SHORT TERM SCIENTIFIC MISSION (STSM) SCIENTIFIC REPORT

This report is submitted for approval by the STSM applicant to the STSM coordinator

Action number: CA18222 – Attosecond Chemistry
STSM title: Probing ultra-fast coherent dynamics in core-excited and Rydberg states with relativistic attosecond transient absorption
STSM start and end date: 03/05/2021 to 18/06/2021
Grantee name: Felipe Zapata Abellán

PURPOSE OF THE STSM:

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(max.200 words)

Attosecond transient absorption spectroscopy (ATAS) is used to study electron coherence and motion in atoms [1-3]. All theoretical studies so far are based on non-relativistic ATAS theory [4]. However, the importance of the spin-orbit coupling was demonstrated already by the first ATAS experiment, which targeted Krypton [2]. The present Short-Term Scientific Mission (STSM) aims to create a strong collaboration between the theoretical groups in Lund and in Madrid around the study of spin-orbit-resolved ATAS experiments triggering the control of ultra-fast coherent dynamics in atoms. Prof. Fernando Martin's group expertise on ATAS, see for example ref. [5-7], together with the RTDCIS method, recently developed in Lund, allow us to explore the relativistic effects on the electron motion at the attosecond time scale.

References

[1] Wirth A et al (2011) Science 334 195–200
[2] Goulielmakis E et al (2010) Nature 466 739–43
[3] Sabbar M et al (2017) Nat. Phys. 13 472–8
[4] Wu M et al (2016) J. Phys. B: At. Mol. Opt. Phys. 49 062003
[5] Petersson C L M et al (2017) Phys. Rev A 96 013403
[6] Cheng Y et al (2016) Phys. Rev. A 94 023403
[7] Ott C et al (2014) Nature 516 374-378

DESCRIPTION OF WORK CARRIED OUT DURING THE STSMS

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(max.500 words)

Benchmark with non-relativistic calculations. Steps:

1.- Selection of the target atom.
2.- Selection of the laser parameters.
3.- Selection of the filter function.
4.- Simulation of an ATAS experiment on neutral Ne with different time delays.
5.- Comparison with Petersson's results.

The required computational time was underestimated. In the second part of the STSM our attention was focused on how to speed the calculations:

1.- I have applied for computational resources to the Swedish National Infrastructure for Computing (SNIC): Project: SNIC 2021/22-503, Theoretical Spin-Orbit Resolved Attosecond Transient Absorption Spectroscopy in Noble Gases. 60.000 CPU hours. Status 28/06/2021: Accepted.

2.- Long-range field-free dipole propagation:
In order to accelerate the simulation, the time-dependent dipole can be split into the short-range contribution and the long-range contribution using the following splitting function:

$$\Phi(t) = \frac{1}{2}\left[1 + \text{erf}\left(\frac{t - t_0}{\sigma\sqrt{2}}\right)\right]$$

The short-range contribution describes the response to the total electric field while the long-range contribution describes the infinite oscillations of the dipole after the interaction with the fields. This fact allows us to reproduce the long-range contribution analytically as follows,

$$\tilde{z}(t) = \sum_{k,j} T_{k,j} \exp\{-i\omega_{k,j} t\}$$

where T_{kj} is the dipole transition matrix and $\omega_{k,j}$ is the energy difference between the states j and k. Then, the Fourier Transform is given by:

$$\text{FT}[\Phi(t)\tilde{z}(t)] = \sum_{k,j} T_{k,j} \frac{e^{-i\omega' t_0}}{2\pi\sigma} \int_{-\infty}^{+\infty} \frac{e^{-s^2/2\sigma^2}}{i\omega'} e^{-i\omega' s} \text{d}s$$

and by using the following expression

$$\int_{-\infty}^{+\infty} e^{(as^2+bs)}\text{d}s = \left(\frac{\pi}{a}\right)^{1/2} e^{b^2/4a} \qquad \text{for} \quad a > 0$$

we finally get

$$\text{FT}[\Phi(t)\tilde{z}(t)] = \sum_{k,j} T_{k,j} \frac{e^{-i\omega' t_0}}{i\sqrt{2\pi}} \frac{e^{-\omega'^2\sigma^2/2}}{\omega'}$$

This expression will allow us to compute the ATAS spectrum.

DESCRIPTION OF THE MAIN RESULTS OBTAINED

The benchmark consisted in the reproduction of an ATAS experiment carried out by Ding et *al* [1] and simulated by Petersson in his PhD thesis with a non-relativistic method [2]. Figure 1 shows the autoionising resonances $2s^2 2p^6$ – $2s2p^6 np$ in neon obtained with our RTDCIS code. Figure 2 shows the temporal profile of the total electric field used in the experiment. The pump creates a coherent superposition of the $2s^2 2p^6$ – $2s2p^6 np$ series. The probe is applied at different time delays in order to explore the dynamics of the system.

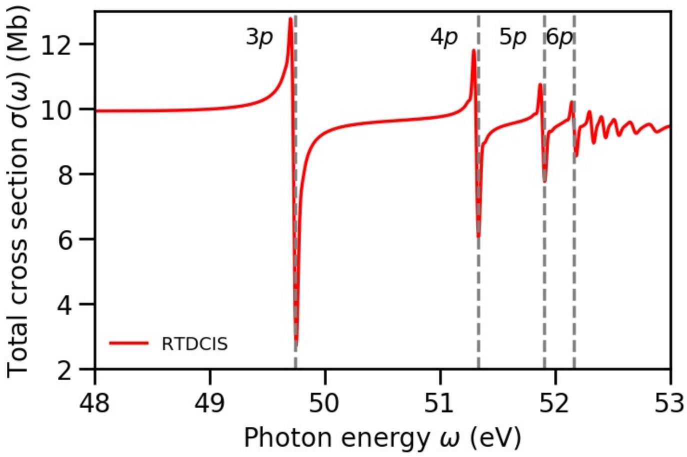

Figure 1. Autoionising $2s^2 2p^6$ – $2s2p^6 np$ series in Ne calculated with RTDCIS.

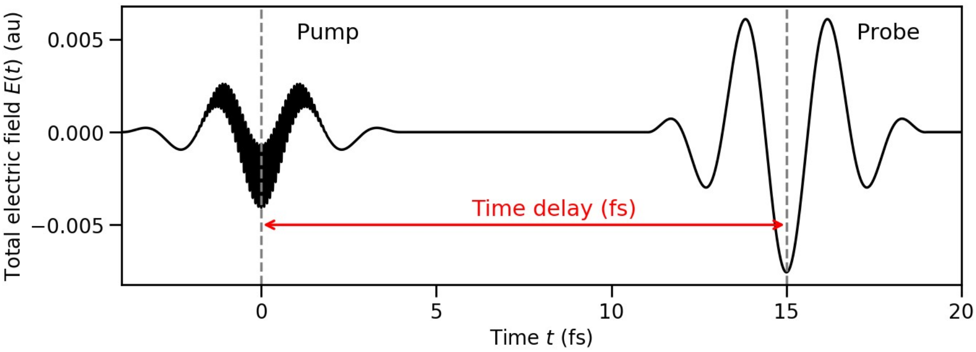

Figure 2. Total electric field used in the ATAS experiment proposed by Ding et *al* [1].

Figure 3 shows the ATAS spectrum in neon simulated for different time delays. The required computational time was 2000 CPU hours. In order to do a full comparison with the results given Petersson in his PhD thesis, we will need to include more time delays and to increase the resolution.

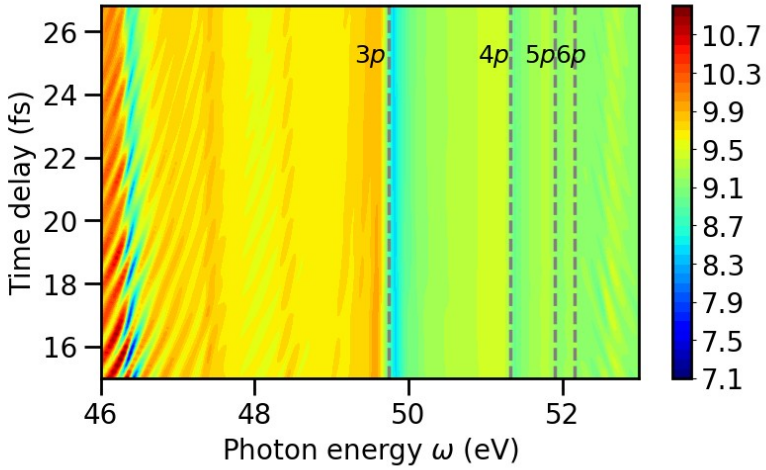

Figure 3. Simulated ATAS spectrum in neon for different time delays obtained with a resolution of 0.2 fs.

As the required computational time is very high, we will use the computational hours provided by the SNIC project "SNIC 2021/22-503, Theoretical Spin-Orbit Resolved Attosecond Transient Absorption Spectroscopy in Noble Gases". In addition, we will accelerate the simulation of the ATAS spectrum by splitting the calculation of the short-range and long-range dipole contributions. Figure 4 shows the splitting of the dipole into the short-range contribution, calculated numerically by solving the RTDCIS equations, and the long-range dipole, obtained analytically.

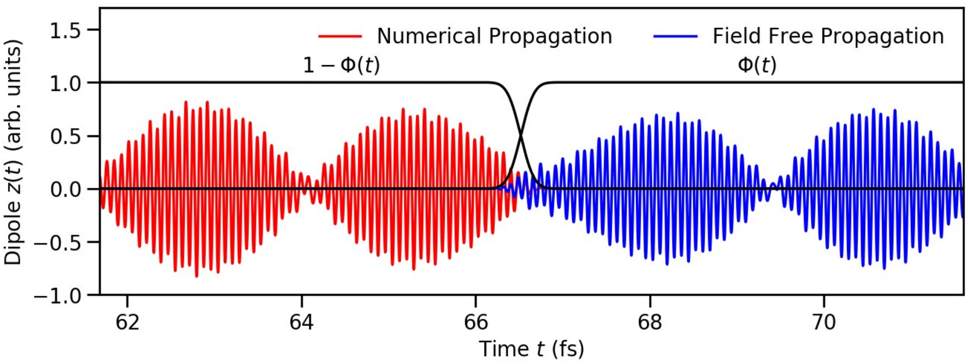

Figure 4. Short-range and long-range dipole contributions.

Reference:

[1] Ding et al. (2016) Opt. Lett. 41, 4 709-712

[2] C. L. M Petersson, *A theoretical study of ultrafast phenomena in complex atoms,* PhD thesis 2019.

FUTURE COLLABORATIONS (if applicable)

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Thanks to the present STSM, a strong collaboration has been established between the theoretical groups in Madrid and Lund around the simulation of ATAS experiments. Future steps will be: (1) full comparison of the ATAS spectrum and (2) simulation of ATAS in argon.