Centre for Theoretical Atomic, Molecular, and Optical Physics

Ph.D. Project 2022-2025

Manipulating high-harmonic generation through the spin-orbit interaction

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State of the art and motivations

In ultrafast physics we study the dynamics of electrons in atoms and molecules which occur on the attosecond (1×10^{-18} s) timescale. One of the key experimental tools in the field– High-harmonic generation (HHG)– underpins the generation of ultra-short pulses[1]. Manipulating the light produced by HHG offers a path to greater control of electron dynamics. In this project, we will use the time-dependent R-matrix approach to investigate how HHG is affected by the spin-orbit interaction and, conversely, how we can use the spin-orbit interaction to tailor ultrashort light pulses.

HHG can be understood as a three-step process [2], in which a strong, long-wavelength laser pulse (a) releases an electron from the atom, and then (b) drives the electron first away from and then back towards the atom. Then (c) upon return, the electron reattaches to the atom by emission of a high-energy photon. This process is highly non-perturbative, and allows the generation of high-order (well above order 100) harmonics of the original driving light. By filtering out all but a small number of harmonics from the full spectrum, this process can be used to generate attosecond pulses.

R-matrix with time-dependence (RMT) is a world-leading code for the description of atoms in strong fields, developed over the last 15 years at Queen's, which builds upon over 50 years of local expertise in theoretical atomic physics. The codes have been found to be surprisingly good at describing high-harmonic generation, including at very challenging mid-IR wavelengths [3]. In the past 5 years, we completed substantial further development on RMT [4], allowing the description of atoms in arbitrarily polarised light fields and the inclusion of spin-orbit interactions. These developments have greatly broadened the range of processes we can study, including, for example, the production of spin-polarised electrons [5].

Although HHG has been studied for 30 years, relatively little attention has been given to understanding the effects of the spin-orbit interaction on the process. However, electron dynamics is strongly affected by the spin-orbit interaction [6], as it couples atomic states with different magnetic quantum numbers. In this project, we will investigate how temporal variations in electron dynamics, caused by the spin-orbit interaction, will lead to temporal variations in the efficiency of HHG. We will then study whether it is possible to use spin-orbit interactions to tailor the generated harmonic radiation.

Objectives & Methodology

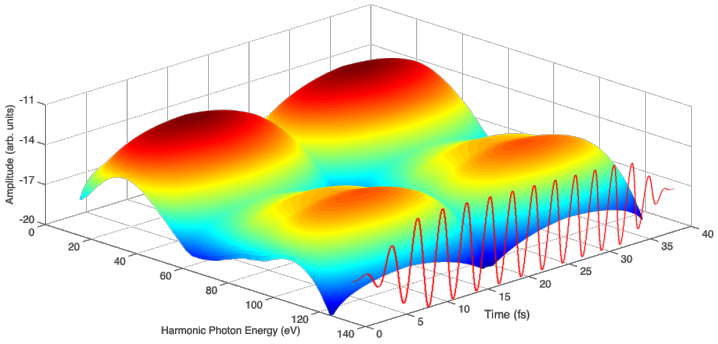

Figure 1: The imagined time-dependent HHG yield from Ar^+ in an 800 nm field (red line)– based on a composition of spectra from $M_L = 0$ and $M_L = 1$ calculations– showing the anticipated time-dependent (fs-scale) fluctuations in yield due to the spin-orbit interaction. The Ar^+ ion evolves via the spin-orbit interaction between two fine-structure states in 12 fs.

In this project, we will be investigating the influence of the spin-orbit interaction on HHG using RMT. The timescale of spin-orbit interactions varies from the light systems to the heavier systems. As a consequence, the spin-orbit interaction may lead to significantly different effects for different atoms. We will first investigate harmonic generation in relatively light systems where previous studies have shown a significant difference in harmonic response between population in different magnetic sublevels [7, 8]. We will elucidate how harmonic generation is affected when the spin-orbit interaction, coupling these sublevels, is taken into account. Once this initial study is completed, we will look at HHG in lighter and heavier systems to investigate the influence of spin-orbit dynamics on a range of timescales.

Collaborations

RMT is developed in collaboration with molecular physics groups at the Open University, University College London and the Charles University in Prague, and with software engineers at the national STFC Daresbury Laboratory. We also collaborate with experimental groups across the world, including at the Max-Planck-Institute in Heidelberg, the Polytechnic University of Milan, the Weizmann Institute and the East China Normal University.

Required skills

The key requirement is a good understanding of quantum-mechanics. RMT is a large-scale code, deployed on the largest high-performance computing research facilities in the world. However, no previous experience in high-performance computing or programming is required.

Further information

For any further questions and/or information, please contact Hugo van der Hart at h.vanderhart@qub.ac.uk.

References

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