Analysing ultrafast atomic and molecular dynamics through sequences of attosecond pulses

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A key technical aim in the burgeoning field of attosecond science is to investigate ultra-fast dynamics in atoms and other systems through attosecond-pump attosecond-probe experiments [1]. Pump-probe spectroscopy has been critical to understanding the physics and chemistry of excited states. However, the associated dynamics of these excited states occurs on a (sub-)femtosecond timescale, and imaging these dynamics thus requires the use of attosecond pulses.

Queen's University Belfast is leading the world in the development of accurate theory to support this upcoming field of science. Attosecond pulses have a large energy bandwidth, and therefore many excited states are involved in the dynamics simultaneously. Steady-state initial states no longer suffice. Instead, we must consider the interaction of light with non-equilibrium ensembles from the outset. Our state-of-the-art R-matrix for time-dependence (RMT) codes [2] already have the capability to describe the atomic and molecular structure underlying the dynamics that occur within a single ultra-short pulse. However, a single calculation will not always suffice for studying the dynamics induced by sequences of ultra-short pulses.

Sequences of ultra-short pulses can be used to investigate the dynamics in residual atoms immediately after ionization. However, the theoretical study of this dynamics requires a new investigation for a new (now ionized) system. We therefore need to connect multiple RMT calculations for different systems with each other. This can be achieved through extending our analysis techniques with a density matrix formalism.

The density matrix keeps track of correlations between different residual-ion states at the end of a calculation. We can then carry out calculations to investigate the subsequent dynamics for each of these residual-ion states separately, and use the density matrix to connect them together. The density-matrix approach can also be used to make calculations more efficient, as they allow a systematic study of the impact of changing polarization directions without a need to perform an extensive calculation for every single polarization direction.

Funding for the project has been applied for in collaboration with the Rutherford-Appleton Laboratory (RAL) in Oxfordshire, but funding has not yet been confirmed. As part of the project, it is expected that the student will spend a significant part of the Ph.D. studies at RAL, with timings to be arranged in mutual agreement.

The project will give the student expertise in modern computing practice using state-of-the-art computing facilities worldwide. The student will gain understanding of leading analysis techniques in atomic physics. The student will also develop other highly valued transferrable skills in research and associated presentation of research, and have opportunity to engage with leading researchers around the world.

[1] UK XFEL Science Case (STFC, Ed. J. Marangos, 2020)
[2] A.C. Brown et al, Computer Physics Communications 250, 107062 (2020).