Metz 2019
COPIAMC

International Symposium on Correlation, Polarization and Ionization in Atomic and Molecular Collisions

The 20th International Symposium on Correlation, Polarization and Ionization in Atomic and Molecular Collisions (COPIAMC) is an official satellite meeting of the XXXI ICPEAC that will be held in Deauville, France.

COPIAMC will take place in Metz (France) and will run over two and a half days, from the 1st to the 3rd of August, 2019. It will comprise plenary talks, progress reports and a poster session.

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Scope

The COPIAMC satellite provides the opportunity to attend high-level specialized talks covering a broad range of hot topics in atomic and molecular physics. Among others, the following areas of research will be discussed:

- Many-body interactions and electron-electron correlation effects in single and multiple ionization processes.
- Alignment and polarization effects in excitation and charge transfer processes.
- Correlation and polarization effects in multiphoton ionization of atoms and molecules.
- Correlations and coherence in the interaction of intense and short-pulsed radiation with gas-phase species.
- Correlated motion in photodissociation processes.
- Coherence effects in multielectron excitation.

Conference Venue

The conference will take place on the campus "Saulcy" of the Université de Lorraine, situated on an island in the centre of Metz.
Oral presentations and poster sessions will be held at the Faculté de Droit, Economie et Administration, Ile du Saulcy, 57045 METZ.

Conference Language

The language of the conference is English.

Social Program

Welcome Reception : 18h30-20h30 on Wednesday 31st July, 2019
Conference Banquet: from 19h30 on Friday 2nd August, 2019

Sponsors

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Webpage : https://copiamc.event.univ-lorraine.fr/

Scientific Program

Wednesday 31st July

17:30 - 20:00   Registration

18:30 - 20:30   Welcome reception

Thursday 1st August

8:00-8:45   Registration

8:45-9:00   Opening by Ugo Ancarani

Session 1 - Chair: Andrew Murray

09:00-09:45   Paola Bolognesi
              Size and environmental effects in molecular photofragmentation
09:45-10:15   Xueguang Ren
              Water acts as a catalyst for electron-driven chemical reaction in biochemically
              relevant hydrogen-bonded systems

10:15-10:50 Coffee break

Session 2 - Chair: Lorenzo Avaldi

10:50-11:20   Kathryn Hamilton
              R-Matrix calculations for ultrafast two-colour spectroscopy of noble gas atoms
11:20-11:50   Etienne Plésiat
              Real-time imaging of ultrafast charge dynamics in CF_4 molecule
              from attosecond pump-probe photoelectron spectroscopy
11:50-12:20   Elena Gryzlova
              Photoelectron spin polarization in bi-chromatic-field ionization of atoms

12:30-14:00 Lunch

Session 3 - Chair: Alexander Dorn

14:00-14:45   Don Madison
              Accuracy of theoretical calculations for electron-impact ionization of molecules
14:45-15:15   Masakazu Yamazaki
              Asymptotic behavior of plane- and distorted-wave theories to their high
              energy limits
15:15-15:45   Carlos Granados
              (e,2e) at intermediate energy on methane

15:45-16:30 Coffee break and Poster session

Session 4 - Chair: Emma Sokell

16:30-17:00   Juan Randazzo
              Double photoionization of water
17:00-17:30   Sven Grundmann
              Separating dipole and quadrupole contributions to single-photon
              double ionization
17:30-18:00   Laura Cattaneo
              Unfolding the physics of photoionization dynamics in simple molecules

Friday 2nd August

Session 5 - Chair: Klaus Bartschat

09:00-09:45   Oksana Travnikova
               Energy dependent cross sections and line shapes in molecular photoionisation
09:45-10:15   Piero Decleva
               The B-spline molecular code: from single photon to strong field ionization

10:15-10:50 Coffee break

Session 6 - Chair: Károly Tőkési

10:50-11:20   Konstantin Kouzakov
               Laser-assisted electron scattering and ionization processes
11:20-11:50   Li Fang
               Investigation of molecular fragmentation subsequent to photoionization
               by synchrotron radiation at core-level
11:50-12:20   Jyoti Rajput
               Three-body dissociation of molecular ions: separating sequential
               and concerted pathways

12:20-12:50 Committee meeting
12:30-14:00 Lunch

Session 7 - Chair: Boghos Joulakian

14:00-14:30   Ziad El Bitar
               A quantum approach to calculate differential and total cross sections
               for electron impact ionization of biologically relevant molecules
14:30-15:00   Matthew Harvey
               Controlling electron spectrometers using LabVIEW and Arduino interfaces
15:00-15:30   Maomao Gong
               Theoretical study of (e,2e) processes for atoms and molecules by multi-center
               three distorted wave method

15:30-16:30 Coffee break and Poster session

Session 8 - Chair: Reika Kanya

16:30-17:00   Anatoli Kheifets
               Attoclock on atomic and molecular hydrogen
17:00-17:30   Daehyun You
               Attosecond-precision coherent control experiment at FERMI
17:30-18:00   Margarita Khokhlova
               Coherent control of Auger decay. Molecular Auger interferometry
From 19:30 Conference dinner

Saturday 3rd August

Session 9 - Chair: Maria Novella Piancastelli

09:00-09:45   Igor Bray
                  Application of the close-coupling method to ionisation processes
09:45-10:15   Daniel Fischer
                  An all-optical atom trap as a target for MOTRIMS-like collision experiments

10:15-10:50 Coffee break

Session 10 - Chair: Alexei Grum-Grzhimailo

10:50-11:20   Tommaso Mazza
                  X-ray spectroscopy of ultrafast-decaying core-excited ionic states in atoms
11:20-11:50   Feng He
                  High-order above threshold dissociation of H_2^+ in strong laser fields
11:50-12:20   Christian Ott
                  Nonlinear XUV absorption spectroscopy of excited-state electron dynamics
                  in atoms

12:20-12:30 Concluding remarks

12:30-14:00 Lunch

End of conference

ABSTRACTS

OF PLENARY TALKS

Size and environmental effects in molecular photofragmentation

P Bolognesi[1*]

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Synopsis. The study of radiation interaction with molecules of biological interest is a valuable tool to determine their spectroscopic characterics, to evaluate the energy transfers and to probe the time evolution of the molecular excited states.

Atomic and molecular physics has developed over the years new and increasingly sensitive tools, in terms of both radiation sources and detection systems, which allow to achieve unprecedent detail in the level of the spectroscopic and dynamic investigation and open up the road towards unexplored areas.

Very diluted gas phase targets like biomolecules, for example, require intense radiation sources in order to catch the finest details of their structure, while the 'real time' observation of the evolution of their excited states demands for ultrashort laser pulses able to follow nuclear as well as electronic dynamics in the femto or even atto-seconds regimes.

The understanding of the physical-chemistry of biomolecules like DNA bases, aminoacids and drugs is extremely interesting. They offer a huge variety in terms of a building block approach to complexity, structure and functions they perform. In addition, the underlaying motivation in the study of biomolecules is to identify properties or mechanisms that connect structure to functions, in order to master their design and exploitation for specific applications. As such, biomolecules pose new challenges but at the same time offer great opportunities to both experimentalist and theoreticians.

Notwithstanding the value of fundamental understanding of biomolecules isolated in the gas phase, the complexity of the biological context in which they will eventually operate calls for a critical evaluation of the representativeness of the results obtained for isolated molecules with respect to their application in real biological media.

In this presentation, a few case studies of biomolecules which extend from the characterization of their electronic structure [1] and radiation interaction [2] to the temporal evolution [3] of some molecular processes will be reported.

For example, the study of the mechanisms of functioning of different classes of radiosensitisers of increasing complexity show how valuable informations can be extracted also for relatively complex molecules, how this information can be tentatively related to their biological functions and how the use of more realistic environment can affect this picture.

In another example it will be shown that pump probe experiments performed with few fs resolution allow to disentangle fast molecular rearrangements that preceed and affect molecular fragmentation.
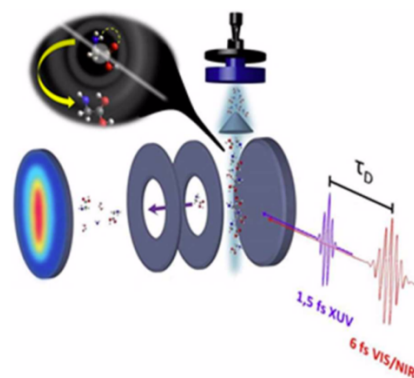

Figure 1. Pump-probe experiment in glycine [2].

References
[1]   Chiarinelli J *et al* 2019 *Front. Chem.* **7** 329; Bolognesi P *et al* 2016 *J. Chem. Phys.* **145** 191102; Castrovilli M C *et al* 2017 *Phys. Chem. Chem. Phys.* **19** 1980
[2]   Maclot S *et al* 2016 *Phys. Rev. Lett.* **117** 073201
[3]   Castrovilli M C *et al* 2018 *J. Phys. Chem. Lett.* **9** 6012

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Accuracy of Theoretical Calculations for Electron-Impact Ionization of molecules

D H Madison[*]

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Synopsis In the last two decades, there have been numerous studies of high-energy electron impact single ionization of molecules. These studies were called EMS (Electron Momentum Spectroscopy) and they were very valuable in determining the accuracy of molecular wavefunctions since the measured cross sections were proportional to the momentum space molecular wavefunction. More recently, fully differential cross sections for lower energy collisions have started to be measured and these cross sections are much more difficult for theory since the detailed kinematics of the experiment become important. In this talk, I will show the current status of agreement between experiment and theory for low and intermediate energy single ionization of small, intermediate and large molecules.

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Energy dependent cross sections and line shapes in molecular photoionisation

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Synopsis I will discuss recents results on the studies of photoionisation and dominant accompanying processes. For instance, coherent photoemission from inner-valence shells gives rise to interference effects, which reflect molecular geometries. Elastic electron scattering is proven to play an important role on inner-shell photoionisation cross sections of small isolated organic molecules. Photoelectron recoil can lead to vibrational and rotational excitations that change and shift profiles of the measured photoelectron lines.

The presented work is a result of several years of research on the influence of the different chemical environments on the core-electron photoionisation cross sections and accompanying processes occurring in isolated molecules. Photoelectron spectroscopy is widely used as a tool for quantitative analysis in solid state physics and surface sciences. Yet, the values of the atomic cross sections are commonly used. In our gas phase study we show that ionisation cross sections can be significantly modulated in molecular environments compared to the atomic ones.

Relative intensities of the C $1s$ photoelectron lines from chemically inequivalent carbon atoms in the same molecule have been measured as function of incident photon energy in the range of 300 – 6000 eV for a series of molecules, such as chloroethanes and ethyl trifluoroacetate and substitutionally related compounds. We observe EXAFS-like oscillations of the photoionisation cross sections of a single molecule. The current findings have been supported by theoretical calculations based on a multiple scattering model. Moreover, the use of soft and hard x-rays gives access to characterisation and separation of intramolecular inelastic scattering and shake processes accompanying photoionisation. Such processes reduce the main line intensities by 20–60%, cf. Figure 1. Intramolecular inelastic scattering relates to the notion of the Inelastic Mean Free Path (IMFP) commonly used in photoemission studies of solids. [1, 2]

Additionally, in the molecules with equivalent atomic sites a photoelectron can be emitted coherently from different centres leading to interference effects, which also result in oscillations of the cross sections. [3, 4]

At sufficiently high photoelectron kinetic energies other effects, such as photoelectron recoil, become important. Recoil-induced vibrational excitations, which are not populated by normal Franck-Condon excitations, cause major changes in the vibrational profiles of the photoelectron lines. At the same time photoelectron-recoil-induced rotational excitations lead to shifts of photoelectron lines, which become apparent in the hard X-ray regime. [5]
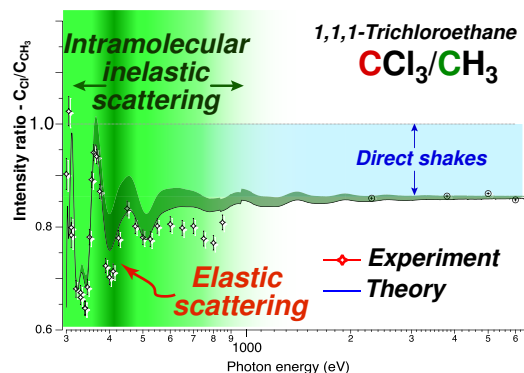

Figure 1. The relative cross section ratios up to 6 keV for C $1s$ ionization of the chemically different carbon atoms in 1,1,1-trichloroethane, CH_3CCl_3.

References

[1] Travnikova *et al submitted*
[2] Söderström *et al* 2012 *Phys. Rev. Lett.* **108** 193005
[3] Kushawaha *et al* 2019 *Phys. Chem. Chem. Phys* DOI: 10.1039/c9cp00723g
[4] Kushawaha *et al* 2013 *PNAS* **110** 15201
[5] Kukk *et al* 2018 *Phys. Rev. Lett.* **121** 073002

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Application of the close-coupling method to ionisation processes

I Bray[1] [*], D V Fursa[1], A S Kadyrov[1], A T Stelbovics[1], A W Bray[2], A S Kheifets[2] and M C Zammit[3]

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Synopsis   The close coupling method was originally designed for calculating electron-impact elastic scattering and excitation of atomic targets. In the last few decades the method has been extended to ionising collisions by electrons, positrons, or photons, and for atomic, ionic and molecular targets.

The close-coupling method for calculating electron-atom scattering dates back to Massey and Mohr [1]. The idea was to expand the total wavefunction of the system using the lowest lying bound states of the target, thereby obtaining scattering amplitudes for eleastic scattering and the included excited states.

With the rapidly increasing computational power during the late 20th century it became possible to take these ideas to completeness by including complete square-integrable state expansions to convergence [2]. Such calculations were labeled convergent close-coupling (CCC), and showed that the method converged to the exact solutions known for elastic scattering and excitation for a model e-H scattering problem. They also showed that the summation of the cross sections for excitation of the positive-energy states was convergent, and identified as the total ionisation cross section.

Subsequent application to the full e-H problem yielded excellent agreement with experiment [3], heralding the birth of applications of close-coupling methods to ionising collisions [4], including in the closely related field of double photoionisation [5]. Most recent CCC applications include the study of electron- and positron-impact near threshold ionisation of atomic hy-

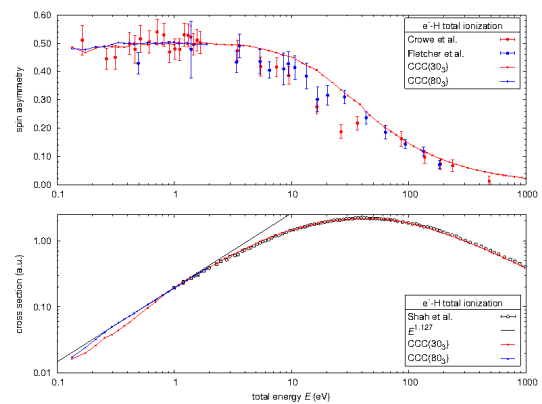drogen [6], see Figure for the e^--H example.

Figure 1. Ionisation cross section and spin asymmetry in e^--H collisions, see [6] for details.

References

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ABSTRACTS OF

PROGRESS REPORTS

Water acts as a catalyst for electron-driven chemical reaction in biochemically relevant hydrogen-bonded systems

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Synopsis   We study the ionization and fragmentation of tetrahydrofuran (THF) cluster and hydrated THF clusters induced by low-energy (65 eV) electron-collision using an (e, 2e + ion) triple coincidence technique in which the momentum vectors and the kinetic energies for all three final-state charged particles are determined. The influence of the environment on the electron induced fragmentation of THF is investigated with the ion fragment correlated binding energy spectra and triple-differential cross sections as well as *ab-initio* dynamical calculations. We find that ring breaking reaction are strongly facilitated in dimers.

Electron-impact ionization of atoms and molecules is a fundamental process which is relevant to understand and interpret a wide range of scientific phenomenon and technological applications including radiation damage in biological tissue [1]. An important role in this respect plays water and hydrated systems which have been the subject of intense studies due to their importance in life and environmental sciences [2].

In the present work, we investigate the electron-collision induced ionization and fragmentation processes in clusters consisting of water and bio-relevant molecules like DNA constituents. We use the multi-particle coincidence technique (reaction microscope) [3, 4] in which the momentum vectors and, consequently, the kinetic energies of all final state charged particles (electrons and ions) are measured in coincidence. The bio-molecule employed here is tetrahydrofuran (THF, C_4H_8O) which is often regarded as being the simplest molecular analog of deoxyribose, part of the DNA backbone linking the phosphate groups and the DNA bases. Figure 1 presents the measured fragment ion time-of-flight (TOF) spectra for both THF and hydrated THF clusters induced by electron ionization (65 eV). Compared to the fragmentation of isolated THF molecules we find in clusters a strong and new ring breaking reaction: $C_4H_8O \rightarrow C_2H_4O^+$ + neutrals. This is manifested by the species $(C_4H_8O) \cdot C_2H_4O^+$ for pure THF clusters and $(H_2O) \cdot C_2H_4O^+$ for hydrated clusters,

see encircled lines in Fig. 1. The binding energy spectra and *ab-initio* dynamical calculations show that in the hydrogen-bonded dimers the activation energy for ring opening channel is reduced. Therefore, we observed that the water environment or a second THF acts as a catalyst for the break-up of THF ring structure. Detailed results on hydrated THF complex and other biochemically relevant clusters will be presented at the conference.
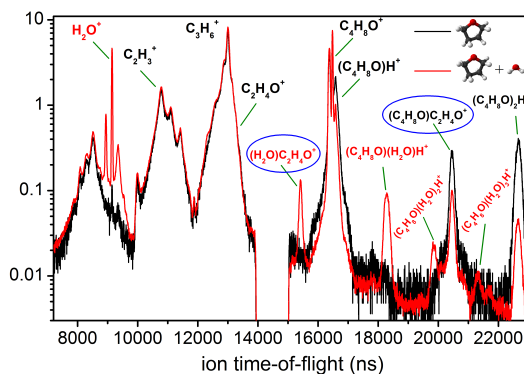

Figure 1. Measured fragment ion time-of-flight spectra of pure THF and hydrated THF clusters.

References

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R-matrix Calculations for Ultrafast Two-colour Spectroscopy of Noble Gas Atoms

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Synopsis   We present results of R-matrix with time-dependence (RMT) method calculations in a variety of two-colour spectroscopic schemes with noble gas targets. The results highlight the capability of RMT to describe multielectron dynamics in complex atomic systems subjected to computationally demanding laser pulses.

We sought to determine the influence of field-induced and multielectron effects on spectroscopic observables of noble gas atoms. To describe such systems requires accurate portrayal of both the atomic structure including electron correlation, and electron dynamics mediated by an IR laser. Only a handful of methods exist which can handle them simultaneously; one such method is the R-matrix with time-dependence method (RMT).

RMT is an *ab-initio* technique which solves the time-dependent Schrödinger equation by employing the R-matrix paradigm, dividing configuration space into two regions [1, 2]. RMT has been previously applied to studies of strong-field processes including electron rescattering [3], XUV-initiated HHG [4] and has recently been extended to describe dynamics in arbitrary light fields [5].

We applied RMT to High Harmonic Generation from neon atoms in a time-delayed laser pulse scheme [6], and XUV-Initiated High Harmonic Generation, Photoelectron Spectroscopy (PES), and Attosecond Transient Absorption Spectroscopy (ATAS) in argon. The ATAS and PES spectra can both be determined from a single RMT calculation (Figure 1), allowing for a direct comparison between the two techniques. In these studies we identified signatures of multielectron dynamics, strong field processes, and, most importantly, examples of the interplay between these two which enhance our ability to use ultrafast laser technology to probe exotic electronic phenomena in atoms and molecules.
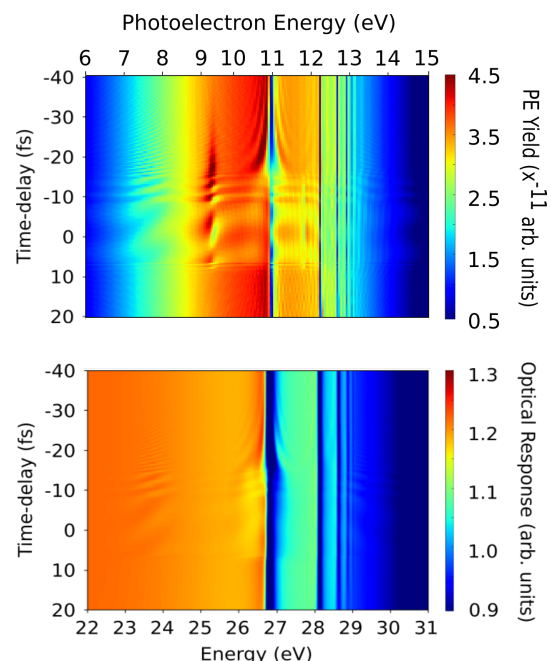
Figure 1. Photoelectron (top) and Attosecond Transient Absorption spectra (bottom) of the $3s3p^6np$ states of argon in a mixed 800nm + 27eV laser pulse scheme.

References

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[8] http://www.archer.ac.uk/

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Real-Time Imaging of Ultrafast Charge Dynamics in CF$_4$ Molecule from Attosecond Pump-Probe Photoelectron Spectroscopy

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Synopsis   Attosecond spectroscopy offers the possibility to track and control electron dynamics in molecules. We are reporting here a theoretical description of an attosecond pump-probe experiment on CF$_4$ showing clear evidences of a complex dynamics due to ultrafast charge fluctuations.

Since the first experimental demonstration of attosecond (as) pulses in 2001 [1], many progresses have been achieved in attosecond technology. In particular, it is now possible to control electron dynamics in atoms, diatomic molecules [2] and polyatomic molecules [3]. A widely used approach to perform such time-resolved studies is the attosecond pump-probe spectroscopy. It consists in analyzing the response of the atom/molecule interacting with two pulses: a pump pulse (generally an attosecond XUV pulse generated by high-harmonic generation) and a probe pulse (IR, VIS, XUV, or X-ray) used to probe the dynamics generated by the former pulse. The temporal information is acquired by varying the time-delay between the two pulses.

In this work, we present the results of a theoretical simulation of an attosecond pump-probe experiment on CF$_4$ molecule with two collinear pulses. A 150 as XUV pump pulse centered at 33 eV ionizes the molecule, ejecting an electron from different molecular orbitals, followed by a time-delayed 3.6 fs VIS pulse (3.2 eV). The laser intensities, 2×10^{11} and 2×10^{12} W/cm^2 respectively, are weak enough to avoid the distortion of the molecular potential.

The theoretical methodology consists in solving the time-dependent Schrödinger equation in a basis of Kohn-Sham orbitals and using the exclusive probability formalism to include interchannel couplings [4]. The basis is obtained in the dipole, fixed-nuclei and static-exchange approximations by solving the Kohn-Sham Hamiltonian using a B-spline multicenter approach and the LB94 functional.

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Figure 1 shows the photoelectron spectra leaving the ion in two different states. In both cases, the photoelectron spectra due to the XUV pump only has been subtracted in order to increase visibility. The two spectra present oscillations with two different behaviors: in the region where pump and probe pulses overlap (τ<2.5fs), the oscillations are approximately twice the frequency of the VIS probe pulse ; for higher time-delays, the oscillations present different frequencies, depending on the photoionization channel, which indicate an ultrafast charge dynamics beating between the CF$_4^+$($1t_1^{-1}$) and the CF$_4^+$($3t_2^{-1}$) states.
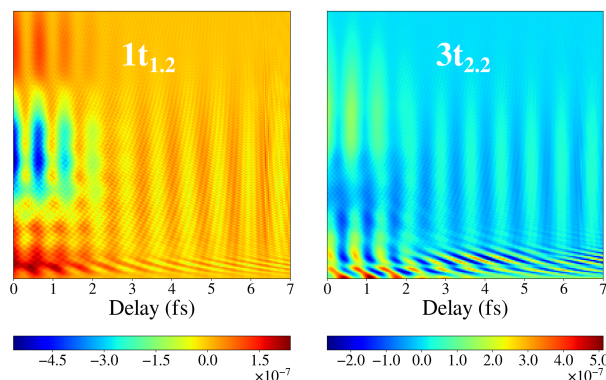

Figure 1. Theoretical pump-probe photoelectron spectra of two photoionization channels of CF4.

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Photoelectron spin polarization in bichromatic-field ionization of atoms

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[1]Skobeltsyn Institute of Nuclear Physics, M.V.Lomonosov Moscow State University, Moscow, 119991, Russia
[2]Faculty of Physics, M.V.Lomonosov Moscow State University, Moscow, 119991, Russia

Synopsis Photoelectron spin polarization may be effectively controlled in ionization induced by the combined action of the fundamental and the second laser harmonics by variation of their relative intensity, phase and polarization. Appropriate choice of the pulse parameters allows moving the maximum of spin polarization towards the maxima in the differential cross section and making it thereby easier to observe.

Spin polarization of photoelectrons has attracted attention for decades as a fundamental phenomenon and as an attribute in applications. Spin polarization shows up due to differences in the wavefunctions of the fine-structure components, which are revealed in this or that way (separating fine-structure levels of the target atom or the photoion, spin-orbit interaction in atomic continuum). The main reasons for the scarce studies of the photoelectron spin polarization in experiments with isolated atoms are related to the crucial decrease of efficiency of spin-sensitive detectors and the fact that essential spin polarization is often reached at photon energies in the minima of the cross sections.

Ionization of an atom by the combined actions of the fields of the fundamental (ω) and the second (2ω) laser harmonics $\mathbf{E}(t) = \boldsymbol{\varepsilon}E_0(t)$ ($\cos \omega t + \eta \cos (2\omega t + \phi)$) provides the opportunity to control the angular distribution and the spin polarization of photoelectrons by manipulating the relative phase ϕ and strength η of the second harmonic. This possibility was recently discussed for alkali atoms in the optical range [1] and the first results are available for photoprocesses in condensed matter [2]. The advent of free-electron lasers producing intense coherent XUV light with variable polarization may open new avenues in studies and control of the photoelectron spin polarization. Current research aimed extending these investigations to the high-frequency domain and to strongly correlated targets, such as noble gases.
Interference between resonant two-photon ionization by the fundamental and direct one-photon ionization by the second harmonic provides a mechanism to control both photoelectron angular distribution [3] and spin polarization. Results of this interference may appear in a crucial way. For instance, if both harmonics are linearly polarized, neither single nor two-photon ionization of an atom produce electrons polarized in the plane spanned by the polarization vector and the emission direction, but the interference does. An example of such a polarization is presented in figure 1. The original cause of the photoelectron spin polarization in our case is spin-orbit interaction in the intermediate excited state leading to fine-structure splitting and term mixing. The spin polarization vanishes after summation over the fine-structure levels.
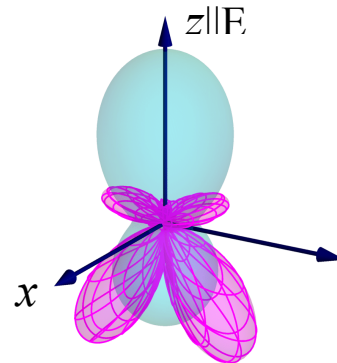

Figure 1. PAD (blue) and absolute value of spin polarization along the x-axis (magenta, dashed) at ω corresponding to the excitation of the lowest $2p^5[^2P_{3/2}]3s_1$ state in neon. The pulse parameters: intensity 10^{12}W/cm^2, $\eta = 0.03$, $\phi = 0$, duration 250 optical cycles; the harmonics are linearly polarized. The maximum of spin polarization is about 20%.

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Asymptotic behavior of plane- and distorted-wave theories to their high energy limits

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Synopsis   We have examined the range of the validity of the plane wave impulse approximation (PWIA) by making comparisons with binary (e, 2e) cross sections measured for rare gas atoms at various incident electron energies (E_0) from 1.2 to 4.0 keV. The comparisons clearly show that the PWIA reproduces the experimental results up to larger momenta at higher E_0 value. These findings strongly indicate that a wider range of spatial distributions of molecular orbitals could be addressed with binary (e, 2e) spectroscopy as the E_0 increases.

Electron momentum spectroscopy (EMS), also known as binary (e, 2e) spectroscopy, is a kinematically-complete electron-impact ionization experiment performed under the high-energy Bethe ridge conditions [1,2]. Of particular note is that within the framework of the plane-wave impulse approximation (PWIA) the triple differential cross section (TDCS) is directly connected to the electron momentum distributions of individual molecular orbitals (MOs). Thus, the knowledge about the range of the validity of PWIA is the key for promoting molecular science with EMS. However, such experimental studies are scarce as yet and the detailed knowledge is among the issues to be further explored. In the present work, we have conducted a series of binary (e, 2e) experiments on rare gases at various incident electron energies (E_0) in order to identify how the range of validity of PWIA depends upon the E_0 value.

The experiments were performed at an E_0 range from 1.2 keV to 4 keV by using one of our symmetric-noncoplanar (e, 2e) spectrometers [3] in which two outgoing electrons having equal energies and making the scattering angle of 45 degrees were dispersed by a spherical analyzer and detected by a position-sensitive detector.

Figure 1 shows one of the experimental results thus obtained, which are the TDCSs for the ionization of the Ne 2p orbital measured at E_0 of 1.2 and 4.0 keV. Also included are associated PWIA calculations as well as distorted wave Born approximation (DWBA) calculations that consider distortion of electron waves due to the static Coulomb field of the residual ion. It is found from comparisons between the

experiment and theory that the range of the validity of the PWIA becomes broader at the higher E_0 value, indicating that a wider range of spatial distributions of MOs could be assessed with EMS.
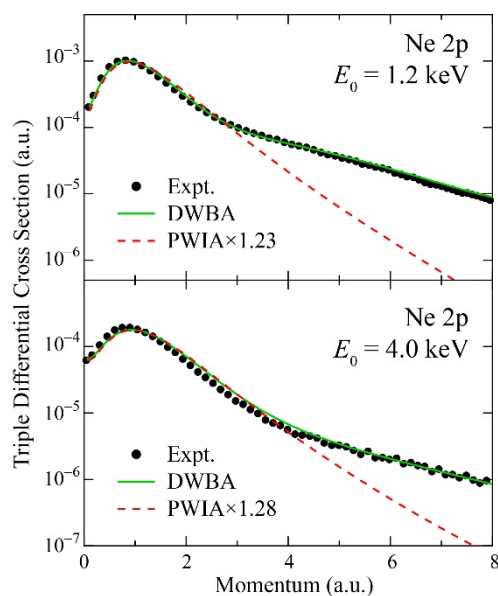

Figure 1. Experimental and theoretical TDCSs of the Ne 2p atomic orbital.

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(e,2e) at intermediate energy on methane

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Synopsis $(e, 2e)$ processes on small molecules such as CH_4, H_2O and NH_3 are investigated at intermediate incident energy and asymmetric coplanar geometries. Triple differential cross sections (TDCS) are presented for electron-impact single ionization of the $1t_2$ and $2a_1$ states of CH_4 at an incident electron energy of 250 eV. Experimental data from two different laboratories are compared with molecular 3-body distorted wave and generalized Sturmian function calculations. Overall good agreement is found. Under some kinematic and geometrical conditions a splitting in the binary lobe is observed for the $1t_2$ state.

Over the last few decades $(e, 2e)$ on small molecules have been studied in a variety of kinematical conditions. Contrary to simple atomic targets, like Hydrogen or Helium, the obtained TDCSs may exhibit more elaborate angular distributions that can be associated to both the more complex molecular structure and to its multicenter nature. Generally speaking, the theory-experiment agreement for molecular targets is not as good as for atoms.

Only in recent years some experimental and theoretical works started to consider an intermediate energy regime with a focus on asymmetric coplanar geometries (see, e.g., [1] for more details, specially on CH_4, H_2O and NH_3). In general, theory-experiment agreement is reasonably good in the binary region but not so much in the recoil region. Moreover, theories present differences in the absolute scale that the relative scale experiments cannot separate. Under particular kinematic conditions and geometrical configurations, a double peak in the binary region has been observed both theoretically and experimentally for several molecules, a feature usually associated to the p−nature of the ionized molecular orbital. In [2], calculations predicted a clear double binary peak for a specific situation for the HOMO $1t_2$ of CH_4. It is the purpose of this contribution to explore the nature of some of these features.

In this joint theoretical-experimental effort we study $(e, 2e)$ processes on CH_4 [3], for both HOMO $1t_2$ and NHOMO $2a_1$ states. We compare two sets of experimental measurements (performed at Manchester, UK and at Afyon, Turkey) with theoretical calculations performed using 3-body distorted wave (M3DW) and generalized Sturmian functions (GSF). All measurements were performed for incident electrons with an incident energy of 250 eV and for ejected electrons with energy of 30 and 50 eV with several different projectile scattering angles, in a coplanar asymmetric geometry.

For ionization of the $1t_2$ state the agreement between experimental measurements and theoretical calculations is fairly good. We see a binary peak that is sometimes doubled with a minimum close to the momentum transfer direction. The evolution of such binary peak vary with the scattering angle, a feature that is explored in detail using both M3DW and GSF calculations.

For ionization of the $2a_1$ state, the theory-experiment agreement is very good. A single maximum is observed in the binary region.

Through theoretical calculations in different kinematical and geometrical configurations, we also investigate the appearance of the double peak in the binary region for other molecular systems, such as H_2O and NH_3.

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Double photoionization of water

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Synopsis   Double ionization of water by single-photon absorption has been studied via the angular resolved coincidence detection of the emitted electrons. Experimental differential cross sections present rich angular distributions. The latter can be, in part, reproduced by a first order treatment of the interaction considering a two active electrons target for which we evaluate the scattering wavefunction, and from it, the cross sections.

Understanding double photoionization (DPI) is of great interest because electronic correlation plays a decisive role in the determination of cross sections, in particular triple differential cross sections (TDCS) which provide the most detailed information on the interactions. Advanced understanding of DPI has been achieved for two-electron atomic systems [1]. For diatomic and polyatomic molecules, on the other hand, experimental and theoretical studies of the DPI mechanisms through electron-electron coincidence angular distribution are scarce.
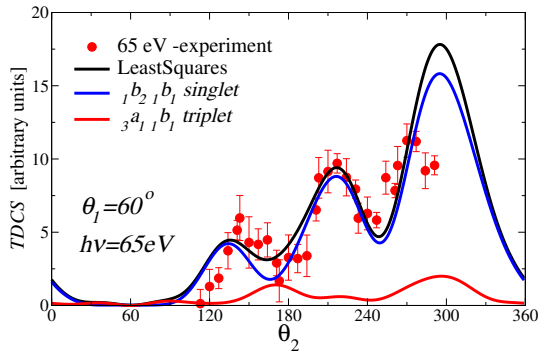

Figure 1. TDCS angular distribution measured and calculated at 65 eV photon energy and $\theta_1 = 60^o$. The contribution of the $(_1b_2)^{-1}(_1b_1)^{-1}$ and $(_3a_1)^{-1}(_1b_1)^{-1}$ states of H_2O^{2+} as well as their combination (black full line) are shown.

In this combined experimental and theoretical work the DPI of water has been investigated. The measurements [2] have been performed at the Gas Phase beamline of Elettra at about 20 eV above the double ionization threshold. Two types of measurements have been performed. In the first one, the electron energies are fixed and

the photon energy scanned, in order to reconstruct the binding energy spectrum of the water dication. In the second type, the photon energy has been fixed and coincidence angular distributions have been measured fixing the direction of one electron at 0°, 30° and 60° with respect to the photon polarization direction. Two photon energies (63 and 65 eV) have been chosen in order to explore dication states with different symmetries.

The measured data have been compared with Generalized Sturmian Functions calculations describing the dynamics of the two electrons emitted in the continuum. The method employed is similar to the one implemented for atomic targets [3] but has been adapted to take into account the multicenter nature of the target and the molecular orientation. Within some approximations and a model potential as in Ref. [4], and for a given molecular orientation, we end up with a three-body Schrödinger equation that we solve 'exactly'. From the numerical double continuum wavefunction we extract the TDCS; a molecular orientation average is then performed.

The experiment-theory comparison is fair, considering some of the theoretical approximations and the mixture of states that are involved at 65 eV. An example of the rich angular distribution is given in Fig. 1.

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Separating Dipole and Quadrupole Contributions to Single-Photon Double Ionization

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Synopsis We report on a kinematically complete measurement of double ionization of helium by a single 1100 eVcircularly polarized photon. By exploiting dipole selection rules in the two-electron continuum state, we observed the angular emission pattern of electrons originating from a pure quadrupole transition.

At high photon energies, the electric dipole approximation is no longer valid and the photon's linear momentum evidently modifies photoelectron angular distributions [1]. In most cases however, the angular distributions are, due to the dominance of the dipole contribution, only slightly modified by the interference term between the quadrupole and the dipole transition (see Ref. [2] for a review). As such, the quadrupole transition amplitude alone has not been directly observed until now.

We succeeded in experimentally isolating the quadrupole contribution to photo-double-ionization and visualize a pure quadrupole pattern in the angular distribution of electrons emitted from a helium atom (Fig. 1). We employed the COLTRIMS reaction microscope technique [3] and intersected a cold supersonic He gas jet with a beam of 1100 eV circularly polarized photons from beamline P04 at Petra III (DESY, Hamburg). The quadrupole contribution to the photoionization process was accessed by exploiting a dipole selection rule for the two-electron continuum. It states that for two electrons of opposite spin the electron pair wave function vanishes for total angular momentum $L = \hbar$ and $\mathbf{k}_a = -\mathbf{k}_b$ (where $\mathbf{k}_{a;b}$ are the momentum vectors of the two electrons a and b) [4]. This pattern corresponds to a nucleus at rest and two electrons receding back-to-back with equal energies. At large distances, this resembles a spatial configuration of the three charges which has only a quadrupole but no charge dipole moment.
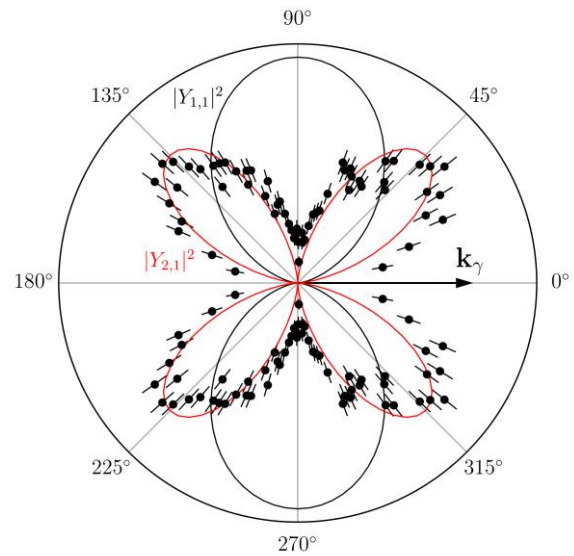

Figure 1. Angular distribution of one of the electrons from photo-double-ionization of He by a single 1100 eV circularly polarized photon. The light propagation axis is horizontal ($\mathbf{k}_\gamma$). Data points: electrons of equal energy ($\Delta E = 0.5 \pm 0.1$) emitted back to back ($\Delta\theta = 180 \pm 20°$, electron mutual angle). For this subset of the data, dipole contributions to the cross section vanish due to selection rules. Black line: dipole contribution ($|Y_{l=1,m=1}|^2$), red line: quadrupole contribution fitted to the data points ($|Y_{l=2,m=1}|^2$).

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Unfolding the physics of photoionization dynamics in simple molecules

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Synopsis In this progress report will be presented the investigation of the dissociative photoionization of three different molecules addressing: autoionizing states and nuclear motion in H_2, the ionization mean position within the asymmetric potential of CO and shape resonances in N_2O. Each experimental result is compared with specific calculations.

Up to now, the observation of photoionization time delays in molecules has been limited to only a few experimental studies [1-3] mostly due to the complexity of molecular targets which makes "clean" and "complete" measurements very challenging.

The three molecular cases described in this progress report: H_2, CO and N_2O, demonstrate how the phase of the escaping electron wavepacket is a sensitive probe not only of the electron dynamics, well studied for atomic cases, but also of the nuclear dynamics in molecules containing light nuclei [4], the electron localization at the moment of ionization [5] and the influence of shape resonances (SR).

All these investigations have been carried out exploiting the XUV pump/IR probe RABBITT interferometric technique combined with a coincidence detection using a COLTRIMS apparatus [6,7].

In H_2 [4] we obtained the first experimental evidence of the importance of the coupled electron-nuclear motion for the attosecond ionization dynamics confirmed by a complete ab initio theoretical study with the group of Fernando Martín.

The study in CO [5], with an asymmetric Coulomb potential, revealed that the accumulated phase of the escaping electron wave packet is not only energy- and molecular orientation dependent, but can give insight into the mean position of the ionization within the CO molecular potential. This dependence is unique to the molecular photoionization process and has been supported by two different theoretical models by S. Patchkovskii's and A. Landsman's groups. We introduced the Stereo-Wigner Time Delay (SWTD) which is the time delay difference between two escaping electrons on opposite sides of the molecule. The unexpected highly negative SWTD can be explained by the localization of the electron within the molecular frame right at the beginning of the electron escape dynamics.

Finally we exploited angular-resolved attosecond measurements in N_2O molecules to reveal new physical insights into the phenomenon of shape resonances (SR) in molecules: a heavily discussed topic in the field of molecular spectroscopy that is still not completely clarified [8]. It has been recently shown by Huppert et al. that SR can induce photoionization delays up to ~160 as in N_2O [2]. In particular we inspected the dependence of the photoionization time delays on the photoelectron emission angle mapping the internal molecular potential. This study is theoretically supported using quantum scattering methods provided by the group of Prof. H. J. Wörner.

All these works confirms the importance of angular resolution and coincidence detection towards the complete understanding of more complex systems.

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The B-spline molecular code: from single photon to strong field ionization

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Synopsis   Theoretical description of molecular continuum procees by the B-spline code will be discussed

A code for molecular electronic continuum processes based on the use of a multicentric (LCAO) B-spline basis, at the DFT and TDDFT level, has been developed over the years for the theoretical description of molecular photoionization processes [1-3]. Recent evolution generalizes to the use of Dyson orbitals for ionization involving multiconfigurational and correlated bound states [4], and the use in TDSE calculation for strong field ionization and HHG, and description of attosecond phenomena [5]. Further current development towards ab initio close coupling formulation will be discussed.

The primitive functions are of the form

$$\chi_{ilm} = (1/r) B_i(r) Y_{lm}(\theta, \varphi)$$
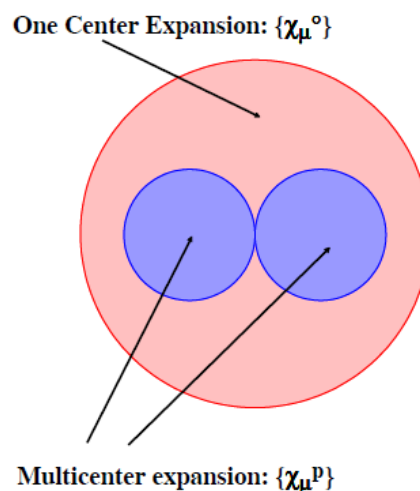
and the full basis is depicted in Figure 1

Figure 1. The LCAO B-spline basis

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Laser-assisted electron scattering and ionization processes

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Synopsis   We develop a quasi-Sturmian-Floquet method to treat electron scattering states in combined Coulomb and laser fields. The robustness and efficiency of our approach is demonstrated with theoretical calculations of the archetypical processes such as laser-modified electron-proton scattering and laser-assisted electron-impact ionization of atomic hydrogen. For this purpose, different frequency and intensity regimes of laser radiation are examined along with different laser-field orientations.

Recently we have formulated a nonperturbative method [1, 2] for the theoretical treatment of laser-modified electron states in various radiation and ionization phenomena in the presence of intense laser fields. Our method is based on the Hermitian Floquet theory in an accelerated, Kramers-Henneberger frame. We use the quasi-Sturmian basis functions [3] taken in parabolic coordinates to solve the system of coupled Lippmann-Schwinger-Floquet equations for the laser-modified Coulomb wave functions of an electron.

In order to develop and test our approach, we investigate theoretically (i) laser-modified nonrelativistic electron scattering on a proton target,

$$\ell\omega + e^- + p \rightarrow p + e^-, \tag{1}$$

and (ii) a laser-assisted $(e, 2e)$ collision on atomic hydrogen,

$$\ell\omega + e^- + H(1s) \rightarrow H^+ + 2e^-, \tag{2}$$

where a net number ℓ of photons of frequency ω can be exchanged between the projectile-target system and the external field.

For process (1), we calculated the ratio of the laser-assisted angular differential cross section $d\sigma_\ell/d\Omega$ to the laser-free, Rutherford cross section $d\sigma_R/d\Omega \propto 1/\sin^4\frac{\theta}{2}$. We explored both the high-frequency ($E_e \ll \omega$) and low-frequency ($E_e \gg \omega$) regimes. For the latter case, Fig. 1 shows the convergence of our numerical results with increasing number $\mathfrak{N}$ of Floquet components of the electron state. An important feature is that the calculated ratio substantially differs, especially at larger angles, from unity, i.e., from the value predicted
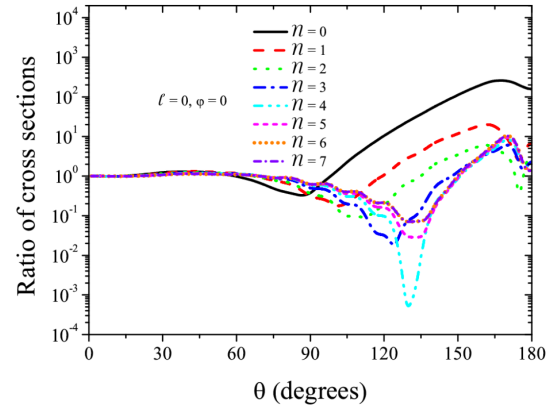
by the customary Volkov and Coulomb-Volkov models.

Figure 1. The ratio $\frac{d\sigma_{\ell=0}}{d\Omega}/\frac{d\sigma_R}{d\Omega}$ as a function of the electron scattering angle when $E_e = 0.43$ a.u., $\omega = 0.05$ a.u., and the laser electric field amplitude is $F_0 = 0.0125$ a.u. The laser field is linearly polarized and is perpendicular to the scattering plane.

For process (2), we studied the asymmetric $(e, 2e)$ kinematics with a laser field either parallel to the incident electron momentum or perpendicular to the scattering plane [1, 2]. We investigated ejected-electron angular distributions in the scattering plane for various ℓ values in the perturbative and nonperturbative regimes of laser radiation. Our calculations using the Born approximation in the projectile-target interaction converge fast with increasing number $\mathfrak{N}$ of Floquet components for the ejected-electron state.

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Investigation of molecular fragmentation subsequent to photoionization by synchrotron radiation at core-level

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Synopsis   We investigated the charge distribution and dissociation dynamics of glycine (NH_2CH_2COOH) molecule irradiated with 310 eV x-rays from the Advanced Light Source synchrotron. With simultaneous measurements of the fragment ion yield, dissociation angle, and kinetic energy, we were able to reconstruct a three-dimensional image of the molecular dissociation. Applying a systematic comparison of properties of ion species, we partially disentangled the fragmentation pathways and identified the most probable fragmentation channels that lead to the observed fragment ions. In addition, we found an association between the initial bond breaking sites and the kinetic energies of the final fragment ions.

We investigated the molecular fragmentation of glycine molecule (NH_2CH_2COOH) subsequent to photoionization using x-rays with a photon energy above the k-edge of atomic carbon. The experiment was performed at the Advanced Light Source, Lawrence Berkeley National Laboratory (USA). Applying 3-D momentum imaging, we carried out co-incidence measurements of the charge state, kinetic energy and angular distribution of the fragment ions. With the information of the multiple variables, we are able to determine the dominant pathways of the fragmentation [1].

At the photon energy above the k-edge of the atomic carbon in the molecule, doubly charge molecular parent ions were dominantly produced due to photoionization and the subsequent Auger decay process. We observed dramatic bond cleavages of the parent molecular ions and the resultant molecular ions. The dominant coincident fragment ions are O^+ and C^+, indicating that the molecular ions tend to break every single bond to release the absorbed photoenergy.

The measured angular distributions for different dissociation channels show that O^+ involving channels are associated with very strong dependence of kinetic energy on the dissociation pathways, in contrast with other channels that show much weaker dependence.

Figure 1 presents the dissociation angular distributions for various dissociation channels, showing a dependence on the dissociation pathways. The dissociation-angle measurement

provides, in addition to the coincidence abundance, important information that we used to determine the fragmentation pathways. For example, [O^+, CO^+] were produced through a different pathway than [C^+, CO^+], since these two channels are associated with different dissociation angular distributions, as seen in Fig. 1.
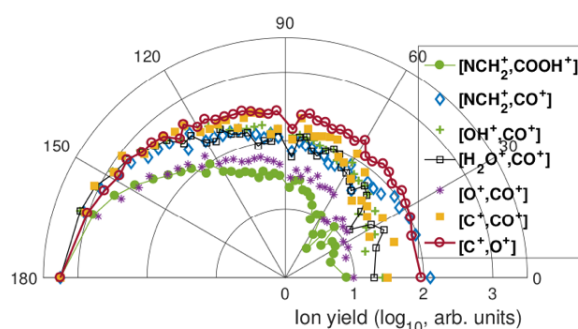

Figure 1. Dissociation angle distribution of co-incident fragment ions for different fragmentation channels, normalized to the highest values. The distributions are normalized to peak values at 180° [1].

Partially distinguishing the fragmentation pathways, we were able to associate the measured kinetic energy release with separate dissociation steps and describe the overall angular distribution of the correspondent fragment ions for a particular fragmentation pathway [1].

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Three-body dissociation of molecular ions: separating sequential and concerted pathways

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Synopsis   The three-body fragmentation of polyatomic molecules often proceeds via two competing pathways, either a concerted fragmentation or a sequential breakup. Since in both cases the final products are the same set of ions, it becomes difficult to disentangle events coming from the two. A novel method based on use of native frames allows for separation of such events and acts as a powerful tool which enables extraction of branching ratios as well as identification of electronic states of the molecular intermediate formed during sequential breakup.

One of the key questions concerning the three-body fragmentation of molecular ions is the distinction of sequential and concerted mechanisms, i.e., the stepwise or simultaneous cleavage of bonds. As the final set of three recoil fragments is the same for sequential and concerted breakup, separation between these two mechanisms becomes difficult. A novel method to enable clear separation of concerted and sequential breakup was demonstrated recently [1] using fragmentation of OCS^{3+} into C^+, O^+ and S^+. These three ionic fragments were detected in coincidence and their respective momenta were determined using the technique of recoil ion momentum spectroscopy. By analyzing the multi-coincident data in two different frames of reference (native frames [1]), events belonging exclusively to the sequential mode of breakup were extracted. This enabled us to identify the electronic states of the intermediate molecular ion which may contribute to the sequential breakup process. The method also allows us to determine the branching ratios for the sequential and concerted processes. The method of separation is applicable in general and does not depend on the mechanism used to generate the multiply charged molecules. We have applied this method to address three body dissociation of OCS molecule, induced by impact of highly charged ions [2]. The results of the experiment will be presented and the features of the native frames method will be discussed.
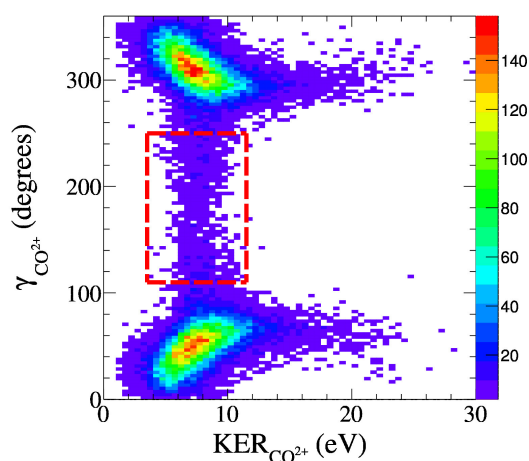

Figure 1. A distribution of all events originating from the dissociation of OCS^{3+} assuming that they all follow sequential breakup pathway with CO^{2+} as the intermediate molecular ion formed during the first step of the breakup. The events within the red rectangle come exclusively from the true sequential events.

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A quantum approach to calculate differential and total cross sections for electron impact ionization of biologically relevant molecules

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Synopsis   We present a theoretical approach to calculate the cross sections for the ionization of molecules by single electron impact. It is based on the First Born approximation (FBA), describing the ejected electron by a distorted wave (DW). The cross sections are calculated for an average molecular orientation with the proper average method. Single-center molecular wave functions are generated using Gaussian making this method easily applicable to any molecule. We show the calculated cross sections of some molecular targets of varying complexity.

We propose a quantum approach to calculate total and differential cross sections for the electron impact ionization of molecules. The developed approach was detailed in [1, 2] where it was used to calculate triply differential cross sections (TDCS) for the electron impact ionization of formic acid and tetrahydrofuran. By integrating the TDCS, we can easily retrieve the doubly differential, simply differential and total cross sections. Another interesting feature of this methodology is that it can be applied to any molecular target. It is particularly adapted for complex molecules for which theoretical cross section data are very scarce but much needed. The theoretical framework is based on the First Born Approximation (FBA) and the ejected electron is described by a distorted wave. The molecular wave functions are generated with a quantum chemistry program and are converted to single center wave functions thereby reducing the complexity of multi-atom systems. All wave functions are written in partial wave expansion making it possible to translate the theory to a parallel program hence reducing the computing time. As an example, we show in the figure below the TDCSs (solid line) for the ionization of phenol. They are in better agreement with the experimental data than the ones calculated with another theoretical approach making an approximation for orientation averaging: the average orientation molecular orbital approximation
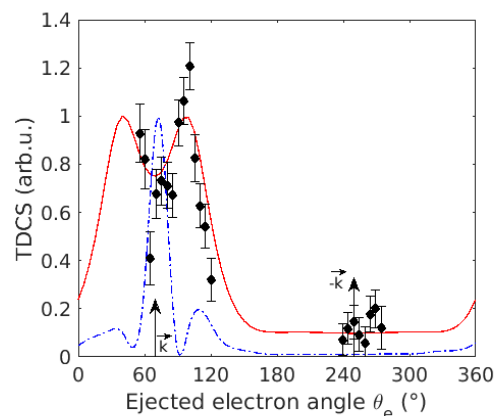
OAMO (dash-dotted line) [3].

Figure 1. Summed TDCSs for the electron impact ionization of the highest occupied and next highest occupied molecular orbitals of phenol calculated using the present model (solid line), compared to theoretical OAMO TDCSs (dash-dotted) and experimental data (solid circles) from [3].

Hence, we demonstrate that it is possible to calculate the cross sections for complex molecular targets at low computational cost without compromising the validity of the theory.

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Controlling Electron Spectrometers using LabVIEW and Arduino Interfaces

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Synopsis As experimental complexity increases, automated systems to control, optimize and handle data acquisition are becoming increasingly necessary in atomic physics. This presentation will detail how LabVIEW and Arduino compatible hardware is used in Manchester to operate & control experiments, thereby providing realtime optimization and live reporting of data.

The (e,2e) spectrometer that has been operating in Manchester since 1992 [1] has recently had the electronics and control systems modernised to measure electron impact-ionization from laser-excited and aligned targets [2]. This requires the laser system to be frequency locked to ~1 part in 10^{10} for long periods of time, while ensuring computer control of the spectrometer.

These new control systems are shown schematically in Fig. 1. They adopt a LabVIEW programme to communicate with a National Instruments Data Acquisition card (DAQcard) and several Arduino compatible microcontrollers [3]. The system drives the spectrometer detectors and optimizes their tuning, and monitors and logs all parameters such as electron beam current and vacuum pressure. By utilizing the DAQcard to acquire signals from an Ortec time to amplitude converter, the LabVIEW interface acts as an always-on multi-channel analyzer to also detect coincidence signals.

The power supplies for the electron gun and analyzers are controlled and monitored by optically isolated Arduinos. The LabVIEW programme communicates with these via error-checked USB serial-based communications. The supplies feature front panels which display the set voltages in real-time, along with digital rotary encoders for manual tuning.

The analyzers are positioned by Arduino controlled stepper motors via closed-loop feedback. LabVIEW communicates with the Arduinos via a similar USB serial interface as for the power supplies. When the LabVIEW programme changes angles, it runs a Nelder-Mead optimisation routine to accurately tune the electron optics, using the DAQcard to monitor the electron scattering rates.

The laser frequency is set by another LabVIEW programme that controls the laser as it monitors wavemeter readings. This communicates with the spectrometer programme across a local area network. The spectrometer programme sets and maintains the laser frequency and is notified of any errors, so that data collection is paused while the laser is re-locked.
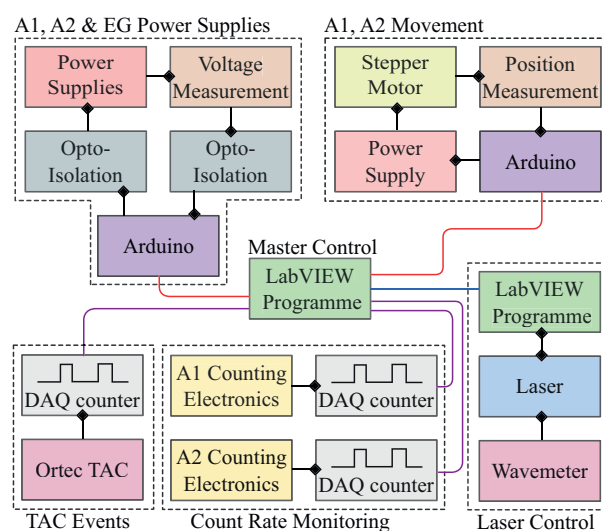

Figure 1. Simplified block diagram of the control and monitoring interface for the experiment. Key: EG = Electron Gun; A1,2 = Analyzers 1,2.

This system has been tested and validated by recent work on CH_4 as part of a joint collaboration [4], and the laser interface will be implemented soon.

Similar control systems are being adopted in all atomic physics experiments in Manchester, and these control systems will also be detailed in the presentation at the conference.

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Theoretical study of (e, 2e) processes for atoms and molecules by multi-center three distorted wave method

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Synopsis   We report a multi-center three distorted wave method to describe the electron impact single ionization processes for atoms and molecules in the perturbative framework. The continuum wave functions of the incident and two outgoing electrons are solved in the multicenter potential of neutral molecule and molecular ion, respectively. As an example, we present the calculations of triple differential cross sections for H_2O molecule by 81 eV electron impact. Good agreement is achieved between present calculations and experimental data.

Electron-impact ionization of atoms and molecules is one of the most basic phenomena in collision physics. The theoretical description of some nonperturbative methods, including exterior complex scaling (ECS) [1], convergent close coupling (CCC) [2], time dependent close coupling (TDCC) [3,4] and B-spline R-matrix (BSR) [5] approaches, have well described the single ionization cross section of some simple atoms and some simple diatomic molecules. As for the electron-impact single ionization of complex atoms and molecules, it remains a big challenge in the theoretical description. Due to the complex electronic structure and multicenter nature, the computational quantity is hardly tolerated for nonperturbative approaches.

Recently, a multicenter three distorted-wave (MCTDW) [6] method is developed to study the electron impact single ionization of molecules within the quantum mechanical perturbative framework. The continuum wave functions of the incident and two outgoing electrons are solved in the multicenter potential of neutral molecule and molecular ion, respectively. It is suitable for all kinds of kinematics for electron impact single ionization process theoretically.

In this talk, we will present some recent calculations of (e, 2e) triple differential cross sections (TDCSs) on atoms and molecules using MCTDW method. As an example, Figure 1 shows the summed TDCS distributions in three dimensional kinematics for summed $1b_1$ and $3a_1$ orbitals of H_2O at $\theta_s = -10°$ and $E_e = 10$ eV with

collision energy of 81 eV. Figure 1.(a) is the MCTDW result and Figure 1.(b) is the corresponding experimental data from the advanced reaction microscope [7]. The results show an overall agreement using one normalization factor compared with the experimental measurements [6].
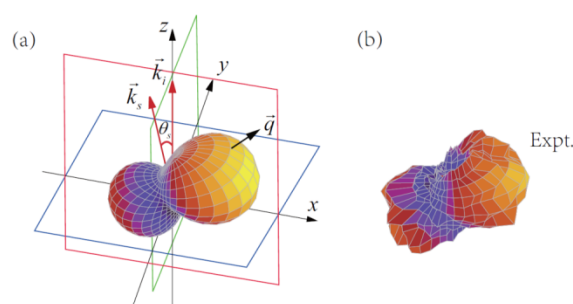

Figure 1. (a) 3D image of summed TDCSs for $1b_1$ and $3a_1$ orbitals of H_2O calculated by MCTDW at impact energy of 81 eV with $\theta_s = -10°$ and $E_e = 10$ eV; (b) the corresponding experimental data carried out by reaction microscope [7].

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Attoclock on atomic and molecular hydrogen

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Synopsis We simulate attosecond angular streaking measurements on atomic and molecular hydrogen by solving the time-dependent Schrödinger equation (TDSE) driven by a few-cycle close-to-circular laser pulse. We compare results for this simulated "numerical attoclock" with experiment and predictions of a semiclassical model based on the saddle point method (SPM). This way we identify the essential physics behind the attoclock measurements related to the concept of tunneling time. In H_2, we give an upper bound on the tunneling time not exceeding 10 attoseconds which is significantly less than prescribed by any definitions of tunneling time.

The attoclock measurements of the tunneling time have attracted very considerable attention [1]. These measurements and further experiments [2] could be interpreted in terms of a finite tunneling time whereas the theory [3, 4] points firmly to instantaneous tunneling. The attoclock technique is based on measuring an offset angle θ_A of the peak photoelectron momentum distribution (PMD) in the polarization plane of a close-to-circularly polarized few-cycle laser pulse. This distribution can be simulated by solving a time-dependent Schrödinger equation (TDSE)

$$i\partial\Psi(\boldsymbol{r})/\partial t = [\hat{H}_{\text{target}} + \boldsymbol{A}(t)\cdot\hat{\boldsymbol{p}}]\Psi(\boldsymbol{r})$$

driven by a laser pulse with a vector-potential

$$\boldsymbol{A}(t) = \frac{A_0}{\sqrt{\epsilon^2+1}}\cos^4(\omega t/2N+\phi)\left[\begin{array}{c}\cos(\omega t)\ \boldsymbol{e}_x \\ \epsilon\sin(\omega t)\ \boldsymbol{e}_y\end{array}\right]$$

where N and ϕ parameterize the number of oscillations and the carrier-envelope phase. Results of such calculations are illustrated on the top panel of Fig. 1 with $N = 2$. In the latest development, the attoclock offset angle was wholly attributed to the Coulomb field effect [5, 6] whereas in the absence of this field the attoclock offset angle $\theta_A = 0$ (bottom panel of Fig. 1). Based on this consideration, the hydrogen atom measurement returned the upper bounds on the tunneling time not exceeding 2 attoseconds [7]. The new set of attoclock measurements and calculations on H_2 was used to give an estimate of the tunneling time that does not exceed 10 attoseconds [8].
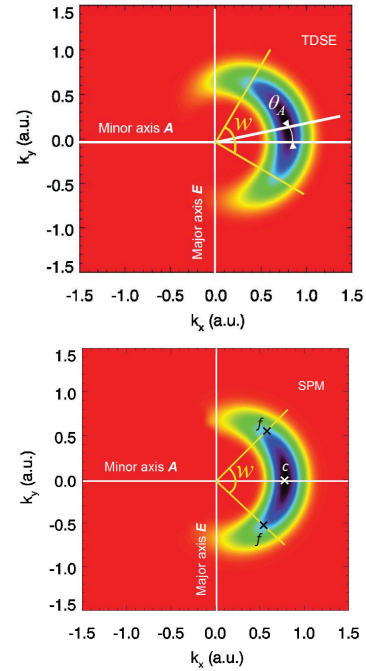

Figure 1. Top: atomic hydrogen PMD in the polarization plane from solution of the TDSE driven by a single oscillation laser pulse. Bottom: same distribution from a strong field calculation employing the saddle point method (SPM) and ignoring the Coulomb field of the nucleus.

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Attosecond-precision coherent control experiment at FERMI

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Synopsis When an electron is ejected from an atom after a photon is absorbed, an extremely short group delay between photon absorption and electron emission occurs in the photoelectron wave packet. It is of the order of a few attoseconds and is known as the Eisenbud-Wigner-Smith delay. Here we introduce a new approach for measuring the delay with coherently-controlled free-electron laser pulses.

The invention of probing methods on a time scale less than femtoseconds has led to the age of attosecond physics [1], and numerous such ultrafast phenomena currently are being investigated. Photoemission is one of those processes that has received a lot of interest, especially concerning the photoemission delay between photon absorption and photoelectron emission known as the Eisenbud-Wigner-Smith (EWS) delay [2].

We are presenting here a new method for measuring the EWS delay. In this method, we use short wavelength EUV light, consisting of phase-locked fundamental (ω) and second harmonic (2ω) pulses. It requires extremely accurate phase control (few attoseconds). Such fine control is available from the Italian free-electron laser (FEL), FERMI [3]. We report, as a demonstration of the new method, the EWS delay difference between one- and two-photon ionization of atomic Ne.

We carried out the measurement at the LDM beam-line, FERMI. The photon energies were set to 14 eV, 16 eV, and 19 eV for ω, which are below the Ne ionization threshold, while the second harmonics 2ω are above the threshold. Phase-locked bichromatic light beams crossed the atomic gas jets of He-Ne mixtures (We used the He for a calibration). A velocity map imaging spectrometer measured ejected electrons.

The target processes are the photoionization of Ne by one or two photon(s):

$$\text{Ne} + 2\omega \rightarrow \text{Ne}^+ + e^-,$$
$$\text{Ne} + \omega + \omega \rightarrow \text{Ne}^+ + e^- \text{ (non-resonant)}.$$
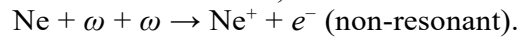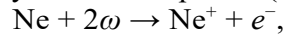
Because these electrons emitted by the two different pathways interfere with each other, the electron angular distribution is correlated to the difference of the unique phase shifts. The phase shift difference can be extracted by scanning the optical phase difference between ω and 2ω FEL pulses. The EWS delay τ is defined as the derivative of phase shift η with respect to the photoelectron kinetic energy E: $\tau = \hbar \cdot \partial\eta/\partial E$ [2]. We have measured the phase shift differences at several photon energies and found their slope to estimate the delay difference.

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Coherent control of Auger decay. Molecular Auger Interferometry

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Synopsis   We introduce a theory of interferometric measurement of a normal Auger decay lifetime in molecules. Molecular Auger interferometry is based on the coherent phase control of Auger dynamics in a two-colour $(\omega/2\omega)$ laser field. We show that, in contrast to atoms, in oriented molecules of certain point groups the relative $\omega/2\omega$ phase modulates the total ionisation yield. A simple analytical formula is derived for the extraction of the lifetimes of Auger-active states from a molecular Auger interferogram, circumventing the need in either high-resolution or attosecond spectroscopy.

We present Auger interferometry, i.e. a theory of coherent control [1] of Auger decay and single-photon laser-enabled Auger decay (spLEAD) [2, 3] in ionised molecules. We show analytically that for oriented molecules belonging to the molecular point groups C_s, C_n, C_{nv}, $C_{\infty v}$, C_{3h}, D_3, D_{3h}, and T_d, one can control not merely the angular distribution, but also the total yield of the Auger or spLEAD electrons or, equivalently, the yield of doubly ionised molecules. The theory is applied to CH_3F ion which has a $2s$-ionised Auger-active state (AAS) with energy only slightly above the double ionisation potential and, as a result, a decay width that is challenging to predict theoretically. We show that attosecond resolution can be gained without using attosecond pulses, by simply controlling the relative $\omega/2\omega$ phase.

Fig. 1 presents Auger interferogram showing the dependence of the population of the doubly ionised state of CH_3F (or the electron yield) as a function of the relative phase ϕ and the energy width Γ_1 of the singly ionised AAS of CH_3F. One can see that for the different decay widths, the depth of the population modulations $\mathcal{M}$ varies. For any value of Γ_1 a high contrast $\mathcal{M}$ can be obtained by tuning the field intensities. $\mathcal{M}$ can be measured experimentally within the relative phase scan of the photoionisation yield and used for extraction of Γ_1 value within a simple analytical expression

$$\Gamma_1 = \frac{1 - \sqrt{1 - \mathcal{M}^2}}{\mathcal{M}} \frac{d_{01}d_{1E}\mathcal{E}_1^2}{d_{0E}\mathcal{E}_2}.$$
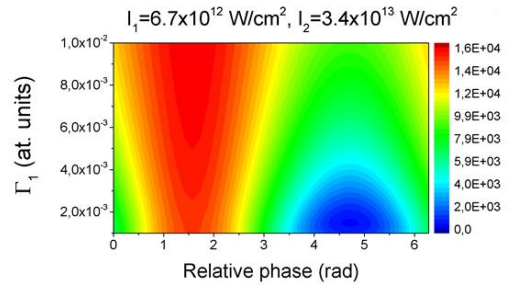

Figure 1. Auger interferograms. The total yield of electrons emitted by a bichromatic laser field for the case of CH_3F^+ as a function of the relative phase ϕ and the AAS width Γ_1 (y-axis).

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An all-optical atom trap as a target for MOTRIMS-like collision experiments

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Synopsis   Momentum-resolved scattering experiments with laser cooled targets have been performed with MOTRIMS (Magneto-Optical Trap Target Recoil Ion Momentum Spectroscopy) for the last two decades. However, the quadrupole magnetic field in a MOT impairs the electron momentum measurement limiting MOTRIMS to ion detection only. Here we report on the first realization of an all-optical lithium atom trap in a reaction microscope which does not require an inhomogeneous magnetic field. The temperature and density of the atom cloud were found to be approximately 2 mK and 10^9 atoms/cm^3 respectively, making it ideally suited for angular resolved photo-electron spectrosocpy and momentum-resolved electron-ion coincidence experiments.

Differential scattering experiments with laser-cooled atomic targets have been performed since almost two decades with MOTRIMS (Magneto-Optical Trap Recoil Ion Momentum Spectroscopy) setups. Compared to experiments with gas-jet targets, MOTRIMS features significantly lower target temperatures allowing for an excellent recoil ion momentum resolution. However, the coincident and momentum-resolved detection of electrons was long rendered impossible due to incompatible magnetic field requirements. A few years ago, the development of MOTReMi (Magneto-Optical Trap Reaction Microscope) [1] made it possible to achieve coincident electron-ion detection by pulsing the inhomogeneous magnetic field. Nevertheless, using this approach comes at the cost of measurement efficiency and a loss of target density.

Here we report on a novel all-optical trap (AOT) with an overall configuration which is very similar to conventional magneto-optical traps. It mainly requires small modifications of laser beam geometries and polarization which makes it easily implementable in other existing MOTRIMS experiments. In contrast to MOTs, the trapping of the atoms is not achieved with the position-dependent Zeeman shifts of the atomic levels, but rather with the optical dipole force along the intensity gradients in the bichromatic field of the cooling lasers [2]. The AOT does not only allow to prepare a lithium target cloud with similar temperaturtes and densities as a MOT, it also enables to prepare a fraction of the atoms (typ. 35%) in an excited and polarized p-state populating a specific magnetic sublevel.
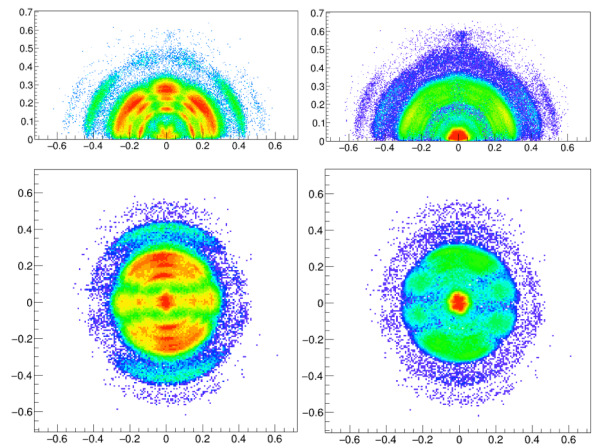

Figure 1. Electron momentum distributions after photionization of lithium from the ground s-state (left) and the excited p-state (right) in an short (7 fs), intense (10^{12} W/cm^2) laser pulse.

First experiments on single and multi-photon ionization of lithium have been performed (see figure 1) and an electron momentum resolution of only 0.01 a.u. has been achieved. The low electronic binding energies in the lithium target make electronic transitions optically accessible which e.g. for noble gas atoms can only be driven using synchrotron-light sources, free electron lasers, or high-harmic attosecond laser pulses. The high level of control of the atomic quantum states combined with the excellent momentum resolution enables to study excitation and ionization dynamics in great detail.

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X-ray spectroscopy of ultrafast-decaying core-excited ionic states in atoms

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Synopsis   Results from the first soft X-ray experiment at European XFEL on non-linear photon-matter interaction will be presented. Electron spectroscopy reveals insight into the character of resonances in highly transient, core ion-ized neon ions and their relaxation dynamics.

Highly intense X-ray pulses from X-ray free-electron lasers (XFELs) allow to study in the short wavelength regime non-linear, photo-induced processes in atoms and molecules. These can include sequential and non-sequential multiphoton ionization and excitation processes. Sequential ionization processes in the X-rays involve core-shells and occur in competition with ultrafast relaxation via Auger emission; when the X-ray intensity is high enough, this competition becomes effective and the population of transient highly excited ions exposed to the radiation becomes significant, so that it is possible to perform a spectroscopic characterization on it. By using the intense pulses from the SASE3 soft X-ray undulators at European XFEL, we could, for the first time, study the core-hole excitation of transient, highly excited atoms by means of resonant Auger electron spectroscopy employing the Small Quantum Systems (SQS) instrument.

Neon was ionized at the 1s shell and, depending on the photon energy, either sequentially ionized to Ne^{2+} $1s^02s^22p^6$ or excited to different Ne^{+*} $1s^02s^22p^6np$ states within the same fs X-ray pulse. Thanks to the extremely efficient tunability of the SASE3 undulator, it was possible to scan the photon energy across the $1s^12s^22p^6 \rightarrow 1s^02s^22p^6np$ resonances with photon energy steps of 1 eV (much smaller than the bandwidth). The resulting Auger electron spectra were recorded by electron time-of-flight spectrometers.

The results of these measurements are compared to previous studies of single-photon processes populating the same final states [2] and with theoretical results based on R-matrix calculations. The comparison of the experimental results with the theoretical model allow to address the relevance of shake-up processes in the resonant Auger decay pattern for the particular case of the double core hole excited $1s^02s^22p^6np$ ionic state in comparison to the neutral $1s^12s^22p^6np$.

These results are benchmarking the performance of the SQS endstation at European XFEL and demonstrate the novel possibilities which have been opened up in particular for investigations of non-linear phenomena and ultrafast decaying transient states in the X-ray domain.

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High-order above threshold dissociation of H_2^+ in strong laser fields

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Synopsis   The dissociation of H_2^+ in strong laser fields has been studied by numerically solving the time-dependent Schrödinger equation. Molecular dissociation via absorbing more than 5 photons is observed.

Electrons bound to atoms or molecules can simultaneously absorb multiple photons via the above-threshold ionization featured with discrete peaks in the photoelectron spectrum on account of the quantized nature of the light energy. Analogously, the above threshold dissociation of molecules is expected to occur. However, till now, no clear dissociative fragments of absorbing more than three photons has been reported. In the joint theory and experimental work, we studied the high-oder above threshold dissociation of H_2^+ in strong laser fields.

Numerically, we solved the time-dependent Schrödinger equation for dissociative ionization of H_2 (atomic units (a.u.) are used)

$$i\frac{\partial}{\partial t}\psi(R, x_1, x_2; t) = \left[\frac{P_R^2}{2\mu} + \frac{[p_1+A(t)]^2}{2} + \frac{[p_2+A(t)]^2}{2} + V(R, x_1, x_2)\right]\psi(R, x_1, x_2; t),$$

where $A(t)$ is the laser vector potential, and the Coulomb potential $V(R, x_1, x_2)$ is modified by two R-dependent soft-core parameters [1], and thus the ground-state energy curve of H_2, the ground state and the first excited state of H_2^+ agree with the real potential curves reasonably well. Experimentally, the measurements were performed in an ultrahigh vacuum reaction microscope setup of the cold target recoil ion momentum spectroscopy (COLTRIMS).

Our results[2] provide an unambiguous evidence that the electron and nuclei of a molecule as a whole absorb multiple photons, and thus above-threshold ionization and above-threshold
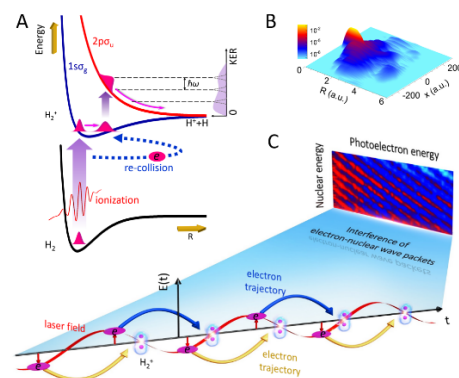

Figure 1. (*A*) The potential energy curves related to the dissociative ionization of H_2 molecules. The rescattering dissociation is sketched. (*B*) The snapshot of wave packet distribution in *R-x* space, where *R* is the internuclear distance of H_2^+ and *x* specifies the position of the ejected electron wave packet propagating along the polarization direction of the laser field. (*C*) The periodical rescattering-induced correlated electron-nuclear wave packets in every optical cycle interfere with each other, contributing to the coexistence of the ATI and ATD.

dissociation must appear simultaneously, which is the cornerstone of the nowadays strong-field molecular physics.

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Nonlinear XUV absorption spectroscopy of excited-state electron dynamics in atoms

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Synopsis Strong XUV fields are generated by the Free-Electron Laser in Hamburg (FLASH) and are employed to both excite and strongly couple resonant transitions in helium and neon atoms. The measurement of the XUV transient absorption spectrum as a function of the FEL intensity provides sensitivity to the nonlinear response of distinct electronic states. We experimentally observe an XUV-induced modification of the Fano profile of an autoionizing state in helium, as well as XUV-induced Stark shifts of bound-bound transitions in the Ne^{2+} ion.

The interaction of intense light with atoms pertains to understand how strong fields are used to measure and control the underlying electron quantum dynamics. For instance, employing weak attosecond pulses derived from high-order harmonic generation together with strong near-infrared (NIR) laser pulses, one can simultaneously excite and couple a manifold of doubly excited states in helium. Hence, a wave-packet of autoionizing Fano resonances can be observed by measuring the laser-dressed XUV absorption response [1]. Furthermore, fully resolving the spectral line shapes in this geometry, NIR-laser-induced and state-resolved phase shifts can be directly mapped onto a measurable change of the asymmetric Fano lineshape [2]. Applying a similar XUV-NIR pulse sequence to neon atoms, the laser-induced coupling of several inner-valence transitions is accessed [3].

While previous experiments have mainly focused on strong-NIR-field-controlled absorption spectroscopy, we will present new results of direct XUV nonlinear effects in both helium and neon atoms. Hereby, intense XUV pulses from the Free-Electron Laser (FEL) in Hamburg (FLASH) are tuned to photon energies of 60 eV and 50 eV, respectively, for the helium and neon absorption targets. The pulse energy ranges up to multiple ten μJ with a photon fluence of several J/cm^2 in the interaction volume. Depending on the stochastic substructure of the FEL pulses, intensities up to 10^{15} W/cm^2 can be reached with these parameters.

With increasing FEL intensity, we observe a significant change of the asymmetric Fano lineshape of the 2s2p resonance in helium to a more symmetric line at 60 eV photon energy. With a quantitative model of this fundamental double excitation in strong fields [4], we show how transient strong-XUV-field-induced energy shifts are directly encoded into the shape of the 2s2p absorption profile [5].

For the case of neon, the FEL photon fluence is sufficiently high to transiently generate a significant abundance of Ne^{2+} ions within the target volume through the absorption of two photons at 50 eV. The Ne^{2+} ions are identified via ^3P–^3D 2p–3d spin-orbit multiplet transitions within the measured FEL absorption spectrum. Depending on the FEL intensity, we observe spectral shifts of these lines due to the ac-Stark effect, which marks, to the best of our knowledge, the first observation of such strong-field effect at this XUV photon energy [6].

New insights on strong-XUV-field light-matter interaction can thus be gained by resolving the resonant line profiles of distinct electronic transitions as a function of the intensity through nonlinear XUV absorption spectroscopy with FELs.

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ABSTRACTS

OF POSTERS

Fitting continuum wavefunctions with complex Gaussians : Computation of ionization cross sections

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Synopsis Real Gaussians are widely used in quantum chemistry calculations to represent bound states. In this work we use complex Gaussians to represent continuum wave functions in order to facilitate the computation of physical quantities such as single ionization cross sections of atoms and molecules.

Gaussians are favorable in quantum chemistry and molecular physics due to some mathematical properties which facilitate the calculation of integrals, especially the Gaussian product theorem.

Atomic and molecular one particle bound functions can be represented by a set of real Gaussians [1] even if the latter are formally inadequate to describe the cusps and the asymptotic behavior. Representing scattering states with Gaussians is much more delicate because of their oscillating nature. Some studies have shown, though, that it is feasible [2] but with a large number of nodeless real Gaussians. On the other hand, complex Gaussians -that is to say Gaussians with complex exponents- have intrinsically an oscillating behavior that renders them more suitable to represent continuum wave functions. In view of scattering applications, this is the lead we wish to follow.

Within a first Born approximation and for a one active electron, computing ionization cross sections involves the calculation of transition matrix elements $\langle \psi_{\mathbf{k}}(\mathbf{r})|\widehat{O}|\psi_i(\mathbf{r})\rangle$, where $\psi_i(\mathbf{r})$ and $\psi_{\mathbf{k}}(\mathbf{r})$ represent, respectively, the initial (bound) and the final (continuum) wave functions. $\widehat{O}$ is the transition operator that connects these two states : $e^{i\mathbf{q}\cdot\mathbf{r}}$ for the case of particle impact ($\mathbf{q}$ is the momentum transfer vector) and $-\hat{\epsilon} \cdot \mathbf{r}$ for photo-ionization in length gauge ($\hat{\epsilon}$ is the polarization vector). If the radial parts of both $\psi_{\mathbf{k}}$ and ψ_i are expanded in Gaussians, then the calculation of the transition matrix elements becomes analytical for both ionization by particle impact [3] and photo-ionization. As a test case to validate our approach we considered the hydrogen atom. Figure 1 shows the photo-ionization cross section versus the photo-electron energy, from its ground state.

The ultimate goal is to apply this Gaussian approach to molecules, using both monocenter and multicenter expansions.
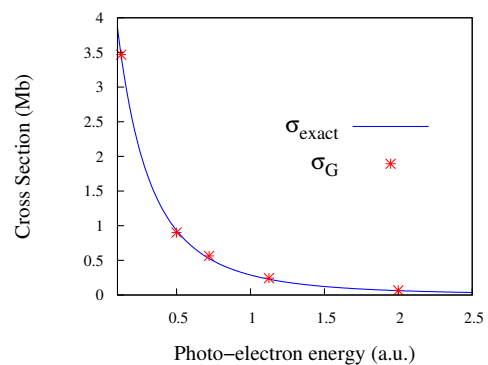

Figure 1. The photo-ionization cross section versus the photo-electron energy, from the ground state $1s$ of H atom. σ_{exact} is the exact theoretical cross section [4] and σ_G is that computed with our complex Gaussians approach.

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PCI effects in multi-step Auger transitions following Kr 1s and 2$p_{3/2}$ photoionizations

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Synopsis   The Kr ($L_3M_{4,5}M_{4,5}$) Auger electron spectra were measured with the photon energy set slightly above the 1s and 2$p_{3/2}$ thresholds. The energy shift and broadening due to the Post-Collision Interaction (PCI) effect were observed in the spectra. Due to the time delay with the KL fluorescence emission process, a tailing toward the lower energy side, which is not seen in the PCI structure of single step Auger decay, appears in the spectrum of 1s photoionization. We propose a time-dependent theory of PCI for these multi-step processes with which we obtain good agreement with experimental results.

Post-Collision Interactions (PCI) upon inner-shell photoionization can occur due to the interactions between the photoelectron and the subsequently emitted Auger electron [1]. In the classical mechanical framework, PCI has been explained as due to the change in the inner ion charge which changes the coulomb potential perceived by the Auger electron, upon the take-over of the slow photoelectron by the faster Auger electron. In the ordinary one-step PCI, the Auger electron tails and shifts towards higher energies. In this work, we studied the case of Kr 1s photoionization which leads to PCI involving multi-step Auger decay, leading to the interplay with multiple Auger electron emissions in sequence as well as the effect of fluorescent decay. These are compared to the simpler case of 2$p_{3/2}$ photoionization. For the first step after 1s photoionization, KL fluorescence emission is stronger than the KLL Auger process [2]. The fluorescence process leads to time delay in the subsequent LMM Auger electron emission and thus affects the PCI effect. In this study the $L_3M_{4,5}M_{4,5}$ Auger spectrum measured close the the Kr 1s threshold and also close to the

Kr 2$p_{3/2}$ threshold were compared to elucidate the effect of KL fluorescence on PCI effects.

The experiment was conducted at the BL19LXU and BL17SU beamlines at the SPring-8 synchrotron radiation facility. The BL19LXU and BL17SU beamlines were utilized for Kr 1s threshold and Kr 2$p_{3/2}$ threshold measurments respectively. The electron energies were measured by the Scienta SES2002 hemispherical electron analyzer.

Close to the Kr 1s photoionization edge, the $L_3M_{4,5}M_{4,5}$ Auger electron peak was found to exhibit a considerably pronounced and longer leading edge tailing towards lower energy compared to its shape at the Kr 2$p_{3/2}$ photoionization edge. This can be explained as the effect of KL fluorescence which causes the delay of LMM Auger emission. Both spectra were found to agree well with our time-dependent PCI theory [3] which takes into accout the time delay due to fluorescence lifetime.

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On the effect of cascade transitions after electron-impact excitation of Zn by spin-polarized electrons

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Synopsis   We investigate the possible effect of cascade transitions from the $(4s5p)^3P_{0,1,2}$ states to the $(4s5s)^3S_1$ state of Zn. Our calculations suggest that the excitation cross sections and the polarization of the cascade radiation cannot explain the serious controversy between two existing experimental datasets below the cascade threshold and raise additional questions regarding the measurements at higher energies.

In a recent paper, Clayburn and Gay [1] reported their measurements of the angle-integrated linear polarization P_2 in the $(4s5s)^3S_1 \rightarrow (4s4p)^3P_0$ transition in Zn after impact excitation by a transversally spin-polarized electron beam. With the light detector placed at right angle to the incident beam direction along the direction of the electron polarization P_e, P_1 and P_2 are the linear polarizations for $(0°, 90°)$ and $(45°, 135°)$ transmission, respectively, while P_3 is the circular polarization. P_1 is independent of P_e, while P_2 and P_3 are proportional to P_e.

Clayburn and Gay noted significant disagreement between their $P_2/P_e \approx 0$ data and the $P_2/P_e \approx -10\%$ measurements of Pravica *et al.* [2] in the cascade-free region of incident electron energies below ≈ 7.6 eV, while relatively good agreement was found above that threshold. According to Bartschat and Blum [3], one would expect $P_1 \approx 0$, $P_2/P_e \approx 0$, and $P_3/P_e \approx -1$ in the cascade-free region. Above the cascade threshold, significant deviations are only possible for sufficiently large excitation cross sections and strong polarization of the cascade radiation.

We performed fully relativistic convergent close-coupling (RCCC-94) and semi-relativistic B-spline R-matrix (BP-BSR-43) calculations with 94 and 43 states, respectively. As seen in Fig. 1, accounting for cascades indeed reduces the magnitude of the circular polarization, in agreement with the measurements. However, we find $P_2/P_e \approx 0$ at all incident energies considered. Our results agree with general theory [3] but are in strong contradiction with both experimental datasets above the cascade threshold.
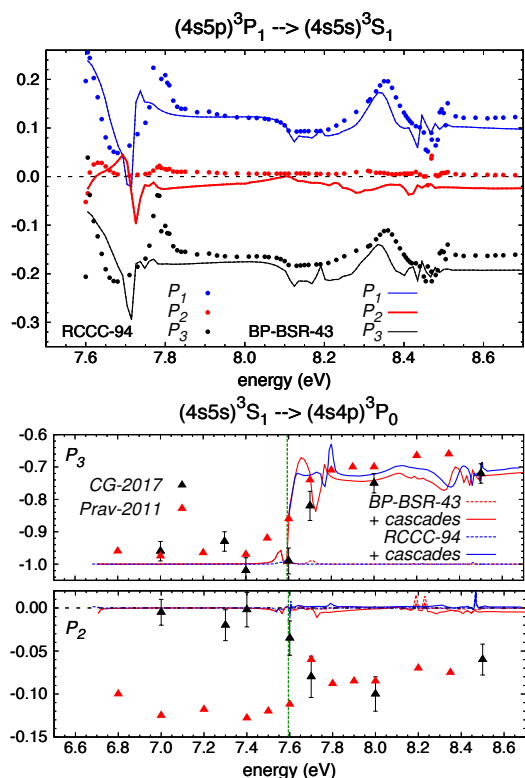

Figure 1. P_1, P_2/P_e, and P_3/P_e for the $(4s5p)^3P_1 \rightarrow (4s5s)^3S_1$ cascade transition (top), as well as P_3 (center) and P_2 (bottom) for $(4s5s)^3S_1 \rightarrow (4s4p)^3P_0$ in Zn. CG-2017 [1]; Prav-2011 [2].

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Frustrated tunnel ionization of argon by intense few-cycle infrared laser radiation

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Synopsis   We report results for strong-field ionization and excitation of argon obtained by solving the time-dependent Schrödinger equation in a single-active-electron (SAE) model and the multi-electron R-matrix with time dependence (RMT) method. We analyze the process of "frustrated tunnel ionization" and its dependence on the potential and a variety of laser parameters, such as the intensity, carrier-envelope phase, and ellipticity. Our calculations support current experimental work at Griffith University.

We carried out calculations for strong-field ionization and excitation of argon by solving the time-dependent Schrödinger equation for the initially bound 3p electron. Our focus is the process of "frustrated tunnel ionization" [1, 2], where the field-ionized electron is driven back towards the nucleus by the changing laser field and ultimately recaptured into an excited bound state without the possibility of escaping again, due to the already weakened few-cycle pulse. This process is currently being investigated experimentally at Griffith University [3]. We discuss the effect of different potentials, the carrier-envelope phase (CEP), the laser intensity, and the ellipticity of the radiation. We also investigate how well our SAE models represent the essential physics by comparing with predictions from the multi-electron RMT approach [5].

Figure 1 shows the probabilities for survival of the Ar($3p^6$) ground state (P_{surv}) as well as for excitation of any Rydberg state (P_{exc}) and ionization (P_{ion}). The bottom part of the figure shows the probabilities normalized to their respective maximum. The results shown were obtained with the static potential suggested by Green *et al.* [4], which was supplemented by a semi-empirical local potential to account for the electric dipole polarizability of the residual core.

Our calculations suggest a particularly strong CEP dependence of the probability for frustrated ionization, while the survival and ionization probabilities remain relatively constant when the CEP is varied. For the parameters chosen here, about 10% of the initially freed electrons are recaptured into bound states.
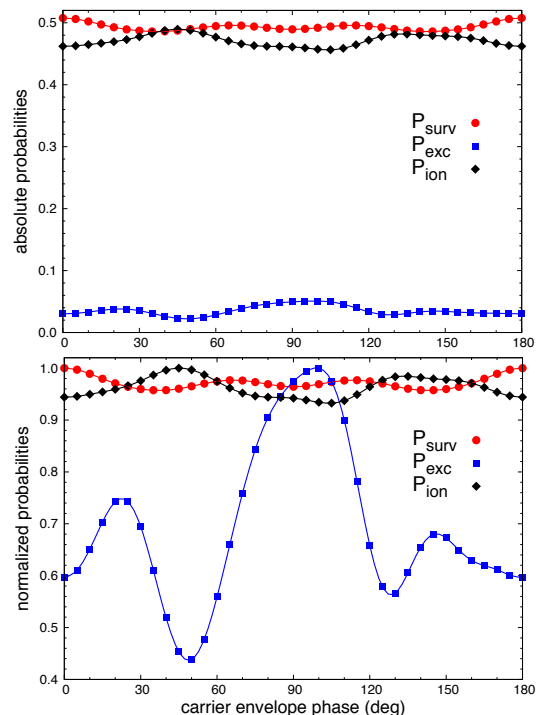

Figure 1. Absolute (top) and normalized (bottom) probabilities for survival of the ground state, excitation, and ionization of Ar(3p) as a function of the CEP for a 6 fs (FWHM) 800 nm laser pulse of peak intensity $4.0 \times 10^{14}\,\text{W/cm}^2$. See text for details.

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Study of interferences in the photoionization of water molecules by linearly polarized radiation

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Synopsis   We study theoretically interference effects in the photoionization of water molecules by linearly polarized radiation. We employ Coulomb continuum wavefunctions for the final state and linear combinations of Slater type orbitals located on the centers of the molecule for the bound states. We calculate total and differential cross sections where we put in evidence three-center interference effects coming from the coherent emission from each center. We verify that these effects may produce a partial suppression of the emission in the classical direction given by the polarization vector.

We analyze the interference effects in the photoionization of water molecules by monochromatic and linearly polarized radiation [1]. We represent the final state wavefunctions by using Coulomb continuum wavefunctions [2]. The water molecule bound states are given by linear combinations of Slater type orbitals located on the centers of the molecule [1, 3]. We calculate total and differential cross sections and we contrast them with more elaborated theoretical results and available experimental data. In particular, for total cross sections, we obtain a very good agreement at enough high energies where there is a lack of predictions [1, 2]. We put in evidence three-center interference effects in the total and differential cross sections coming from the coherent emission from each center of the molecule [1]. As an example of interference effects in the total cross sections, we show in figure 1 the interference factor as a function of the photon energy for the $3a_1$ water orbital. This factor is obtained as the ratio between the molecular total cross sections, where we take into account the multicenter nature of the molecule, and the effective monocentric total cross sections [1]. Interference effects are difficult to detect in total cross sections as they decrease quickly with the photon energy [4]. The interference factor exposes them through deviations from unity, i.e., indicating constructive or destructive interferences produced by the emission of the ionized electron from the different centers of the molecule. On the other hand, we find that the interference effects may be directly detected in the angular distributions of photoelectrons corresponding to randomly oriented molecules. Specifically, we observe that the interference effects under certain conditions may produce a partial suppression of the emission of photoelectrons in the classical direction given by the polarization vector [1]. Results for angular distributions will be presented during the conference.
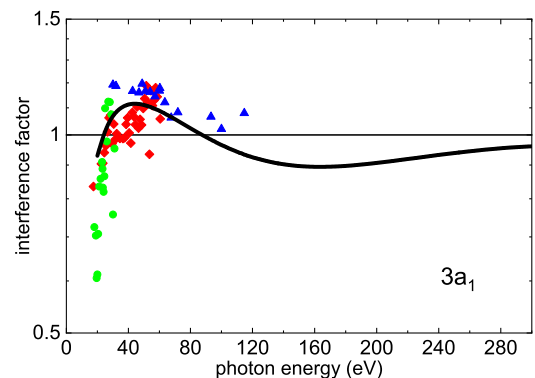

Figure 1. Interference factor as a function of the photon energy for photoionization of the $3a_1$ water orbital (━━━) [1]. Ratio between the experimental data and the effective monocentric cross sections for [5] (◆), [6] (●) and [7] (▲).

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Angular correlation in two-photon single ionization of helium

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Synopsis We theoretically explore the contribution of radial and angular electron correlation terms in the two-photon single ionization of helium, by a full dimensional numerical solution of the time dependent Schrodinger equation, over a wide photon energy range.

We perform *ab initio* simulations to extract angularly-resolved two-photon single ionization yields of helium subject to attosecond XUV pulses, by numerically solving the full dimension time-dependent Schrodinger equation [1]. We extract the photoelectron angular distributions (PADs) for two-photon single ionization, which reveal the different mechanisms that dominates specific photon energy ranges, depending on the laser parameters employed. We first verify that a single active electron picture is only a qualitatively valid approach for low photon energies [2], but angular electron correlation plays a detectable role in the above-threshold region. However, for photon energies close to or above the excitation energy of He^+, the process can be rationalized as a sequential and uncorrelated ionization-excitation process.

Very distinct features appear at those photon energies where the lowest series of doubly excited states are populated upon two-photon absorption. In Fig. 1, we show the asymmetry parameters $\beta_{2,4}$ (top panel) characterizing the two-photon single ionization angular distributions of He, together with the total and partial ($^1S^e$ and $^1D^e$) cross sections (middle panel). Subplots (a) to (d) show the evolution of the PADs along the typical Fano profile, appearing in the ionization cross section, of the lowest $^1S^e$ doubly excited state, where the phase of the amplitude associated to a given channel undergoes a π shift, resulting in a rapid evolution of the angular distributions. We discuss the origin of an almost pure $s-$wave behavior in panel (b), where the photoelectron is ejected independently of the

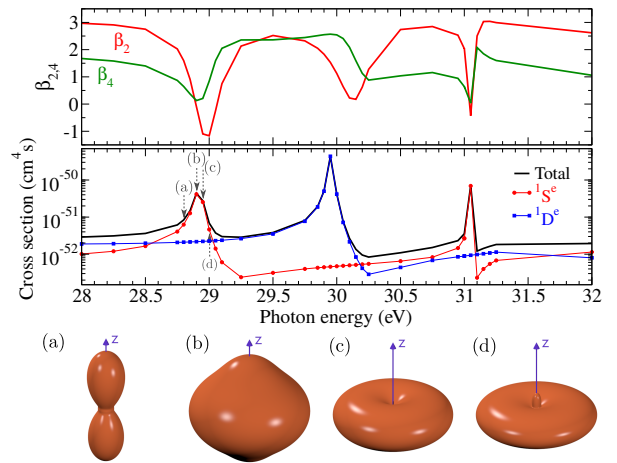electromagnetic-field direction, i.e. losing memory of the initial light-induced transition.

Figure 1. Asymmetry parameters and cross sections around the first series of autoionizing states.

At slight larger energies, the electron ejection along the light polarization direction is almost fully suppressed, resulting into an unexpectedly large electron emission in the plane perpendicular to the light polarization [see subplots (c) and (d)]. This strong variation of the PADs with photon energy, in the autoionizing region, is mostly independent of the pulse parameters, i.e. it is the result of the correlation in the final state. In contrast, the PADs at lower photon energies, in resonance with one-photon excitations, are significantly dependent on the pulse duration employed [2].

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Deep core photoionization of iodine in CH₃I and CF₃I molecules: how deep down does the chemical shift reach?

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Synopsis   This study reports for the first time a chemical shift in a very deep core level. The experimental observations have been supported by a combination of molecular and atomic calculations taking into account different relativistic effects corrections.

The availability of hard X-ray synchrotron radiation has opened up new avenues in gas-phase molecular photoelectron spectroscopy. The possibility to create a very deep core hole via single photon excitation or ionization enables studies of multitude of interesting phenomena like ultrafast electronic and molecular relaxation dynamics.

The present work aims to answer to what extend the created deep core-hole feels its molecular environment. Is there still an observable chemical shift in the binding energy of iodine 1s and 2s levels in two different molecules, CH3I and CF3I? Indeed, a chemical shift is observed, indicating that deep core levels are not purely atomic in nature[1], thus widespread concepts such as electronegativity and charge distribution inside a molecule extend down to very deep levels.

The subsequent Auger spectra are also studied. It is observed that CH₃I and CF₃I have virtually identical Auger spectra and the overall spectral features and their relative intensities are close to the recently reported corresponding Auger spectrum in Xe[2]. High-level theoretical simulations including quantum electrodynamics (QED) effects have been performed for neutral iodine atom and Xe-like iodine negative ion, and it is found that the experimental spectrum lies in-between these two extremes. Using Z+1 and Z+2 approximations, a charge distribution analysis was carried out, and it was observed that the slightly positive iodine in neutral molecule gains more negative charge (withdraws charge from the rest of the molecules) as a function of the excess positive charge in the core.
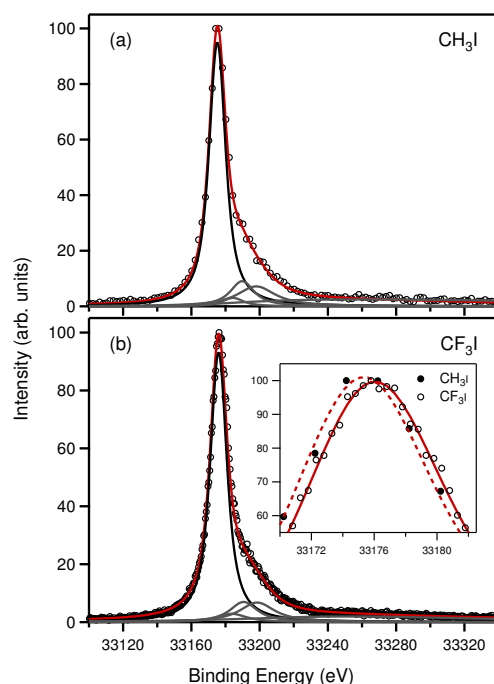

Figure 1. Experimental iodine 1s photoelectron spectrum of CH₃I (a) and CF₃I (b), measured at photon energy of 35.5 KeV.

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Two-body fragmentation dynamics of $C_2H_2^{2+}$ induced by electron impact at 1 keV

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Synopsis   Two-body fragmentation of $C_2H_2^{2+}$ induced by electron impact is investigated. Channel $H^+ + C_2H^+$ and channel $H_2^+ + C_2^+$ are clearly identified, while channel $CH^+ + CH^+$ and channel $C^+ + CH_2^+$ cannot be separated because of the mixture of the events. Taking the advantage that the kinetic energy release (KER) distribution is independent on the detection angle, we are able to disentangle the events of the latter two channels from the mixture. KER distributions and branching ratios for these four channels are deduced.

Acetylene (C_2H_2) is one of the simplest stable hydrocarbons widely existed in planetary atmospheres and interstellar medium. The dication $C_2H_2^{2+}$ has been extensively studied both theoretically and experimentally as a prototype for superfast deprotonation [1], molecular isomerization [2] and H_2^+ formation [3].

In the present work, the experiment was performed utilizing a newly built ion momentum spectrometer. The precursor ion $C_2H_2^{2+}$ is created by the collisions of C_2H_2 molecule with 1 keV electrons. The measured ion-ion coincidence time of flight (TOF) spectrum is shown in Figure 1(a). Channel $H^+ + C_2H^+$ and channel $H_2^+ + C_2^+$ can easily be observed, while channel $CH^+ + CH^+$ and channel $C^+ + CH_2^+$ are too close with each other and mixed on TOF coincidence map, which cannot to be separated.

Figure 1(b) and (c) exhibit KER distributions for channel $H^+ + C_2H^+$ and channel $H_2^+ + C_2^+$. Taking the advantage that the KER distribution is independent on the detection angle, we are able to disentangle the events of channel $C^+ + CH_2^+$ and channel $CH^+ + CH^+$ from the mixture. The details will be presented on the poster. Figure 1(d) and (e) show the thus obtained KER distributions for channel $CH^+ + CH^+$ and channel $C^+ + CH_2^+$. This method also enables us to deduce relative branching ratios for these four channels, which are of 66.7% for $H^+ + C_2H^+$, 0.03% for $H_2^+ + C_2^+$, 29.9% for $CH^+ + CH^+$ and 3.67% for $C^+ + CH_2^+$. The KER distributions and branching ratios obtained in present work agree well with those in the literature.
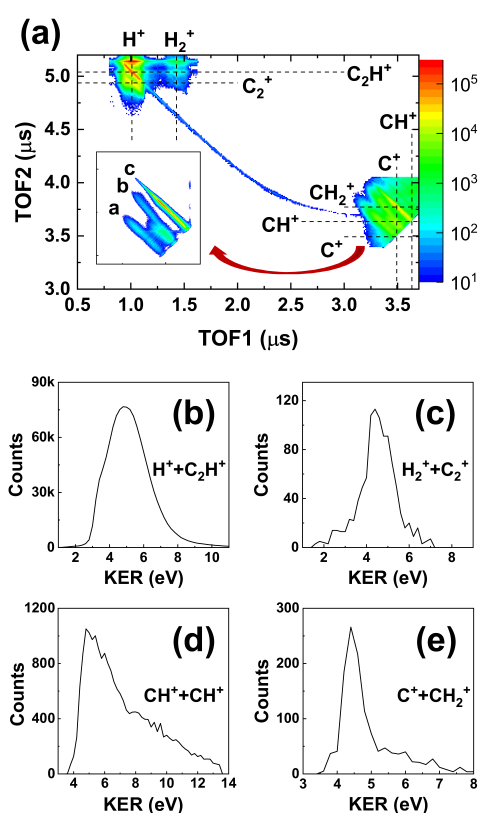

Figure 1. (a) Ion-ion coincidence TOF spectrum. The inset is the enlarged image of C-C bond breaking channels. (b) and (c) are KER distributions for channel $H^+ + C_2H^+$ and channel $H_2^+ + C_2^+$. (d) and (e) are KER distributions for channel $CH^+ + CH^+$ and channel $C^+ + CH_2^+$. (see text for details)

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Three center two electron correlated wave functions: Application to the ground and excited states of equilateral H$_3^+$

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The ground $^1A_1'$ and the first five excited states $^3E'$, $^1E'$, $^3A_1'$, $^1A_2'$, $^3A_2'$ of the equilateral triangular H$_3^+$ molecule are determined by an original three center wave functions constructed by the use of the irreducible representations of the D$_{3h}$ point group. The Hamiltonian which describes the ion H$_3^+$ under the assumption that the protons are infinitely massive and located at the vertices of an equilateral triangle of side ρ is written as follows:

$$\mathcal{H} = -\frac{1}{2}\sum_{i=1}^{2}\Delta_{\mathbf{r}_i} - \sum_{j=1}^{2}\sum_{k=a,b,c}\frac{1}{r_{jk}} + \frac{1}{r_{12}} + \frac{3}{\rho}, \quad (1)$$

where

$$\mathbf{r}_{jk} = \mathbf{r}_j - \mathbf{k}, \; \mathbf{r}_{12} = \mathbf{r}_1 - \mathbf{r}_2, \; j=1,2, \quad (2)$$

and $\mathbf{k} = \mathbf{a}$, $\mathbf{b}$, $\mathbf{c}$ are the positions of the three protons. We seek the wave functions using the extended basis set with 14 parameters [1], [2]

$$\Psi_Q \equiv \Psi_Q(\mathbf{r}_1,\mathbf{r}_2) = (1 + P_{12})$$
$$\times\Big(\sum_{\text{perm}\{a,b,c\}} c_i \exp(-\alpha_1 r_{1a} - \alpha_2 r_{1b} - \alpha_3 r_{1c})$$
$$\times \exp(-\alpha_4 r_{2a} - \alpha_5 r_{2b} - \alpha_6 r_{2c} - \alpha_7 r_{12}) \quad (3)$$
$$+ \sum_{\text{perm}\{a,b,c\}} d_i \exp(-\alpha_8 r_{1a} - \alpha_9 r_{1b} - \alpha_{10} r_{1c})$$
$$\times \exp(-\alpha_{11} r_{2a} - \alpha_{12} r_{2b} - \alpha_{13} r_{2c} - \alpha_{14} r_{12})\Big).$$

The computational schemes, which will deliver the wave functions and the energy values of the desired levels are based on the Rayleigh-Ritz variational functional with the electronic energy given by

$$\varepsilon_Q = \langle\Psi_Q|\mathcal{H}|\Psi_Q\rangle/\langle\Psi_Q|\Psi_Q\rangle, \quad (4)$$

where Q represents the energy levels under consideration. To minimize the energy ε_Q for each levels under consideration by parameters α_i, we have used a sequential quadratic programming method for several variables. Also we needed to calculate first derivatives by the parameters of the energy with respect to the Hellmann-Feynman theorem

$$\partial_{\alpha_i}\varepsilon_Q = 2\langle\partial_{\alpha_i}\Psi_Q|\mathcal{H} - \varepsilon_Q|\Psi_Q\rangle/\langle\Psi_Q|\Psi_Q\rangle,$$

All matrix elements – 6D integrals which appear in the functional of energy were calculated numerically using a globally adaptive subdivision scheme. All numerical integrations were done with a absolute accuracy $10^{-6} - 10^{-7}$.

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Table 1. The ground and excited energies of H$_3^+$ at the equilibrium internuclear distance $\rho = 1.65$ au of ground state. [a] From [4] at $\rho = 1.63332$ au.

	$^1A_1'$	$^3E'$	$^1E'$	$^3A_1'$	$^1A_2'$	$^3A_2'$
	-1.342 520	-0.792 082	-0.626 728	-0.510 758	-0.209 164	-0.035 843
[3]	-1.343 835		-0.633 512	-0.511 569		
[4]		-0.776 695[a]	-0.622 277	-0.496 986[a]		

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Magnetoelectric response of quantum systems driven by optical vector beams

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Synopsis   We explore the interaction of quantum matter with optical vector beams. Such beams have a spatially inhomogeneous polarization state. Of special interest are cylindrical vector beams that may have a radial or azimuthal polarizations. It is shown that radially polarized beams induce a radially breathing charge-density oscillations and in atoms they trigger tunable quadrupole transitions. The interaction with azimuthally polarized beams triggers in the system a dynamic Aharonov–Bohm effect.

Spatiotemporally modulated electromagnetic fields (EM) are currently in the focus of theoretical and experimental studies and are promising for a wide range of practical applications [1]. One example of such EM fields are the orbital angular momentum (OAM) carrying vortex beams, which possess an inhomogeneous azimuthal phase distribution and a homogeneous polarization [2, 3]. Another example are the vector beams (VB), which are characterized by inhomogeneous phase and polarization in the plane perpendicular to the propagation of the field. VBs interact with quantum matter in a fundamentally different manner. In this contribution we communicate our findings on the characteristics of the dynamics triggered by vector beams in on low-dimensional quantum systems (nanorings) and atoms.

Irradiating with cylindrical VBs with radial (RVB) or azimuthal (AVB) polarizations a spin-active quantum ring we observe radial charge oscillations via electric transitions [4] when RVB are used, while for AVBs an oscillating magnetic moment is induced via a magnetic interaction. The latter is interpreted as a dynamic Aharonov–Bohm effect [4].

In the second part of this contribution, we study the interaction of VBs with Ca^+ atoms. When RVBs are considered, we show that such interactions induce angular-momentum-conserving quadrupole transitions which can be manipulated in magnitude by rotating the field relative to the quantization axis set by an external static magnetic field [4]. When a AVBs are used, different magnetic sublevels in the final state can be selected by rotating the laser field relative to the quantization axis of the target [4].

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Triple Differential Cross Sections for electron impact ionization of methane at intermediate energy

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Synopsis Triple differential cross sections are presented for electron-impact single ionization of the $1t_2$ and $2a_1$ states of CH_4 at an incident electron energy of 250 eV. Experimental data from two different laboratories are compared with molecular 3-body distorted wave and generalized Sturmian function calculations. A splitting in the binary lobe is observed for the $1t_2$ state, at some kinematic conditions. This work is a joint experimental and theoretical collaboration.

We present triple differential cross sections (TDCS) for 250 eV electron-impact single ionization of the HOMO $1t_2$ and NHOMO $2a_1$ states of CH_4, for several different projectile scattering angles and for ejected electron energies of either 30 eV or 50 eV. Measurements were made for coplanar asymmetric scattering.

This work is product of a joint theoretical-experimental effort. We compare measurements taken at Manchester, UK and Afyon, Turkey with theoretical 3-body distorted wave (M3DW) and generalized Sturmian function (GSF) calculations.

For 50 eV data were collected for five different projectile scattering angles. An example, at 25°, is shown in Fig. 1. For each theory, results calculated at the centroid angle and convoluted over the experimental uncertainty are presented. The agreement between the two measurements is very good, and the overall agreement between the two theoretical calculations and the experiment data is also very good.

For 30 eV ejected electrons, the agreement between experiment and theory is reasonable, but not as satisfactory as found for the 50 eV case. Overall the M3DW results are in a little better shape agreement with experiment than the GSF calculations.

For ionization of the $2a_1$ state a single maximum is observed in the binary region while for the $1t_2$ state this peak is sometimes doubled with a minimum close to the momentum transfer di-rection. The evolution of the single or double binary peak with varying scattering angle will be explored in detail using both M3DW and GSF calculations, and conclusions drawn as to the origins of rapidly changing features. More details are given in Ref. [1] and in references therein.
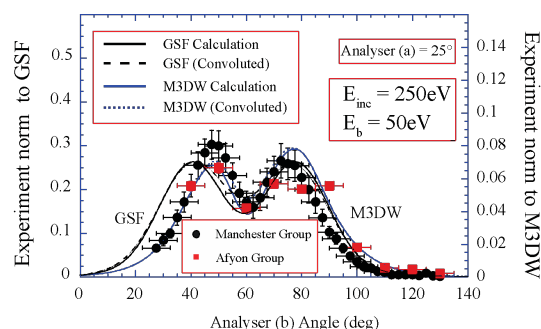

Figure 1. Cross section data from the Manchester (black circles) and Afyon (red squares) groups, compared to the GSF and M3DW theories for ionization from the $1t_2$ orbital of CH_4. The M3DW and GSF calculations yield TDCS with a factor of ~ 4 difference in magnitude, so that two vertical scales are used to represent them. The experimental data are normalized to theory.

The full set of measurements for both the HOMO and NHOMO states will be presented, together with details of the theoretical models and of the experiments.

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Two-center electron-impact ionization via collisional excitation-autoionization

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Synopsis   Electron-impact ionization of an atomic center in the vicinity of another atom is theoretically studied. First, collisional excitation of the neighbouring atom occurs by high-energy electron impact. Then, the other atom (or ion) receives the excitation energy via a radiationless two-center Auger process and is ionized. We show that the energy-differential cross section of electron-impact ionization is strongly influenced by the neighbouring atom and exhibits a pronounced peak on resonance. The process is also examined concerning changes to the angular distribution of ejected electrons.

Electron-impact ionization of atoms or ions represents one of the basic processes studied by atomic physics [1]. Aside from direct electron-impact ionization, more complex ionization mechanisms exist involving autoionizing resonances. Electron impact can excite a bound electron into an autoionizing state which afterwards deexcites via Auger decay. Indirect (e,2e) processes relying on intra-atomic electron correlations can exceed the direct ionization channels.

Those autoionizing transitions can also be caused by electrons positioned at two spatially well separated atomic centers. A resonantly excited atom may transfer its excitation energy radiationlessly to a neighbouring atom via interatomic electron-electron interaction. This neighbouring atom can then be ionized, given the energy transfer is sufficently large. Consequently, radiationless decay is possible even when a single-center Auger decay is energetically forbidden. This process is called interatomic Coulombic decay (ICD) [2]. While photoabsorption represents the common method to generate autoionizing states in experimental research of ICD, there have been recent experiments utilizing electron impact instead [3, 4].

In this contribution we study electron-impact ionization of two-center atomic systems where excitation-autoionization can occur (see Fig. 1). Collisional excitation populates an excited state of an atom B. Its decay back to the initial states of B via two-center Auger decay (or ICD) leads to the ionization of a neighbouring atom A. Contrary to [3, 4] the incident electron causes here excitation (instead of ionization) of atom B. The two-center electron-impact ionization process, which we denote as 2C(e,2e) interferes with the single-center direct ionization of atom A. We show that 2C(e,2e) qualitatively modifies the electron-impact ionization, particularly by strongly enhancing the emission of electrons in a narrow range of energies [5]. Moreover, we study the influence of atom B on the angular dristribution of electron ejected from atom A.
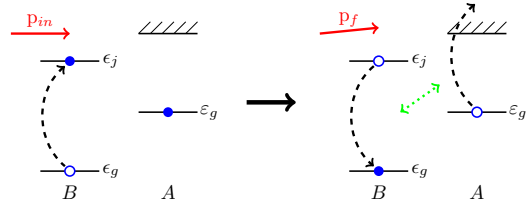

Figure 1. Schematic representation of 2C(e,2e). Collisional excitation of atom B by electron impact creates an autionizing state (left). Radiationless energy transfer to atom A leads to its ionization via two-center Auger decay or ICD (right).

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An R-matrix study of the resonant Auger decay of double K-vacancies produced in sequential X-ray ionization of neon

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Synopsis   Excitation and decay of a double K-vacancy during ionization of the Ne^{+}(1s^{1}2s^{2}2p^{6}) ion in the resonance Ne^{+}(1s^{0}2s^{2}2p^{6}np) region is theoretically studied by an R-matrix method. Photoionization cross sections, photoelectron spectra and angular distributions of the electrons are obtained.

Intense pulses from X-ray free-electron lasers make it possible to study nonlinear photoprocesses in the short-wavelength domain. This includes, in particular, sequential and direct multiphoton ionization and excitation. Sequential ionization of the core electrons competes with the fast relaxation of vacancies via the Auger decay [1]. With intense X-ray beams, more than one photon can be absorbed within a few femtoseconds before the Auger decay of a vacancy occurs. Thus, in sequential photoabsorption of core electrons the relative population of the ionic states with two, and possibly more holes, increases significantly in comparison with the single-photon ionization, where double core holes are produced via shake-up/off processes from the inner shells [2].

We consider photoionization

$$\gamma + \text{Ne}^{+}(1s^{1}2s^{2}2p^{6}) \rightarrow \text{Ne}^{++} + e$$
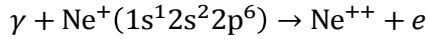
at photon energies around 980 eV, i.e. in the region of the Ne^{+}(1s^{0}2s^{2}2p^{5}np) double core hole resonances. Calculations were performed by the B-spline R-matrix method with non-orthogonal orbitals [3]. Both the direct ionization channels 1s^{1}2s^{2}2p^{5}, 1s^{1}2s^{1}2p^{6}, 1s^{0}2s^{2}2p^{6} ('participator Auger decay') and the shake up ionization channels 1s^{1}2s^{2}2p^{4}np, 1s^{1}2s^{1}2p^{5}np, 1s^{1}2s^{0}2p^{6}np ('spectator Auger decay') were taken into account. The spectator channels were found to dominate the widths of the resonances (Fig. 1a).

Angular distributions of the emitted electrons

$$I(\theta) = I_0(1 + \beta P_2(\cos\theta)),$$

where θ is the polar angle with respect to the electric field vector, contain the anisotropy parameter β. Fig. 1b demonstrates the β-parameters for ionization into selected states of the doubly charged ion. Energy shifts and broadening of resonances in the β-parameters [4] are remarkable.
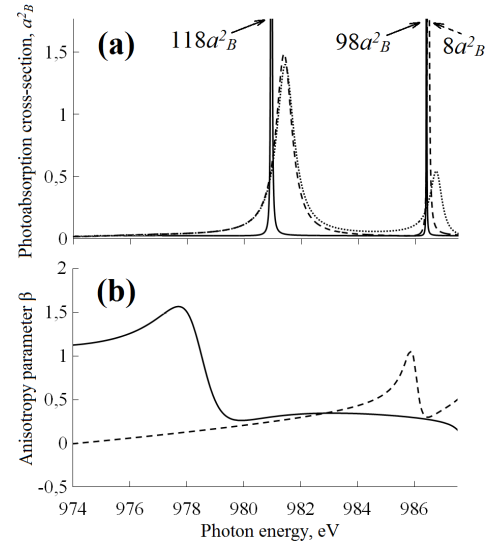

Figure 1. (a) Photoabsorption cross section of Ne^{+}(1s^{1}2s^{2}2p^{6}): without spectator channels (solid); including 3p-electron spectator channels (dashed); including 3p- and 4p-electron spectator channels (dotted). (b) β-parameters for ionization to Ne^{++}(1s^{1}2s^{2}2p^{4} ^{1}D 4p ^{1}F) (solid) and Ne^{++}(1s^{1}2s^{2}2p^{4} ^{1}D 3p ^{3}F) (dashed).

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On a possibility to observe and control spin polarization of photoelectrons produced by bi-chromatic coherent XUV radiation

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Synopsis   Photoelectron spin polarization is generated in the ionization by combined fundamental and second laser harmonics. It has various origins and values for photoelectrons ejected in different directions and can be controlled by tuning the relative intensity, phase and polarization of the harmonics.

Spin polarization of photoelectrons occurs due to spin-orbit interaction. In discrete spectrum it causes fine-structure splitting and term mixing. Essential spin polarization of photoelectrons is often reached at photon energies in the minima of the cross sections and is difficult to be observed due to low efficiency of spin-sensitive detectors.

The way to cope with such difficulties is finding processes with spin polarization occurring in the maxima of cross sections and non-vanishing after summing over fine-structure levels of the residual ion. The latter is crucial for experiments with light noble-gas atoms. Resonant two-photon ionization is one of such processes [1]. The origin of the photoelectron spin polarization in this case is the fine-structure splitting of the intermediate excited atomic state and term mixing.

When it comes to simultaneous one- and two-photon ionization by a field

$$\mathbf{E}(t) = \boldsymbol{\varepsilon} E_0(t) (\cos \omega t + \eta \cos (2\omega t + \phi))$$

with fundamental (ω) and second (2ω) laser harmonics causing two- and one-photon ionization, respectively, spin polarization can originate both from resonant two-photon ionization and interference between one- and two-photon pathways. The interference provides a mechanism to control both the photoelectron angular distribution (PAD) [2] and the spin polarization. Current research aims at studying this possibility for noble gases. Such a control was recently discussed for alkali atoms in the optical range [3]. Recent progress of producing intense coherent XUV light with variable polarization by free-electron lasers can stimulate experimental studies in the gaseous targets.

Results of the interference may appear in a crucial way. For example, in the case of circularly polarized counter-rotating fields, the spin polarization in the direction of their propagation $\mathbf{k}$ takes the form of the PAD (the latter strongly modified by the interference itself), rotated about $\mathbf{k}$ by an angle which depends on the atomic structure (Fig. 1). The spin polarization component along $\mathbf{k}$ originates both from the resonant two-photon ionization and the interference, while the component opposite to $\mathbf{k}$ originates only from the interference.
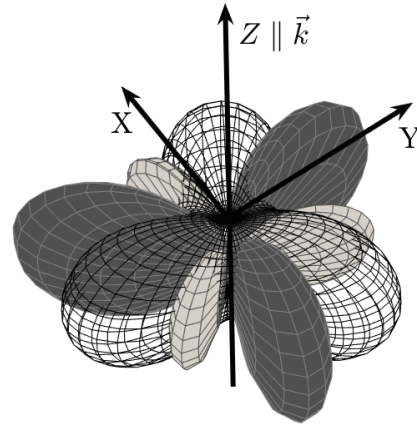

Figure 1. PAD (transparent) and values of positive (dark gray) and negative (light gray) spin polarization along the z-axis at ω corresponding to excitation of the $2p^5[^2P_{3/2}]4s_1$ state in neon. The pulse parameters: intensity 10^{12} W/cm^2, $\eta = 0.05$, $\phi = 0$, duration 250 optical cycles; the harmonics are circularly polarized. Maximum spin polarization is about 20%.

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A combined R-Matrix approach for investigating time-dependent multielectron dynamics in complex atoms

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Synopsis   We present a combined R-matrix approach for describing multielectron dynamics in complex atomic systems. The method results from the consolidation of two pre-existing R-matrix codes: the time-independent B-spline atomic R-matrix code (BSR) and the time-dependent R-matrix with time-dependence method (RMT).

As the time-scales on which we can observe electron behaviour steadily decrease, there is a great need for theoretical support, or even prediction, for the growing number of ultrafast experiments that have become possible. However, these experiments are becoming increasingly focused on the role of multielectron effects [1] and the capabilities of mid-IR lasers [2], both of which present considerable challenges to current computational methods. A successful time-dependent multielectron approach will therefore need to contain two things: an accurate but compact atomic structure description, and an efficient propagation scheme.

One set of methods which have achieved much success in the arenas of time-dependent and time-independent computational atomic physics are R-matrix methods. Introduced by Burke and colleagues to the field of atomic physics [3], at their core these approaches employ the R-matrix paradigm of dividing configuration space into two regions, allowing the most suitable numerical methods to be employed in each one. Among R-matrix methods, two approaches have distinguished themselves in their respective fields: the time-independent B-spline atomic R-matrix code (BSR) [4] and the R-matrix with time-dependence method (RMT) [5].

BSR differs from other time-independent R-matrix approaches in two significant ways. Firstly, non-orthogonal orbitals can be used to represent both the bound and continuum one-electron orbitals, allowing a high level of accuracy to be achieved with a compact configuration expansion and largely eliminating the appearance of pseudo-resonances. Secondly, a set of B-splines, which have many desirable numerical properties, are used to define the R-matrix basis functions.

The time-dependent RMT method divides configuration space into two separate regions, in this case over the radial coordinate of an ejected electron. In the inner region the time-dependent wave function is represented over a B-spline R-matrix basis with time-dependent coefficients, while, in the outer region, it is described in terms of residual-ion states coupled with the radial wave function of the ejected electron and expressed explicitly on a finite-difference grid. At variance with other R-matrix-based approaches, the wave function itself is matched explicitly at the boundary, rather than via an R matrix. The RMT code is optimized to run on massively parallel machines, thus making the extension to challenging physical systems tractable.

Given their individual desirable characteristics and common B-spline R-matrix basis, BSR and RMT are natural choices to provide, respectively, the atomic structure description and propagation scheme for a method probing time-dependent behaviours in general atomic systems. It is hoped that this combined approach could enable the investigation of phenomena such as auto-ionization and spin-orbit dynamics in multielectron systems, and that preliminary results will become available in the coming months.

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Atomic and molecular suite of R-matrix codes for ultrafast dynamics in strong laser fields and electron/positron scattering

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Synopsis   We describe and illustrate a number of recent developments of the atomic and molecular *ab initio* R-matrix suites for both time-dependent calculations of ultrafast laser-induced dynamics and time-independent calculations of photoionization and electron and positron scattering.

The field of attosecond science has presented computational physics with a set of new challenges to describe the dynamics of matter in strong femtosecond lasers. The recent move to study complex multielectron systems makes the need for a matching *ab initio* theoretical description even more pressing. In the more mature field of electron-molecule collisions, the need to study more complex targets has also required significant software improvements. Our suites of codes [1, 2] have recently undergone a major upgrade and been made available to the community.

A set of atomic and the molecular time-independent codes form their core. On the atomic side these are the well-established RMATRXI and RMATRXII codes: both use a B-spline representation of the continuum; RMATRXI allows the inclusion of relativistic (Breit-Pauli) corrections. On the molecular side, a major development has led to the release of UKRmol+, based on code originally developed for electron/positron - molecule scattering. This suite has been extended to enable photoionization calculations and to include B-splines in the representation of the continuum via the new GBTOlib integral library. Significant improvements in performance of the molecular codes have been achieved developing a new parallel Hamiltonian build algorithm and a parallelized version of the R-matrix propagator.

The second layer of the R-matrix suite comprises R-matrix with time (RMT) [3] codes which can now be used to propagate in time both the atomic and molecular (in the fixed-nuclei approximation) wavefunctions. Recent developments allow the use of arbitrary polarized light and interfacing with the atomic RMATRXI suite thus enabling studies of spin-orbit dynamics in ultrafast laser fields.

The recent developments will be described, including illustrations of the results on generation of electron vortices from argon atoms, relativistic effects in the vicinity of the giant resonance in Xenon and validation tests of molecular RMT on photoionization of water. The benefits for the electron-molecule scattering community will be illustrated with the results for inelastic electron scattering from thiophene.

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NH₃ ultrafast dissociation probed by Auger electron-ion coincidences

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Synopsis   Ultrafast dissociation of core-excited NH_3 molecule is revisited by using Auger electron-ion coincidences. The results give new insights to fragmentation mechanism and its efficiency.

Resonant excitation of a core electron to an anti-bonding molecular orbital may lead to ultrafast nuclear dynamics and even dissociation, which occurs on the same time-scale and, therefore, competes with radiative or non-radiative Auger decay [1].

In this work we have revisited ultrafast dissociation (UFD) phenomenon, which occurs in core-excited NH_3 molecule and competes with Auger decay on a few femtosecond time-scale, using energy-selected resonant Auger electron-ion coincidence techniques. ES-AEPICO is a unique complementary tool to high-resolution photoelectron spectroscopy as it allows disentangling fragmentation mechanisms [2]. This is because the observed resonant Auger final states can be directly correlated to the fragment ions and their behavior can be tracked as a function of the photon energy.

Narrow bandwidth photon energies across the N1s->$4a_1$ resonance were used to produce dissociative N1s core-excited states of NH_3. ES-AEPICO spectra were recorded at several energies across the resonance using EPICEA coincidence setup, permanently installed at PLEIADES beamline of the SOLEIL synchrotron. Detuning the photon energy allows varying the so-called effective scattering time [3] and hence influence the relative yield of the fragmentation occurring within the lifetime of the N1s core hole ($\tau \sim 6$ fs).

The 2D resonant Auger electron-photoion coincidence maps, as presented in Fig.1, at different photon energies allow correlating Auger final states to different fragmentation patterns and confirming that the Auger states (381-383 eV) are related to NH_2^+ fragments, as first observed in a previous study [3] by single-channel resonant Auger spectroscopy. Moreover, other final states of the fragments, which are hidden in resonant Auger measurements, could be identified by the coincidence measurements.

We also observe increased efficiency of UFD for positive energy detunings, which is in line with the previously calculated potential energy surface of the N1s^{-1}$4a_1$^{1} core-excited state of NH_3 [4]. The experiments were also performed at MAXIV synchrotron in Sweden at FinEst beamline in March 2019, revealing the so-called Auger Doppler effect [5], which was previously observed only for atomic fragments.
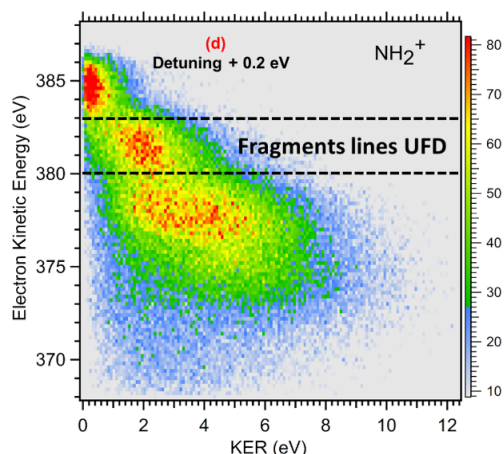

Figure 1. 2D Auger-electron-ion coincidence maps for $NH_3 + h\nu -> NH_2^+ + H° + e^-$ dissociation pathway.

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Electron-impact ionization of molecules

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Synopsis   Triple differential cross section for ionization of molecular targets by electron impact is calculated using approximate three body waves. The post-collisionnal interaction (PCI) between the outgoing electrons as well as the distortion effects of the ejected electron are included. The effect of the short range potential is also investigated. Results are compared with available theoretical results and experimennts.

Electron impact ionization of atoms and molecules has been a topic of large number of experimental and theoretical investigations. The triple differential cross section (TDCS), which represents the most sensitive test for theoretical models, is particularly investigated, the process has then gained a lot of interest during the past years. Much emphasis has been devoted to cross sections calculations because of basic importance in many practical applications such as astrophysics, plasma physics and irradiation of living matter. The ionization process has in fact been widely dedicated to atomic targets in contrast to molecules where further calculations   are still to be developed because of the encountered problems in theory and experiments.

We consider here an approach which includes post-collisionnal interaction (PCI) between the outgoing electrons as well as the distortion effects induced by the residual ion to investigate the ionization mechanism for some atoms and small molecules. (i) firstly the BBSR model where the outgoing electrons are represented by Coulomb waves taking into account their interactions with the residual ion. (ii) secondly the BD model where the ejected electron is represented by a distorted wave while the scattered one is described by a Coulomb wave. (iii) and in a third step the BDSR model which includes of the short range potential in the of BD model . In all cases the impact electron is described by a plane wave and a Coulomb interaction is used to account for the PCI, a full description of the inclusion of distortion effects can be found in our previous works [1]. The method is applied to study the ionization process in the case of some molecules.

In figure 1 the TDCS is displayed in the case of $1t_2$ and $2a_1$ molecular orbitals of CH_4 molecule at 250 eV projectile energy. Our results are compared with recent experiments [2] and the well known M3DW model,

where theory and experiments experiments are normalized to BBSR model in the binary region. It is seen that the BBSR model reproduces quite well the data as done by M3DW. For $1t_2$ orbital, the double peak structure experimentally exhibited in the binary region is reproduced by our theory. Likewise the recoil region is even better described by our BBSR where it is observed in slightly better agreement with the data. For the inner $2a_1$ orbital the shape of the TDCS is still well reproduced and closer to the data then M3DW. For these kinematics BBSR model seems to be able to describe the (e,2e) reaction.
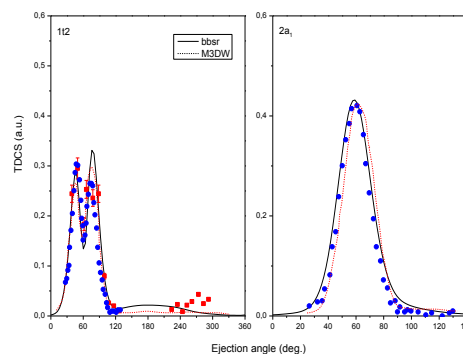

Figure 1. TDCS vs ejection angle for the ionization of CH_4. Kinematical parameters are: $\theta_S = 25°$, *Ei=250 eV* and *Ee=50 eV* [2]. Theory is compared with the Afyon data (red squares) and Manchester data (blue circles).

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An investigation of double photoionization in Benzene

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Synopsis A double photoionization (DPI) coincidence technique has been used to study electron correlation as a function of photon impact energy on Benzene (C_6H_6). The simultaneous emission of two photoelectrons from the Benzene π orbital has been measured using two sets of hemispherical electron energy analysers. The experiment was performed at the Gas Phase Photoemission (GAPH) beamline at Elettra with photon energies in the range 30-70 eV. Angle resolved single and coincidence photoelectron spectra are reported, providing new data on the DPI mechanism.

Benzene is a prime example of an aromatic hydrocarbon, a molecule with a planar arrangement where the π molecular orbitals overlap and the electrons are completely delocalised. Aromatic hydrocarbons have shown some interesting properties, like superconductivity at high temperatures [1] and excellent thermal properties. Hydrocarbons play a central role in biological process, astronomical process and combustion.

This cross-beam experiment was completed on the GasPhase Beamline at Elettra. The liquid benzene was placed in a test tube outside the vacuum chamber, freeze-pump-thawed to remove gases, and then admitted to the interaction region via a gas line and a gentle heating . The sample gas pressure was on the order of 10^{-6} mbar. A VASGM (Variable Angle Spherical Grating Monochromator) allowed energy tuning from 2 to 1000eV with an extremely fine resolution ($E/\Delta E >= 10000$). The linearly polarised beam delivered a small spot to the interaction region (200 μm x 200 μm) with a very small angular divergence of <3mrad [2].

The 10 hemispherical electron energy analysers host in the multi-coincidence experimental chamber are mounted on two independent frames, holding respectively 3 and 7 analysers separated by 30 degrees of each other. In the present experiments, the two frames were placed in the perpendicular plane and used to detect electron coincidences from the DPI process. A detailed description of the apparatus and the related methods of the acquisition of the triple differential cross section (TDCS) and its analysis are described in ref. [3].
At a resonance photon energy, it has been proposed [4] that, benzene allows the formation of a two-electron pseudo-particle, which has been likened to a 'Cooper pair'. This is only possible when the C-C distance matches the de Broglie wavelength. In this model the electron pair leaving the molecule breaks up and the electrons are emitted back-to-back. Previous results [5] have shown small enhancements in the single electron photoelectron spectra at angles of 70 and 138 degrees [5]. However, other experiments [6] do not support this interpretation and the mechanism responsible for the enhancement is an open question.

The present results, investigate the case of equal energy sharing of the excess kinetic energy recorded at photon energies of (i) 45.75 eV (electron kinetic energy ke1=ke2=10 eV) and (ii) 65 eV (electron kinetic energy ke1=ke2=20 eV) This provides new information on the DPI process.

We also report prelimianry results where the two electrons share the excess kinetic energy unequaly. Two cases were invesitagted. (i) excess energy of 38eV with an unequal sharing of ke1=9.5eV ke2=28.5 eV and (ii) excess energy of 30eV with an unequal energy sharing of ke1=7.5 ke2=22.5eV. This provides further information on the process.

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Anisotropic excitation of highly charged ions by electron impact

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Synopsis   The relativistic distorted-wave program of the Flexible Atomic Code is extended to allow calculations of multipole collision strengths for anisotropic excitation of highly charged ions by an electron beam. These collisional parameters are required in analysing the intensity and polarization of lines emitted from hot plasmas with cylindrically symmetric electron distributions. Illustrative numerical results relevant to tokamak plasmas are obtained for excitation of the Be-like O^{4+} ion between levels of $2s^2$, $2s2p$ and $2s3p$ configurations.

Multipole collision strengths for excitation of highly charged ions by impact with unidirectional electrons are necessary in interpreting polarized line emissions from hot plasmas characterized by an anisotropic electron distribution with cylindrical symmetry [1, 2]. These collisional parameters denoted $\Omega_q^{kk'}$ with $k \leq 2J$, $k' \leq 2J'$ and $-\min(k,k') \leq q \leq +\min(k,k')$, J and J' being the total angular momenta of initial and final levels of transition, can be deduced from the collision strengths for transitions between magnetic sublevels only for $q = 0$, but not for $q \neq 0$. The $q \neq 0$ components of $\Omega_q^{kk'}$ arising when both k and k' are non zero, are required in the calculation of the multipole rates for plasmas with arbitrary electron velocity distributions.

Recently, we introduced some modifications to the relativistic distorted-wave (RDW) program of the Flexible Atomic Code (FAC) [3] to calculate the multipole collision strengths for isotropic colliding electrons [4]. Here we derive a RDW formula for $\Omega_q^{kk'}$ in the framework of the factorization method, and modify accordingly the RDW program of FAC to get $\Omega_q^{kk'}$ for all values of q. Numerical calculations are performed for a large number of transitions in the Be-like ion O^{4+} at different incident electron energies. The 46 levels of the nine configurations $2s^2$, $2s2p$, $2p^2$ and $2l3l'$ ($l = 0,1; l' = 0,1,2$) were included in these calculations. Owing to the lack of space, we selected in table 1 only the results for three transitions between $2s2p$ and $2s3p$ triplet levels at an energy slightly above the excitation threshold. Note that Ω_0^{00} which is given in the top line of the table is related to the conventional collision strength Ω by $\Omega_0^{00} = \Omega/\sqrt{(2J+1)(2J'+1)}$.

The values of Ω_q^{44} ($q = 1 - 4$) for the transition $^3P_2 \rightarrow {}^3P_2$ are not shown in the table because they are found to be practically equal to Ω_0^{44}.

Table 1. Values of $\Omega_q^{kk'}$ for excitation of O^{4+} from $2s2p\,{}^3P_J$ to $2s3p\,{}^3P_{J'}$ levels at the electron energy 5 Ry. $x[y]$ means $x \times 10^y$.

$kk'q$	$^3P_1 \rightarrow {}^3P_1$	$^3P_1 \rightarrow {}^3P_2$	$^3P_2 \rightarrow {}^3P_2$
000	1.47[−1]	1.98[−2]	1.56[−1]
020	3.14[−3]	−6.00[−3]	−8.68[−3]
200	1.04[−3]	2.85[−3]	−3.91[−3]
220	1.32[−1]	−2.11[−3]	1.35[−1]
221	1.32[−1]	−1.80[−3]	1.34[−1]
222	1.33[−1]	−3.16[−3]	1.34[−1]
040		2.15[−4]	−5.54[−5]
400			−5.55[−5]
240		1.53[−3]	−1.45[−3]
241		1.26[−3]	−1.29[−3]
242		5.98[−4]	−8.51[−4]
420			−1.71[−3]
421			−1.53[−3]
422			−1.02[−3]
440			1.31[−1]

The present calculations may be useful in the interpretation of UV line emission observed in tokamak experiments, e.g. [2].

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Observation of laser-assisted electron scattering signals with sub-10 fs laser pulses

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Synopsis We observed laser-assisted elastic electron scattering (LAES) processes induced by near-IR few-cycle laser pulses ($\lambda \sim 750$ nm, $\Delta t = 7.4$ fs). An accelerated (1 keV) electron beam was scattered by Xe atoms under the presence of the few-cycle laser field. The LAES signals with a one-photon energy gain were identified in the energy and angular distributions of the scattered electrons recorded by an angle-resolved time-of-flight type electron analyzer.

In electron-atom elastic collision processes in a laser field, electrons can gain or lose their energy by multiples of the photon energy. This scattering process is called laser-assisted elastic electron scattering (LAES). It was demonstrated recently that the LAES processes induced by femtosecond laser pulses ($\Delta t = 200$ fs) can be used for investigating ultrafast responses of electrons within an atom in an intense laser field [1] as well as for determining instantaneous geometrical structures of molecules [2]. Because the temporal resolution of the LAES measurements is determined by the laser pulse duration, LAES measurements with much shorter laser pulses have been awaited for probing ultrafast responses of atoms and molecules. In the present study, we have performed measurements of LAES processes induced by sub-10 fs laser pulses by an angle-resolved time-of-flight (ARTOF) type electron analyzzer.

The sub-10 fs laser pulses were generated by the pulse compression method using a gas-filled hollow-core fiber. The output of a Ti:sapphire laser system ($\lambda = 800$ nm, 5 kHz, $\Delta t = 40$ fs) transported through a beam-pointing stabilization module was focused into the hollow-core fiber filled with an Ar gas (0.5 atm), so that the spectral bandwidth was broadened by the self-phase modulation. The group delay dispersion induced by the self-phase modulation was compensated by chirped mirrors and wedged fused silica plates. The compressed pulses were characterized by a two-dimensional spectral shearing interferometer. The pulse duration of the compressed pulses was determined to be 7.4 fs (FWHM), and the spectrum covers the wavelength range (630 - 870 nm) centered at ~750 nm.

The electron signals scattered by Xe atoms were recorded as a function of the kinetic energy and the two-dimensional scattering angles using the ARTOF-type LAES apparatus. Figure 1 shows the kinetic energy distribution of the scattered electrons measured with the laser field whose intensity at the interaction point is 5.2×10^{11} W/cm^2 (red filled circles) and that measured without the laser field (black open squares), both of which were recorded after the accumulation for 13 hours. In the area in the range between 1.3 and 2.1 eV, whose width corresponds to the bandwidth of the laser pulse, the intensities of the signals recorded with the laser field are larger than those without the laser field, showing that the LAES process with one-photon energy gain was induced by the sub-10 fs laser pulses.
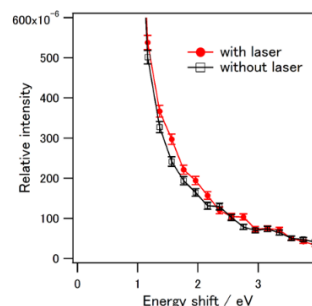

Figure 1. Energy distributions of electrons scattered by Xe recorded with the laser field (~750 nm, 7.4 fs, 5.2×10^{11} W/cm^2) (red filled circles) and those recorded without the laser fields (black open squares).

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Excited Electronic States in Thiophene and Thiolane using Resonant Auger Spectroscopy

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Synopsis   Sulfur-based organic molecules are important building units for polymers used applied in photovoltaics. In this work, we used absorption spectroscopy to excite the sulfur K-edge core electron from thiophene and thiolane to an empty molecular orbital. Resonant Auger spectroscopy is used to monitor delocalization from an initially localized core-excited state around the absorption atom. Visualization of the data in the form of two-dimensional Auger maps made it easier the analysis and highlighted the qualitative difference in the behavior of the spectral lines.

In this work, we present an experimental and theoretical study in thiophene and thiolane gas-phase molecules around the resonance excitation of the Sulfur K-shell to the lowest unoccupied molecular orbital (LUMO) and further beyond the ionization threshold using high-resolution high-energy resonant Auger spectroscopy. Data visualization in the form of two-dimensional (2D) Auger maps make the analysis significantly easier and highlight the qualitative difference in the behavior of the spectral lines [1]. The change in the order of the two lowest orbitals in thiophene when compared to thiolane was observed from 2D maps.

To improve the interpretation of the experimental measurements, theoretical chemistry models were applied to obtain an explanation for the differences between the spectral lines in 2D Auger maps. We chose the multi configurational wave functions to describe the excited states, applying a specific routine for inner-shell states (IS-CASSCF) [2]. In this method, high energy states can be reached by constraining the occupation of the core orbitals and optimizing them during an isolated SCF step. Relaxation effects are included in our description and they play an important role in the accurate description of excitations from S 1s to σ^* and π^* orbitals, as well their correlate states in Auger $S2p^{-1}$ states.

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Gaussian Processes Optimization of atomic structure for collisional problems calculation

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Synopsis The accurate description of the target is required when describing collisional processes. Generally, the atomic structure is obtained through the optimization of electronic configurations and parameters to minimize the differences between the numerical and experimental values of the energies and the oscillator strengths. By implementing a machine learning approach through a Bayesian Optimization with Gaussian Processes, we can reproduce the experimental atomic structure with high accuracy. Results for neutral beryllium are presented.

A highly accurate target description has proven to be of importance to solve collisional problems with the close-coupling expansion [1, 2, 3]. Nevertheless, the determination of an adequate atomic structure often requires expertise and significant computational resources. In general, the wavefunction of the target is expressed using the configuration interaction (CI) method. Moreover, the radial part of the one-electron wavefunctions can be obtained with model potentials containing scaling parameters. The accuracy of the calculations is improved by incrementing the number of configurations in the CI, which in turn increases the number of parameters to be varied. However, there is neither a systematic nor logical prescription for this procedure.

We are currently studying machine learning techniques in order to perform such kind of optimizations automatically. To illustrate one of these implementations, we considered the neutral beryllium atom. The atomic structure is calculated with the AUTOSTRUCTURE code [4], and the orbital radial wavefunctions are obtained with a Slater–type–orbitals containing λ_{nl} scaling parameters. These parameters are then optimized within a Gaussian Process Regression using a squared exponential kernel and an expected improvement acquisition function. In the present contribution, we used GPyOpt codes [5], for calculating the best set of hyperparameters that produces the minimum cost function

$$J = \sum_{nl} \left| \frac{E_{nl} - E_{nl}^{\text{A.S.}}}{E_{nl}} \right|, \qquad (1)$$

i.e., the total relative difference between computed $E_{nl}^{\text{A.S.}}$ and benchmark energies E_{nl} [6, 7].

The target expansion considered the $2snl$ configurations up to $nl = 5g$, and $2p^2$. Our *search space* included only the first three orbitals, and they were varied around $\lambda_{nl} = 1$. The results obtained after 18 evaluations are given in Fig. 1. The agreement found for the $2s^2\,^1S$, $2s2p\,^3P$ and $2s2p\,^1P$ absolute energies is better than the 0.5%.
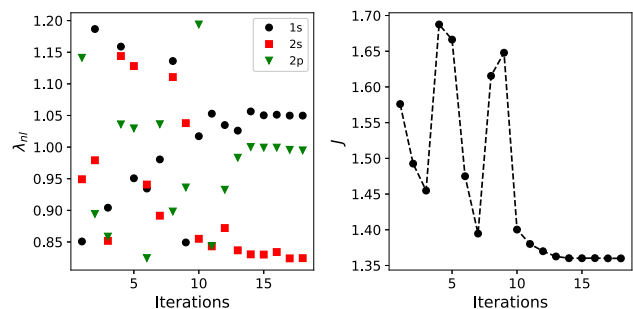

Figure 1. Left: Results of the Gaussian Process Bayesian optimization for the scaling parameters of Be. Right: Evolution of the cost function J.

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Dynamical investigation of the electron momentum distributions of adamantane

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Synopsis A new analysis of the outer valence (e, 2e) momentum distributions of adamantane has been performed using two complementary approaches. The first relies upon the harmonic oscillator model, while the second uses the principles of the Born-Oppenheimer Molecular Dynamics (BOMD).

In this study an in-depth investigation of vibrational effects on the electron momentum distributions of the outer valence orbitals of adamantane ($C_{10}H_{16}$) is reported [1]. To this aim, two different methodologies have been used. The first one, known as Harmonic Oscillator Quantum Mechanical (HAQM) approach [2], allows to quantitatively evaluate the role of each normal mode of vibration on a given momentum distributions. The second one uses the principles of Born-Oppenheimer Molecular Dynamics (BOMD) simulations [3] and allows to average the effects of the temperature over a large number of structures and to take into account non-harmonic effects. The obtained theoretical results have been compared with newly acquired experimental (e, 2e) momentum distributions with improved statistics as compared with that of the available data on the same compound [4].

Despite their intrinsic difference and the intricate nature of the vibrational structure of the target, both methods provide consistent results and give quantitative insights into the results of the new (e, 2e) experiment, except for the $\{1t_1 + 6t_2\}$ ionization band. Comparisons between experiment and theory have shown that taking into account vibrational effects leads to a strong enhancement of the low p component of the $7t_2$, $\{2t_1 + 3e\}$, and $\{5t_2 + 5a_1\}$ momentum profiles and considerably improves the agreement with experiment, showing also the deficiency of the previous explanation that ascribed the discrepancies mostly to distorted waves [4]. Further analysis based on the HAQM approach has revealed, in line with similar studies [5, 6], the significant role of vibrational modes associated with the C-H bonds.

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A Double Slit Photoionization Experiment in a Single Atom

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Synopsis   A new type of 'double slit' experiment is presented where the interference proceeds via photoionization of Rb atoms subjected to two resonant laser fields. The data from experiments compare well to quantum models that detail the interaction, and further results in this area will be discussed at the conference.

The 'double-slit' experiment is one of the key experiments that demonstrates wave-particle duality in quantum physics. Experiments showing this duality using *single* electrons passing through slits have only recently been performed [1-3], and these provide convincing evidence of these interference processes. Figure 1a shows the types of experiments that are performed. The particles are constrained to pass through two slits defining paths 1 and 2. Quantum mechanically the paths are represented by wavefunctions φ_1 and φ_2 that must be added coherently at the detector to produce the resulting particle distribution. This leads to interference fringes, with the single particles being detected with a distribution that emulates the classic interference structure first observed by Young in the early 19th century.

In the current work (figure 1b) the pathways that interfere are created in a single Rubidium atom, with the individual paths being defined using high resolution laser beams that select the intermediate 5P and 6P states. These excited states hence are equivalent to the 'slits' in a conventional experiment. Since the 5P state can be ionized by radiation that excites the 6P state and vice-versa, it is not possible to know which pathway creates the photoelectron in the continuum. The wavefunctions ψ_1 and ψ_2 must again be added coherently to define the probability of ionization, leading once more to interference.

In this case the measurements are carried out by selecting photoelectrons with 0.36eV energy, and their angular distribution is determined. By varying the detuning of the laser beams individual pathways can also be selected, and a systematic study of these processes allows both the amplitude and phase of the interference term to be experimentally determined.

A quantum model of the interaction developed in Halle, Germany agrees well with the results of experiment [4], and this model has now been extended to predict how the interference term varies as different states of the atom are selected. The model further predicts how the phase and amplitude of the interference term evolves in time under ultra-short laser conditions [5], and these results will also be discussed at the meeting.
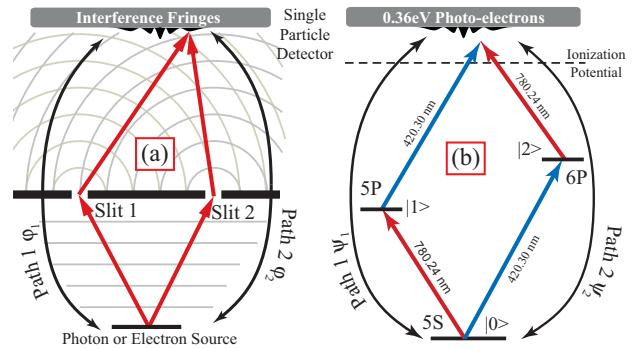

Figure 1. (a) shows the conventional 'double-slit' experiment that demonstrates wave-particle duality using single particles. (b) shows the new experiments where laser beams define the pathways to ionization in a single atom, leading to interference in the angular distribiution of the emerging electrons.

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Study of the electron impact ionization of the intermediate valence state of NH_3

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Synopsis A study of the ionization of the intermediate valence state of NH_3 by electron impact is reported. Measurements are performed in an asymmetric coplanar geometry under kinematics which are unexplored to date. The data are compared to predictions from the first order approaches and BBK model.

Electron impact ionization presents an important tool for the study of the structure of the considered target or to understand the dynamincs of the (e,2e) reactions. Performing electron-electron coincidence techniques experiments on molecules targets becomes a big challenge mainly because of the complex nature of molecules.

Various theoretical calculations for noble gas atoms such as neon show good agreement with the experimental results. But it is not the case for methane, even that neon and methane are isoelectronic targets. Thus, the study of the ionization process of ammonia by electron impact comes to test the validity of the theoretical models. Those targets are isoelectronic targets, and what really differs is the structure of each one. Thus, we can investigate the influence of the molecular structure on the ionization process.

Few experiments were performed on NH_3 for low incident energy [1], in addition to EMS experiments [2] for high incident energy. Triple differential cross sections for the electron impact ionization of the three valence states of ammonia in an intermediate energy regime were reported [3]. We performed new experimental and theoretical results for ionization of the intermediate valence state 1e of ammonia molecule, under moderate incident energy. This second energy level is a doubly degenerate bonding level with $1e1$ symmetry. It is found to have significant p-like character from EMS measurements [4]. Scattered and ejected electrons have been detected with 500 eV and 12 eV,

respectively. The experimental parameters are listed in table 1. θ_k /θ_{-k} indicates the forward and backward directions of the momentum transfer.

Table 1. Kinematical parameters for the SI process considered in this study.

Ea=500 eV	θ_a=-6°	Eb=12ev
Target	NH3	
E_0 (eV)	K (a.u)	θ_k /θ_{-k} (deg)
528.6	0.66	72/252

The chosen kinematics do not satisfy neither Bethe ridge nor dipolar conditions. In this situation, the recoil scattering of the ejected electron is enhanced relative to the binary.

The data have been compared to theoretical calculations using first Born approach and BBK model. Both theories reproduce quite well the shape of the binary region for each molecular orbital, however the recoil peak is largely underestimated.

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A Computer-Controlled Atomic Beam Oven for Alkaline-Earth Metals

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Synopsis   A new computer-controlled atomic beam oven for use in laser-excited (e,2e) measurements has been designed and constructed. The oven delivers an atomic beam with an angular divergence of ~5°, which minimizes the Doppler profile of the beam while ensuring a good beam density. The oven has been tested using the alkaline-earth metals Mg, Ca and Sr in preparation for new ionization studies on these laser-excited targets.

Many experiments require a highly collimated beam of atoms, including electron-impact ionization and excitation studies [1] and the creation of highly excited Rydberg states [2]. In these experiments it is necessary to reduce Doppler broadening of the atomic beam to increase the excited state population created by resonant laser radiation, and it is also desirable to reduce or eliminate unwanted metal deposition onto the vacuum chamber surfaces in the apparatus.

A new atomic beam oven and associated control electronics has hence been designed to satisfy these criteria. This oven is to be used in new laser-excited (e,2e) measurements in Manchester which are to be carried out using different alkaline-earth targets, and follows a similar design to that described in reference [3]. The design of the new atomic beam oven and the associated computer controlled power supply is shown in Figure 1.
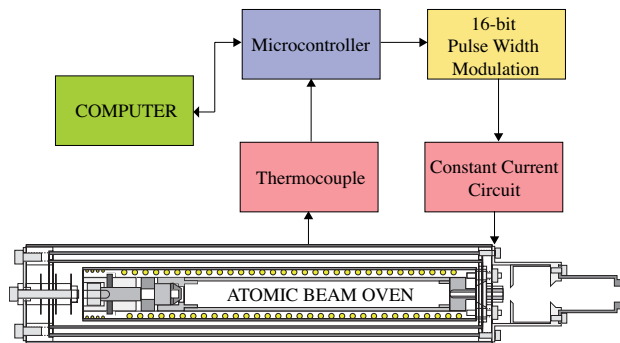

Figure 1. Diagram showing the cross section of the atomic beam oven and a block diagram of the electronics used to control the temperature. Directional arrows show the flow of data to and from the experiment and control systems. The Thermocoax heater element surrounding the crucible is shown in yellow.

The oven is composed of five main parts: a heating element, an outer jacket surrounding this element, removable crucibles for holding the atomic species, a back-plate heat-shield, and a front skimmer system to collimate the beam. Ceramic insulators were placed between the heating element and outer heat-shields to provide thermal insulation. The 2mm diameter Thermocoax element that surrounds a copper furnace is heated using a constant current power supply as shown. The oven has been tested with Mg, Sr, and Ca. Temperatures up to 700°C have been achieved during these tests, and in each case the average beam divergence was measured to be ~5°.

The constant current supply is computer-controlled, as shown in Figure 1. The microcontroller produces a filtered 16-bit pulse-width modulated signal that adjusts the high-current circuit that feeds the Thermocoax heater. A type-K thermocouple measures the crucible temperature and provides feedback to the microcontroller, so as to set and maintain the required temperature. An algorithm within the controller increases or decreases the temperature of the oven in a controlled manner, so that the oven is heated or cooled so as to prevent blocking of the exit aperture of the crucible. The microcontroller can also be connected to an external computer using a USB interface, allowing the oven temperature to be monitored and adjusted during data acquisition.

Full details of the control system and oven design will be presented at the conference.

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Study of charge dependent effects in the electron and positron impact single ionization of inert gas targets

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Synopsis   Electron impact and positron impact differential cross sectons are reported for the Ar and Xe targets in modified distorted wave Born approximation. Present attempt is helpful to analyze the recent experimental [PRA **95**, 062703 (2017)] and theoretical study [PRA **97**, 062702 (2018)] aimed to search the charge dependent effects in the ionization of the inert gas targets. The present attempt will be helful to further probe the collision dynamics of the inert gas targets in terms of charge dependent effets.

The study of electron impact ionization of atoms, ions and molecules has been of interest since the early days of quantum physics. Particularly, study of electron impact ionization cross section of various targets is important as it gives information about the delicate collision dynamics and wave functions [1]. Detailed information about this kind of collision process is obtained from the triple differential cross sections (TDCS) obtained through a coincidence study. A coincidence study of TDCS has been of particular interest since it provides full information about the collision dynamics and momentum vectors of all free particles involved in the ionization are determined. There has been demand for differential cross section studies with positrons for a long time, which has mainly remained confined to single differential and double differential investigations [2-3]. The projectile target interaction is better probed by the differential ionization data; they give information about energy and momentum transfer, and in particular the positron impact studies are desirable to probe the role of projectile charge or mass on the collision dynamics in comparison to the electrons having similar kinematics conditions and also has important applications in characterization of materials, PET imaging in medicines, etc. [4]. Very few experimental and theoretical TDCS data are available for the positron impact ionization.

We present triple differential cross sections for the electron and positron impact ionization cross sections of the Ar and Xe targets in Coplanar asymmetric geometry. We calculate TDCS for the inert gas targets in distorted wave Born approximation (DWBA) formalism including target polarization and post collision interaction (PCI) effects. We compare our results with the available measurements [5] and theoretical results [6-7] and discuss the salient features of TDCS for inert gas targets which are projectile charge dependent.
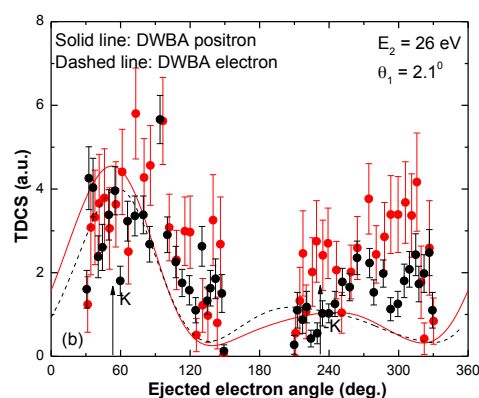

Figure 1. TDCS plotted as a function of ejected electron angle for the ionization of Ar (3p) at ejected electron energy 26 eV. Red solid line: DWBA result for positron and Black dashed line: DWBA results for electron impact, Red circles: experimental data [5] for positron impact; black circles: experimental data [5] for electron impact. experimental data have been normalized to the solid curve / dashed curve in the binary peak region for best visual fit

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Task-Parallelized Numerical linear algebra methods to solve the Few Body Coulomb Problem

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Synopsis We apply a reformulated numerical method for the solution of large linear systems related to the numerical treatment of Helium-like double photoionization systems. This method allows the use of multi-processor computers to solve problems where the amount of basis functions to obtain an accurate representation of the particle's physics is not known *a priori*, being able to update solutions of incremental linear systems.

In atomic physics simulations, the formulation of partial derivative equations (PDEs) leads to the final task of solving a linear algebra system. Since we want to be more accurate when describing a phenomenon, this results in needing incrementally more variables to obtain a satisfactory solution. Because of this, developing fast numerical linear algebra methods is a top priority in computational physics.

In this work, we attempt to solve the Schrodinger equation for continuum states that results from collisions in a three-body system, that can be obtained from the solution of the time-independent problem:

$$(H - E)\Psi = W\Psi_0, \qquad (1)$$

where Ψ_0 is the initial state of the process and E is the total energy of the system. The operator W is responsible for the transitions from state Ψ_0 to the collisional unknown state Ψ.

A widely used tool to obtain this solution is the spectral method, where we represent Ψ and Ψ_0 as an infinite sum of base functions:

$$\Psi(r_1, r_2) = \sum_{n,m}^{NM} x_{nm}\Omega_{nm}^L(r_1, r_2) \qquad (2)$$

The proposed model for the solution of these problems is based on a previous publication, where we describe a task-parallelization approach to the solution of a linear system with an unknown final dimension, using *latent matrices* [1]. For this work, we adapted the method to compute cross-sections of the double photoionization of helium, using Generalized Sturmian
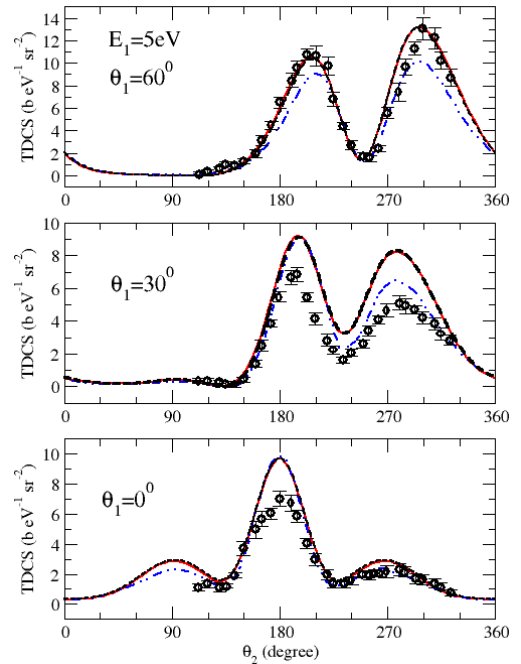
Functions to describe the function space.

Figure 1. Cross sections of double photoionization of helium for photon energy 40 eV above threshold (red curve), compared with experimental data (circles and dash-point curve) and calculations from McCurdy et al [2] (dashed curve).

The obtained results coincide with the state of the art for atomic simulations. Because of its characteristics, we conclude that this method can be useful for calculations that can't be done with the current state of the art.

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The Sturmian eigenvalue problem in two dimensions for bound states and collision problems

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Synopsis We define a family of two dimensional eigenvalue problems of the Sturmian type, where the energy is fixed and the weight of a given potential is considered as the eigenvalue. The differential equation involves other potentials and centrifugal barriers related to the partial wave decomposition of the three-body equation. By choosing negative energies, we get adequate basis sets to study bound states. By choosing positive ones and outgoing flux boundary conditions, we obtain solutions suitable as basis elements to describe three-body break-up processes.

For systems involving more than two particles, solving the Schrödinger equation still constitutes a challenging numerical task, in particular for continuum states. For the simplest three body Coulomb systems, 'numerically exact' solutions to the Schrödinger equation for bound and continuum states have been obtained, but require extensive computational resources.

In order to accurately describe the two electron continuum with respect to a fixed reference frame (atomic core), a partial wave expansion of the wave function transforms the equation into a N_L coupled set of two dimensional differential equations for the radial parts. N_L corresponds to the dimension of the angular basis employed in the expansion (partial waves), and generally depends on the energy regime of the problem (for electron atom collisions, for example, N_L increases with energy). The size of the related linear system of equations grows, roughly speaking, as the square of the one dimensional radial basis size (matrix representation), multiplied by N_L. For atomic scattering problems, extended spatial domains must be span, requiring large radial basis sets and leading to computation costs that can become prohibitive. The situation is even worse for polyatomic molecules, for which the additional issue of molecular orientation further increases the number of required calculations.

With the aim of tackling always more computationally demanding scattering problems, it is important to gain in efficiency by looking for the best basis functions. With this purpose in mind we explore here a new strategy that considers a two dimensional basis instead of products of one dimensional ones.

Within the Generalized Sturmian Functions spectral scheme [1], developed for both bound states (*e.g.*, [2]) and scattering (*e.g.*, [3, 4]) problems, the one dimensional basis functions (1DSF) are constructed as to contain some physical aspects, as for example an adequate asymptotic behavior.
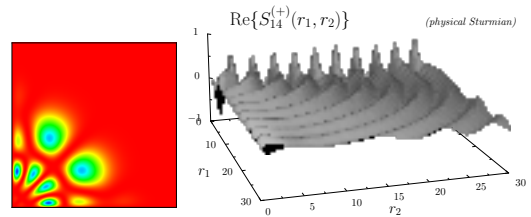

Figure 1. Left: density plot of a bounded 2DSF. Right: Real part of a positive energy outgoing-flux 2DSF for continuum processes .

In the present spectral approach we introduce a 2D Sturmian eigenvalue problem (2DSF). This give us wave functions (see Fig. 1) with adequate features of the problem under consideration. This ultimately allows us to reduce the block size of the coupled set of equations. We compare the two approaches (1DSF and 2DSF) by exploring their performance in describing the solution of the Helium atom bound states, and the continuum wave functions arising in the single photon double ionization of Helium.

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Low Energy (e,2e) Measurements from CH_4 from a Coplanar Symmetric Geometry to the Perpendicular Plane

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Synopsis   (e,2e) ionization cross section measurements have been carried out from a methane target at low incident energies, where the outgoing electrons are detected with equal energy and equal azimuthal angles. Results were obtained from a coplanar symmetric geometry where the incident, scattered and ejected electron momenta (k_0, k_a, k_b) were all in a plane, through intermediate geometries to the perpendicular plane where k_0 is orthogonal to both k_a and k_b.

Measurements of electron impact ionization provide maximum information when all kinematic conditions of the scattering process are determined. This is achieved by measuring the scattered electron and the electron ejected from the target in a coincidence experiment. In these experiments the incident electron momentum k_0 is well-defined, and the momenta of the outgoing electrons k_a and k_b are measured. By time-correlating events from the collision, a fully differential cross-section (DCS) is determined for comparison to quantum models.

These processes are now well understood for lighter atomic targets, but some unsolved problems remain for heavy targets and for excited atoms. By contrast, measurements from molecular targets in the low energy regime are in their infancy, and so much can still be learned about these interactions. This work is challenging due to the complexity of molecular targets which may rotate and vibrate, and which are often oriented randomly in space. The nuclear distribution in molecules adds further complexity since the spherical symmetries applicable to atomic targets are no longer valid.

Recent work was carried out on CH_4 at an intermediate incident electron energy of 250eV in a joint collaboration between a number of groups [1]. The experimental measurements were compared to two different models which both agreed well with the data. By contrast, at lower energies (from 3 eV to 60 eV above the ionization threshold) a large difference is found between the models and experiment, which warrants further investigation [2,3]. These previous low energy measurements were carried out in both a coplanar geometry and in the perpendicular plane, and so to further explore this region new measurements are underway to measure the DCS in intermediate geometries.
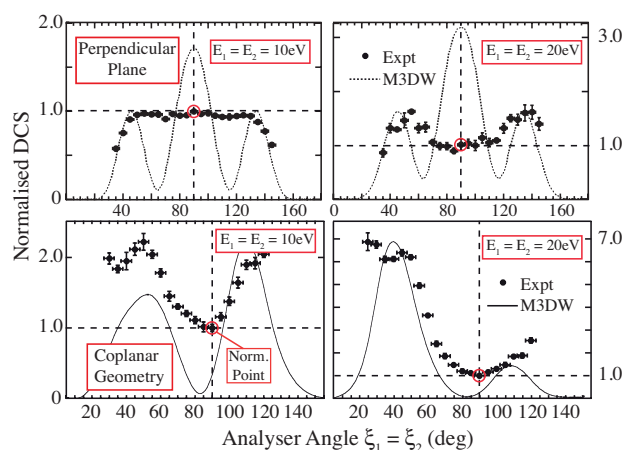

Figure 1. DCS measurements at 10eV and 20eV outgoing electron energies in a coplanar symmetric geometry and in the perpendicular plane. A common point at $\xi_1 = \xi_2 = 90°$ in all geometries allows the data to be inter-normalized at each energy. Results from an M3DW model are also shown.

Figure 1 shows examples of previous measurements in both planes, demonstrating the variation seen as k_0 is changed. Calculations from the M3DW model are also shown for comparison.

The results of this new experimental survey will be presented at the conference.

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Rearrangement of the atomic core in the two- and three-photon ionization of noble gas atoms

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Synopsis   Two- and three-photon ionization of the outer np- shell of the rare gas atoms in the below- and above- threshold ionization (BTI and ATI) regions was calculated. Large impact of many-electron correlations on the computed generalized photoionization cross sections (GPICS) is illustrated.

Since the increased interest to the investigation of the multiphoton ionization processes (see the review of early work in [1]) the precise calculation of the multiphoton PICSs becomes desirable.

Recently, a strong influence of the core polarization and many-electron correlations on the computed G2PICSs of Ar and Xe was determined in the BTI regions of Ar [2] and Xe [3].

Investigation of the two-photon ionization in the ATI region revealed strong influence of the Auger-like correlations on the near threshold G2PICSs of Ar [4, 5].

In the present paper, the reasons leading to the large impact of the core polarization and many-electron correlations on the near-threshold G2PICS of the rare gas atoms are analyzed. In case of the three-photon ionization many-electron correlations influence the PICS even larger than in the case of two-photon ionization. In Fig. 1, we illustrate an influence of the core relaxation on the G3PICS of the Kr $4p^5_{1/2}n'\ell$ autoionizing resonances comparing the frozen-core (FC)(b) and relaxed-core (RC)(c) results. One can recognize that core relaxation 'suppresses' the $nd'[5/2]_3$ resonances and changes the sign of the q parameter of the $nd'[3/2]_1$ resonances providing fairly well agreement between present theory and experiment [6] (see Fig. 1(a)).

Previous calculation within the multichannel quantum defect theory (MQDT) [7] (dotted-line in Fig. 1(b)) describes experiment much worse even with the MQDT parameters chosen from the energy position of resonances. The reasons resulting in the strong influence of the many-electron correlations on the G3PICS are analyzed.
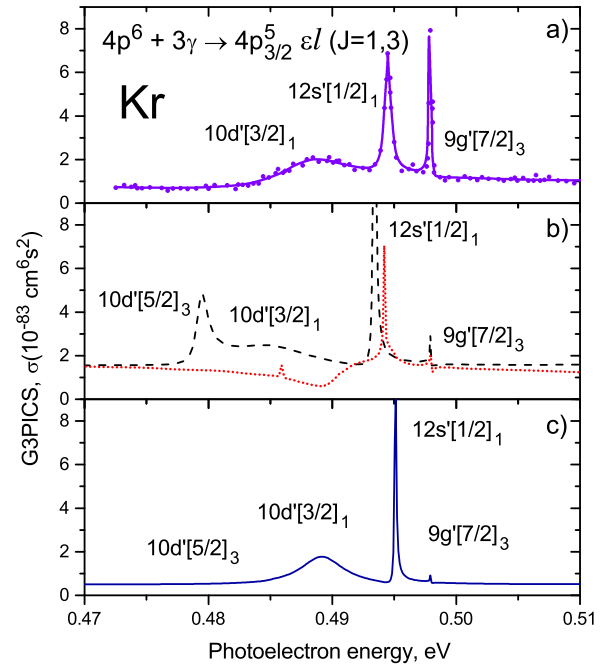

Figure 1. G3PICS of Kr computed in the FC (dashed-line in (b)) and RC (solid-line in (c)) approaches. Experimental data from [6] (a) and MQDT result from [7] (dotted-line in (b)) are shown in arbitrary units.

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Impact of the continuum electronic states on the van der Waals interactions

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Synopsis   In the present work, we have studied the impact of continuum electronic states on the van der Waals interactions. The results from our analytical and numerical calculations show considerable contribution from the unbound states to the van der Waals interaction between to atoms.

As two neutral atoms approach each other a correlation between the atomic states arises due to the mutually induced electric dipole moments. At leading order in the inverse distance between the atoms this is a dipole-dipole interaction. Such kind of weak interactions, known as van der Waals or dispersive forces, are originated from the spontaneous charge fluctuations of the electron cloud. Dispersive forces are the weakest and at the same time the most persistent of all electromagnetic interactions. For this reason they play an important role across molecular physics, self-assembly processes, protein folding, crystal formation.

The fluctuations of the charge density contain transitions not only to the bound excited electronic states but to the continuum states as well. The non-negligible role of the continuum states has already been pointed out by Dalgarno and Lews six decades ago [1]. This contribution is almost one-fifth of the total van der Waals energy between two hydrogen atoms. This continuum part is typically neglected in atomic models, e.g. quantum oscillators, which are widely used for evaluating van der Waals dispersion interactions between atoms in preceding studies [2]. Nevertheless, the contribution from the continuum states are neither included in general electronic structure calculations (DFT, Hartree-Fock) nor in the high-level correlated methods (CCSD(T), MRCI), because the computation of the continuum states is a big challenge due to the unbound nature of the problem.

In the present work, our purpose was to model atomic systems with simple one dimensional potentials possessing both bound and continuum electronic states. Then for two of such model atoms, we calculate van der Waals dispersion interaction energy by second-order perturbation theory. The results from our analytical and numerical investigations show non-negligible contributions from the continuum states to van der Waals interaction between atoms.

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Energy distribution of photoelectrons emitted from silver nanoparticles after laser irradiation

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Synopsis   We present theoretical study of photoelectron emission from silver nanoparticles after laser irradiation based on the classical simulation of the electron trajectories. The individual electron trajectories were analized with and without the image force between the emitted electron and its mirror charge taken into account. We show the calculated energy spectra in comparison with the available experimental data. Our theoretical curve is in agreement with the experimental observations. Moreover, we observed a significant effect of the image accelerationon the calculated electron spectra.

Electron emission from nanoparticles induced by laser excitation attract great attention during the last years [1,2]. It is not only because it comprises very interesting and challenging fundamental and basic science both in experimental and theoretical points of views, but these researches also promise many technical applications. Among them it can be used in plasmonic biosensors, optoelectronics, photovoltaics, construction of ultrafast nanoemitters.

We developed a computer simulation code based on the classical dynamics to calculate the electron trajectories emitted from a semi-ellipsoid protrusion of an Ag surface. At first, the time dependent electric field, generated by the incident laser pulse in Kretschmann configurationwas calculated using finite difference time domain simulation [3]. The laser pulse was an 800 nm, 5 fs plane wave. The angle of incidence was 44.6°.

We performed a large number of classical trajectory simulations in the pre-calculated electric field. During the simulations we are able to calculate the electron trajectories with and without the image force between the emitted electron and its mirror charge taken into account. We found that the image acceleration toward the surface plays a dominant role in shaping the electron trajectories [4]. We found also that the calculated energy distributions of photoelectrons are in agreement with the recent experiments (see Fig.1.). The experimentally obtained double peak structure was also identified in our calculated spectra.
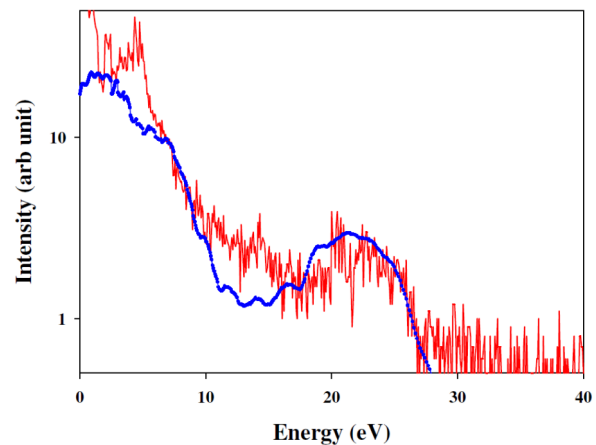

Figure 1. Comparison of the energy distribution of photoelectrons ejected from silver nanoparticles after laser irradiation at 230 GW/cm^2 focused beam intensity. Result of Monte Carlo simulation (thin solid line, red in color), the experimental data from ref. [22] (thick solid line, blue in color).

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Acousto-optic couplings in Sapphire phoXonic cavities

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Synopsis We present theoretical study of acousto-optic couplings mechanisms in phoXonic crystal. In our recent work two dimensional calculations were performed for Sapphire phoXonic cavities composed of air cylinders arranged periodically in a square arrangement on a plate of Sapphire. We show that both the elastic and the electromagnetic energy can be confined inside a cavity simultaneously. We also show that the optomechanical coupling for the moving interface and photoelastic effects can be in phase or not in phase thereby producing strengthening or weakening effects in the total optomechanical coupling.

The electronic technology arrived today at a limit of performances compared to the requirements imposed by the new communication and information technologies services. A great number of research turns their attention to a new materials and components allowing going up at frequencies inaccessible to the electronics field and which have micrometric dimensions in order to be integrated in embarked systems. The components containing photonic crystals seem a promising solution for the development of photonic integrated circuits.

The photonic crystals are structures formed of different dielectric mediums whose index of refraction is periodically modulated [1-3]. These new structures constitute a very promising solution for the design of high performance components of micrometric size, such as multiplexers, filters or photonic wave guides which can transport the light along strongly-curved trajectories.

In this work the acousto-optic couplings mechanisms in photonic crystal is studied. In this work two dimensional calculations were performed for Sapphire phoXonic cavities composed of air cylinders arranged periodically in a square arrangement on a plate of Sapphire [4]. Taking sapphire as a matrix component allows us to take advantage of the anisotropy of the dielectric tensor in the microwave regime. The goal is to study the acousto-optic coupling, based on both photoelastic and opto-mechanical mechanisms, in periodic structures with simultaneous photonic and phononic band gaps. The acousto-optic interaction generates a phonon thanks to the excitation of the cavity by the confinement of the optical wave. The aim in this field being to seek a maximum coupling of this interaction, and this also due to a strong confinement of the waves in the microcavities.

In our investigations, we have focused on the acousto-optic couplings inside a phoXonic cavity by taking into account two coupling mechanisms, i.e. the photoelastic effect and effect of movement of the interfaces. We discuss the importance of the symmetry of modes to distinguish those that don't interfere in an effcient way. We calculate the modulation of the frequency of the photonic mode during a period of acoustic oscillations. We show that both the elastic and the electromagnetic energy can be confined inside a cavity simultaneously. We also show that the optomechanical coupling for the moving interface and photoelastic effects can be in phase or not in phase thereby producing strengthening or weakening effects in the total optomechanical coupling.

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Semiclassical two-step model and ionization of the hydrogen molecule by a strong laser pulse

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Synopsis We extend the semiclassical two-step model for strong-field ionization to molecules. For the hydrogen molecule oriented along the polarization direction of the linearly polarized laser pulse we predict significant deviations of the electron momentum distributions and energy spectra from the case of atomic hydrogen. We find that at the same parameters of the pulse the energy spectrum falls off slower with increasing energy, and the holographic interference fringes are more pronounced for the hydrogen molecule than for the hydrogen atom.

The semiclassical two-step model for strong-field ionization (SCTS) allows to describe quantum interference effects and accounts for the ionic potential beyond the semiclassical perturbation theory [1]. We extend the SCTS model to ionization of the H_2 molecule by a strong linearly polarized few-cycle laser pulse [2]. In our simplest implementation of the model, the molecule is oriented along the laser polarization direction, and the electron moves in the laser field and in the Coulomb fields of two fixed atomic nuclei with equal effective charges of 0.5 a.u.

The comparison of the two-dimensional photoelectron momentum distributions calculated for H_2 and H shows that for the same laser parameters the distributions generated from the molecule are more extended along the polarization axis than the ones from the hydrogen atom. Therefore, the electron energy spectrum for the hydrogen molecule falls off slower than for the hydrogen atom. We also show that the holographic interference fringes in the electron momentum distributions are more pronounced for H_2 than for H.

Furthermore, we compare our results with the predictions of the quantum trajectory Monte Carlo model (QTMC) [3] for H_2. We find that similarly to the atomic case the QTMC predicts fewer radial nodal lines in the fanlike interference structure that is characteristic for the low-energy part of the momentum distributions. We attribute this to the fact that QTMC underestimates the Coulomb potential in the phase associated with each classical trajectory.
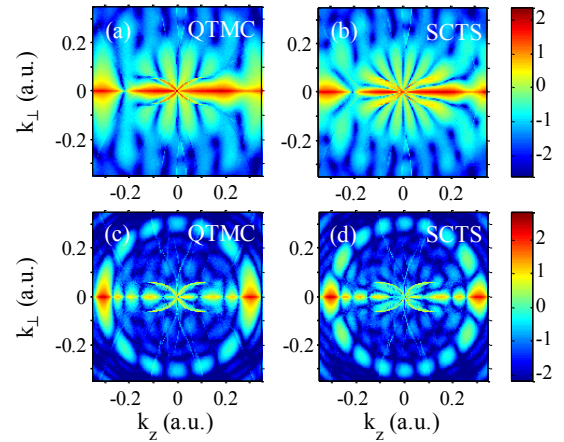

Figure 1. The low-energy part of the two-dimenstional photoelectron momentum distribution for H_2 ionized by a laser pulse with duration of 8 cycles, wavelength of 800 nm, and intensity of 2.0×10^{14} W/cm^2. Panels (a, c) show the predictions of the QTMC model. Panels (b, d) displays the results of the SCTS model. Panels (a, b) and (c, d) correspond to the sine squared and trapezoidal pulse, respectively. The distributions are normalized to the total yield. The laser pulse is linearly polarized along the z axis.

The present SCTS model for H_2 can be extended to an arbitrary orientation of the molecule, polarization of the field, as well as to other molecules, e.g., heteronuclear or polyatomic molecules.

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Studies of Cold ^{39}K Rydberg Atoms in an AC Magneto-Optical Trap

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Synopsis   Cold ^{39}K atoms are studied using an AC-Magneto-Optical Trap (AC-MOT) whose trapping field can be switched on and off over 300 times faster than a conventional DC-MOT. Rydberg excitation of ^{39}K is carried out during the time the field is off, the atoms being detected using field and collisional ionization. The experiment can study Rydberg atoms in states with principal quantum numbers ranging from n = 5 to n > 200.

An alternating current magneto-optical trap (AC-MOT) [1] is used to study potassium atoms excited to high Rydberg states using combined radiation from two continuous wave (CW) Ti:Sapphire (Ti:S) lasers and a CW blue laser operating around 450 – 460 nm. One of the Ti:S lasers is tuned to ~766.701 nm for cooling and trapping of atoms using the closed $4^2S_{1/2}$(F=2) to $4^2P_{3/2}$(F=3) transition in ^{39}K. The second Ti:S laser is tuned to the $4^2P_{1/2}$ state at ~770.1 nm, and the blue laser is resonant either from the $4^2P_{3/2}$ or $4^2P_{1/2}$ state to the Rydberg states under study.

In these experiments a re-circulating oven produces a quasi-collimated beam of potassium atoms emitted with an rms speed ~580 m/s. The atomic beam is injected into a Zeeman slower which cools the atoms so they exit at a speed of ~30 m/s. The atoms load the AC-MOT at a rate of ~1.4×10^8 atoms/s. The AC-MOT magnetic field is produced by an alternating current supply and the polarization of the optical molasses is switched from σ^+ to σ^- at the same frequency to ensure efficient trapping of atoms in the MOT.

The AC-MOT traps ~2×10^{10} atoms/cm^3 at a temperature of ~350 µK with a trap lifetime up to 400 s. The trapping magnetic field can be switched off in less than 20 µs, over 300 times faster than for a conventional DC-MOT. The optical molasses beams can also be switched on and off using a custom-built optical chopper [2] which has a phase jitter of below 0.5° when operated at the rate used in these experiments. Stray electric fields in the MOT are eliminated using electrostatic plates.

During the AC-MOT off state (600 µs), a second Ti:S laser (operating at ~770.1 nm) is switched on to excite the trapped atoms. Blue light from a Matisse dye laser or diode laser is then injected to excite Rydberg atoms from the $4^2P_{1/2}$ or $4^2P_{3/2}$ states. Ions produced by collisional or field ionization are subsequently extracted by a pulsed field and are detected by a channel electron multiplier (CEM) as a function of the blue laser frequency.

Figure 1 shows a simplified diagram of the apparatus. Cold Rydberg atoms in states ranging from 5D to 225S have been observed [3]. For atoms in states n > 100, the spectra shows evidence of perturbations due to the interaction of Rydberg atoms in the trap, with the dipole forbidden $n^2P_{1/2}$ and $n^2P_{3/2}$ peaks being clearly observed together with atoms in higher nL states (with $L \geq 2$).

In our current experiments we will explore these dipole forbidden Rydberg states to ascertain the origin of the perturbations. We will further study the spectral region beyond n ~ 200, since in this region there is a significant overlap between cold atoms in the trap due to their very large size (individual atoms in n = 225 have a diameter of more than 5 µm). The status of these new experiments will be presented at the conference.
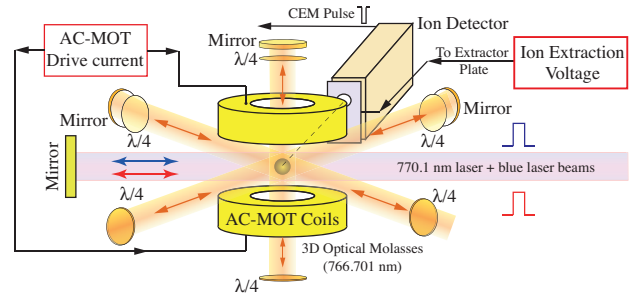

Figure 1. Rydberg excitation in the AC-MOT. A custom-built chopper switches the molasses beams into the experiment. In this example Rydberg excitation uses combined 770.1nm and blue radiation retro-reflected through the interaction region. Ions from collisional or field ionization are extracted using a pulsed extraction-field and are detected using a channel electron multiplier (CEM).

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Triple-differential cross sections for electron-impact ionization of tetrahydrofuran at low projectile energy

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Synopsis Triple differential cross sections (TDCSs) for the electron-impact ionization of the highest occupied molecular orbital (HOMO) of tetrahydrofuran (THF) have been measured utilizing the reaction microscope at projectile energy E_0 = 91 eV. The experimental data are compared with predictions from the multi-center distorted-wave (MCDW) and molecular 3-body distorted-wave (M3DW) approaches.

Radiation damage of biorelevent molecules has captured considerable interest in the past decade due to its importance in medical radiation therapy [1]. It is accepted that the electrom impact process plays an important role in damaging biomolecules. Thereby, electron impact triple differential cross sections (TDCS) are highly demanded for considering the ionization and subsequent fragmentation processes in biological tissue.

Tetrahydrofuran (THF, C_4H_8O) is often regarded as a molecular analog of the deoxyribose sugar-ring in the DNA backbone for the modelling [2]. In this work, we carried out a low energy (91 eV) electron-impact ionization experiment of THF using the Reaction Microscope (ReMi) technique [3]. The TDCSs are determined by covering a large part of the full solid angle for the ejected electrons. The ionization of the HOMO of THF is identified by coincidence detection of two outgoing electrons and the stable parent ion, i.e., THF$^+$ [4]. The experimental data are compared with theoretical predictions from two different approaches, i.e., the MCDW and M3DW models. In MCDW, the continuum wavefunction of the slow ejected electron is considered as a distorted wave and plane waves are adopted to describe the incoming and scattered ptojectile. In M3DW, all of the three continuum electron are described by distorted-waves.

In Fig. 1 the experimental results as well as MCDW in which the post-collision interaction (PCI) effect is not included and MCDW-Nee considering the PCI effect using the Gamow factor are shown. The results are selected in the scattering plane which is defined by the momentum vectors of the incident and scattered projectile. MCDW-Nee gives overall better agreement than MCDW indicating that the PCI effect is important. The details of this work will be discussed atthe conference.
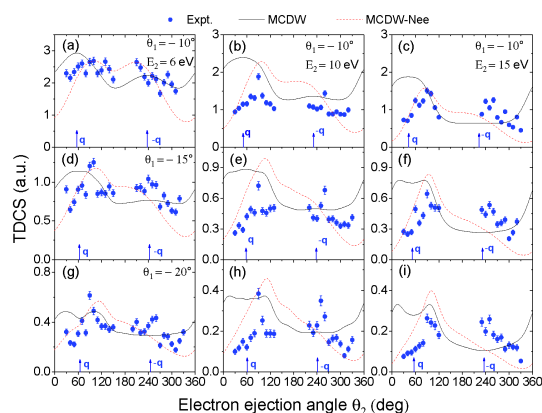

Figure 1. Comparison between experimental TDCS and MCDW as well as MCDW-Nee calculations for the scattering plane geometry. The arrows marked as **q** and –**q** indicate the angles of the momentum transfer and its opposite direction.

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Production of molecular dications by electron impact investigated by (e, e+ion) measurement

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Synopsis   We present (e, e+ion) measurements for double ionization of nitrogen and oxygen molecules employing the reaction microscope in which the momentum vectors for final-state charged reaction products are determined experimentally. By measuring the final state electrons in coincidence with the molecular dication, the related electronic states and, thus, the production mechanisms of these dications could be identified and studied in detail.

When a molecules is populated to doubly charged states, it is usually unstable and may undergo fragmentation. However it is also possible that the molecular dication is metastable and could survive for a quite long time. The mechanisms for formation of metastable dications are of great interest in many applied fields, such as plasma devices, radiation chemistry and biology, planetary atmospheres and the physics of cometary tails. In this respect, studies of electron-molecule collisions have made tremendous contributions to our understanding of the reaction mechanisms [1].

In an electron-impact ionization process, the molecular dications can be produced by two different pathways, (1) the direct double ionization process with both electrons being removed in the collision with the projectile and (2) the Auger process following single ionization of an inner shell electron. In Refs. [2,3] these two pathways are identified according to the ratio between doubly charged and singly charged parent molecules as function of the incident electron energy. If both the parent dication and the scattered/emitted electron from one collision event could be measured in coincidence, then the related electronic state information could be obtained via the energy spectra of scattered projectile and emitted electron. This will provide direct information to determine the pathways for formation of metastable dications.

We have obtained such a measurement using the reaction microscope technique in which the momentum vectors and, consequently, the kinetic energies for all final-state charged reaction products are determined in coincidence. In our measurements the molecular nitrogen (N_2) or oxygen (O_2) dication produced in collisions with electrons of 120 eV impact-energy is detected in coincidence with either the scatted electron or one of the emitted electrons, in a so-called (e, e+ion) measurement. During the off-line data analysis, the related electronic states could be determined through the energy loss spectrum while the energy distribution of the emitted electron may present the typical features of an Auger process if the dication is produced through pathway (2). Figure 1 presents the time-of-flight (TOF) vs. position 2D plot of the doubly charged and singly charged parent cations of N_2. The background signal around the N_2^{2+} dication is quite weak, indicating that N^+ fragments are well eliminated in our measurement. Details of this study will be presented at the conference.
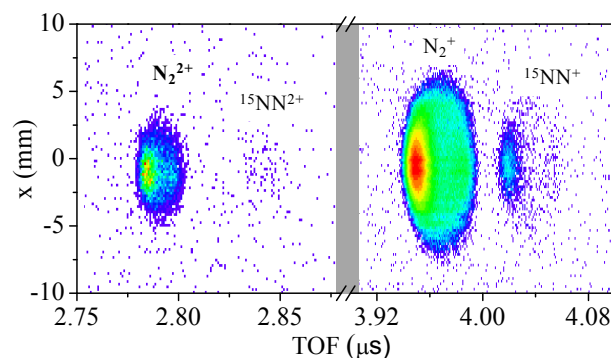

Figure 1. TOF vs. x-coordinate of the impact position for doubly and singly charged parent cations of N_2.

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Application of 'dressed' convoluted quasi-Sturmian functions to the double ionization of atoms

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Synopsis   The two electron continuum problem is dealt with an *ab initio* approach based on an expansion on convoluted quasi Sturmian functions (CQS) equipped with an appropriately built electron-electron phase factor. Such 'dressed' CQS functions constitute a suitable basis to solve Coulomb three-body scattering problems as illustrated through numerical applications to double ionization processes on Helium. They put in evidence, once more, the importance of electron-electron correlation over the whole space.

We are interested in the Coulomb three-body scattering problem which results from the double ionization of Helium by impact of fast projectiles or the double photoionization; to first order, such a process reduces to solving a non-homogeneous Schrödinger equation

$$\left[E - \hat{H}\right] \Phi^{(+)}(\mathbf{r}_1, \mathbf{r}_2) = \hat{W}_{fi}\Phi^{(0)}(\mathbf{r}_1, \mathbf{r}_2) \quad (1)$$

with a three-body Helium Hamiltonian $\hat{H}$. Assumed to be initially in the Helium ground state $\Phi^{(0)}$, after the double ionization process the two electrons escape from the nucleus with total energy $E = k^2/2$; $\hat{W}_{fi}$ is the interaction operator related to the studied process.

We propose to solve equation (1) with a truncated expansions in CQS functions [1, 2]. To make the expansion converge, we dress the 'bare' CQS functions with a phase factor $e^{i\mathcal{W}}$, such that $\mathcal{W}$ approaches the Coulomb phase $-\frac{1}{k}\frac{\rho}{r_{12}}\ln(2k\rho)$ in the limit $\rho \to \infty$ ($\rho = \sqrt{r_1^2 + r_2^2}$); r_{12} is the interelectronic distance. To test our approach and compare it with other *ab initio* methods, we calculate fully differential cross sections (FDCS) for $(\gamma, 2e)$ and $(e, 3e)$ processes on Helium. We demonstrate that our 'dressed' CQS functions constitute a suitable basis which allows us to provide converged and sensible partial ionization amplitudes, and thereafter cross sections [1, 2]. This is in contrast with 'bare' CQS which also yield cross sections but with amplitudes having a divergent phase.

In the $(e, 3e)$ example shown in Fig. 1, our FDCS calculated with the 'dressed' CQS are compared with experimental data [3], and with results obtained with the CCC [4] and GSF [5] methods. The comparison between such *ab initio* approaches, all solving exactly the same problem, illustrates the mathematical and numerical difficulties associated with the Coulomb three-body problem.
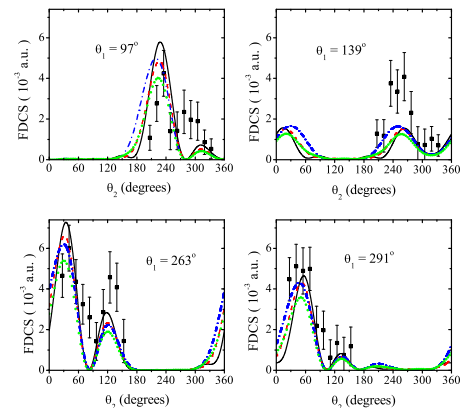

Figure 1. FDCS as a function of θ_2 for four fixed θ_1 values, in the coplanar geometry and kinematical situation of the Orsay experiments [3, 4]: $E_1 = E_2 = 10$ eV and $q = 0.24$ a.u.. Experimental data in absolute scale [3, 4]; the 'bare' CQS functions representation (solid dots); 'dressed' CQS (solid lines); CCC [4] (dashed lines); GSF [5] (dot-dashed lines). Al theoretical results are upscaled by a factor 2.2.

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Laser-assisted electron scattering and ionization processes in a quasi-Sturmian-Floquet approach

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Synopsis We demonstrate the robustness and efficiency of our recently formulated quasi-Sturmian-Floquet method to treat light-dressed electron states. For this purpose we study the archetypical processes such as laser-modified electron scattering on a Coulomb potential and laser-assisted electron-impact ionization of atomic hydrogen. For the latter, we identify modifications due to the laser field that could be experimentally observed.

We present our recently formulated non-perturbative method [1, 2] for the theoretical treatment of laser-modified electron states in scattering and ionization phenomena in the presence of intense laser fields. Our method is based on the Hermitian Floquet theory in the Kramers-Henneberger representation. We use the quasi-Sturmian functions [3] taken in parabolic coordinates to solve the system of coupled Lippmann-Schwinger-Floquet equations for the laser-modified electron Coulomb wave function.

We tested our approach by investigating theoretically (i) laser-modified electron scattering on a Coulomb potential and (ii) a laser-assisted $(e, 2e)$ collision on atomic hydrogen:
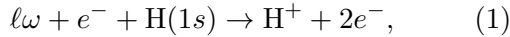
$$\ell\omega + e^- + H(1s) \rightarrow H^+ + 2e^-, \qquad (1)$$

where a net number ℓ of photons of frequency ω can be exchanged between the projectile-target system and the external field. Other laser-assisted processes, such as photoionization, will be considered in a near future.

As an example, for process (1), the expected typical pattern of the ejected-electron angular distribution in the scattering plane (with a binary and a recoil peak in the $\pm\mathbf{q}$ directions) is illustrated in Fig. 1 for two regimes of laser radiation. The figure shows (i) how the calculated laser-assisted triple differential cross section (TDCS) converges very fast, especially for the perturbative regime, with increasing the number $\mathfrak{N}_f$ of Floquet components of the ejected-electron state; (ii) that the modification due to the presence of laser radiation should be observable ex-perimentally in the non-perturbative regime.
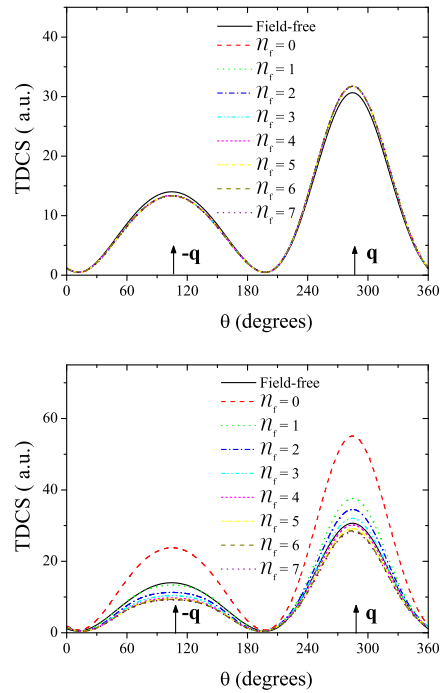
Figure 1. Laser-assisted TDCS as a function of the ejected-electron angle θ for the coplanar geometry $(e, 2e)$ process when $\ell = 0$ photons are exchanged: (top panel) perturbative regime (quiver amplitude $a_0 = 0.1$) and (bottom panel) nonperturbative regime ($a_0 = 5$) of laser radiation with frequency $\omega = 0.1$ and the polarization vector perpendicular to the scattering plane. The scattered electron is fixed at $\theta_s = 0.44°$ and has energy $E_s = 5$ keV, so that the momentum transfer $q = 0.15$.

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Coulomb explosion and fragmentation Dynamics of CH$_3$I following single photon absorption by synchrotron radiation

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Synopsis   We observed the concerted photodissociation of CH$_3$I producing multiply-charged atomic ions. The mean stretching of the I–C bond during the relaxation was determined by a Coulomb explosion model.

When a high energy photon is absorbed in the inner-shell of molecules, many electrons can be ejected via cascade Auger relaxation. As a result, highly charged ion can be formed which can lead to the dissociation into atomic or molecular ions. The total charge created will be distributed on the induced fragments.

In the present work, the photodissociation processes in gas-phase Methyl iodide (CH$_3$I) by tender X-ray photons are studied. CH$_3$I has become a model system for studying photoinduced charge transfer due mainly to the relative simplicity of this small organic molecule containing a heavy halogen atom, its C$_{3V}$ geometry, the importance of fragmentation dynamic in physico-chemistry [1, 2].

Our measurements were obtained by single photon absorption on the GALAXIES beamline at SOLEIL synchrotron. We have measured the momentum vector of the ionic fragments using time and position sensitive detector [3], after ionization of iodine at 2s and 2p edges. The supersonic molecular jet of CH$_3$I crosses perpendicularly with the photon beam. We applied electron – ions coincidence technique (PE3PICO) to elucidate the fragmentation mechanisms. We demonstrate that photoabsorption of in deep inner-shell L of iodine followed by multiple Auger decay leads to the creation of highly charged atomic ions (C^{m+}, I^{n+} and H$^+$) where the total charge of the ions produced can reach up to +15. This indicates that efficient (and fast) electron rearrangement mechanisms are at work in polyatomic systems.

Using different types of analysis, such as the correlation of linear moments and kinetic energies shared between three ions measured in coincidence, we characterized the photodissociation dynamic and we deduced a concerted fragmentation mechanism for all the ionic pathways obtained.

We also used a Coulomb explosion model of instantaneous charge creation to provide more insight into N-body fragmentation dynamics [4]. These simulations made it possible to support the concerted break up observed in CH$_3$I. From a detailed analysis of the experimental kinetic energies released and simulated ions we were able to give an estimate of the average stretching of I – C bond during the relaxation. In addition, it has also been found that the direction of carbon ejection is affected by the early departure of hydrogen.

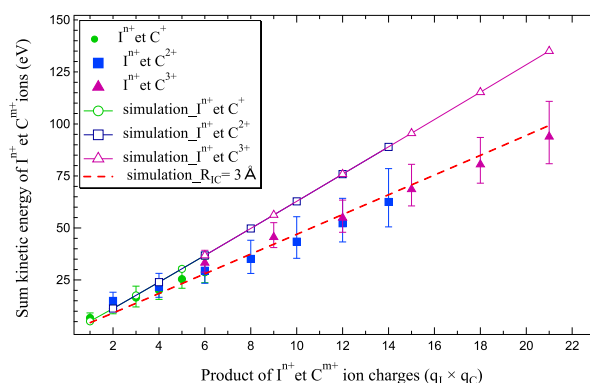**Figure 1.** Measured and calculated sum kinetic energy of I and C ions as a function of the product of their charge states.

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