A Quantum-compute Algorithm for the Exact Laser-driven Electron Dynamics in Molecules

Fabian Langkabel[1,2]
Annika Bande[1]

[1]*Helmholtz-Zentrum Berlin, Hahn-Meitner-Platz 1, 14109 Berlin, Germany*

[2]*Freie Universität Berlin, Arnimallee 22, 14195 Berlin, Germany*

fabian.langkabel@helmholtz-berlin.de

With the advent of new experimental techniques, the theoretical description of ultrafast electron dynamics in molecular systems plays an increasingly important role in understanding photo-induced processes such as energy transfer [1] or electron solvation. Unlike for solving the time-independent Schrödinger equation in "regular" quantum chemistry [2], very little thought has been given to the use of quantum computers in the field of electron dynamics.

In this work, we have investigated the capability of known quantum computing algorithms for fault-tolerant quantum computing to simulate the laser-driven electron dynamics in small molecules such as lithium hydride and performed these simulations on a quantum computer simulator. Results were compared with the time-dependent full configuration interaction method (TD-FCI). In addition to the actual wave packet propagation, which was successfully reproduced using the Jordan-Wigner transformation and the Trotter product formula, the time-dependent dipole moment was also satisfactorily determined as an example of time-dependent expectation values using the Hadamard test [3]. In order to be able to include non-Hermitian operators in the dynamics, a similar approach to the quantum imaginary time evolution (QITE) algorithm [4] was employed to translate the propagator used into quantum gates. Thus, ionization in a hydrogen molecule was also reproduced using a complex absorption potential. All these calculations were performed with the group's own program Jellyfish. While TD-FCI scales exponentially, all quantum computer algorithms used scale polynomially, which may lead to an enormous progress in the understanding of electron dynamics of increasingly large molecular systems in the future.

References

[1] F. Langkabel et al., J. Chem. Phys. 2021, 154, 045111
[2] S. McArdle, et al., Rev. Mod. Phys. 2020, 92, 015003
[3] D. Aharonov, et al., Algorithmica 2008, 55, 395-421
[4] M. Motta, et al., Nat. Phys. 2019, 16, 205-210
[5] F. Langkabel et al., Chem. Phys. 2022, 557, 111502

Figures

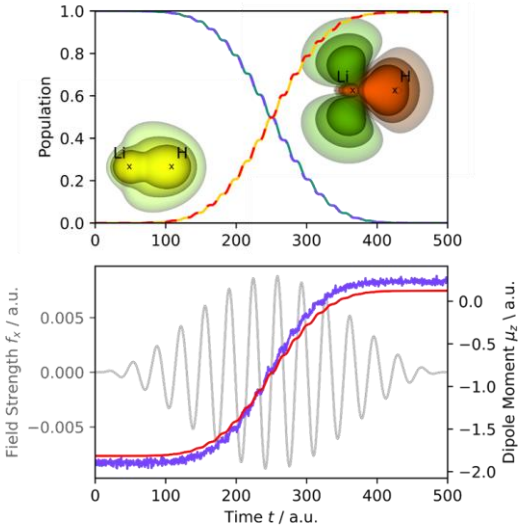

Figure 1: Comparison of population and dipole moment with TD-FCI (solid top and red bottom) and quantum computer dynamics (dashed top and purple bottom) of charge transfer in lithium hydride during a resonant π-pulse excitation (gray bottom), as well as the electron density at the beginning and the difference density [5] at the end.