A 20: Interaction with strong or short laser pulses II

Time: Wednesday 11:00–13:00 Location: C/HSW

Invited Talk A 20.1 Wed 11:00 C/HSW

Strong-field ionization of molecules in circularly polarized fields — Ingo Petersen, Jost Henkel, and •Manfred Lein — Institut für Theoretische Physik, Leibniz Universität Hannover, 30167 Hannover

The electron momentum distribution from strong-field ionization of molecules contains rich information about the molecular electronic structure. Circularly polarized light is particularly advantageous because (i) it delivers a 360 degree scan of the molecule, (ii) the absence of recollisions makes the ionization process easier to interpret, and (iii) enantiomers of chiral molecules can be distinguished by exploiting photoelectron circular dichroism. Our calculations for diatomic and polyatomic molecules show that the width of the momentum distribution in the direction perpendicular to the polarization plane is a robust observable that is useful for imaging of aligned molecules. In particular, a strong variation of the width as a function of emission angle indicates contributions of multiple orbitals to the total ionization. For chiral molecules, we implement an extension of the strong-field approximation where the plane-wave continuum states are replaced by the Born approximation. The calculated distributions from a gas of randomly oriented molecules (using the examples camphor and fenchone) exhibit a forward-backward asymmetry as expected due to the dichroism effect, but they are not in quantitative agreement with recent experiments.

A 20.2 Wed 11:30 C/HSW

Wavelength and intensity dependent fragmentation of H_2^+ in the mid-infrared — •Wustelt P.[1,2], Möller M.[1,2], Rathje T.[1,2], Sayler A.M.[1,2], and Paulus G.G.[1,2] — [1]Institute of Optics and Quantum Electronics, Friedrich Schiller University Jena, Germany — [2]Helmholtz Institute Jena, Germany

The wavelength and intensity dependent ultra-short laser-induced ionization and dissociation of an H_2^+ molecular ion beam target is measured at mid-infrared wavelength between 0.8 and 2.2 μm and at intensities up to 10^{16} W/cm^2. A three-dimensional coincidence imaging system allows kinematically complete coincidence measurements of both fragmentation channels and enables an unprecedented look into laser-driven fragmentation dynamics of the simplest molecule in strong mid-infrared laser fields.

A 20.3 Wed 11:45 C/HSW

Differential spectra for dissociative ionization of H_2^+ — •Volker Mosert and Dieter Bauer — Institut für Physik, Universität Rostock, 18051 Rostock

We compute and analyze correlated spectra for the dissociative ionization (DI) of the H_2^+ molecular ion using the tSURFF method. A low dimensional model of H_2^+ allows us too treat both nuclear and electronic degree of freedom exactly. In our TDSE simulations the molecular ion interacts with intense short laser pulses in the near IR regime. We find a pronounced modulation in the DI yield with respect to the kinetic energy of the protons. This modulation is almost independent of the electronic energy and could - erroneously - be attributed to vibrational excitations. However, by comparison with calculations for fixed internuclear distances the modulation can be traced back to an electronic interference effect that depends on the ionization potential.

A 20.4 Wed 12:00 C/HSW

Quantum Optimal Control of Photoelectron Spectra. — Esteban Goetz[1], Antonia Karamatskou[2], Robin Santra[2], and •Christiane Koch[1] — [1]Theoretische Physik, Kassel Universität, Kassel, Deutschland — [2]Center for Free-Electron Laser Science, Hamburg, Deutschland

Photoelectron spectra obtained in photoionization reveal information on charge transfert or hole coherence in the parent ion. Optimal control can be used to enhance certain desired features in the photoelectron spectrum. To this end, we combine Krotov's optimal control algorithm[1], time-dependent configuration-interaction singles formalism and the time-splitting method [2] for the calculation and optimization of photoelectron spectra.

The optimization target can be formulated to include specific desired properties in the angle-resolved photoelectron spectrum (PES) alone, in the energy-resolved PES, or both. As an example, we demonstrate our algorithm to maximize the difference between the number of electrons emitted into the upper hemisphere and the number of electrons emitted into the lower hemisphere for Argon.

[1] D. Reich, M. Ndong, and C.P. Koch, J. Chem. Phys. 136, 104103 (2012). [2] A. Karamatskou, S.Pabst, Yi-Jen Chen, and R. Santra, Phys. Rev. A 89, 033415 (2014).

A 20.5 Wed 12:15 C/HSW

Electron-correlation effects in laser-intensity dependent ionization processes observed by transient-absorption spectroscopy of helium atoms — •Andreas Kaldun, Veit Stooss, Christian Ott, Alexander Blättermann, Thomas Ding, Andreas Fischer, and Thomas Pfeifer — Max-Planck-Institut für Kernphysik, Saupfercheckweg 1, 69117 Heidelberg, Germany

In our transient-absorption spectroscopy setup we simultaneously observe the line shapes of the singly-excited states of helium ($1sn$p) and the doubly exited states of helium ($sp_{2,n+}$). Both line series are coherently excited by an attosecond-pulsed extreme-ultraviolet(XUV)field. A strong near-visible (VIS) laser pulse is co-propagating with the XUV. Both pulses overlap spatially and temporally. Scanning the intensity of the VIS laser pulse we observe different ionization thresholds for the uncorrelated ($1sn$p) and correlated ($sp_{2,n+}$) electronic wave function, respectively. Several different ionization channels (direct photo ionization, autoionization, tunneling, barrier-suppressed ionization) complicate the situation.

A 20.6 Wed 12:30 C/HSW

In situ characterization of few-cycle laser pulses in transient absorption spectroscopy — •Alexander Blättermann[1], Chrsitian Ott[1,2], Andreas Kaldun[1], Thomas Ding[1], Martin Laux[1], Veit Stooss[1], Marc Rebholz[1], Maximilian Hartmann[1], and Thomas Pfeifer[1] — [1]Max-Planck-Institut für Kernphysik, Saupfercheckweg 1, 69117 Heidelberg , Germany — [2]Chemistry Department, University of California, 94720 Berkeley CA, United States

We demonstrate a method that allows to extract the pulse duration and intensity of few-cycle laser pulses directly from a measured extreme ultraviolet transient-absorption spectrum. Since we analyze the signature of the laser pulse interacting with the spectroscopy target, the pulse is characterized in situ. By doing so, we combine laser pulse characterization and strong-field driven quantum dynamics measurement in a single experiment. The precise knowledge of the driving laser pulse is important to fully understand the highly nonlinear optical response of quantum systems to strong laser fields.

A 20.7 Wed 12:45 C/HSW

Description of the hydrogen molecular ion with time-dependent renormalized-natural-orbital theory — •Adrian Hanusch, Julius Rapp, Martins Brics, and Dieter Bauer — Institut für Physik, Universität Rostock, 18051 Rostock, Germany

Time-dependent natural-orbital theory is a promising approach to overcome the "exponential wall" in solving time-dependent many-body quantum problems numerically. Time-dependent renormalized-natural-orbital theory (TDRNOT) has already proven good performance using an exactly solvable 1D Helium model atom. Highly correlated phenomena — not accessible via practicable time-dependent density functional theory — have been successfully described with TDRNOT [1-3]. In this work, we extend TDRNOT to investigate the hydrogen molecular ion exactly, i.e., beyond the Born-Oppenheimer approximation, and in intense laser fields. The correlation between electron and nuclear degree of freedom is taken into account by considering electronic and nuclear natural orbitals, and their coupling.

[1] M. Brics, D. Bauer, Phys. Rev. A 88, 052514 (2013).
[2] J. Rapp, M. Brics, D. Bauer Phys. Rev. A 90, 012518 (2014).
[3] M. Brics, J. Rapp, D. Bauer, Phys. Rev. A 90, 053418 (2014).