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Disentangling spectral phases of interfering autoionizing states from attosecond interferometric measurements

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Synopsis The spectral phase of the autoionising neon continuum has been probed via interferometric pump-probe measurements as well as ab initio, full-electron, time-dependent, theoretical calculations in energy intervals containing several auto-ionising. Theory and experiment are in excellent agreement. Despite the complex energy-dependence of the phase, a very simple model has been used to disentangle the contribution by different resonances. This work extends the applicability of reconstruction methods to multi-resonance regions of autoionising continua - a common scenario in multi-electron systems.

With the advent of attosecond science, the development of interferometric techniques has proved itself a highly useful avenue for the study of electron dynamics on their natural time scale. One such method is the pump-probe method known as reconstruction of attosecond beatings by interference of two-photon transitions (RABITT), which has been successfully used to study phenomena such as spectral phases [1]. Although this method has proven itself able to probe not just atomic [1] system, but also more complex system systems, such as molecules [2], solids [3]; it has so far been restricted to regions of autoionising continua containing one or no resonances. This is due to the difficulty in accurately describing electron correlation of multi-resonance regions, making accurate, full-electron, time-dependent ab initio theory, necessary to guide experiment inaccessible. This dearth of accurate calculations leaves simple models able to describe such regions a highly desired prospect.

In this communication, RABITT is used to probe neon below the second ionisation threshold, scanning the spectral phase both in a single- and a multi-resonance regions. Time-dependent, full-electron, ab-initio calculations were used to provide a theoretical prediction; enabled using an implementation of the XCHEM methodology [4], itself designed to accurately describe correlated processes in many-electron systems. Experimental data is also presented, with which the theoretical calculations show excellent result, providing further support for the XCHEM approach. In the multi-resonance region, the spectral phase has a complex dependence on the energy. Despite this, a very simple model based on a recent extension of the Fano model for two-photon ionisation induced by ultrashort pulses [5] is presented, capable of disentangling the contributions of different autoionising states and accurately predicting the energy-dependence of the spectral phase in the studied multi-resonance region.

This works extends the applicability of interferometric methods to multi-resonance regions of atomic and molecular systems, thus paving the way for reconstruction of electron wave packets coherently generated in systems previously unavailable due to their complexity.

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