# Ab initio simulations of the interaction of helium with ultrashort laser pulses

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The availability of ultrashort XUV pulses of about a hundred attoseconds duration allows to probe the ultrafast electronic processes occurring in atoms, molecules and solids on their natural timescale. However, the theoretical and computational treatment of the interaction of atoms with strong electromagnetic fields is still a challenging problem. Consequently, helium is virtually the only multi-electron atom which one can handle exactly by numerical calculation. Understanding the dynamics in this simple two-electron system is of crucial importance for the understanding of more complex atoms and even simple molecules.

We have developed an ab initio simulation method for solving the time-dependent Schrödinger equation for helium in its full dimensionality (six spatial and one temporal degree of freedom). The wave function is represented in a time-dependent close-coupling (TDCC) scheme, where the angular variables are represented in the basis of coupled spherical harmonics. Time integration is performed using the short iterative Lanczos method, where the time evolution operator is approximated in a Krylov subspace, giving a high-order, explicitly unitary and well-parallelizable time propagation algorithm. The spatial discretization is performed using a finite element discrete variable representation (FEDVR) basis, which provides high accuracy, but produces sparse matrices and thus lends itself to effective parallelization with linear scaling across large numbers of processors. Even with optimal algorithms, the solution of this six dimensional problem leads to matrices $2\text{x}10^8$ in size. Consequently, these calculations could not have been performed without access to large scale computing resources available at the TeraGrid and DoE laboratories.

We will present our recent results on the interaction of attosecond XUV pulses with a helium atom, in particular focusing on the effects of electron-electron interaction and correlation. One prototypical process is the two-photon double ionization (TPDI) of helium, which strongly depends on the photon energy. At low energies, TDPI only occurs when the electrons share the energy absorbed from the photons. As this can only happen when the electrons are close together, the photons are absorbed quasi-simultaneously. Therefore this regime is called *nonsequential* TPDI. Because of the energy exchange, the electrons are also strongly correlated, i.e., the full wave function can not be separated into independent single-particle wave functions [1]. For higher photon energy, TPDI is dominantly *sequential*, i.e., proceeds in two separate steps – first ionization of the neutral He atom and then ionization of the remaining $He^+$ ion. In long pulses, this leads to uncorrelated emission of the two electrons. However, by employing ultrashort XUV pulses, the TPDI process is confined to within the duration of the pulse. The influence of the electron-electron interaction can then be tuned by varying the pulse duration. This can be used to probe and induce electronic correlation [2]. In pulses of a few femtoseconds duration, as produced by X-ray free-electron lasers, we have discovered a novel interference between nonsequential and sequential contributions to the total double ionization [3].

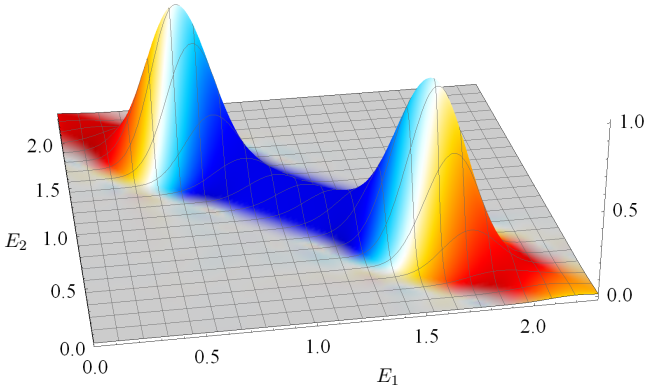


FIG. 1: Combined double ionization probability $P^{DI}(E_1, E_2)$ and forward-backward asymmetry $\mathcal{A}(E_1, E_2)$ of the two electrons after TPDI by an XUV pulse at $\hbar\omega = 70\,\text{eV}$ with a duration of 450 as. The $z$-axis gives $P^{DI}(E_1, E_2)$ (in arbitrary units), while the color encodes the asymmetry. The asymmetry quantifies angular correlation by encoding the tendency for both electrons to be emitted in the same direction, with cyan to blue signifying negative values (ejection in opposite directions) and yellow to red signifying positive values (ejection in the same direction). Vanishing $\mathcal{A}$ corresponds to white.

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[2] J. Feist, S. Nagele, R. Pazourek, E. Persson, B. I. Schneider, L. A. Collins, and J. Burgdörfer, arXiv.org:0812.0373 (2008).
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