Centre for Theoretical Atomic, Molecular and Optical Physics
CTAMOP
Queen's University Belfast

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Attosecond Two-Dimensional Spectroscopy

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State of the art and motivations

Taking pictures of fast moving objects requires short pulses of light: instead of opening and closing a camera shutter at speeds beyond their mechanical capability, photographers in the 1960s hit on the idea of leaving their cameras 'open' in a dark room. They would then switch the lights on and off at the moment that, say, a bullet passed in front of the camera, allowing them to capture a still image of a fast moving object.

Similar schemes are used every day in physics labs: with recent improvements in laser technology, experimentalists are routinely able to generate laser pulses which are mere attoseconds (1 as $= 1 \times 10^{-18}$s) in duration. This has facilitated several incredible experiments which have been able to 'photograph' (or even 'video') electron dynamics which evolve on the attosecond scale [1]. Many such experiments are actually based on a very old idea, a 'pump-probe' scheme, where one laser pulse is used to start a process, and a second pulse, arriving after some time delay, images the system. By varying the time delay, one can elucidate the dynamics.

More recently, a scheme proposing 3 pulses, with two independently varied time-delays has been used to elucidate more complex dynamics [2]. While this 'two-dimensional spectroscopy' (2DS) scheme has been used extensively to address 'slow' (pico-femtosecond) electron-dynamics in molecules, it is hoped that attosecond duration XUV and X-ray pulses will enable 2DS to measure attosecond scale electron-dynamics [3]. Extending 2DS into the attosecond regime will enable the precision measurement of the exotic coupled dynamics of multiple electrons, but it faces several substantial technological challenges.

It is not, however, beyond the capabilities of current theory to address attosecond scale 2DS in atomic systems, and take steps toward leading future experiments. Indeed, we have recently demonstrated the capabilities of our world leading theory in an experiment involving so-called 'Attosecond Transient Absorption Spectroscopy', an experimental stepping stone towards attosecond scale 2DS [4].

Objectives & Methodology

The purpose of this project will be to implement a 2DS scheme in the recently developed R-matrix with time-dependence (RMT) codes, and to use these extended techniques to investigate 2DS across a range of systems. While the code is already well placed to provide theoretical support for of ongoing experiments, in this project we will lead the field: investigating new schemes yet to be realised experimentally. The nature of the investigation may also require the use of some of the worlds leading atomic structure codes, such as CIV3 and RMATRX-I, and so the successful candidate will gain broad experience in computational physics. The physics

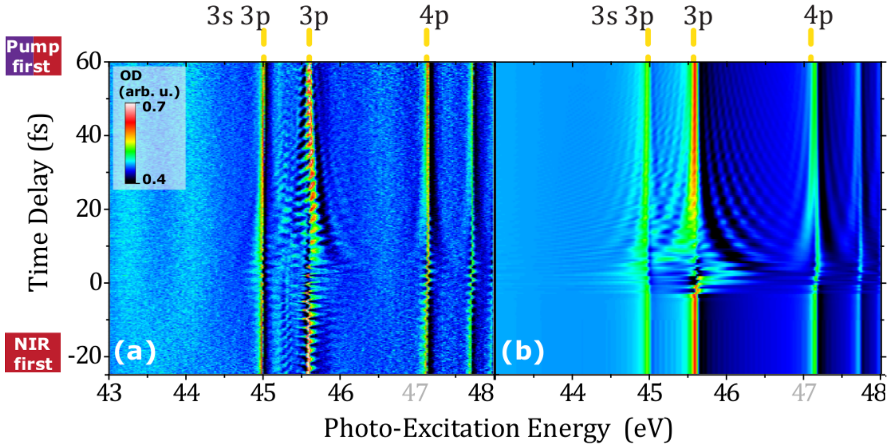

Figure 1: Absorption spectrogram of neon showing the coupled dynamics of core- ($2s2p^6np$) and doubly-excited ($2s^22p^43s3p$) states in neon as measured in experiment (left) and calculated using the R-matrix with time-dependence codes at Queen's (right) (adapted from [4]).

addressed in this project will address advanced measurement techniques, and will certainly make a valuable contribution to the understanding of 2DS, and the field of attosecond science in general.

Required skills

The successful candidate should have a solid grounding in mathematics and physics, with a firm grasp of quantum mechanics. A decent level of programming ability, in particular using Fortran and/or parallel computing techniques, is desirable, but training in these aspects of the project will be provided.

Further information

Queens University Belfast hosts an active group in the study of ultrafast processes using R-matrix theory. This group presently comprises 2 academic staff and 3 Ph.D. students. The group has a good record of Ph.D. student and PDRA success, both during their studies and in their further career.

The group is one of the leading users of massively parallel computing in the UK. Through development of our own software, researchers trained in CTAMOP have gone on to become industry leaders in the development and exploitation of parallel software.

For further information, please contact Dr Andrew Brown: andrew.brown@qub.ac.uk.

References

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[3] I. V. Schweigert and S. Mukamel, Phys. Rev. Lett. **99**, 163001 (2007)

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