A 15: Poster: Interaction with strong or short laser pulses

Time: Monday 16:30–18:30 Location: Spree-Palais

A 15.1 Mon 16:30 Spree-Palais

Time-Dependent Generalized Active Space Configuration Interaction Approach to Ultrafast Dynamics in Atoms and Molecules — •Sebastian Bauch[1,2], Lasse Kragh Sørensen[2], and Lars Bojer Madsen[2] — [1]Institut für Theoretische Physik und Astrophysik, Christian-Albrechts-Universität zu Kiel, Leibnizstrasse 15, 24098 Kiel, Deutschland — [2]Institut for Fysik og Astronomi, Aarhus Universitet, Ny Munkegade 120, 8000 Aarhus C, Danmark

The progress in experiments using ultrashort and strong laser pulses demands for the development of theoretical tools for the time-dependent description of multi-electron targets including electron-electron correlations. In this contribution, we address one approach to the time-dependent many-electron problem: the time-dependent generalized active space configuration interaction (TD-GAS-CI) approach [1] within a mixed basis set consisting of a localized bound part and a discretized continuum [2]. We demonstrate the capabilities of the method by comparing to exact solutions of the multi-particle time-dependent Schrödinger equation and address the question of the choice of an appropriate single-particle basis set, in particular Hartree-Fock and natural orbitals. Photoionization cross-sections and higher-harmonic generation in multi-electron model systems are discussed.

[1] T. Fleig et al., J. Chem. Phys. 114, 4775 (2001)
[2] D. Hochstuhl and M. Bonitz, Phys. Rev. A 86, 053424 (2012)

A 15.2 Mon 16:30 Spree-Palais

Time resolved investigation of non-sequential double ionization using two-color laser-pulses — •Nicolas Camus[1], Lutz Fechner[1], Andreas Krupp[1], Joachim Ullrich[1,2], Thomas Pfeifer[1], and Robert Moshammer[1] — [1]Max-Planck-Institut für Kernphysik, Saupfercheckweg 1, 69117 Heidelberg — [2]Physikalisch-Technische Bundesanstalt, Bundesallee 100, 38116 Braunschweig

We present an experiment where two laser pulses (800+400 nm) with controllable time-delay are used to investigate double ionization of atoms. The polarization of the weaker blue field is chosen to be perpendicular to the one of the more intense IR field. Due to the different intensities, ionization is assumed to result from the red component with the blue field causing a streaking of the electrons in the transverse direction. Information about the time the electrons escape can be extracted from the momentum component parallel to the blue field and its variation with the relative phase between the laser pulses. Using a Reaction Microscope, we are able to measure the three-dimensional momentum vectors of all particles in coincidence and determine the streaking traces as a function of the time-delay. Applied to non-sequential double ionization we are able to distinguish and to identify different ionization pathways.

A 15.3 Mon 16:30 Spree-Palais

Orthohelium in laserfields beyond TDDFT: doubly-excited states made simple using TDRNOT — •Julius Rapp, Martins Brics, and Dieter Bauer — Institut für Physik, Universität Rostock

Time-dependent renormalized natural orbital theory (TDRNOT) is a novel many-particle method [1] to investigate correlated dynamics of quantum systems. We explore the capabilities of TDRNOT and benchmark its performance regarding the application to an exactly solvable one-dimensional helium model atom in both singlet and triplet configuration. Despite being in an early stage of development, TDRNOT outperforms "mature" techniques such as practicable time-dependent density functional theory (TDDFT) concerning the description of highly correlated phenomena such as doubly-excited states, autoionization, and Fano profiles. Moreover, the treatment of, e.g., Rabi oscillations is more intuitive than it is within TDDFT, considering the exotic features of the exact exchange-correlation (XC) functional there such as inital-state dependence, non-locality in time, and similar.

We discuss the result of a recent modification to the TDRNOT scheme [2] which substantially improves the description of the spin-triplet in particular. One remarkable feature compared to TDDFT is the presence of doubly-excited states in the linear response spectrum, whose energies approach the correct values if enough natural orbitals are included in the calculation. As an outlook, we also address dynamics induced by laser fields beyond linear response.

[1] M. Brics, D. Bauer, Phys. Rev. A **88**, 052514 (2013).
[2] J. Rapp, M. Brics, D. Bauer (submitted).

A 15.4 Mon 16:30 Spree-Palais

Line shape modifications of singly-excited Helium states coupled to a ponderomotive continuum — •Veit Stooss, Andreas Kaldun, Christian Ott, and Thomas Pfeifer — Max-Planck-Institut für Kernphysik, 69117 Heidelberg, Germany

In order to understand and control the dynamics of correlated electron systems as they appear in atoms and molecules it is instructive to study the most fundamental case, namely two interacting electrons bound to an atom. Here, we investigate the special case of singly-excited states in Helium and their laser coupling to a ponderomotive continuum. The line shapes observed in attosecond transient absorption spectra reveal signatures of both bound-to-continuum and bound-to-bound laser coupling. To better understand the full laser coupling dynamics a model simulation was carried out in which the states of the system are excited by an extreme ultraviolet (XUV) laser pulse and coupled by a femtosecond infrared (IR) pulse. The simulation includes the s-, p-, and d-series of singly-excited helium. The completed simulation allows for varying the infrared intensity of the coupling pulse or the relative time delay between XUV- and IR-pulse as control parameters. In addition the coupling between the states as well as the coupling to the continuum can be activated separately to study the relative contributions of the different coupling channels. The results are discussed within the recently developed Fano-phase formalism [1] which shows that resonance line shapes can be used to extract the laser intensity.

[1] C. Ott et al., Science 340, 716 (2013)

A 15.5 Mon 16:30 Spree-Palais

Coulomb effects in two-color atomic ionization and "the phase of the phase" in the photoelectron yield — •Mohammad Adel Almajid and Dieter Bauer — Institut für Physik, Universität Rostock, 18051 Rostock

The plain strong field approximation (SFA) fails in reproducing *ab initio* spectra obtained by solving the time-dependent Schrödinger equation (TDSE) exactly. There have been various attempts to include Coulomb effects into the direct SFA, one of them being the Coulomb-Volkov approximation (CVA). In our work, we compare TDSE and CVA-SFA photoelectron spectra, in particular for two-color, colinearly polarized pulses, where the relative phase between the two laser fields affects the photoelectron dynamics. A pronounced disagreement between exact TDSE and CVA-SFA results is found for the so-called direct electrons that move towards the detector without hard rescattering at the parent ion. The rescattered electrons instead are well described already by the SFA, extended for the rescattering matrix element. We analyze the photoelectron spectra by Fourier-transforming the momentum-dependent yield as a function of the relative phase between the two pulses, thus obtaining "the phase of the phase". Plotted vs the momenta parallel and perpendicular to the laser polarization direction, this entity tells how the yield of photoelectrons with a certain final momentum is synchronized with respect to changes of the relative phase.

A 15.6 Mon 16:30 Spree-Palais

Ionization of small Argon clusters in intense laser pulses at different wavelengths — •Mehrdad Baghery, Ulf Saalmann, and Jan-Michael Rost — Max Planck Institute for the Physics of Complex Systems, Dresden, Germany

Electrons in clusters, after absorbing energy from a laser field, may not be bound to a specific atom any more, but may still be bound to the cluster as a whole. These electrons may then absorb further energy through inverse Bremsstrahlung and cause other electrons to ionize through collisions. These phenomena make the dynamics of clusters non-trivial.

Since hybrid quantum-classical calculations are usually computationally less expensive than those of a full quantum description, a hybrid quantum-classical model which reproduces quantum predictions may be beneficial to future research. We are presenting such a model for small Argon clusters interacting with intense laser pulses.

A 15.7 Mon 16:30 Spree-Palais

A Two-Color XUV-IR Pump-Probe Scheme for the Study of Xenon Double Ionization — •Alexander Sperl[1], Andreas Fischer[1], Philipp Cörlin[1], Michael Schönwald[1], Arne

Senftleben[2], Thomas Pfeifer[1], Joachim Ullrich[3], and Robert Moshammer[1] — [1]Max-Planck-Institut für Kernphysik, Heidelberg — [2]Universität Kassel — [3]Physikalisch-Technische Bundesanstalt, Braunschweig

The ionization of rare gas atoms with extreme-ultraviolet (XUV) attosecond laser pulses in the presence of a strong infrared (IR) laser field has been studied frequently and, moreover, it can be used to characterize both the XUV and IR laser fields [1]. Here, we combine a XUV-IR laser system with a reaction microscope and we apply this technique to xenon. With the available XUV-photon energies ranging from 17-40 eV double ionization of xenon (Ip = 33.1 eV) becomes possible and thus enables the investigation of correlated two-electron transitions as a function of the relative phase-shift between the XUV and IR pulses. In the experiment we detect event by event the doubly-charged Xe++ ions in coincidence with the two electrons and we fully reconstruct the three-dimensional momentum vectors and the final kinetic energies of all particles [2]. In the poster we present data on double ionization of Xe in combined XUV and IR laser fields and draw conclusions about the two-electron emission process by analyzing electron angular distributions as well as energy correlation spectra.

[1] H. G. Muller et. al., Appl. Phys. B 74, 2002
[2] O. Guyétand et. al., Appl. Phys. B 41, 065601, 2008

A 15.8 Mon 16:30 Spree-Palais

Imaging collective and uncorrelated electron motion in an FEL-induced nanoplasma — •M Müller[1], J-P Müller[1], M Sauppe[1], L Flückiger[1], A Ulmer[1], B Langbehn[1], T Gorkhover[1,2], C Bostedt[2], I Barke[3], H Hartmann[3], S Toleikis[4], S Düsterer[4], I Rosenow[4], K-H Meiwes-Broer[3], D Rupp[1], and T Möller[1] — [1]TU Berlin — [2]LCLS@SLAC — [3]Uni Rostock — [4]FLASH@DESY

Free-electron lasers (FELs) enable for the first time imaging of single non-crystallizable nanosized particles by elastic light scattering. Due to the high power density involved, the target is almost immediatly strongly ionized. The subsequent formation of a nanoplasma within the sample on the timescale of the pulse implies changes on the scattering picture.

To disentangle the influence of the nanoplasma on the imaging process we performed two-color pump-probe experiments on single large xenon clusters at the FLASH FEL. An intense IR-pulse prepares a highly ionized plasma state with uncorrelated electron motion which is then imaged by an FEL pulse. As a next experimental step, an IR-double pulse can be used to preexpand a cluster to resonant density and drive a collective motion of the quasi-free electrons while a simultaneously incident FEL pulse images the so called Mie plasmon. The experimental setup and preliminary results of this most recently performed experiment will be presented.

A 15.9 Mon 16:30 Spree-Palais

Comparison of split-operator methods for solving the three-dimensional time-dependent Schrödinger equation — •Paul Struszewski, Julius Rapp, and Dieter Bauer — Institut für Physik, Universität Rostock

We study numerical properties of different split-operator methods such as exponential operator splitting (EOS) and alternating direction implicit (ADI) for solving the three-dimensional time-dependent Schrödinger equation (TDSE) in Cartesian coordinates. The aim is to obtain accurate ab initio results for the interaction of a lithium model system with an intense laser field in order to benchmark methods such as time-dependent density functional theory (TDDFT) in several approximations. The operator splitting allows for a high-order time propagation via the solution of *tridiagonal* linear equations only, which performs very well and can be directly parallelized. In addition, a highly adjusted numerical grid is used to minimize storage space and computing time.

The results provide general indications which operator splitting method is most suitable for solving the TDSE in three dimensions. As expected, the unitary EOS is perfectly norm-conserving, whereas the ADI is not. However, for ADI the variation of norm over time is orders of magnitude smaller in comparison to an explicit scheme. For cases in which ADI is faster than EOS, ADI offers a good compromise between numerical effort and norm-conservation.

A 15.10 Mon 16:30 Spree-Palais

Reconstructing the shape of single nanoclusters imaged with highly intense X-ray pulses — •A. Ulmer[1], L. Flückinger[1], T. Gorkhover[1], B. Langbehn[1], J.P. Müller[1], M. Müller[1], D. Rupp[1], M. Sauppe[1], A. Schreider[1], C. Bostedt[2], I. Barke[3], H. Hartmann[3], K.H. Meiwes-Broer[3], S. Toleikis[4], S. Düsterer[4], R. Treusch[4], and T. Möller[1] — [1]TU Berlin — [2]LCLS@SLAC — [3]Uni Rostock — [4]DESY Hamburg

Free-Electron Lasers (FELs) provide coherent highly intense and short pulses which make it possible for the first time to analyze the morphology of non-periodic or non-crystallizable nanoparticles by elastic light scattering. In addition to shape and structural information the optical properties (dielectric function) and their changes during the light pulse can be imaged.

We recently performed single shot experiments on single large clusters at the Free-Electron LASer Hamburg (FLASH) in a NIR/XUV pump-probe configuration. Reconstruction of the scattering patterns will give insight in both the growth process of the particles as well as the ionization dynamics in the irradiated clusters.

As the phase information is lost due to the imaging process it has to be retrieved using sophisticated techniques [1]. The information in the scattering patterns is limited by experimental constraints such as a necessary center hole and the limited detector size. Possible approaches combining scattering simulations and phase retrieval algorithms will be discussed.

[1] S. Marchesini et. al., Phys. Rev. B 68, 140101 (2003)

A 15.11 Mon 16:30 Spree-Palais

Implementation of Infinite-Range Exterior Complex Scaling in B splines — •Alvaro Magana and Alejandro Saenz — AG Moderne Optik, Institut für Physik, Humboldt-Universität zu Berlin, Newtonstraße 15, 12489 Berlin, Germany

The traditional box-discretization method for describing atoms and molecules in strong laser fields can lead to an inacurrate description of the system due to unphysical reflections at the finite box boundary. The problem can be cured by choosing a sufficiently large box size to describe correctly all the dynamics of the system, but this in turn demands very large computational efforts. The Infinite-Range Exterior Complex Scaling method (Ir-ECS) [1] has been proposed as a way of solving exactly the time-dependent Schrödinger equation (TDSE) within the physically most relevant region, dramatically reducing the numerical effort. In contrast to previous work in which the inner part of the wavepacket was described with a finite-element approach, we successfully implemented Ir-ECS using a mixed B-spline/Legendre basis in order to solve the TDSE. We report a drastic reduction of the total number of states needed for performing a time propagation using the spectral *ansatz*.

[1] A. Scrinzi, Phys. Rev. A 81, 053845 (2010).

A 15.12 Mon 16:30 Spree-Palais

Contributions from different orbitals to the high-harmonic spectra of N_2 — •Étienne Plésiat[1], Piero Decleva[2], and Alejandro Saenz[1] — [1]AG Modern Optics, Institut für Physik, Humboldt-Universität zu Berlin, Newtonstraße, 15, 12489 Berlin, Germany — [2]Dipartimento di Scienze Chimiche, Universita' di Trieste, Via L. Giorgieri 1, 34127 Trieste, Italy

In the last decade, high-harmonic generation (HHG) is supposed to become a fantastic tool for obtaining information about the electronic and geometric structure of molecules on the ultrafast time scale. Since the rate of the tunnel ionization depends exponentially on the ionization potential, the HHG spectra were thought to reflect the contribution of the highest occupied molecular orbital (HOMO) only. However, in 2008 the predominance of the HOMO-1 of N_2 in the cut-off region has been observed for the first time [1]. From then on, a particular attention has been payed to the contributions of molecular orbitals with lower binding energies. They have been shown to be responsible for dynamical minima in the plateau region of CO_2 [2]. Theoretical studies generally employ models based on the strong-field approximation (SFA) which are computationally fast but sometimes also inaccurate. In this work, we investigated the multiple-orbital contributions to the HHG spectra of N_2 by solving the TDSE in the single-determinant approximation in a basis of Kohn-Sham orbitals. Accurate results obtained with this method will be presented during the conference.

[1] B. K. McFarland *et al.*, *Science* **322**, 1232 (2008).
[2] O. Smirnova *et al.*, *Nature* **460**, 972, (2009).

A 15.13 Mon 16:30 Spree-Palais

Two-color pump–probe momentum spectroscopy on dissociative photo-ionization of molecular nitrogen. — •Philipp Cörlin[1], Alexander Sperl[1], Andreas Fischer[1], Michael Schönwald[1], Arne Senftleben[2], Thomas Pfeifer[1], Joachim

Ullrich[3], and Robert Moshammer[1] — [1]Max-Planck-Institut für Kernphysik, Saupfercheckweg 1, 69117 Heidelberg — [2]Universität Kassel, Heinrich-Plett-Str. 40, 34132 Kassel — [3]Physikalisch-Technische Bundesanstalt, Bundesallee 100, 38116 Braunschweig

Molecular nitrogen (N_2) has been investigated in an extreme-ultraviolet (XUV)–infrared (IR) pump–probe experiment using attosecond pulse trains with photon energies between 20 eV and 40 eV to ionize N_2 into several binding and anti-binding electronic states. An additional, time delayed 15 fs IR pulse with a center-wavelength of 780 eV is used to modify and probe these states.

The three dimensional momenta of coincident photo-electrons and ions N_2^+ and N^+ are detected over the full solid angle using a Reaction Microscope. This allows to study electron-ion energy-correlations and molecular frame photo-electron angular distributions for dissociative channels and thereby identify the involved N_2^+ states by energy and symmetry. Of particular interest is the ionization into vibrational levels of the bound $C^2\Sigma^+{}_u$ state that for $k \geq 3$ dissociate into $N^+ + N$ via predissociation which has been studied in synchrotron experiments for many years (compare [1] and references therein). In our experiment the energies of these vibrational states have been measured.

[1] L.-E. Berg *et al.*, *Phys. Scr.* **44**, (1991), 131-137

A 15.14 Mon 16:30 Spree-Palais

Krylov subspace methods in relativistic quantum dynamics — •Randolf Beerwerth, Heiko Bauke, and Christoph H. Keitel — Max-Planck-Institut für Kernphysik Saupfercheckweg 1, 69117 Heidelberg

The theoretical investigation of relativistic quantum dynamics of electrons, atoms, or ions that interact with strong or short laser pulses often necessitates nonperturbative approaches which rely on numerical methods, e. g., the numerical solution of the time-dependent Dirac equation. Krylov subspace methods, e. g., the Lanczos algorithm, allow to calculate efficiently few approximate eigenvalues and eigenvectors of very large matrices and have become a standard method for solving the nonrelativistic Schrödinger equation. Here we transfer these methods into the domain of relativistic quantum dynamics. Since Krylov subspace methods usually converge to eigenstates with extremal eigenvalues and relativistic Hamiltonians are not bounded it is not self-evident that these methods are applicable in the relativistic domain.

We focus on bound states in relativistic systems and we demonstrate that Krylov subspace methods are well suited to calculate the bound states of the Dirac Hamiltonian. For this purpose, we combine the Lanczos algorithm with the pseudospectral method, where the wave function is expanded into harmonic oscillator eigenfunctions. Due to the exponential convergence of the pseudospectral approach this yields precise results using only a moderate number of basis functions. Time-dependent propagation of Dirac wave functions will also be demonstrated.

A 15.15 Mon 16:30 Spree-Palais

Tunneling theory of H_2 in presence of strong linear and circular polarized fields — •Abdou Mekky Hussien and Alejandro Saenz — AG Moderne Optik, Institut für Physik, Humboldt-Universität zu Berlin, Newtonstraße 15, 12489 Berlin, Germany

The ratio $\Re$ of the angular ionization probabilities of H_2 between parallel and perpendicular molecular orientations in a linear and a circular polarized fields using a molecular tunneling theory (MO-ADK [1]) is studied. It is found that the ratio $\Re$ for an H_2 molecule in a strong field generally decreases with increases laser intensity, and the pattern of decrease depends on the duration of the laser pulse. A reasonable agreement with the experimental data in [2] is observed. These results invalidate the main result in [3] and corresponding statements in other works, where it is claimed that MO-ADK theory fails to predict the intensity-dependent anisotropy of H_2. Furthermore, the effect of the focal-volume averaging on the anisotropy is studied. Finally, an extention of the MO-ADK model into the barrier-suppression regime is also given. A further improvement of the results at high intensities is found, if this correction is considered.

[1] X. M. Tong *et al.*, *Phys. Rev. A*, **66**, 033402 (2002).
[2] A. Staudte *et al.*, *Phys. Rev. Lett.* **102**, 033004 (2009).
[3] Y.-J. Jin *et al.*, *Phys. Rev. A*, **83**, 063409 (2011).

A 15.16 Mon 16:30 Spree-Palais

Reconstructing scattering patterns of single xenon clusters — •Alexander Schreider[1], Leonie Flückiger[1], Fengling Wang[2], Daniela Rupp[1], and Thomas Möller[1] — [1]TU Berlin — [2]Universität Hamburg

Structure determination of small particles with conventional optical microscopy is significantly limited by diffraction. Coherent Diffraction Imaging (CDI) with short wavelengths brings a solution for imaging such samples by recording diffraction patterns and subsequently reconstructing the lost phase information by iterative algorithms.

We used algorithms based on the Hybrid-Input-Output (HIO) algorithm [1], the Error-Reduction (ER) algorithm [2] and the Shrink-Wrap method [3] to develope a programm for reconstructing measured diffraction patterns of xenon clusters ab initio. This programm was tested and optimized for the particular difficulties of the cluster scattering patterns due to restrictions introduced by the experimental setup. The challenges and possible solutions will be discussed.

[1] J.R. Fienup, Optics Letters 3, 27 (1978), [2] J.R. Fienup, Applied Optics 21, 2758 (1982), [3] S. Marchesini et al., Physical Review B 68, 140101 (2003)

A 15.17 Mon 16:30 Spree-Palais

Influence of the laser waveform of near single-cycle laser pulses on the dissociative ionization of N2O — •Christian Jendrzejewski[1,2], Matthias Kübel[1,2], and Matthias Kling[1,2] — [1]Ludwig-Maximilians-Universität München, Garching, Germany — [2]Max-Planck-Institut für Quantenoptik, Garching, Germany

Within the past few years, the development of near-single cycle laser pulses allowed for various studies on electron dynamics in the femto- and attosecond time scale which are fundamental for chemical reactions. In order to investigate the ionization behavior of triatomic molecules in such laser fields laughing gas (N2O) is used as a model system. Thereby, the focus lies on the non sequential double ionization (NSDI) as it is highly important for correlated electron processes. The ionization products of the N2O molecule are temporally and spacially resolved within a reaction microscope. As such photoionization processes are strongly dependent on the laser waveform of the pulse, especially on the carrier-envelope phase (CEP), the CEP is detected for each laser pulse. Combining these two methods, strong dependencies of the ionization yield on the CEP and polarization of the laser pulse are observed.

A 15.18 Mon 16:30 Spree-Palais

Photoelectrons close to threshold — •Elias Diesen, Ulf Saalmann, and Jan-Michael Rost — Max Planck Institute for the Physics of Complex Systems, Dresden, Germany

The 2009 discovery of the low energy structure (LES) [1] of photoionization spectra in the long-wavelength, high-intensity tunneling regime spurred a large experimental and theoretical activity in the last years. The role of classical so-called soft recollisions was investigated in [2,3]. Recent experiments at unprecedented resolution reveal more spectral features [4] above and below threshold, whose explanation is not yet complete. We present theoretical results on the formation of these features that typically appear at energies well below 1 eV.

[1] Blaga et al, Nature Phys., 5, 335 (2009)
[2] Kästner et al, Phys. Rev. Lett. 108, 033201 (2012)
[3] Kästner et al, J. Phys. B 45, 074011 (2012)
[4] Dura et al, Sci. Rep. 3, 2675 (2013)

A 15.19 Mon 16:30 Spree-Palais

Velocity Map Imaging of Trajectory Controlled Above-Threshold Ionization Spectra of Xenon Using the Two-Color Field Technique — •Daniel Würzler, Max Möller, Frank Meyer, Max Sayler, and Gerhard G. Paulus — HIJ IOQ, Max-Wien-Platz 1, 07743 Jena

Recent research shows that adding a weak second harmonic field perpendicular to a strong fundamental beam provides the ability to control high harmonic generation by suppressing recombination of certain electron trajectories. In this study, a collinear interferometer is build in order to combine an ultrashort laser pulse with its second harmonic field in the target area. The setup is used to demonstrate control over electron trajectories of Xenon in above-threshold ionization.