Centre for Theoretical Atomic, Molecular, and Optical Physics

Ph.D. Project 2021-2024

Attoclock: timing photoionization on the sub-femtosecond timescale

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State of the art and motivations

Over the past two decades, great strides have been made in the investigation of ultra-fast atomic and molecular dynamics (see eg. [1]). The typical timescale for electron motion around a nucleus is on the order of 10^{-16} s (or 100 attoseconds), and light pulses shorter than this have been generated experimentally [2]. This allows experiment to study fundamental processes in physics with new levels of detail. It allows, for example, electron motion to be tracked inside an atom. A recent example of the new physics that can be studied is provided by [3]. In this study, it was shown that the direction in which light travels across a molecule generates asymmetries in the ejected photoelectron spectra.

To fully comprehend the results generated by these exciting new experiments, theoretical interpretation is crucial. A key experiment in attosecond science has been the measurement of photoionization time delays [4]. Advanced theoretical methods, such as the R-matrix with Time-dependence (RMT) approach developed here at Queen's [5, 6], were needed to explain that the time delays observed experimentally were an interplay between the main photoionization process, and the resonant excitation of many electrons simultaneously to a highly excited state [7].

The RMT code developed at Queen's over the last 14 years is widely regarded as one of the most advanced methods for the description of multi-electron systems in arbitrary light fields. Substantial external funding has been attracted over the last decade to develop this code, expanding capability to describe molecular dynamics [8], arbitrarily polarised light [6], and atomic systems in which relativistic effects need to be taken into account [9]. These developments put QUB in a unique position to answer state-of-the-art questions on atomic dynamics in ultra-short light fields.

Objectives & Methodology

In this project, we will be using the RMT codes to study photoionization time delays in a range of systems, including systems where relativistic effects need to be taken into account. The codes have been designed to make efficient use of the most advanced computational facilities available in the UK and world-wide. Initial studies will normally involve linearly polarised light, and these typically use around 500 cores working together on a single problem. Subsequent calculations may involve arbitrarily polarised light, which require significantly larger computational resources, beyond 10,000 cores. The codes are now well established, so that there is no requirement that you should be involved with code development. However, the project offers significant opportunities to learn about massively parallel computing.

In the first instance, we will be using the RMT codes to investigate attosecond time delays in heavier atomic systems, such as Kr and Xe. In these systems, relativistic effects cause a small splitting of the lowest ionization threshold. We will be investigating how this splitting affects the observed photoelectron spectra, and how this leads to changes in time delays with photoelectron energy. Once we have obtained a good understanding for the methods and techniques, we will be investigating open-shell systems to investigate circular dichroism, the difference in atomic response to left- and right-circularly polarised light, including effects on time delays.

Collaborations

The RMT codes have been developed in collaboration with the Open University, where researchers are interested in the application of the codes to molecular systems. Ongoing development of the codes is also likely to involve the Charles University in Prague. The RMT codes are under further development for the application to double ionization processes in a collaboration with UCL. The codes are increasingly used in support of experimental research with collaborations with the Max-Planck-Institute in Heidelberg, as well as research groups in Italy and China.

Required skills

Typical skills required are those acquired by a first-class Masters-level graduate in Mathematics and/or Physics. A good core understanding of Quantum Mechanics is important. Good computational skills are beneficial, but are not essential.

Further information

For further information, please do not hesitate to contact Prof. Hugo van der Hart. In the current circumstances, it is best for initial contact to be through email, h.vanderhart@qub.ac.uk.

References

[1] F. Krausz and M.Yu. Ivanov, Rev. Mod. Phys. **81**, 163 (2009).

[2] J. Li *et al*, Nature Comm. **8**, 186 (2017).

[3] S. Grundmann *et al*, Science **370**, 339 (2020).

[4] M. Schultze *et al*, Science **328**, 1658 (2010).

[5] L.R. Moore, M.A. Lysaght, J.S. Parker, H.W. van der Hart and K.T. Taylor, Phys. Rev. A **84**, 061404 (2011).

[6] D.D.A. Clarke, G.S.J. Armstrong, A.C. Brown and H.W. van der Hart, Phys. Rev. A **98**, 053442 (2018).

[7] M. Isinger *et al*, Science **358**, 893 (2017).

[8] J. Benda, J.D. Gorfinkiel, Z. Mašin, G.S.J. Armstrong, A.C. Brown, D.D.A. Clarke, H.W. van der Hart and J. Wragg, Phys. Rev. A **102**, 052826 (2020).

[9] J. Wragg, D.D.A. Clarke, G.S.J. Armstrong, A.C. Brown, C.P. Ballance and H.W. van der Hart, Phys. Rev. Lett. **123**, 163001 (2019).