoretically that the electron density in the triatomic molecular ion H_3^{2+} can be controlled by applying a sub-cycle-shaped circularly polarized few-cycle laser pulse. We suggest that in a first step, the stable molecular ion H_3^+ is ionized to H_3^{2+}. Then, the remaining electron in the dissociating, unstable molecular ion H_3^{2+}, is driven by a few-cycle laser pulse so as to control the final location of the electron in the dissociated limit: Which one of the reactions $H_3^{2+}\rightarrow H+H^++H^+$, $H_3^{2+}\rightarrow H^++H^++H$, and $H_3^{2+}\rightarrow H^++H+H^+$ that occurs can be selected by tuning the parameters of the laser pulse.

Method of calculation

In the project "Creation of core-hole atoms by recolliding electrons", the classical trajectory Monte Carlo method is used. In this method, the classical equations of motion for the system under study are solved many times (in our case, up to 10^7 times) for slightly different initial conditions. Each trajectory can be calculated independently of the others, which makes this method suitable for implementation on a cluster such as the RICC. Experimentally measurable quantities such as photoelectron energy spectra, relative yields of different ionization channels, and so on, are calculated by counting how many trajectories that led to a particular outcome. There is a difficulty in constructing a suitable classical Hamiltonian providing a stable ground state which does not autoionize. We have solved this issue by adopting the "Fermionic molecular dynamic" model [C. L. Kirschbaum and L. Wilets, Phys. Rev. A **21**, 834 (1980)].

In the project "Control of electron localization in a laser-driven polyatomic molecule", we solve the time-dependent Schrödinger equation on a three-dimensional grid, using a finite-difference approximation for the kinetic energy operator. We have made use of the MATLAB software, recently installed on RICC, which includes many useful libraries for numerical computation.

Results and conclusions

In the first project, "Creation of core-hole atoms by recolliding electrons", we could show that it is indeed possible to create core-hole atoms with near-infrared laser fields. The recolliding valence electron acts as a transmitter of energy in that it absorbs a lot of energy from the laser field, which is subsequently transferred to the bound core electron. We also found interesting trajectories where the recolliding electron is recaptured into a bound state after the recollision.

In the second project, "Control of electron localization in a laser-driven polyatomic molecule", we demonstrated for the first time that the location of an electron in a triatomic molecule can be controlled by a few-cycle laser pulse. This demonstration paves the way towards the ultimate goal of controlling the positions of one (or several) electrons in a general molecule, implying that chemical bonds can be created and destroyed at will by laser light.

Future prospects

The current RICC project "Atoms and molecules driven by relativistic laser fields" is considered to be finished. We will not ask for extension of this project in FY2015, but instead apply for a new, different one.

During FY2014, no jobs were actually executed on the RICC system, mostly due to lack of time because of a relocation of the project leader (E. Loetstedt) to a new institution (The University of Tokyo). The calculations leading to the results described in this report were performed at RICC during FY2013, but were written up and published during FY2014.

Fiscal Year 2014 List of Publications Resulting from the Use of RICC

[Publications]

Erik Lötstedt and Katsumi Midorikawa, *Ejection of innershell electrons induced by recollision in a laser-driven carbon atom*, Physical Review A **90**, 043415 (October 2014).

Erik Lötstedt and Katsumi Midorikawa, *Carrier-envelope phase control of electron motion in laser-driven* H_3^{2+}, Journal of Physics B: Atomic, Molecular and Optical Physics **47,** 204018 (October 2014) . [No copy provided due to copyright embargo period]

Erik Lötstedt and Katsumi Midorikawa, *Nuclear Reaction Induced by Carrier-Envelope-Phase Controlled Proton Recollision in a Laser-Driven Molecule*, Physical Review Letters **112**, 093001 (March 2014). [This article was published in FY2013, and described in the RICC report for FY2013, but the volume and page numbers were not available at the time of submission of the FY2013 report].

[Oral presentation at an international symposium]

Erik Lötstedt, *Nuclear reaction induced by proton recollision in a laser-driven molecule*, International Conference on High Energy Density Sciences 2014 (HEDS 14), April 22–25, 2013, Yokohama, Japan.

Erik Lötstedt, *Molecules and clusters in intense laser fields*, International Symposium on Ultrafast Intense Laser Science 13 (ISUILS 13), October 5–10, 2014, Jodhpur, India.

[Others]

Erik Lötstedt, Poster presentation: *Classical trajectory modeling of atoms and molecules in intense laser fields*, International Symposium on Ultrafast Intense Laser Science 13 (ISUILS 13), October 5–10, 2014, Jodhpur, India.

Erik Lötstedt, Poster presentation: *Laser-induced electron localization in* H_3^{2+}, The 5th Shanghai-Tokyo Advanced Research Symposium on Ultrafast Intense Laser Science (STAR 5), May 21–24, 2014, Miyazaki, Japan.