MO 9: Molecules in Intense Fields and Quantum Control (joint session MO/A)

Time: Wednesday 14:30–16:30 Location: F102

Invited Talk MO 9.1 Wed 14:30 F102

Full Angle-Resolved Mapping of Electron Rescattering Probabilities in the Molecular Frame — Federico Branchi[1], Lingfeng Ge[2], Felix Schell[1], Killian Dickson[3], Mark Mero[1], Horst Rottke[1], Serguei Patchkovskii[1], Marc Vrakking[1], Varun Makhija[3], and •Jochen Mikosch[2] — [1]Max-Born-Institut, Berlin, Germany — [2]Universität Kassel, Kassel, Germany — [3]Univ. of Mary Washington, Fredericksburg, USA

A reaction microscope experiment on strong-field ionization and laser-driven electron rescattering of the asymmetric top molecule 1,3-butadiene is presented. Importantly, by virtue of the ion-electron coincidence detection, our experiments separate the ground-state (D0) and first excited state (D1) ionization channel. In this way two scattering experiments on the same target are performed simultaneously with two very different continuum electron wavepackets.

By analyzing lab frame coherent rotational wavepacket evolution following a non-adiabatic alignment laser pulse we achieve both polar and azimuthal angle-resolved molecular frame information.

Our results indicate that the nodal structure of the ionizing orbitals is more strongly reflected in the electron rescattering probability rather than in the ionization probability. Propagation of the wavepacket influences the differential cross section that is measured for the two channels. Experimental results are compared with results from a TD-RIS ab-initio simulation.

MO 9.2 Wed 15:00 F102

Pulse length dependence of photoelectron circular dichroism — •Hangyeol Lee[1], Simon Ranecky[1], Sudheendran Vasudevan[1], Nicolas Ladda[1], Tonio Rosen[1], Sagnik Das[1], Jayanta Ghosh[1], Hendrike Braun[1], Daniel Reich[2], Arne Senftleben[1], and Thomas Baumert[1] — [1]Institut für Physik, Universität Kassel, Heinrich-Plett-Str. 40, 34132 Kassel, Germany. — [2]Dahlem Center for Complex Quantum Systems and Fachbereich Physik, Freie Universität Berlin, Arnimallee 14, D-14195 Berlin, Germany.

We studied the dependence of photoelectron circular dichroism (PECD) of fenchone on the duration of ionizing laser pulses from 30 fs to 5 ns. The laser pulses were centered at 380 nm to induce 2+1 resonant-enhanced multiphoton ionization of fenchone via 3s and 3p intermediate states. The photoelectrons from each intermediate state were distinguished by their different kinetic energies. As the pulse duration increases, the effect of relaxation dynamics was observed as a change in the ratio of photoelectron contributions from the 3s and the 3p intermediate states. The PECD measured via the 3s intermediate resonance was about 15 % and robust despite ongoing molecular dynamics such as rotation, vibration, and internal conversion. We simulated the observed relaxation dynamics using a simplified model system and estimated the lifetimes of the intermediate states.

MO 9.3 Wed 15:15 F102

Influence of laser properties on the high-order harmonic generation process in benzene — •Samuel Schöpa and Dieter Bauer — Institute of Physics, University of Rostock, Rostock, Germany

We solve the Schrödinger equation for benzene by expanding the wave function in a linear combination of ground-state Kohn-Sham orbitals. Those have been calculated previously via ground-state density functional theory. This method is orders of magnitude faster than comparable full time-dependent density functional theory simulations but neglects the update of the Hartree-exchange-correlation potential during time evolution. The selection rules stemming from the 6-fold symmetry of the benzene molecule as well as the opposite polarization of each harmonic couple are observed for a laser field at normal incidence that is circularly polarized in the molecular plane. We investigate how ellipticity and angle of incidence of the laser influence the spectrum. The selection rules are broken already for small deviations from normal incidence.

MO 9.4 Wed 15:30 F102

Nondipole time delay and double-slit interference in tunneling ionization — •Peilun He, Karen Hatsagortsyan, and Christoph Keitel — Max-Planck-Institut für Kernphysik, Saupfercheckweg 1, 69117 Heidelberg, Germany

The photon takes zeptoseconds time to travel through the bond length of a molecule, which results in the fringe shift of the photoelectron momentum distribution. We investigate the possibility of decoding this nondipole time delay signal in tunneling ionization. With the newly developed Coulomb-corrected nondipole molecular strong-field approximation [PRL **128**, 183201 (2022)], we derive and analyze the photoelectron momentum distribution, the signature of nondipole effects, and the role of the degeneracy of the molecular orbitals. We show that the ejected electron momentum shifts and interference fringes efficiently imprint both the molecule structure and laser parameters. The corresponding nondipole time delay value significantly deviates from that in single-photon ionization. In particular, when the two-center interference in the molecule is destructive, the time delay is independent of the bond length. We also identify the double-slit interference in tunneling ionization of atoms with nonzero angular momentum via a nondipole momentum shift.

MO 9.5 Wed 15:45 F102

Strong coupling to a phonon bath enhances adiabatic population transfer — •Frank Grossmann and Michael Werther — Technische Universität Dresden, Institut für Theoretische Physik, 01062 Dresden

We present a study on the influence of an environmental heat bath on the rapid adiabatic passage scheme for optimal population transfer in a two-level system, originally invented in nuclear magnetic resonance [1].

To cope with strong coupling to an external phonon bath with super-ohmic spectral density, we are solving the time-dependent Schrödinger equation of the extended system, including a carefully chosen finite number of bath modes, using the multi-Davydov D2-Ansatz [2], which will be briefly reviewed. This Ansatz allows for the treatment of the non-Markovian reduced dynamics of the two-level subsystem. We find that strong system-bath coupling stabilizes the transition probability from the lower to the upper level as a function of the area under the laser pulse. This dissipative engineering effect could only be uncovered by a non-Markovian treatment. For strong coupling, the transition probability then becomes a monotonically increasing function of the pulse area at zero temperature of the heat bath. Finite temperatures break the monotonicity in the range of pulse areas that we studied but not the stability of the observed effect.

[1] M. Werther and F. Grossmann, Phys. Rev. A 102, 063710 (2020)
[2] M. Werther and F. Grossmann, Phys. Rev. B 101, 174315 (2020)

MO 9.6 Wed 16:00 F102

Optimization of selective two-photon absorption in cavity polaritons — •Edoardo Carnio[1,2], Andreas Buchleitner[1,2], and Frank Schlawin[3,4,5] — [1]Physikalisches Institut, Albert-Ludwigs-Universität Freiburg, Hermann-Herder-Straße 3, D-79104, Freiburg, Germany — [2]EUCOR Centre for Quantum Science and Quantum Computing, Albert-Ludwigs-Universität Freiburg, Hermann-Herder-Straße 3, D-79104, Freiburg, Germany — [3]Max Planck Institute for the Structure and Dynamics of Matter, Luruper Chaussee 149, D-22761 Hamburg, Germany — [4]The Hamburg Centre for Ultrafast Imaging, Luruper Chaussee 149, D-22761 Hamburg, Germany — [5]Clarendon Laboratory, University of Oxford, Parks Road, Oxford OX1 3PU, United Kingdom

We investigate optimal states of photon pairs to excite a target transition in a multilevel quantum system. From the optimal control theory of entangled two-photon absorption we infer the maximal population achievable by optimal entangled vs. separable states of light. Interference between excitation pathways, as well as the presence of nearby states, may hamper the selective excitation of a particular target state, but we show that quantum correlations can help overcome this problem, and enhance the achievable "selectivity" between two target energy levels, i.e. the relative difference in population transferred into each of them.

[1] E. G. Carnio, A. Buchleitner, F. Schlawin, J. Chem. Phys. 154, 214114 (2021).

MO 9.7 Wed 16:15 F102

Quantized fields for optimal control in the strong coupling

regime — •Frieder Lindel[1], Edoardo Carnio[1], Stefan Yoshi Buhmann[2], and Andreas Buchleitner[1] — [1]University of Freiburg, Germany — [2]University of Kassel, Germany

The control of quantum systems lies at the core of many quantum technologies. In the field of coherent control, classical fields coherently drive the quantum system from a given initial state into a target state. Exploiting the quantum nature of the field to improve these control protocols has so far been mostly limited to the weak coupling regime. Here we will discuss how the quantum statistics of a bosonic field can be optimally tailored in order to drive a weakly or (ultra-)strongly coupled quantum system, such as an atom or a molecule in a cavity, towards a desired target state. This extends optimal control theory to control and target systems that are both quantized and strongly coupled.