

Molecular Physics Division Fachverband Molekülphysik (MO)

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Overview of Invited Talks and Sessions

(Lecture halls F102 and F142; Poster Empore Lichthof)

Invited Talks

MO 4.1	Tue	11:00–11:30	F102	Revealing chiral charge migration in UV-excited molecules — •VINCENT WANIE, ETIENNE BLOCH, ERIK P. MÅNSSON, LORENZO COLAIZZI, SERGEY RYABCHUK, KRISHNA SARASWATHULA, ANDREA TRABATTONI, VALÉRIE BLANCHET, NADIA BEN AMOR, MARIE-CATHERINE HEITZ, YANN MAIRESSE, BERNARD PONS, FRANCESCA CALEGARI
MO 7.1	Wed	11:00–11:30	F142	Augmenting basis with normalizing flows for solving Schrödinger equations: theoretical analysis — •YAHYA SALEH, ARMIN ISKE, ANDREY YACHMENEV, JOCHEN KÜPPER
MO 9.1	Wed	14:30–15:00	F102	Full Angle-Resolved Mapping of Electron Rescattering Probabilities in the Molecular Frame — FEDERICO BRANCHI, LINGFENG GE, FELIX SCHELL, KILLIAN DICKSON, MARK MERO, HORST ROTTKE, SERGUEI PATCHKOVSKII, MARC VRAKING, VARUN MAKHIJA, •JOCHEN MIKOSCH
MO 15.1	Thu	11:00–11:30	F142	Excited state dipole moments from rotationally resolved Stark spectroscopy — •MICHAEL SCHMITT, MATTHIAS ZAJONZ, MARIE-LUISE HEBESTREIT
MO 22.1	Fri	11:00–11:30	F142	A QED Theory of Mediated RET Between a Pair of Chiral Molecules — •AKBAR SALAM

Invited Talks of the joint Symposium Precision Physics with Highly Charged Ions

See SYHC for the full program of the symposium.

SYHC 1.1	Mon	11:00–11:30	E415	First experiments at CRYRING@ESR — •ESTHER BABETTE MENZ, MICHAEL LESTINSKY, HÅKAN DANARED, CLAUDE KRANTZ, ZORAN ANDELKOVIC, CARSTEN BRANDAU, ANGELA BRÄUNING-DEMIAN, SVETLANA FEDOTOVA, WOLFGANG GEITHNER, FRANK HERFURTH, ANTON KALININ, INGRID KRAUS, UWE SPILLMANN, GLEB VOROBYEV, THOMAS STÖHLKER
SYHC 1.2	Mon	11:30–12:00	E415	Testing quantum electrodynamics in the simplest and heaviest multi-electronic atoms — •MARTINO TRASSINELLI
SYHC 1.3	Mon	12:00–12:30	E415	Indirect measurements of neutron-induced reaction cross-sections at heavy-ion storage rings — •BEATRIZ JURADO
SYHC 1.4	Mon	12:30–13:00	E415	Laboratory X-ray Astrophysics with Trapped Highly Charged Ions at Synchrotron Light Sources — •SONJA BERNITT
SYHC 2.1	Mon	17:00–17:30	E415	Observation of metastable electronic states in highly charged ions by Penning-trap mass spectrometry — •KATHRIN KROMER, MENNO DOOR, PAVEL FILIANIN, ZOLTÁN HARMAN, JOST HERKENHOFF, PAUL INDELICATO, CHRISTOPH H. KEITEL, DANIEL LANGE, CHUNHAI LYU, YURI N. NOVIKOV, CHRISTOPH SCHWEIGER, SERGEY ELISEEV, KLAUS BLAUM
SYHC 2.2	Mon	17:30–18:00	E415	Towards extreme-ultraviolet optical clocks — •JOSÉ R. CRESPO LÓPEZ-URRUTIA

SYHC 2.3	Mon	18:00–18:30	E415	Coupling atomic and nuclear degrees of freedom in highly charged ions — ●ADRIANA PÁLFFY
SYHC 2.4	Mon	18:30–19:00	E415	Laser Spectroscopy at the Storage Rings of GSI/FAIR — ●WILFRIED NÖRTERSCHÄUSER

Invited Talks of the joint Symposium SAMOP Dissertation Prize 2023

See SYAD for the full program of the symposium.

SYAD 1.1	Mon	14:30–15:00	E415	Quantum gas magnifier for sub-lattice resolved imaging of 3D quantum systems — ●LUCA ASTERIA
SYAD 1.2	Mon	15:00–15:30	E415	From femtoseconds to femtometers – controlling quantum dynamics in molecules with ultrafast lasers — ●PATRICK RUPPRECHT
SYAD 1.3	Mon	15:30–16:00	E415	Particle Delocalization in Many-Body Localized Phases — ●MAXIMILIAN KIEFER-EMMANOULIDIS
SYAD 1.4	Mon	16:00–16:30	E415	Feshbach resonances in a hybrid atom-ion system — ●PASCAL WECKESSER

Invited Talks of the joint Symposium Machine Learning in Atomic and Molecular Physics

See SYML for the full program of the symposium.

SYML 1.1	Tue	11:00–11:30	E415	Imaging a complex molecular structure with laser-induced electron diffraction and machine learning — ●KATHARINA CHIRVI, XINYAO LIU, KASRA AMINI, AURELIEN SANCHEZ, BLANCA BELSA, TOBIAS STEINLE, JENS BIEGERT
SYML 1.2	Tue	11:30–12:00	E415	Physics-inspired learning algorithms for optimal shaping of atoms with light — ●MAXIMILIAN PRÜFER
SYML 1.3	Tue	12:00–12:30	E415	Machine-Learning assisted quantum computing and interferometry — ●LUDWIG MATHEY, LUKAS BROERS, NICOLAS HEIMANN
SYML 1.4	Tue	12:30–13:00	E415	Efficient quantum state tomography with convolutional neural networks — ●MORITZ REH, TOBIAS SCHMALE, MARTIN GÄRTTNER

Prize Talks of the joint Awards Symposium

See SYAS for the full program of the symposium.

SYAS 1.1	Tue	14:35–15:05	E415	The Reaction Microscope: A Bubble Chamber for AMOP — ●JOACHIM ULLRICH
SYAS 1.2	Tue	15:05–15:35	E415	Quantum Computation and Quantum Simulation with Strings of Trapped Ca⁺ Ions — ●RAINER BLATT
SYAS 1.3	Tue	15:35–16:05	E415	Amplitude, Phase and Entanglement in Strong Field Ionization — ●SEBASTIAN ECKART
SYAS 1.4	Tue	16:05–16:35	E415	All-optical Nonlinear Noise Suppression in Mode-locked Lasers and Ultrafast Fiber Amplifiers — ●MARVIN EDELMANN

Invited Talks of the joint Symposium Molecules in Helium Droplets

See SYHD for the full program of the symposium.

SYHD 1.1	Wed	11:00–11:30	E415	Structure and field-induced dynamics of small helium clusters — ●MAKSIM KUNITSKI, JAN KRUSE, QINGZE GUAN, DÖRTE BLUME, REINHARD DÖRNER
SYHD 1.2	Wed	11:30–12:00	E415	Coherent Diffraction Imaging of isolated helium nanodroplets and their ultrafast dynamics — ●DANIELA RUPP
SYHD 1.3	Wed	12:00–12:30	E415	Clustering dynamics in superfluid helium nanodroplets: A theoretical study — ●NADINE HALBERSTADT, ERNESTO GARCÍA ALFONSO, MARTÍ PI, MANUEL BARRANCO
SYHD 1.4	Wed	12:30–13:00	E415	Messenger spectroscopy of molecular ions – Development of a new experimental setup — ●ELISABETH GRUBER

Invited Talks of the joint Symposium From Molecular Spectroscopy to Collision Control at the Quantum Limit

See SYCC for the full program of the symposium.

SYCC 1.1	Thu	11:00–11:30	E415	The unity of physics: the beauty and power of spectroscopy — ●PAUL JULIENNE
SYCC 1.2	Thu	11:30–12:00	E415	Using high-resolution molecular spectroscopy to explore how chemical reactions work — ●JOHANNES HECKER DENSCHLAG
SYCC 1.3	Thu	12:00–12:30	E415	Monitoring ultracold collisions with laser light — ●OLIVIER DULIEU
SYCC 1.4	Thu	12:30–13:00	E415	The birth of a degenerate Fermi gas of molecules — ●JUN YE

Invited Talks of the joint PhD-Symposium – Many-body Physics in Ultracold Quantum Systems

See SYPD for the full program of the symposium.

SYPD 1.1	Thu	14:30–15:00	E415	Entanglement and quantum metrology with microcavities — ●JAKOB REICHEL
SYPD 1.2	Thu	15:00–15:30	E415	Many-body physics in dipolar quantum gases — ●FRANCESCA FERLAINO
SYPD 1.3	Thu	15:30–16:00	E415	Quantum Simulation: from Dipolar Quantum Gases to Frustrated Quantum Magnets — ●MARKUS GREINER
SYPD 1.4	Thu	16:00–16:30	E415	Quantum gas in a box — ●ZORAN HADZIBABIC

Invited Talks of the joint Symposium Quantum Optics and Quantum Information with Rigid Rotors

See SYQR for the full program of the symposium.

SYQR 1.1	Fri	11:00–11:30	E415	Femtosecond timed imaging of rotation and vibration of alkali dimers on the surface of helium nanodroplets — ●HENRIK STAPELFELDT
SYQR 1.2	Fri	11:30–12:00	E415	Quantum toolbox for molecular state spaces — ERIC KUBISCHTA, SHUBHAM JAIN, IAN TEIXEIRA, ERIC R. HUDSON, WESLEY C. CAMPBELL, MIKHAIL LEMESHKO, ●VICTOR V. ALBERT
SYQR 1.3	Fri	12:00–12:30	E415	Coherent rotational state control of chiral molecules — ●SANDRA EIBENBERGER-ARIAS
SYQR 1.4	Fri	12:30–13:00	E415	Optically levitated rotors: potential control and optimal measurement — ●MARTIN FRIMMER
SYQR 2.1	Fri	14:30–15:00	E415	Rotational optomechanics with levitated nanodumbbells — ●TONGCANG LI
SYQR 2.2	Fri	15:00–15:30	E415	Quantum rotations of nanoparticles — ●BENJAMIN A. STICKLER
SYQR 2.3	Fri	15:30–16:00	E415	Quantum control of trapped molecular ions — ●STEFAN WILLITSCH
SYQR 2.4	Fri	16:00–16:30	E415	Full control over randomly oriented quantum rotors: controllability analysis and application to chiral observables — ●MONIKA LEIBSCHER

Sessions

MO 1.1–1.8	Mon	11:00–13:00	F102	Cold Molecules (joint session MO/Q)
MO 2.1–2.8	Mon	11:00–13:00	F142	Photochemistry
MO 3.1–3.7	Mon	11:00–13:00	F303	Interaction with Strong or Short Laser Pulses I (joint session A/MO)
MO 4.1–4.7	Tue	11:00–13:00	F102	Ultrafast Dynamics I (joint session MO/A)
MO 5.1–5.8	Tue	11:00–13:00	F142	Electronic Spectroscopy
MO 6.1–6.23	Tue	16:30–19:00	Empore Lichthof	Poster I
MO 7.1–7.7	Wed	11:00–13:00	F142	Machine Learning and Computational and Theoretical Molecular Physics
MO 8	Wed	13:15–14:00	F142	Members' Assembly
MO 9.1–9.7	Wed	14:30–16:30	F102	Molecules in Intense Fields and Quantum Control (joint session MO/A)
MO 10.1–10.7	Wed	14:30–16:15	F142	Collisions (joint session MO/Q)
MO 11.1–11.7	Wed	14:30–16:30	E214	Quantum Technologies (joint session Q/MO/QI)

MO 12.1–12.6	Wed	14:30–16:15	F107	Interaction with Strong or Short Laser Pulses II (joint session A/MO)
MO 13.1–13.19	Wed	16:30–19:00	Empore Lichthof	Poster II
MO 14.1–14.8	Thu	11:00–13:00	F102	Ultrafast Dynamics II (joint session MO/A)
MO 15.1–15.7	Thu	11:00–13:00	F142	Rotational- and Vibrational-resolution Spectroscopy
MO 16.1–16.7	Thu	11:00–13:00	F107	Atomic Clusters (joint session A/MO)
MO 17.1–17.8	Thu	14:30–16:30	F102	Quantum Optics and Quantum Information with Rigid Rotors (joint session MO/Q/QI)
MO 18.1–18.8	Thu	14:30–16:30	F142	Cluster and Experimental Techniques (joint session MO/A)
MO 19.1–19.5	Thu	14:30–16:00	F107	Interaction with Strong or Short Laser Pulses III (joint session A/MO)
MO 20.1–20.19	Thu	16:30–19:00	Empore Lichthof	Poster III
MO 21.1–21.8	Fri	11:00–13:00	F102	Molecular Physics with X-rays
MO 22.1–22.8	Fri	11:00–13:15	F142	Theoretical and Computational Molecular Physics
MO 23.1–23.8	Fri	14:30–16:30	F102	Ultrafast Dynamics III (joint session MO/A)

Members' Assembly of the Molecular Physics Division

Wednesday 13:15–14:00 F142

MO 1: Cold Molecules (joint session MO/Q)

Time: Monday 11:00–13:00

Location: F102

MO 1.1 Mon 11:00 F102

A Continuous Source of Aluminium Monofluoride Molecules

— ●MAXIMILIAN DOPPELBAUER¹, SIDNEY C. WRIGHT¹, SIMON HOFSSÄSS¹, JOSÉ EDUARDO PADILLA-CASTILLO¹, SEBASTIAN KRAY¹, RUSSELL THOMAS¹, BORIS SARTAKOV¹, STEFAN TRUPPE^{1,2}, and GERARD MEIJER¹ — ¹Molecular Physics, Fritz-Haber-Institut der Max-Planck-Gesellschaft, Faradayweg 4-6, 14195 Berlin, Germany — ²Centre for Cold Matter, Blackett Laboratory, Imperial College London, Prince Consort Road, London SW7 2AZ, United Kingdom

The aluminium monofluoride (AlF) molecule is a unique candidate for laser cooling and trapping experiments. As a starting point, we require a high-density molecular source. In our original setup, we can generate AlF by reaction of laser-ablated aluminium atoms with NF₃ in a pulsed cryogenic buffer gas source with more than 10¹² molecules per steradian per ablation shot. By exploiting the reaction of AlF₃ and Al in a UHV oven above 600°C, we can generate a continuous thermal AlF beam with a total brightness of about 10¹⁶ molecules per steradian per second.

In this contribution, we present spectroscopic information on vibrational levels up to $v'' = 4$ and rotational levels to above $J'' = 80$ in the $X^1\Sigma^+$ electronic ground state that we obtained using the oven source as well as first experiments laser cooling AlF.

MO 1.2 Mon 11:15 F102

Cryo-cooled beams of “small” macromolecules

— ●JINGXUAN HE^{1,2,3}, LENA WORBS^{1,2}, SURYA KIRAN PERAVALI^{1,4}, ARMANDO D. ESTILLORE¹, AMIT K. SAMANTA^{1,3}, and JOCHEN KÜPPER^{1,2,3} — ¹Center for Free-Electron Laser Science, Deutsches Elektronen-Synchrotron DESY, Hamburg, Germany — ²Department of Physics, Universität Hamburg, Germany — ³Center for Ultrafast Imaging, Universität Hamburg, Germany — ⁴Fakultät für Maschinenbau, Helmut-Schmidt-Universität, Germany

We have demonstrated the preparation of cold and controlled dense beams of nanoparticles and macromolecules designed for x-ray single-particle diffractive imaging (SPI). We exploit buffer-gas cell cooling and aerodynamic focusing techniques [1-2]. We are extending the cooling and control techniques developed for SPI to experimental investigations of ultrafast electron dynamics in complex biomolecules. We aim at disentangling charge and energy transfer following electronic excitation, which still has important open questions [3].

Here, we present our approach to prepare appropriate samples of cryogenically-cooled proteins to study these also biologically important elementary processes, for instance, using photofragmentation mass spectrometry and velocity map imaging.

- [1]A. K. Samanta, et al., *Structural dynamics* **7**, 024304 (2020)
- [2]L. Worbs, et al., *In preparation*, (2022)
- [3]H. Duan, et al., *PNAS* **114**, 8493 (2017)

MO 1.3 Mon 11:30 F102

Zeeman slowing of CaF

— ●MARIIA STEPANOVA, TIMO POLL, PAUL KAEBERT, SUPENG XU, MIRCO SIERCKE, and SILKE OSPELKAUS — Institut für Quantenoptik, Leibniz Universität Hannover

Our Zeeman slowing scheme for laser-cooling of molecules with favorable Franck-Condon factors promises a substantial increase in molecular number in the velocity range of under 20 m/s, which is required for loading a Magneto-Optical Trap (MOT). The scheme stands out in its ability to not only lower the initial mean velocity of the molecular beam, but also to compress the velocity distribution in a continuous fashion. In this talk, we will present our most recent status on the experiment to achieve the goal of slowing and cooling CaF molecules generated from a buffer gas cell source, followed by our efforts to implement a dual-frequency MOT without sub-Doppler heating, discussed in [1].

[1] S. Xu, P. Kaebert, M. Stepanova, T. Poll, M. Siercke and S. Ospelkaus, DOI: <https://doi.org/10.1103/PhysRevResearch.4.L042036>

MO 1.4 Mon 11:45 F102

Ortho ground state preparation of cooled and trapped formaldehyde molecules

— ●MAXIMILIAN LÖW, MARTIN IBRÜGGER, MARTIN ZEPPENFELD, and GERHARD REMPE — Max-Planck-Institut für Quantenoptik, Hans-Kopfermann-Str. 1, 85748 Garching

Methods to directly cool polar molecules to ultracold temperatures saw

remarkable progress in recent years. One of the most promising techniques in this field is optoelectrical Sisyphus cooling which can provide a large number of electrically trapped molecules at the sub-millikelvin level [1]. However, molecules in their absolute ground state cannot be addressed with this approach.

Cold ground state molecules can still be created by first applying Sisyphus cooling to, e.g., formaldehyde (H₂CO) molecules in the rotational states $|J=3, K_a=3, K_c=0\rangle$ and $|4, 3, 1\rangle$. Then, we use optical pumping to transfer them via a vibrational transition to their ortho ground state $|1, 1, 0\rangle$. In a proof-of-principle experiment trapped ground state molecules with a temperature of 65 mK and trapping times of several seconds were obtained. There is no fundamental obstacle to achieving lower temperatures in the future.

As formaldehyde in this state is stable against inelastic two-body collisions this fulfills an important requirement for evaporative or sympathetic cooling of this species in, e.g., a microwave trap which takes one a step closer to the long-term goal of quantum degeneracy.

[1] A. Prehn *et al.*, *Phys. Rev. Lett.* **116**, 063005 (2016).

MO 1.5 Mon 12:00 F102

Towards direct laser cooling of barium monofluoride

— ●MARIAN ROCKENHÄUSER, FELIX KOGEL, EINIUS PULTINEVICIUS, TATSAM GARG, and TIM LANGEN — UNI Stuttgart, 5. Physikalisches Institut, IQST

We report on our progress towards the laser cooling of BaF molecules. This molecular species shows high promise for various types of precision measurement applications. However, due to its high mass, complex hyperfine structure and branching losses through intermediate states, it is also notoriously difficult to cool. In an effort to realize laser cooling, we have performed high-resolution absorption spectroscopy of the lowest rovibrational states to determine an improved set of molecular constants. This has allowed us to identify missing cooling and repumping transitions necessary to realize laser cooling of BaF, as well as to realize near background-free fluorescence imaging of a cold molecular beam

MO 1.6 Mon 12:15 F102

Optical properties of the Si₂O⁺ cation

— ●EMIL MICKELIN, TAARNA STUEMUND, KAI POLLOW, MARKO FÖRSTEL, and OTTO DOPFER — Institut für Optik und Atomare Physik, Technische Universität Berlin, Berlin, Germany

The emission of SiO from stars is well-known and proven. Moreover, the existence of different μm -sized silicate grains in interstellar dust is observed, but the formation pathway is unknown and information concerning larger molecules and their ions is missing.

In our project we are looking for transitions of cluster and characterize them via their measured optical spectrum.

In this talk, experimental data and quantum chemical calculations on the absorption and dissociation properties of Si₂O⁺ are presented. The spectrum of Si₂O⁺, which are created in a laser vaporization source, was obtained by photodissociation of mass-selected Si₂O⁺ cations in a tandem mass spectrometer. The experimental results are discussed and compared with theoretical results of TD-DFT calculations.

Significantly, our optical spectrum provides the first spectroscopic information for this simple triatomic cation.

MO 1.7 Mon 12:30 F102

Threshold photodetachment spectroscopic studies of C₂⁻

— ●SRUTHI PURUSHU MELATH, CHRISTINE MARIA LOCHMANN, MARKUS NÖTZOLD, ROBERT WILD, and ROLAND WESTER — Institut für Ionenphysik und Angewandte Physik, Universität Innsbruck, Austria

Photodetachment spectroscopy is a powerful spectroscopic technique for determining the internal state distribution of a molecular anion. The dicarbon anion, our current molecule of interest, is a well-studied system due to its stable electronic level structure and potential laser cooling transition [1].

Here we present the photodetachment spectroscopy of C₂⁻ near threshold in a radiofrequency 16-pole wire trap at 8 K. The main goal of the experiment is to analyze the behavior of the cross section near the threshold, determine the electron affinity more precisely than previously measured [2,3], and if possible, obtain a rotationally resolved photodetachment signal as a function of photon energy. The status of

the project will be presented.

[1]. M. Nötzold *et al.*, Phys. Rev. A 106, 023111 (2022) [2]. K. M. Ervin, *et al.*, J. Phys. Chem. 95, 2244 (1991) [3]. B. A. Laws *et al.*, Nat. Commun. 10, 1(2019)

MO 1.8 Mon 12:45 F102

Theoretical study of photoassociation of ultracold $^{23}\text{Na}^{39}\text{K}$ and ^{39}K — ●BARAA SHAMMOUT¹, CHARBEL KARAM², LEON KARPA¹, EBERHARD TIEMANN¹, SILKE OSPELKAUS¹, and OLIVIER DULIEU² — ¹Institut für Quantenoptik, Universität Hannover — ²Université Paris-Saclay, CNRS, Laboratoire Aimé Cotton

Understanding the physics underlying ultracold alkali atom-diatom

collisions is essential for full quantum control on ultracold molecules. The long-range photoassociation (PA) process of loosely-bound ultracold trimers from a scattering state of atom-diatom is a possible pathway to investigate their collisional properties. In this work, we present a long-range model for modeling photoassociation of ultracold $^{23}\text{Na}^{39}\text{K}$ and ^{39}K close to the molecular resonant excitation $\text{NaK}(X^1\Sigma) \rightarrow \text{NaK}(b^3\Pi)$. We have calculated potential energy surfaces (PESs) for the low-lying doublet excited states of NaK_2 up to the $\text{NaK}(b^3\Pi)+\text{K}(4s)$ dissociation limit. We extracted the energy of vibrational-rotational levels using the time-independent close-coupling method, restricted to the long-range PESs. Finally, we demonstrate the possibility of experimental observation of trimer photoassociation by estimating trimer PA-rates.

MO 2: Photochemistry

Time: Monday 11:00–13:00

Location: F142

MO 2.1 Mon 11:00 F142

Time-resolved transient absorption spectroscopy of oxindole-based molecular motors and switches — ●CAMILO GRANADOS^{1,2}, MATTHEW MGBUKWU², DAN DOELLERER³, DAISY POOLER³, ALINA KHODKO^{1,4}, BEN L. FERINGA³, JÉRÉMIE LÉONARD², OLEG KORNILOV¹, and STEFAN HAACKE² — ¹Max Born Institute, Max-Born-Straße 2A, 12489 Berlin, Germany — ²Université de Strasbourg, CNRS, Institut de Physique et Chimie des Matériaux de Strasbourg, UMR 7504, F-67034 Strasbourg, France — ³Stratingh Institute for Chemistry, Zernike Institute for Advanced Materials, University of Groningen, Nijenborgh 4, 9747 AG Groningen, The Netherlands — ⁴Institute of Physics NAS of Ukraine, Nauky Ave, 46, 03028, Kyiv, Ukraine

The design and characterization of synthetic molecules (motors and switches) is of vital importance for the construction of larger artificial structures that can be used for harvesting light energy and for other light-induced functionality [1]. However, the isomerisation quantum yields of the existing synthetic systems [1], are still low compared to the natural chromophores. Time-resolved transient absorption spectroscopy (TAS) can map the evolution of the relaxation of the molecule along the potential energy surfaces [1] and reveal the mechanisms affecting the isomerisation quantum yields. We will present recent TAS experiments on oxindole-based molecular motors and switches dissolved in different solvents. We will further present plans to investigate these synthetic molecular systems using TRPES. [1] DRS Pooler *et al.*, Chem. Sci., 12, 7486 - 7497 (2021).

MO 2.2 Mon 11:15 F142

Unraveling the photochemistry of $\text{Ti}^{IV}(\text{Cp})_2(\text{NCS})_2$ — ●JONAS SCHMIDT, LUIS IGNACIO DOMENIANNI, and PETER VÖHRINGER — University of Bonn, DE

Recently, the complex, $\text{Cp}_2\text{Ti}(\text{Cl})_2$, has been used as a photo-redox-catalyst for atom economical transformations in one-electron steps.^[1] We investigated the initial excitation and quenching of the catalyst using time-correlated single-photon-counting and femtosecond ultraviolet-pump mid-infrared-probe spectroscopy (UVmIR). The chlorido ligands were substituted with isothiocyanato ligands to render the catalyst amenable to UVmIR.

We recorded the emission spectrum and determined luminescence lifetimes of $\text{Cp}_2\text{Ti}(\text{NCS})_2$ in liquid tetrahydrofuran solution at room temperature. The bi-exponential nature of the luminescence decay is highly indicative of thermally activated delayed fluorescence in addition to prompt fluorescence from the optically prepared singlet excited state. The triplet state was successfully quenched with triphenylamine and a Stern-Vollmer quenching constant of $9.4 \times 10^9 \frac{\text{L}}{\text{mol s}}$ was determined

UVmIR data obtained continuously from 50 femtoseconds to several microseconds supported this interpretation and the vibrational signatures of the S_1 - and T_1 -states were obtained by performing a target analysis of the time- and frequency-dependent pump-probe data set.

References:

[1] Z. H. Zhang *et al.*, *Angew. Chem. Int.* **2020**, *59*, 9355–9359.

MO 2.3 Mon 11:30 F142

Ultrafast Photochemistry of Metallo-Nitrenes — ●MARKUS BAUER¹, TILL SCHMIDT-RÄNTSCH², LUIS DOMENIANNI¹, SVEN SCHNEIDER², and PETER VÖHRINGER¹ — ¹Clausius Institut

für physikalische Chemie, Rheinische Friedrich-Wilhelms-Universität Bonn, Deutschland — ²Institut für Anorganische Chemie, Georg-August-Universität Göttingen, Deutschland

Metallo-nitrenes, formed by the photochemical decomposition of metallo-azides, have recently shown to be promising complexes for chemical catalysis, specifically for nucleophilic metallo-nitrene C-H insertion.^[1] The exact reaction pathways, as well as the electronic structures of intermediate species remain so far largely unknown.

In this work, the photochemistry of the square-planar $[\text{M}(\text{N}_3)(\text{PNP})]$ ($\text{PNP}=\text{N}(\text{CHCHP}^t\text{Bu}_2)_2$, $\text{M}=\text{Pd}$, Pt) complexes after excitation with 320 nm light were investigated using ultrafast UV-pump mIR-probe and time resolved Fourier transform IR-spectroscopy.

The data reveal that dinitrogen cleavage from the photolabile azide group occurs from the triplet state on a time scale in excess of 1 ns. The quantum yield for nitrene formation depends on the nature of the metal.

[1] T. Schmidt-Räntsch, H. Verplancke, J. N. Lienert, S. Demeshko, M. Otte, G. P. Van Trieste, K. A. Reid, J. H. Reibenspies, D. C. Powers, M. C. Holthausen, S. Schneider, *Angew. Chem. Int. Ed.* **2022**, *61*, e202115626;

MO 2.4 Mon 11:45 F142

Photodynamics of arylazopyrazole derivatives: new insights from molecular dynamics studies — HELENA OSTHUES, ●MARCUS BÖCKMANN, and NIKOS DOLTSINIS — Institut für Festkörpertheorie, Westfälische Wilhelms-Universität, Münster, Germany

In the realm of photoswitches, azobenzene is an archetype for studying photodynamics primarily due to its fatigue resistance, simplicity, and tuneability by substitutions at the phenyl rings [1]. Here, we could demonstrate that the time scale of photoisomerisation can be drastically changed upon chemical modification [2]. Recently, the discovery of quantitative arylazopyrazole (AAP) photoswitches with long thermal lifetimes has pushed into focus this class of azobenzene derivatives, where one phenyl ring is replaced by a less bulky five-membered ring [3]. In this contribution, we report on dynamical photoisomerisation simulations of AAP derivatives elucidating the effect of different substituents and solvents on nonradiative lifetimes [4].

[1] H. M. D. Bandara and S. C. Burdette, *Chem. Soc. Rev.* **41**, 1809-1825 (2012).

[2] M. Böckmann, N. L. Doltsinis, and D. Marx, *J. Chem. Phys.* **137**, 22A505 (2012).

[3] L. Stricker, M. Böckmann, T. M. Kirse, N. L. Doltsinis, and B. J. Ravoo, *Chemistry - A European Journal*, **24**, 8639-8647 (2018).

[4] H. Osthues and N. L. Doltsinis, *J. Chem. Phys.* in press.

MO 2.5 Mon 12:00 F142

Multireference Chlorophyll Nuclear and Electron Q-Band Dynamics: a Theoretical XAS Study — ●LENA BÄUML, SEBASTIAN REITER, FLORIAN ROTT, BASTIAN MICHELS, and REGINA DE VIVIE-RIEDLE — Department of Chemistry, LMU Munich, Germany

Chlorophylls play a vital role during photosynthetic light-harvesting. Here, the nonradiative relaxation of high-energy excited states to the lowest excited state is of central importance.

We simulate the ultrafast relaxation process in the Q-bands of chlorophyll in a representative 2D space using grid-based wave packet quantum dynamics. The excited state energies and potential energy

surfaces are computed at the XMS-CASPT2 level of theory to capture the multi-reference character of chlorophyll excitations. We propose a possibility to observe the wave packet dynamics, as well as the strong coupling between the Q_x and Q_y state via magnesium K-edge X-ray absorption spectra. Following the RASPT2 procedure outlined by Rott *et al.*^[1] our results show from a fully quantum mechanical point of view how the Q_x and Q_y band are strongly coupled by internal vibrations,^[2] in contrast to the Gouterman model. Thus the absorption intensity should be spread over the whole Q-band, influencing charge and energy transfer in photosynthetic light-harvesting complexes, such as photosystem 1.

[1] F. Rott *et al.*, *Struct. Dyn.*, **8**, 034104 (2021).

[2] L. Bäuml *et al.*, *Phys. Chem. Chem. Phys.* **24**, 27212 (2022).

MO 2.6 Mon 12:15 F142

The role of dephasing for dark states and polaritonic chemistry — ●ERIC DAVIDSSON and MARKUS KOWALEWSKI — Department of Physics, Stockholm University, Albanova University Center, SE-106 91 Stockholm, Sweden

Common quantum-mechanical models for chemistry in optical cavities lack a mechanism to populate collective dark states. In this work, we explicitly model a process that does populate these states; i.e. disorder from loss of phase information (decoherence) in the matter sub-systems. Such processes arise due to local environment interactions, and the effect enters into the equations of motion as dephasing operators. Viewed through the lens of polaritonic states, a reservoir of previously inaccessible states has thus opened up. Since these states are superpositions of excitations in only matter sub-systems, one would expect that dephasing can protect excited states from photon decay. In this work, we find that dephasing indeed does that quite effectively. We also discuss how to understand the same physical result in a standard product basis, where there are no dark states.

MO 2.7 Mon 12:30 F142

Cavity-induced effects on the ground-state chemical reactivity of the click reaction — ●THOMAS SCHNAPPINGER and MARKUS KOWALEWSKI — Department of Physics, Stockholm University, Sweden

If a molecule interacts with the vacuum field of a nanoscale cavity, strong coupling reshapes the potential energy surfaces, forming hybrid light-matter states, termed polaritons. Recent experiments show that this strong coupling between light and matter is capable of modifying chemical and physical properties. The situation in which the quantized cavity modes are coupled via their characteristic frequency to vibrational degrees of freedom of molecules is called vibrational strong coupling (VSC). In the VSC regime, the chemistry of a single electronic state (mostly the ground-state) and its vibrational spectroscopy are influenced by the cavity interaction.

In this theoretical contribution, we study different aspects to see to what extent and how the chemical reactivity can be altered by VSC in the single-molecule case. As an illustrative example, we are investigating the azide-alkyne Huisgen cycloaddition, which is better known as the prototypical click reaction. We describe the hybrid light-molecule matter system with the help of an extended Jaynes-Cummings-like model taking into account the counter-rotating terms and the dipole-self-energy terms. In this setup, we can realize coupling to multiple cavity modes and study the cavity-induced changes of ground-state energies, geometries, and activation energies.

MO 2.8 Mon 12:45 F142

Suppressing non-radiative decay of photochromic organic molecular systems in the strong coupling regime — ●MARKUS KOWALEWSKI and RAFAEL C. CUOTO — Stockholm University, Stockholm, Sweden

Organic solar cells and related optoelectronic applications rely on molecules with long-lived electronic states. Non-radiative decay channels, which are caused, for example, by non-adiabatic processes in the molecule can have a significant impact on the efficiency of these devices. More favorable lifetimes are in practice, achieved by chemical substitution of particular base compound.

In this contribution, we investigate meso-tert-butyl-BODIPY, which is known to for its low fluorescence yield, caused by the non-radiative decay through a conical intersection [1]. We show theoretically that strong light-matter coupling by means of an optical nano-cavity may be used to modify the excited state lifetime.

[1] R. C. Couto, M. Kowalewski, *Phys. Chem. Chem. Phys.*, **24**, 19199 (2022).

MO 3: Interaction with Strong or Short Laser Pulses I (joint session A/MO)

Time: Monday 11:00–13:00

Location: F303

Invited Talk

MO 3.1 Mon 11:00 F303

Time-resolved Kapitza-Dirac effect — ●KANG LIN, MAKSYM KUNITSKI, SEBASTIAN ECKART, ALEXANDER HARTUNG, QINYING JI, LOTHAR SCHMIDT, MARKUS SCHÖFFLER, TILL JAHNKE, and REINHARD DÖRNER — Goethe University

The Kapitza-Dirac effect describes that that an electron beam can be diffracted when passing through a light standing wave. In analogy to optical diffraction, the incident electron beam behaves like a wave, while the light standing wave plays the role of the grating. The Kapitza-Dirac effect serves as an optical diagnosis of the electron property in frequency domain. However, with the advent of pulsed laser technique, the ultrafast time information is imprinted in both the electron wavepacket and the light standing wave. It is totally unclear how an electron wavepacket will be diffracted by an ultrafast light standing wave. Here, a principle new phenomenon, termed as time-resolved Kapitza-Dirac effect, is discovered. We track the spatiotemporal evolution of an electron wavepacket diffracted by an ultrafast femtosecond (10-15 seconds) light standing wave. By scanning the time delay between the electron wavepacket and the standing wave, we observe so far unseen quantum interference effects. We show that the momentum spacing between diffraction peaks decreases continuously with the time delay increasing, which can be fractions instead of multiply integers of 2-photon momenta. The time-resolved Kapitza-Dirac effect can directly measure the chirp of the electron wavepacket optically.

MO 3.2 Mon 11:30 F303

Laser-Driven Acceleration of Gold Ions — ●LAURA DESIREE GEULIG, ERIN GRACE FITZPATRICK, MAXIMILIAN WEISER, VERONIKA KRATZER, VITUS MAGIN, MASOUD AFSHARI, JÖRG SCHREIBER, and PETER G. THIROLF — Ludwig-Maximilians-Universität München

The efficient acceleration of gold ions is a first step towards the 'fission-fusion' reaction mechanism, which aims at investigating the rapid neutron capture process in the vicinity of the N=126 waiting point [1]. In our recent measurement at the PHELIX laser with a pulse length of 500fs, for the first time, the laser-based acceleration of gold ions above 7 MeV/u was demonstrated. Additionally, individual gold charge states were resolved with unprecedented resolution [2]. This has allowed the investigation of the role of collisional ionization using a developmental branch of the particle-in-cell simulation code EPOCH [3], showing a much better agreement of the simulated charge state distributions with the experimentally measured ones than when only considering field ionization. This work is continued at the Centre for Advanced Laser Applications (CALA), using the ATLAS3000 laser (800nm central wavelength, 25 fs pulse length).

[1] D. Habs *et al.*, *Appl. Phys. B* **103**, 471-484 (2011)

[2] F.H. Lindner *et al.*, *Sci. Rep.* **12**, 4784 (2022)

[3] M. Afshari *et al.*, *Sci.Rep.* **12**, 18260 (2022)

MO 3.3 Mon 11:45 F303

Transfer learning and visualization of a convolutional neural network for recognition of the internuclear distance in a molecule from electron momentum distributions — ●NIKOLAY SHVETSOV-SHILOVSKI and MANFRED LEIN — Leibniz Universität Hannover

We use a convolutional neural network (CNN) to retrieve the internuclear distance in the two-dimensional H_2^+ molecule ionized by an intense few-cycle laser pulse from the photoelectron momentum distributions [1]. We study the effect of the carrier-envelope phase on the retrieval of the internuclear distance with a CNN [2]. By using the transfer learning technique, we make our CNN applicable to mo-

mentum distributions obtained at the parameters it was not explicitly trained for. We compare the CNN with alternative approaches that are shown to have very limited transferability. Finally, we use the occlusion sensitivity technique to extract features of the momentum distributions that allow a CNN to predict the internuclear distance.

[1] N. I. Shvetsov-Shilovski and M. Lein, Phys. Rev. A, 105, L021102 (2022).

[2] N. I. Shvetsov-Shilovski and M. Lein, submitted to Phys. Rev. A, arXiv:2211.01210.

MO 3.4 Mon 12:00 F303

Holographic Single-Shot Imaging and Reconstruction of ultrafast laser-driven dynamics in thin films — ●RICHARD ALTENKIRCH, FRANZISKA FENNEL, CHRISTIAN PELTZ, THOMAS FENNEL, and STEFAN LOCHBRUNNER — Institute of Physics, Universität Rostock, Germany

Well controlled laser material processing with a spatial resolution on the scale of the laser wavelength is significant to a large variety of both research and industrial applications. A full characterization of the spatial and temporal evolution of the ultrafast laser-induced plasma dynamics will be key to future developments in the respective fields. So far, the established diagnostic methods are mostly sensitive to the target absorption and luminescence. Here, we present an experimental and numerical approach based on coherent diffractive imaging (CDI), a technique well known from free particle characterization at XFEL's [1], also providing access to the phase delay caused by the target. In a two-color pump-probe experiment, a thin film is excited by a short pump pulse and the resulting plasma dynamics are imaged by a frequency doubled probe pulse. The corresponding complex near field behind the target is reconstructed from the recorded scattering image via a phase retrieval approach [2]. We also present a thorough characterization of the method and a first successful application to experimental data.

[1] H. Chapman et al., Nature Physics 2 839-843 (2006)

[2] J. Fienup, Appl. Opt. 21, 2758-2769 (1982)

MO 3.5 Mon 12:15 F303

Modeling controlled sub-wavelength plasma formation in dielectrics — ●JONAS APPORTIN, CHRISTIAN PELTZ, BJÖRN KRUSE, BENJAMIN LIEWEHR, and THOMAS FENNEL — Institute for Physics, Rostock, Germany

Laser induced damage in dielectrics due to short pulse excitation plays a major role in a variety of scientific and industrial applications, such as the preparation of 3D structured evanescently coupled waveguides [1]. Here the irreversible modifications originate from higher order nonlinearities like strong field ionization and plasma formation. Improving user control over these material modifications, e.g. permanent refractive index modifications, therefore strongly relies on a better understanding of the underlying interaction dynamics, in particular the early phases of interaction. To this end we developed and utilized a numerical model, that combines a local description of the dynamics in terms of corresponding rate equations for ionization, collisions and heating with a fully electromagnetic field propagation via the Finite-Difference-Time-Domain method, adding self-consistent feedback ef-

fects like the sudden buildup of plasma mirrors. Here we present first numerical results regarding the creation and control of sub-wavelength gratings formed at the rear side of fused silica films.

[1] D. Blömer et al., Opt. Express 14, 2151-2157 (2006)

MO 3.6 Mon 12:30 F303

Ultrafast two-electron correlations from metal needle tips — ●JONAS HEIMERL, STEFAN MEIER, and PETER HOMMELHOFF — Department Physik, Friedrich-Alexander-Universität Erlangen-Nürnberg (FAU), 91058 Erlangen

When two electrons are emitted in a very confined space-time volume on the nanometer-femtosecond scale, strong Coulomb interaction is present. With the advent of multi-hit capable electron detectors, the field of ultrafast light matter interaction around metal needle tips is venturing into correlated multi-electron dynamics. Here we show the Coulomb-induced energy anti-correlation of two electrons emitted from nanometer-sized tungsten needle tips triggered by femtosecond laser pulses [1]. We extract two important key parameters: (1) the mean energy splitting of 3.3 eV and (2) the correlation decay time of 82 fs. Both parameters are essential for modern ultrafast electron microscopes, as shown in similar work from the Göttingen group [2]. We demonstrate that by filtering the electrons energetically, clear sub-Poissonian distributed electron beams can be achieved, highly relevant for beating the shot-noise limit in imaging applications. Furthermore, we show that in the strong field regime, where ponderomotive effects of the laser field become important, the anti-correlation gap is strongly influenced.

[1] S. Meier, J. Heimerl and P. Hommelhoff, arXiv:2209.11806 (2022)

[2] R. Haindl et al., arXiv:2209.12300 (2022)

MO 3.7 Mon 12:45 F303

Testing Born's rule via photoionization of helium — ●PETER ROBERT FÖRDERER¹, DAVID BUSTO^{1,2}, ANNE L'HULLIER², ANDREAS BUCHLEITNER^{1,3}, and CHRISTOPH DITTEL^{1,3,4} — ¹Physikalisches Institut, Albert-Ludwigs-Universität Freiburg, Hermann-Herder-Straße 3, 79104 Freiburg, Germany — ²Department of Physics, Lund University, Box 118, 22100 Lund, Sweden — ³EUCOR Centre for Quantum Science and Quantum Computing, Albert-Ludwigs-Universität Freiburg, Hermann-Herder-Straße 3, 79104 Freiburg, Germany — ⁴Freiburg Institute for Advanced Studies, Albert-Ludwigs-Universität Freiburg, Albertstraße 19, 79104 Freiburg, Germany

We propose a protocol to test Born's rule (a fundamental axiom of quantum mechanics) via the Sorkin test, applied to photoionization of helium, induced by the combination of an ultrashort extreme ultraviolet pulse with a trichromatic infrared laser pulse. We numerically simulate the outcome of the Sorkin test, the Sorkin parameter κ , for realistic parameters and randomly sampled, typical experimental imperfections. The latter do not only lead to a spread, but also to a systematic offset of κ from its ideal value $\kappa = 0$. A determination of κ with an achievable precision of the order $10^{-3} - 10^{-4}$ is predicted, which is comparable to the precision of 10^{-3} reached in the best optical experiments [Kauten, et. al., New J. Phys. 19, 033017 (2017)] in the quantum regime.

MO 4: Ultrafast Dynamics I (joint session MO/A)

Time: Tuesday 11:00–13:00

Location: F102

Invited Talk

MO 4.1 Tue 11:00 F102

Revealing chiral charge migration in UV-excited molecules — ●VINCENT WANIE¹, ETIENNE BLOCH², ERIK P. MÅNSSON¹, LORENZO COLAIZZI^{1,3}, SERGEY RYABCHUK³, KRISHNA SARASWATHULA^{1,3}, ANDREA TRABATTONI^{1,4}, VALÉRIE BLANCHET², NADIA BEN AMOR⁵, MARIE-CATHERINE HEITZ⁵, YANN MAIRESSE², BERNARD PONS², and FRANCESCA CALEGARI^{1,3} — ¹DESY, Germany — ²Université de Bordeaux - CNRS - CEA, CELIA, France — ³Universität Hamburg, Germany — ⁴Leibniz Universität Hannover, Germany — ⁵CNRS, France

Electron-driven charge migration occurs following photoexcitation of a molecule, leading to a charge density traveling rapidly along the molecular structure. We report our most recent works devoted to the investigation of charge migration in neutral molecules and its applications to manipulate the outcome of photochemical and photophysical processes. We exploited our new light source delivering few-femtosecond UV pulses in order to photoexcite below the ionization threshold and

trigger electronic dynamics in chiral methyl-lactate. We used time-resolved photoelectron circular dichroism (TR-PECD) to image electronic coherences driving charge migration and disclose - for the first time - their impact on the molecular chiral response, allowing for an ultrafast chiroptical switching effect where the amplitude and direction of the photoelectron current generated by PECD can be controlled on a sub-10 fs timescale. The results provide important perspectives to exploit charge-directed reactivity for controlling the chiral properties of matter at the molecular scale. [1] V. Wanie et al., 'Ultrafast chiroptical switching in UV-excited molecules,' (under review, 2022).

MO 4.2 Tue 11:30 F102

UV and Mid-IR Photo-induced Dissociation Dynamics of Solvated (Bio)Molecular Complexes — ●MUKHTAR SINGH^{1,2,3}, MATTHEW SCOTT ROBINSON^{1,2,3}, HUBERTUS BROMBERGER^{1,2}, JOLIJN ONVLEE^{1,3}, SEBASTIAN TRIPPEL^{1,2}, and JOCHEN KÜPPER^{1,2,3} — ¹Center for Free-Electron Laser Science CFEL, Deutsches

Elektronen-Synchrotron DESY, Hamburg — ²Center for Ultrafast Imaging, Universität Hamburg — ³Department of Physics, Universität Hamburg

We present the imaging of ultrafast UV- and thermal-energy-induced chemical dynamics of micro-solvated (bio)molecular complexes probed with strong-field ionization techniques [1]. We produce a pure gas-phase indole-water sample using a combination of a cold molecular beam and the electrostatic deflector [2]. To study the induced dynamics, we set up both a UV and a mid-IR pump-probe experiment, in which a 269 nm and 2.9 μm laser pulses were used to excite the system, respectively. A 1.3 μm laser pulses was used for ionizing the system. First experiments focused on the ion imaging of the UV and mid-IR-triggered systems. Furthermore, we will report on efforts to use laser-induced electron diffraction (LIED) [3,4] to probe the molecular dynamics to obtain structural information with atomic resolution.

[1] J Onvlee, *et al.*, *Nat Commun.*, DOI: 10.1038/s41467-022-33901-w

[2] S. Trippel, *et al.*, *Rev. Sci. Instrum.* **89**, 096110 (2018)

[3] J. Wiese, *et al.*, *Phys. Rev. Research* **3**, 013089 (2021)

[4] E. T. Karamatskos, *et al.*, *J. Chem. Phys.* **150**, 244301(2019)

MO 4.3 Tue 11:45 F102

Supramolecular dynamics investigated on hydrogen bonded pyrrole-water clusters upon site-specific x-ray photoionization — ●IVO S. VINKLÁREK¹, HUBERTUS BROMBERGER¹, WUWEI JIN¹, REBECCA BOLL², MICHAEL MEYER², SEBASTIAN TRIPPEL¹, and JOCHEN KÜPPER^{1,3,4} — ¹Center for Free-Electron Laser Science, Deutsches Elektronen-Synchrotron DESY, Hamburg — ²European XFEL GmbH, Schenefeld — ³Department of Physics, Universität Hamburg — ⁴Center for Ultrafast Imaging, Universität Hamburg

Solvation of molecules crucially affects their photostability and opens additional pathways for the relaxation dynamics compared to isolated molecules. We intend to get molecular-level insight into the solvation effect in photofragmentation dynamics of a supramolecular system through our molecular beam experiments with stoichiometrically well-defined pyrrole-water (Pyr-H₂O) clusters [1]. Concretely, the dissolution dynamics of the spatially separated pure sample of Pyr-H₂O clusters prepared by the electric deflector was investigated through an IR-pump-x-ray-probe experiment. The singly ionizing IR-pulse triggers the (Pyr-H₂O)⁺ fragmentation, which is then site-specifically probed by x-ray free-electron laser pulses [2] at different times of the pyrrole-H₂O separation. The study of the hydrogen-bound Pyr-H₂O system is especially relevant to abundant pyrrole-containing biomolecules and establishes a novel approach for investigating the key role of intermolecular interactions in supramolecular dynamics.

[1] Johny, M. et al. *Chem. Phys. Lett.*, **2019**, 721, 149-152. [2] Onvlee, J. et al., *Nat. Commun.*, in press, arXiv:2103.07171 [physics]

MO 4.4 Tue 12:00 F102

Real time tracking of ultrafast dynamics in liquid water — ●GAIA GIOVANNETTI¹, AMMAR BIN WAHID¹, SERGEY RYABCHUK¹, HUI-YUAN CHEN², VINCENT WANIE¹, ANDREA TRABATTONI^{1,3}, ERIK MAANSSON¹, HUGO MARROUX⁴, MAJED CHERGUI², and FRANCESCA CALEGARI^{1,5} — ¹Center for Free-Electron Laser Science, DESY, Notkestr. 85, 22607 Hamburg, Germany — ²Ecole Polytechnique Fédérale de Lausanne, Rte Cantonale, 1015 Lausanne, Switzerland — ³Institute of Quantum Optics, Leibniz Universität Hannover, Welfengarten 1, 30167 Hannover, Germany — ⁴Laboratoire Interactions, Dynamiques et Lasers, CEA-Saclay, 91191 Gif-sur-Yvette, France — ⁵The Hamburg Centre for Ultrafast Imaging, Universität Hamburg, 22761 Hamburg, Germany

Understanding the properties of water is key to determinate the effects of the liquid environment on the dynamics of biological systems. In our experiment, a 3 fs visible pump impulsively creates a vibrational wave-packet, whose evolution is probed by a time-delayed sub-2 fs UV pulse [1]. As a result of the wave-packet dynamics, the probe signal is modulated in time and the vibrational spectrum can be obtained by a Fourier analysis of the temporal interferogram. A preliminary analysis of our data shows a transient signal whose oscillation period (~ 11 fs) and decay time (~ 70 fs) match the values expected for the O-H stretching mode in the ground electronic state of liquid bulk water [2]. Further theoretical insights will allow us to assign specific contributions from the ground, excited and ionized states. [1] Opt. Lett. **44**, 1308-1311 (2019) [2] J. Chem. Phys. **135**, 244503 (2011).

MO 4.5 Tue 12:15 F102

Systematic variation of triplet chromophore energies in

iron(II) complexes linked to organic chromophores — ●MORITZ LANG¹, PHILIPP DIERKS², MIGUEL ARGÜELLO CORDERO¹, MATTHIAS BAUER², and STEFAN LOCHBRUNNER¹ — ¹Institute for Physics, University of Rostock, Germany — ²Faculty of Science, CSSD, Paderborn University, Germany

For the efficient conversion of solar light, photosensitizers with appropriate absorption properties and long living excited states are crucial. Iridium and ruthenium complexes stand out due to their extraordinary stable triplet metal-to-ligand charge transfer (3MLCT) excited states but are expensive and toxic. To find sustainable alternatives, iron-based metal complexes are intensely studied. But due to an efficient internal conversion pathway mediated by metal centered states, the MLCT lifetime and therefore the performance of these types of complexes are still limited. In a systematic study the influence of various chromophores attached to homoleptic iron complexes was investigated. The excited state dynamic was studied by ultrafast transient absorption spectroscopy. For particular chromophores an additional decay component was observed, exceeding the lifetime of the otherwise predominant 3MLCT state by an order of magnitude. With fitting energy levels, a triplet state of the chromophore is populated during the relaxation process, achieving a comparably stable intermediate configuration. A better understanding and further improvement of these systems will contribute to the ultimate goal of developing efficient iron based photosensitizers for solar energy conversion.

MO 4.6 Tue 12:30 F102

Ultrafast dynamics of photochemical nitrile imine formation — ●STEFAN FLESCH and PETER VÖHRINGER — Clausius-Institut für Physikalische und Theoretische Chemie, Rheinische Friedrich-Wilhelms-Universität Bonn, Wegelerstr. 12, D-53115 Bonn

The chemical reactivity of nitrile imines is of great utility in organic synthesis with applications rapidly expanding into the materials and life sciences.¹ Yet, our understanding of the electronic and molecular structures of nitrile imines remains incomplete and the elementary mechanism of their photoinduced generation is entirely unknown. Here, femtosecond infrared spectroscopy after 266 nm-excitation of 2,5-diphenyltetrazole has been carried out to temporally resolve the formation and structural relaxation dynamics of the nascent diphenyl-nitrile imine in liquid solution under ambient conditions.² An initial sequence of intersystem crossings within 250 fs is followed by the cleavage of N₂ with formation of a structurally relaxed nitrile imine on the adiabatic ground-state singlet surface within a few tens of picoseconds. The infrared spectrum supports the notion of a "floppy" nitrile imine molecule whose equilibrium character ranges from fully propargylic to fully allenic under these conditions.

References:

1 G. Bertrand, C. Wentrup, *Angew. Chem. Int. Ed. Engl.* **1994**, *33*, 527-545.

2 S. Flesch, P. Vöhringer, *Angew. Chem. Int. Ed.* **2022**, e202205803.

MO 4.7 Tue 12:45 F102

Investigating the oxidation states of a perylene bisimide cyclophane with ultrafast spectroelectrochemistry — ●REBECCA FRÖHLICH¹, JESSICA RÜHE², MICHAEL MOOS², FRANK WÜRTHNER², CHRISTOPH LAMBERT², and TOBIAS BRIXNER¹ — ¹Institut für Physikalische und Theoretische Chemie, Universität Würzburg, Am Hubland, 97074 Würzburg — ²Institut für Organische Chemie, Universität Würzburg, Am Hubland, 97074 Würzburg

From photosynthesis to optoelectronic devices, charged species fulfill essential roles in our everyday world. With spectroelectrochemistry the oxidation states of molecules can be generated and investigated under potential control. In our setup we combine spectroelectrochemistry with femtosecond transient absorption spectroscopy to investigate the dynamics of charged species on an ultrafast timescale [1].

Here we show new data on the oxidation states of a perylene bisimide cyclophane [2]. A fit of the cyclic voltammetry data of the molecule shows four reduction steps with closely lying redox potentials. The four reduced states show a change in absorption which is highlighted by the deconvolution of the absorption spectroelectrochemistry data. Through the fits of the absorption spectra the charged species can be distinguished in a set of transient absorption spectroelectrochemistry maps. The excited state lifetimes of the reduced molecule are analyzed with global fitting and change according to their oxidation state.

[1] J. Heitmüller et al., *Spectrochim. Acta Part A*, **253**, 119567 (2021)

[2] J. Rühle et al., *Organic Materials*, **2**, 149-158 (2020)

MO 5: Electronic Spectroscopy

Time: Tuesday 11:00–13:00

Location: F142

MO 5.1 Tue 11:00 F142

Highly-resolved Stark effect measurements of Rydberg states in nitric oxide — ●FABIAN MUNKES^{1,4}, PATRICK KASPAR^{1,4}, PHILIPP NEUFELD^{1,4}, ALEXANDER TRACHTMANN^{1,4}, YANNICK SCHELLANDER^{2,4}, LARS BAUMGÄRTNER^{3,4}, ROBERT LÖW^{1,4}, TILMAN PFAU^{1,4}, and HARALD KÜBLER^{1,4} — ¹Physikalisches Institut, Universität Stuttgart, Pfaffenwaldring 57, 70569 Stuttgart — ²Institut für Großflächige Mikroelektronik, Universität Stuttgart, Allmandring 3b, 70569 Stuttgart — ³Institut für Intelligente Sensorik und Theoretische Elektrotechnik, Universität Stuttgart, Pfaffenwaldring 47, 70569 Stuttgart — ⁴Center for Integrated Quantum Science and Technology (IQST), Universität Stuttgart

We demonstrate Stark effect measurements at room temperature of high-lying Rydberg states in nitric oxide. These states are generated using a three-photon continuous-wave excitation scheme. The readout is based on the detection of charged particles created by collisional ionization of Rydberg molecules. A theoretical discussion of the gained experimental results is given.

MO 5.2 Tue 11:15 F142

Threshold Photoelectron Spectra of fragments of AsMe3 and SbMe3 — ●EMIL KARAEV¹, MARIUS GERLACH¹, PATRICK HEMBERGER², and INGO FISCHER¹ — ¹Julius-Maximilians-Universität, Würzburg, Germany — ²Swiss Light Source, Villigen, Switzerland

Our group already investigated the pyrolysis products of methylated group V compounds $X = N^{[1]}$, $P^{[2]}$, $Bi^{[3]}$. While for the single methylated isomers of nitrogen we observed H-N-CH₂, N-CH₂ and H-N-CH, bismuth showed only Bi-CH₃. For phosphorus the isomers H-P-CH₂, P-CH₃ and P-CH₂ were detected. To complete our investigation of the 5th main group, trimethylarsenic and trimethylantimony were pyrolyzed. The emerging reactive species were characterized with the PEPICO setup of the VUV beamline of the synchrotron SLS in Switzerland. The resulting mass-selected threshold photoelectron spectra were interpreted using quantum chemical calculations and Franck-Condon simulations.

Our results show that antimony behaves similarly to bismuth, forming only Sb-CH₃. Arsenic on the other hand showed H-As-CH₂, As-CH₃ and As-CH₂, which is analogous to phosphorus.

MO 5.3 Tue 11:30 F142

Optical Absorption and Photodissociation Properties of Si_nO_m⁺ — ●TAARNA STUEMUND, MARKO FÖRSTEL, KAI POLLOW, EMIL MICKELIN, and OTTO DOPFER — IOAP, TU Berlin, Germany

The formation mechanisms of interstellar dust grains are still poorly understood. It is known, however, that these contain a significant amount of μm -sized silicate material [1]. So far only silicon monoxide (SiO) is identified in the interstellar medium [2]. We compare experimental data to quantum chemical calculations to understand photodissociation and optical absorption behavior, structures, and energies of possible Si_nO_m⁺ molecules that are potential and promising precursors. The experimental setup relies on action spectroscopy via mass spectrometry and resonant laser photodissociation of size-selected Si_nO_m⁺ clusters. These are generated via laser vaporization in a molecular beam expansion coupled to a quadrupole/time-of-flight tandem mass spectrometer and a broadly tuneable UV/VIS-OPO laser [3]. Preliminary data reveal competing fragmentation channels, their appearance energies and branching ratios, and the abundance and stability of neutral fragments. We highlight especially the spectrum of Si₃O₂⁺ measured by photodissociation and discuss it in an astrophysical context.

[1] K. Nagashima et al. *Nat.*, 2004, 428, 921-924[2] R. Wilson et al. *Astrophys. J.*, 1971 167, L97[3] M. Förstel et al., *Rev. Sci. Instrum.*, 2017, 88, 123110

MO 5.4 Tue 11:45 F142

The Electronic Spectrum of Si₂⁺ — ●KAI POLLOW, TAARNA STUEMUND, SOPHIE VERHOEVEN, EMIL MICKELIN, OTTO DOPFER, and MARKO FÖRSTEL — IOAP, TU Berlin, Germany

The first absorption lines of neutral Si₂ were measured 75 years ago. However, experimental information on the cation is rare. We recently

measured the first optical spectrum of Si₂⁺ via photodissociation spectroscopy[1]. We observe two vibronic band systems near 430 and 270 nm that are in very good agreement with high-level quantum-chemical calculations[2]. The measured vibronic transitions allow for determination of molecular constants in the ground and respective excited states. The optical spectrum of Si₂⁺ may enable astrophysical searches for this potential building block of interstellar silicate dust. We present the measured optical spectrum and compare it to quantum-chemical calculations.

[1] T. Studemund, K. Pollow, S. Verhoeven, E. Micken, O. Dopfer and M. Förstel *J.Phys.Chem.Lett.* 2022, 13 (33), 7624-7628.[2] Y. Liu; H. Zhai; X. Zhang; Y. Liu Ab initio calculation on the low-lying excited states of Si₂⁺ cation including spin-orbit coupling. *Chem. Phys.* 2013, 425, 156-161.

MO 5.5 Tue 12:00 F142

State selective diagnostics and spectroscopy of H₃⁺ in a heavy ion storage ring — ●AIGARS ZNOTINS¹, LUKAS BERGER¹, FLORIAN GRUSSIE¹, DAMIAN MÜLLER¹, OLDRICH NOVOTNY¹, FELIX NUSSLEIN¹, ANDREAS WOLF¹, ARNAUD DOCHAIN², XAVIER URBAIN², and HOLGER KRECKEL¹ — ¹Max-Planck-Institut für Kernphysik, Heidelberg, Germany — ²Institute of Condensed Matter and Nanoscience, Louvain-la-Neuve, Belgium

The significance of the triatomic hydrogen ion H₃⁺ for astrochemistry is well-established. It is a key contributor in a network of ion-neutral reactions that govern interstellar chemistry at low temperature and density. Additionally, as the simplest polyatomic molecule, H₃⁺ is an important benchmark system for theoretical calculations.

Due to a lack of a permanent dipole moment, laboratory spectroscopy of H₃⁺ remains a non-trivial endeavor. The highest-lying H₃⁺ levels experimentally reported have been identified by transitions from the H₃⁺ ground state to energies around 16500 cm⁻¹. Considering that the dissociation limit of H₃⁺ is at approximately 35000 cm⁻¹, more than half of the energetic landscape remains unexplored.

In this work, we discuss the possibility and requirements to extend H₃⁺ spectroscopy into the energy region above 20000 cm⁻¹. An approach for multi-color action spectroscopy is proposed to state-selectively investigate highly excited states of H₃⁺ in a cryogenic ion storage ring environment. We present model calculations describing the laser diagnostic schemes, based on a comprehensive H₃⁺ linelist.

MO 5.6 Tue 12:15 F142

Electronic Photodissociation Spectroscopy of Diamondoid Cations in a Cryogenic Trap — ●PARKER CRANDALL, SIMONE STAHL, VIKTORIA LOVASZ, MARKO FÖRSTEL, and OTTO DOPFER — Technische Universität Berlin, Berlin, Germany

Similarities have been observed between the infrared spectra of diamondoids and unidentified IR emission bands seen in the spectra of young stars with circumstellar disks.^{1,2} It is also suggested that their radical cations could contribute to features in the largely unassigned diffuse interstellar bands due to their low ionization energy and absorption in the visible range.³ However, the optical spectra of these cations have only recently begun to be measured experimentally, which is required for astronomical identification. Here, we present the optical spectra of the radical cations of adamantane (C₁₀H₁₆⁺)⁴, diamantane (C₁₄H₂₀⁺)⁵, and 1-cyanoadamantane (C₁₁H₁₅N⁺). These spectra were recorded by photodissociation of mass-selected ions in the gas phase at 5 K using a tandem mass spectrometer coupled to a cryogenic 22-pole ion trap. The experimental results are compared to photoelectron spectra and time-dependent DFT calculations for interpretation. All spectra reveal broad structures that are attributed to lifetime broadening and Franck-Condon congestion arising from geometric changes and/or Jahn-Teller distortion. The astrophysical implications of these ions will also be discussed.

MO 5.7 Tue 12:30 F142

Measuring fluorescence-detected two-quantum photon echoes using cogwheel phase cycling — ●AJAY JAYACHANDRAN, STEFAN MÜLLER, and TOBIAS BRIXNER — Institut für Physikalische und Theoretische Chemie, Universität Würzburg, Am Hubland, 97074 Würzburg, Germany

We describe two-quantum photon echo spectroscopy, a new technique that enables one the selective characterization of doubly excited states. This technique is analogous to the popular ‘photon echo’ experiment [1], which encodes the dynamic information associated with singly excited states.

We transfer the core principle, i.e., a rephasing contribution that removes the effect of inhomogeneous broadening, to doubly excited states, and we extract the signal experimentally using cogwheel phase cycling of three-pulse sequences and fluorescence detection. We verify the applicability of cogwheel phase cycling, which has proven useful in two-dimensional nuclear magnetic resonance spectroscopy to reduce overall measurement time in comparison to nested phase cycling [2], by extracting the two-quantum photon echo without any signal aliasing.

We study how exciton–exciton annihilation of squaraine molecular aggregates of varying chain length [3] can be quantified by accessing the homogenous linewidth of the biexciton state which is obtained from the two-quantum photon echo.

[1] S. Asaka et al., *Phys. Rev. A* **29**, 2286-2289 (1984).

[2] M. H. Levitt et al., *J. Magn. Reson.* **155**, 300-306 (2002).

[3] P. Malý et al., *Chem. Sci.* **11**, 456-466 (2020).

MO 5.8 Tue 12:45 F142

Near-field scanning optical microscopy of topologically protected excitons in molecular aggregates — ●SIDHARTHA NAYAK,

CHRISTOPHER W. WÄCHTLER, and ALEXANDER EISFELD — MPIPKS, Dresden, Germany

Delocalized excitonic eigenstates of molecular aggregates are responsible for the energy transfer from an incoming radiation into the aggregate. Static disorder, which can arise from an imperfect environment of each molecule, reduces the exciton transport and large disorders can even localize the exciton. It has been shown theoretically that a two-dimensional periodic array of tilted and interacting molecules in a homogeneous magnetic field shows topologically protected edge states [1] which are robust under local disorder. With a scattering scanning near-field optical microscope setup, one can not only record position dependent absorption spectra [2] but also reconstruct the wavefunctions from these spectra [3]. In this contribution we study theoretically the near field spectra of the aforementioned 2D aggregates in which the molecules experience a disordered environment because of the probing metallic tip. Due to the topological protection, the edge states are robust even in the presence of the metallic nanoparticle, such that the recorded spectrum shows clear signatures of these edge states.

[1] J. Y. Zhou, S. K. Saikin, N. Y. Yao and A. Aspuru-Guzik, *Nature materials* **13**, 1026-1032 (2014)

[2] X. Gao and A. Eisfeld, *J. Phys. Chem. Lett.* **9**, 6003 (2018)

[3] F. Zheng, X. Gao and A. Eisfeld, *Phys. Rev. Lett.* **123**, 163202 (2019)

MO 6: Poster I

Time: Tuesday 16:30–19:00

Location: Empore Lichthof

MO 6.1 Tue 16:30 Empore Lichthof

Boundary effects in sensory adaptation & interacting sensory systems — ●VANSH KHARBANDA — Cell Biophysics and Statistical Physics, Faculty of Veterinary Medicine, LMU Munich

Sensory adaptation is vital to all living organisms. An adaptive sensory system can be modelled as a stochastic, nonlinear feedback network. Using a generic framework, we study the accuracy of adaptive mechanisms and its energetic cost. Recently, it has been suggested that the steady-state dissipation rate associated to maintenance of an adaptive state increases logarithmically with the adaptation accuracy. We present results that demonstrate that this logarithmic scaling does not hold generally, but appears to be linear when the state of the system is close to the phase-space boundaries. Our numerical results also suggest that boundaries in the phase space of system variables limit the capacity of the system to dissipate. Moreover, we conjecture a new empirical expression relating the steady-state dissipation rate and the strength of the input signal if the state lies in the vicinity of the boundaries. Finally, the combined adaptation accuracy of two linearly coupled systems is studied. We show that a coupling the outputs of the systems deteriorates the overall adaptation accuracy while the associated energy cost is also reduced. In contrast, a coupling of the control elements reduces the dissipation rate without compromising on the adaptation accuracy.

MO 6.2 Tue 16:30 Empore Lichthof

Time-resolved study of the photogeneration of the phenylselenium cation from diphenyl diselenide and subsequent use for covalent activation in organic reactions — ●DANIEL GREINDA¹, ANNA TIEFEL², CARINA ALLACHER¹, ALEXANDER BREDER², and PATRICK NÜRNBERGER¹ — ¹Institute of Physical and Theoretical Chemistry, University of Regensburg, Regensburg — ²Institute of Organic Chemistry, University of Regensburg, Regensburg

Using a pump-probe setup on a μ s-timescale with a streak-camera detector [1], one can investigate light-induced chemical reactions with long-lived intermediates. We apply this technique to organoselenium compounds, which are useful catalysts in organic synthesis due to their stability towards oxygen and their rich radical chemistry [2]. Especially diphenyl diselenide is a versatile precursor for organic synthesis, as it is often used to generate phenylselenenyl radicals by photolytic cleavage of the selenium-selenium bond [3]. However, we demonstrate that given the right reaction conditions, the phenylselenium cation can be generated in a similar way, which then is utilizable for covalent activation of allylic selenium species [4].

[1] R. J. Kutta et al., *Appl. Phys. B* **111**, 203-216 (2013).

[2] A. Breder et al., *Tetrahedron Lett.* **56**, 2843-2852 (2015).

[3] O. Ito et al., *J. Am. Chem. Soc.*, **105**, 850-853 (1983).

[4] M. Tingoli et al., *J. Org. Chem.* **61**, 7085-7091 (1996).

MO 6.3 Tue 16:30 Empore Lichthof

Photoemission delays in similarly sized molecules — ●MAXIMILIAN FORSTER, CHRISTIAN SCHRÖDER, MAXIMILIAN POLLANKA, PASCAL SCIGALLA, MICHAEL MITTERMAIR, ANDREAS DUENSING, and REINHARD KIENBERGER — Chair for laser and x-ray physics, E11, Technische Universität München, Germany

In the gas phase we measure the relative photoemission delay between the Helium 1s and the Iodine 4d state in Iodobenzene and Iodocyclohexane. This allows us to determine the absolute timing of the Iodine 4d photoelectrons.

Iodine as a substituent was chosen for its giant resonance and therefore high cross section in the I4d state which is expected to be unaffected by its chemical surrounding. Measurements on a variety of different iodoalkanes have shown an unexpected variation of the I4d delay for different molecules, with no clear correlation with the molecular size, showing that the chemical environment may play a role in forming the observed photoemission delay. Therefore performing measurements on molecules with similar size and yet significant differences in their bonding structure yields further information about the possible underlying effects causing the delay.

MO 6.4 Tue 16:30 Empore Lichthof

Femtosecond Transient Absorption Spectro-Microscopy with High Spatial Resolution on Dye Microcrystals — ●MAGNUS FRANK, CHRIS REHHAGEN, and STEFAN LOCHBRUNNER — Institute for Physics and Department of Life, Light and Matter, University of Rostock, 18051 Rostock, Germany

Solid organic materials open a promising pathway for further improvement of optoelectronic devices like OLEDs or organic photovoltaics. The interaction of light and matter in such systems is essential for potential applications. For instance, in organic solar cells, excitons need to be diffusive to an interface for charge separation - a dynamic process competing with other decay channels on the femto- and picosecond timescale. Femtosecond transient absorption is still the hallmark of dynamics determination in molecular systems, but though a lot of research has been done on the dynamics of organic solids, many fundamental aspects are still unclear. This is because measuring not only dynamics on the femtosecond timescale but also ensuring high spatial and spectral resolution is challenging. In this work, we combine classical transient absorption with microscopy allowing for spectroscopy with both high spatial and time resolution, 2.5 μ m and sub-100fs respectively. We demonstrate the performance by measuring the dynamics of single microcrystals of a perylene dye, a material class known for

its outstanding potential in optoelectronics.

MO 6.5 Tue 16:30 Empore Lichthof

Phase-modulated transient-absorption spectroscopy in the liquid phase — ●JAKOB GERLACH, YILIN LI, ARNE MORLOK, ULRICH BANGERT, FRANK STIENKEMEIER, and LUKAS BRUDER — Institute of Physics, University of Freiburg, Germany

In our group a setup for two-dimensional electronic spectroscopy has previously been developed and used to study dynamics of nanosystems trapped in helium nanodroplets [1]. The action-detected spectroscopy is based on phase modulation of the laser pulses [2]. Many processes relevant in the field of photochemistry take place in a liquid environment, which significantly impacts the investigated dynamics. Therefore, a new setup is assembled to allow the examination of a probe in the liquid phase using transient-absorption spectroscopy. We plan to combine this setup with the phase modulation technique in order to improve the signal-to-noise. The design of the setup and a first characterization of its implementation will be presented.

[1] L. Bruder et al., *Nat Commun* 9, 4823 (2018).

[2] P. F. Tekavec, G. A. Lott, and A. H. Marcus, *J. Chem. Phys.* 127, 214307 (2007).

MO 6.6 Tue 16:30 Empore Lichthof

Dependence of photoelectron circular dichroism on the distance between marker atom and stereocenter in chiral molecules — ●EMILIA HEIKURA¹, FLORIAN TRINTER², LUTZ MARDER¹, CATMARN KÜSTNER-WETEKAM¹, DANA BLOSS¹, NILS KIEFER¹, JOHANNES VIEHMANN¹, CHRISTINA ZINDEL¹, MARKUS ILCHEN², ANDREAS HANS¹, and ARNO EHRESMANN¹ — ¹Institut für Physik und CINSaT, Universität Kassel, Heinrich-Plett-Str. 40, 34132 Kassel, Germany — ²Deutsches Elektronen-Synchrotron, DESY, Notkestraße 85, 22607 Hamburg, Germany

Photoelectron circular dichroism (PECD) is one of the most powerful methods to investigate molecular chirality in the gas phase. PECD arises from the asymmetry of the angular distribution of photoelectrons (probe electrons) scattered on the chiral backbone of the molecule even from randomly oriented molecules after interaction with circularly polarized light. One still unclear aspect of PECD is, how the distance between the probe electron emitter site and the center of a point-chiral molecule affects the magnitude of the PECD asymmetry. For detection of these forward-backward asymmetries of emitted photoelectrons a velocity map imaging (VMI) electron spectrometer was used. Measurements were performed on sec-butyl trimethylsilylether and its derivatives.

MO 6.7 Tue 16:30 Empore Lichthof

Ultrafast Transient Absorption Spectroscopy of Metal Complexes with 10 fs Probe Pulses — ●MARVIN KRUPP, MORITZ LANG, CHRIS REHHAGEN, and STEFAN LOCHBRUNNER — Institut für Physik, Uni Rostock, Deutschland

Most of the photoactive complexes used in chemical applications are based on rare metals like iridium or ruthenium. Replacing them with abundant metals e.g. iron is of long-standing interest, but the resulting complexes are currently limited by their photoactive properties. To analyze newly prepared iron complexes femtosecond transient absorption spectroscopy is typically used to determine the dynamics upon photoexcitation with short pump pulses and a few hundred femtoseconds long white light continuum for probing. Increasing the time resolution by reducing the pulse duration of the probe can make it possible to observe extremely fast processes below the current time resolution and give insights into the very first relaxation steps in the metal complexes such as intersystem crossing. In this work a broadband noncollinear optical parametric amplifier (NOPA) is used to generate broadband pulses with a duration below 10 fs. These pulses are used as probe light improving the time resolution in comparison to the usual CaF₂ white light. The setup was characterized with transient grating frequency-resolved optical gating (TG FROG) and first measurements on Fe-complexes are compared with respect to the two probe sources.

MO 6.8 Tue 16:30 Empore Lichthof

THz-Streaking for Detecting Inner-valence-shell Correlation-induced Time-delays in the Ionization of PAHs. — ●MOHAMED ABDELRASOUL, MARK PRANDOLINI, MAREK WIELAND, and MARKUS DRESCHER — Institute of Experimental Physics, University of Hamburg, Luruper Chaussee 149, 22761 Hamburg, Germany.

Photoionization delays (Wigner time delay) in outer molecular shells

occur in the range of a few ten attoseconds. This delay increases for inner valence shell photoionization due to strong correlation effects, namely, shake-up/down and knock-up/down processes, leading to markedly longer relaxation times, and raising an interesting question: how long does it take to remove an electron from the molecular inner shell relative to the outermost shell? Most polycyclic aromatic hydrocarbons (PAHs) have relatively high photoelectron emission cross sections in the energy range typically between 15 and 25 eV, and to access both inner and outer photoelectron spectra, an Extreme Ultraviolet (XUV) photon energy in that range is required. A laser-based setup is presented, combining a high harmonic XUV source with THz streaking for measuring the photoelectron emission delay in PAHs of different sizes.

MO 6.9 Tue 16:30 Empore Lichthof

Photodissociation dynamics of CHCl₂ — ●JONAS FACKELMAYER, CHRISTIAN MATTHAEI, and INGO FISCHER — Julius-Maximilians-Universität Würzburg, 97074 Würzburg, Germany

Recent studies suggest that the depletion of atmospheric O₃ is not only caused by the banned CFCs and HCFCs but also catalysed by commonly used solvents such as dichloromethane.^[1] Photodissociation of these compounds often results in the release of highly reactive halogen radicals. While the dissociation dynamics of molecular halocarbons have been studied in detail in the past, less is known about their open shell counterparts.

The photofragmentation of the open shell CHCl₂, generated by pyrolysis from the bromide precursor CHCl₂Br, was investigated in a free jet utilising time-of-flight mass-spectrometry and velocity map imaging. Photodissociation was achieved by a pulsed dye laser in the range of 230 - 250 nm mainly producing CHCl and Cl fragments, while ionisation was provided by either a second dye laser (REMPI) or a frequency multiplied solid state laser at 118 nm (SPI). First insights into the involved dissociation mechanisms are discussed.

[1] Hossaini, R. et al., *Nat. Commun.* 2017, 8, 15962.

MO 6.10 Tue 16:30 Empore Lichthof

Probing conical intersection dynamics in the dissociative photoionization of formaldehyde — ●DAVID CHICHARRO VACAS, WEIYU ZHANG, THOMAS PFEIFER, and ROBERT MOSHAMMER — Max-Planck-Institute für Kernphysik, Heidelberg, Germany

Conical intersections (CI) between electronic states often govern the photochemistry of molecules and radicals. Their role and characteristics have been largely studied before both theoretically and experimentally. It has been proven that CI are crucial in different photochemical reactions and are characterized by coupled electronic and nuclear dynamics, breaking the Born-Oppenheimer approximation. The neutral photofragmentation of formaldehyde has been widely studied in the past years mainly because of a great interest for the roaming pathway producing H₂. However, the photoionization and dissociative photoionization (DPI) has been less studied. Experimentally, the DPI of formaldehyde has been studied and two main DPI processes were found: The H-atom loss and the molecular channel. More recently, experimental and theoretical findings allowed to predict the mechanism involved in the DPI of formaldehyde, suggesting the presence of a conical intersection that controls this mechanism. The aim of this work is to directly observe the presence of this conical intersection by using pump-probe schemes along with the reaction microscope. The 3D-ion-electron momentum coincidence measurements in a pump-probe experiment provides enough information to fully understand this process and to directly visualize the presence of the conical intersection in the experimental results.

MO 6.11 Tue 16:30 Empore Lichthof

Ultrafast coherent control of single molecules via two-photon excitation at room temperature — ●XINPENG XU¹, ULLRICH SCHERF², and RICHARD HILDNER¹ — ¹Zernike Institute for Advanced Materials, University of Groningen, The Netherlands — ²Institut für Polymertechnologie, Universität Wuppertal, Germany

Quantum coherent control has been a powerful technique to understand and manipulate ultrafast photoinduced processes occurring at the inter-/intra-molecular level for more than twenty years. In a coherent control experiment, one can exploit quantum interference between competing pathways of multiphoton transitions toward the desired outcome by tailoring the spectral phase, amplitude, or polarization of the electromagnetic field of the exciting laser. For larger functional molecules in condensed phase, the influence of the (often disordered) surrounding environment varies between molecules. Hence, ensemble

measurements do typically not allow to exert full control over competing pathways. Here, we demonstrate that the two-photon transition of single molecules can be controlled by a sequence of shaped pulses at room temperature. Varying the spectral phase of the pulses, we observe phase-dependent photoluminescence signals corresponding to the two-photon excitation probability. We assign this phase dependence to the combination of quantum photon interference and coherence between the incident laser spectrum and the single molecule absorption spectrum. Notably, cancellation of the transition probability by so-called ‘dark pulses’ is observed in some molecules, which shows the ability to fully coherent control of single molecules in condensed phase.

MO 6.12 Tue 16:30 Empore Lichthof

Attosecond Chronoscopy of organic iodine compounds on Pt111 — ●SVEN PAUL¹, PASCAL SCIGALLA¹, CHRISTIAN SCHRÖDER¹, PETER FEULNER², and REINHARD KIENBERGER¹ — ¹Chair for laser and x-ray physics, E11, Technische Universität München, Germany — ²Surface and Interface Physics, E20, Technische Universität München, Germany

We report on attosecond streaking measurements of the relative photoemission delay between the Iodine 4d state in organic iodine compounds like Iodoethane and -methane adsorbed on a Pt111 surface in respect to the Pt valence band. Iodine was chosen as a substituent in these organic molecules because of its giant dipole resonance in the 4d state, which results in a high cross section that is mostly unaffected by the rest of the molecule. The surface coverage of the organic iodine compounds on the Pt crystal is controlled via thermal programmed desorption. This is important because the orientation, i.e. horizontal and vertical configuration, of these adsorbents on the surface depends on the surface coverage. By using those two effects, we can ensure whether the detected photoelectrons have been perturbed by the whole organic chain or only by parts of it. A change in photoemission delay between both configurations is expected, as suggested by previous measurements for Iodoethane. Similar measurements are now made for Iodomethane as part of my master thesis, as we want to study the correlation of the photoemission delay with the length of the organic chain.

MO 6.13 Tue 16:30 Empore Lichthof

Lamellar and amorphous Alucones as Nanoscaffolds for Cellular Response; A Route for Building Nature-Inspired Materials. — ●MABEL MORENO¹, ANGÉLICA ZACARIAS², SIMÓN GUERRERO¹, LUIS VELASQUEZ¹, YUSSER OLGUIN^{3,4}, and EBERHARD GROSS⁵ — ¹Universidad SEK, Instituto de Investigación Interdisciplinar en Ciencias Biomédicas SEK (I3CBSEK). — ²Max Planck Institute of Microstructure Physics, Weinberg 2, D 06120, Halle, Germany, and ETSEF. — ³Universidad Técnica Federico Santa María, Centro de Biotecnología, Avenida España 1680, Valparaíso, Chile. — ⁴Universidad Técnica Federico Santa María, Departamento de Física, Avenida España 1680, Valparaíso, Chile — ⁵Fritz Haber Research Center for Molecular Dynamics and Institute of Chemistry, The Hebrew University of Jerusalem, Jerusalem 91904, ISRAEL.

We present our study of the growth of alucone thin films (AIO-T and AIO-A, T: terephthalate and A: adipate) by atomic layer deposition (ALD) and molecular layer deposition (MLD). Stoichiometric thin films with large area uniformity were obtained in both cases, as shown in SEM/Focus Ion Beam (FIB), transmission electron microscopy (TEM), X-ray diffraction (XRD), X-ray reflectometry (XRR), Atomic force microscopy (AFM), reflectance diffuse by UV-Visible, and attenuated total reflectance (ATR) data. Quantum information (QI), C2C12 cells cultured test and antibacterial activity were performed to prove the QI and biocompatibility concepts on such thin films, making them possible candidates for bioinspired-quantum devices.

MO 6.14 Tue 16:30 Empore Lichthof

Long-lasting XUV-induced ignition of avalanche ionization of helium nanodroplets — ●C. MEDIA¹, A. Ø. LAEGDSMAND², L. BEN LTAIEF², Z HOQUE³, A. H. ROOS³, M. JURKOVIC³, O. HORT³, O. FINKE³, M. ALBRECHT³, J. NEJDL³, F. STIENKEMEIER¹, E. KLIMESOVA³, M. KRIKUNOVA³, A. HEIDENREICH⁴, and M. MUDRICH² — ¹Institute of Physics, University of Freiburg, Freiburg, Germany — ²Department of Physics and Astronomy, Aarhus University, Aarhus, Denmark — ³ELI Beamlines Centre, FZU- Institute of Physics of the Czech Academy of Sciences, Na Slovance 2, 182 21 Prague, Czechia — ⁴IKERBASQUE, Basque Foundation for Science, 48011 Bilbao, Spain

We study the dynamics of avalanche ionization of pure helium nanodroplets activated by a weak extreme-ultraviolet (XUV) pulse and

driven by an intense near-infrared (NIR) pulse. In addition to a transient enhancement of the droplet ignition probability at short delay times ~ 200 fs, long-term activation of the nanodroplets lasting up to a few nanoseconds is observed. Molecular dynamics simulations suggest that the short-term activation is caused by the injection of seed electrons into the droplets by XUV photoemission. Long-term activation is due to electrons remaining loosely bound to photoions which form stable ‘snowball’ structures in the droplets. Thus, we show that XUV irradiation can induce long-lasting changes of the strong-field optical properties of nanoparticles, potentially opening new routes to controlling avalanche-ionization phenomena in nanostructures and condensed-phase systems.

MO 6.15 Tue 16:30 Empore Lichthof

Bidirectional photorearrangement reaction of a xanthine derivative — ●THOMAS RITTNER¹, KARINA HEILMEIER¹, RAFAEL E. RODRÍGUEZ-LUGO², SIMON DIETZMANN², SVENJA WORTMANN¹, ROGER JAN KUTTA¹, ROBERT WOLF², and PATRICK NUERNBERGER¹ — ¹Institut für Physikalische und Theoretische Chemie, Universität Regensburg, 93053 Regensburg — ²Institut für Anorganische Chemie, Universität Regensburg, 93053 Regensburg

7-Aryl-8-pyridyl substituted theophyllines exhibit a remarkable photoreaction. Our studies show that upon excitation with ultraviolet light, the anisyl substituted 7-(4-methoxyphenyl)-1,3-dimethyl-8-(pyridin-2-yl)-3,7,8,9-tetrahydro-1H-purine-2,6-dione undergoes a rearrangement. The anisyl moiety migrates from the 7-nitrogen on the purine skeleton of the theophylline to the 1'-nitrogen of the pyridine, yielding a zwitterionic isomer. Both isomers could be isolated and subsequently characterized by NMR spectroscopy and single-crystal X-ray scattering. Time-resolved and steady-state absorption and emission spectroscopies in the visible spectral range were used to investigate the photorearrangement mechanism. Together with quantum-chemical calculations a detailed picture on a molecular level could be obtained. We further demonstrate that also a photo-induced back-isomerization is feasible, enabling in principle a general design for photo-switchable molecular structures.

MO 6.16 Tue 16:30 Empore Lichthof

Coverage-dependent agglomeration in molecular films on atomically flat surfaces — ●ERIK VON DER OELSnitz, TIM VÖLZER, JULIAN SCHRÖER, RICO SCHWARTZ, TOBIAS KORN, and STEFAN LOCHBRUNNER — Institute of Physics, Albert-Einstein-Straße 23-25, 18059 Rostock, Germany

The deposition of a thin, monomeric molecular layer on an atomically flat surface, as it is provided by Van-der-Waals crystals, is crucial to the functionalization of such layered materials and the fabrication of corresponding hybrid structures. In this work, Perylene Orange (PO) molecules were coated onto a thin exfoliated hexagonal boron nitride layer using thermal vapor deposition (TVD) at various evaporation temperatures. The resulting hybrid structures were examined by a fluorescence lifetime imaging microscope, revealing a biexponential fluorescence decay, as well as by micro-photoluminescence spectroscopy, proving for low evaporation temperatures the monomeric nature of the PO layer. Upon increasing the temperature, the fluorescence intensity rises and at the same time the fast decay component becomes dominant and a low-energy band emerges in the emission spectrum. Interestingly, the slow decay time remains pretty constant. Therefore, this slow component can be assigned to the fluorescence decay of PO monomers, whereas the fast decay component can be attributed to agglomerates that form at higher coverage and provide additional decay channels such as the relaxation into excimer states. From these findings, an optimum for the vaporization temperature can be determined, which will be used in future applications.

MO 6.17 Tue 16:30 Empore Lichthof

Effect of vibropolariton formation on ground-state reactivity of Diels-Alder cycloaddition reaction — ●BERNA ARSLANOGLU, THOMAS SCHNAPPINGER, and MARKUS KOWALEWSKI — Stockholm University, Sweden

It has been experimentally demonstrated that ground-state reaction mechanisms can be modified by an optical cavity at room temperature. It has been shown by Thomas et al. [1], that coupling a Si-C bond resonantly to a quantized field mode can change the chemical reaction rate of the silyl bond cleavage reaction.

We present ab initio simulations of the ground state Diels-Alder reaction in the presence of an optical cavity field. The vibrational modes of the reactants are strongly coupled to the light field. The change of

the reaction barrier in the presence of the field is studied for varying vacuum field strengths as well as for different resonance conditions. Additionally, we study how the dipole self-energy term influences stationary points of the reaction.

[1] A. Thomas et al., *Science* 363, 615 (2019)

MO 6.18 Tue 16:30 Empore Lichthof

Classification of noisy spectra using machine learning — ●ARITRA MISHRA and ALEXANDER EISFELD — Max Planck Institute for the Physics of Complex Systems, Dresden, Germany

A general problem in quantum mechanics is to obtain information of the eigenstates from the experimental measured data which consists inherent noises. In particular, we consider molecular aggregates, where information about excitonic eigenstates is vitally important to understand their optical and transport properties [1,2]. It has been shown that it is possible to reconstruct the underlying delocalised aggregate eigenfunctions from near-field spectra using convolution neural networks [3].

In this work, we also use a convolution neural network but ask a question related to the eigenstate based classification of the spectra in the presence of noise. Given that each eigenstate correspond to a distinct spectrum, we can assign a class to each of the eigenstate. We add a random noise to these spectra and build a network that can classify the spectra into these classes, in the presence of the noise. We find that the network is also able to classify the spectra of different noise strengths along with the one it has been trained for.

[1] X. Gao and A. Eisfeld, *J. Phys. Chem. Lett.* 9, 6003 (2018)

[2] S. Nayak, F. Zheng and A. Eisfeld, *J. Chem. Phys.* 155, 134701 (2021)

[3] F. Zheng, X. Gao and A. Eisfeld, *Phys. Rev. Lett.* 123, 163202 (2019)

MO 6.19 Tue 16:30 Empore Lichthof

Photoelectron Circular Dichroism for Chiral Helium — ●MAREC HEGER, MANEL MONDELO-MARTELL, and DANIEL REICH — Dahlem Center for Complex Quantum Systems and Fachbereich Physik, Freie Universität Berlin, Arnimallee 14, D-14195 Berlin, Germany

Chiral molecules, i.e. molecules that cannot be superimposed by its mirror image by rotations and translations, show different reactions to other chiral systems depending on their handedness. A prominent example is the interaction of chiral molecules with left and right-circularly polarized light which leads to differences in the photoelectron angular emission spectrum. This difference is called photoelectron circular dichroism (PECD) which can be used as a tool to detect and distinguish chiral signatures of molecules in the gas phase.

Understanding the precise origin and the relationship between the chiral molecular scaffold and the PECD signal is still an ongoing theoretical challenge, particularly when it comes to the role of electron correlation. Numerical simulations for large molecules are particularly challenging in this context. For this reason we perform *ab initio* simulations for an elementary system - helium with an artificial external chiral potential - and investigate the relationship between PECD and the chiral potential. Finally, by considering theoretical simulations involving various degrees of electron correlation - from single-active electron approaches to full configuration interaction - we also aim to elucidate the role of electron correlation systematically.

MO 6.20 Tue 16:30 Empore Lichthof

Towards understanding the enhancement of the circular dichroism in the ion yield of 3-methylcyclopentanone by tailored femtosecond laser pulses — ●SAGNIK DAS, JAYANTA GHOSH, SUDHEENDRAN VASUDEVAN, HANGYEOL LEE, NICOLAS LADDA, SIMON RANECKY, TONIO ROSEN, ARNE SENFTLEBEN, THOMAS BAUMERT, and HENDRIKE BRAUN — Institut für Physik, Universität Kassel, Heinrich-Plett-Strasse 40, 34132 Kassel, Germany

One of the methods to differentiate between the two enantiomers of a chiral molecule is Circular Dichroism (CD). It arises due to the difference in absorption of left and right circularly polarised light. The difference in absorption can also be mapped to the difference in ionisation of the enantiomers and is known as CD in ion yield [1]. We use our home-built Time of Flight (ToF) mass spectrometer with twin peak [2] measurement setup to study the effect of linear chirp (GDD) on the anisotropy. The candidate molecule for this experiment is 3-methylcyclopentanone (3-MCP). We perform all the experiments at 309 nm, where 3-MCP shows enhancement of anisotropy, upto 10%. At this wavelength, a 1+1+1 resonance-enhanced multi-photon ioni-

sation (REMPI) takes place in 3-MCP through the $\pi^* \leftarrow n$ transition. We observed enhancement of anisotropy for chirped pulses, which we have compared to bandwidth limited pulses of equal peak intensity. Furthermore, we perform a pump-probe experiment to investigate the intermediate state dynamics.

[1] U. Boesl and A. Bornschlegl, *ChemPhysChem*, 7, 2085, 2006

[2] T. Ring et al., *Rev. Sci. Instrum.*, 92, 033001, 2021

MO 6.21 Tue 16:30 Empore Lichthof

Excited-state dynamics of aqueous aminoazobenzene Metanil Yellow studied by time-resolved transient absorption spectroscopy — ●ALINA KHODKO^{1,5}, MATTHEW MGBUKWU⁴, CAMILO GRANADOS^{1,4}, EVGENII TITOV^{2,3}, STEFAN HAACKE⁴, OLEG KORNILOV¹, and JÉRÉMIE LÉONARD⁴ — ¹Max Born Institute, Max-Born-Straße 2A, 12489 Berlin, Germany — ²Institute of Physical and Theoretical Chemistry, University of Würzburg, Germany — ³Institute of Chemistry, University of Potsdam, Germany — ⁴Institut de Physique et Chimie des Matériaux de Strasbourg, Université de Strasbourg, CNRS UMR 7504, Strasbourg, France — ⁵Institute of Physics NAS of Ukraine, Nauky Ave, 46, 03028, Kyiv, Ukraine

The excited-state dynamics of the aminoazobenzene Metanil Yellow in aqueous solutions was studied using ultrafast time-resolved transient absorption spectroscopy in both the UV-visible and the NIR regions. The different solutions were studied with different excitation wavelengths to investigate the protonated and non-protonated forms of the molecule and reveal differences in the corresponding dynamics. The relaxation dynamics of the non-protonated form was previously studied by time-resolved photoelectron spectroscopy and TDDFT calculations and revealed transitions from the bright S2 to the ground state S0 via the dark S1 state. As a first interpretation, the present results for the protonated form could be explained by direct excitation of S1 followed by the proton ejection during the first 0.5-1.0 ps. After 1 ps the excited state absorption looks similar for both forms and reflects the internal conversion to the trans ground state along the torsional coordinate.

MO 6.22 Tue 16:30 Empore Lichthof

Ultrafast dynamics and reversible switching of azobenzene-copper complexes — ●MARCEL J. P. SCHMITT¹, JUSTIN HORNBOGEN², RAPHAEL I. PETRIKAT¹, SABINE BECKER¹, ROLF DILLER², and CHRISTOPH RIEHN^{1,3} — ¹Dept. of Chemistry, RPTU Kaiserslautern — ²Dept. of Physics, RPTU Kaiserslautern — ³Forschungszentrum OPTIMAS, 67663 Kaiserslautern (Germany)

Azobenzenes and their derivatives are known for their reversible *E-Z*-photoisomerization around the N=N bond.[1,2] Their usage as photoswitchable ligands in metal complexation has also been explored.[3] We present preliminary spectroscopic and ultrafast dynamic results in solution and gas phase of a newly synthesized cyclic [Cu₂L₂]²⁺ complex, comprised of two pyridyl-substituted azobenzene ligands (L) in conjunction with Cu(I). We focus on the photoswitching dynamics and possible cooperative effects involving the ligands and metal cores. Therefore, we have examined the dynamics in parallel in solution by transient absorption and in gas phase by femtosecond transient photodissociation using an electrospray ionization mass spectrometer. The resulting ultrafast dynamic spectra reveal multiexponential electronic decay with lifetimes in the sub-ps and ps time ranges for both *E* and *Z* configurations. For comparison with UV/Vis and mIR spectra, the binding situation of the copper centers (tetrahedral vs. planar) in the (*E,E*), (*E,Z*) and (*Z,Z*) complexes was quantum chemically modelled by RI-DFT/TD-DFT calculations.

[1] *Nat. Rev. Chem.* 2019, 3, 133. [2] *Chem. Soc. Rev.* 2012, 41, 18091825. [3] *J. Phys. Chem. Lett.* 2019, 10, 6048.

MO 6.23 Tue 16:30 Empore Lichthof

Femtosecond Spectroscopy of a highly strained benzene isomer — ●LUKAS FASCHINGBAUER¹, TOBIAS PREITSCHOPF¹, JENS PETERSEN¹, LOU BARREAU², LIONEL POISSON², ROLAND MITRIC¹, and INGO FISCHER¹ — ¹Institut für Physikalische und Theoretische Chemie, Würzburg, Germany — ²Institut des Sciences Moléculaires d'Orsay, Orsay, France

As a highly strained benzene isomer, 3,4-dimethylenecyclobutene (DMCB) forms an intriguing system to study the dynamics of its excited states, both from an experimental and a theoretical point of view. The formally Woodward-Hoffmann allowed electrocyclic ring opening to 1,2,4,5-hexatetraene is not observed, in contrast to the thermal counterpart, which has been observed in pathways to benzene in flames. Nevertheless, as indicated by a very broad and diffuse gas-phase UV/VIS absorption spectrum, ultrafast non-radiative decay

is observed by femtosecond time-resolved photoelectron and time-of-flight mass spectrometry. To explore the underlying mechanism, ab

initio calculations and mixed quantum-classical surface hopping molecular dynamics simulations have been performed.

MO 7: Machine Learning and Computational and Theoretical Molecular Physics

Time: Wednesday 11:00–13:00

Location: F142

Invited Talk

MO 7.1 Wed 11:00 F142

Augmenting basis with normalizing flows for solving Schrödinger equations: theoretical analysis — ●YAHYA SALEH^{1,2}, ARMIN ISKE², ANDREY YACHMENEV^{1,4}, and JOCHEN KÜPPER^{1,3,4} — ¹Center for Free-Electron Laser Science CFEL, Deutsches Elektronen-Synchrotron DESY, Hamburg — ²Department of Mathematics, Universität Hamburg — ³Department of Physics, Universität Hamburg — ⁴for Ultrafast Imaging, Universität Hamburg

Spectral methods are a popular class for solving time-independent Schrödinger equations. Here, one approximates the wavefunctions in the linear span of standard basis sets of L2. In spite of the well-posedness and the convergence guarantees of such methods, they suffer from the curse of dimensionality, as the computational expenses grow exponentially with the size of the quantum system.

Recently, nonlinear functions, e.g., neural networks have been proposed to model ground states and low-lying excited states of Schrödinger equations. Although they promise accurate results with smaller scaling than standard methods, extensions of such models to the simultaneous computation of many states are still lacking.

Here, we propose to model excited states of Schrödinger equations via augmented basis sets, where standard basis sets are composed with normalizing flows. We show that such a numerical scheme is well-posed and defines a richer approximation space than standard methods. Moreover, we provide convergence guarantees.

MO 7.2 Wed 11:30 F142

A machine learning full dimensional potential energy surface for AIF-AIF: lifetime of the intermediate complex — WEIQI WANG¹, ●XIANGYUE LIU¹, and JESÚS PÉREZ-RÍOS² — ¹Fritz-Haber-Institut der Max-Planck-Gesellschaft, Faradayweg 4-6, 14195 Berlin, Germany — ²Institute for Advanced Computational Science, Stony Brook University, Stony Brook, NY 11794-3800, USA

AIF is a promising candidate in the quest of finding the most efficient molecule for laser cooling. In this work, a full-dimensional potential energy surface of AIF-AIF dimer has been constructed by machine learning methods. In particular, we analyze the reliability and efficiency of different active learning schemes developed for this system. The potential energy surface has been employed in calculating the four-body complex lifetime relevant to the stability of molecules in the ultracold regime via molecular dynamics simulations.

MO 7.3 Wed 11:45 F142

Quantum flows neural network for variational solutions of the Schrödinger equation — ●ÁLVARO FERNÁNDEZ^{1,3}, YAHYA SALEH^{1,4}, ANDREY YACHMENEV^{1,2}, ARMIN ISKE⁴, and JOCHEN KÜPPER^{1,2,3} — ¹Center for Free-Electron Laser Science, Deutsches Elektronen-Synchrotron DESY, Hamburg — ²Center for Ultrafast Imaging, Universität Hamburg — ³Department of Physics, Universität Hamburg — ⁴Department of Mathematics, Universität Hamburg

Recently, a few deep neural network models for solving the electronic Schrödinger equation were developed, demonstrating both outstanding computing efficiency and accurate results.

Here, we present a new quantum-flow-neural-network approach for obtaining variational solutions of the stationary Schrödinger equation. At the core of the method is an invertible neural network composed with the general basis of orthogonal functions, which provides a more stable framework for simultaneous optimization of the ground state and excited states. This approach is applied in calculations of the vibrational energy levels of polyatomic molecules as well as of electronic energies in a single-active-electron approximation. The results show a considerable improvement of variational convergence for the ground and the excited states. In addition, we extend our approach for solving the time dependent problems using recurrent flows.

MO 7.4 Wed 12:00 F142

Electronic excited states in deep variational Monte Carlo — ●MIKE ENTWISTLE¹, ZENO SCHÄTZLE¹, PAOLO ERDMAN¹, JAN

HERMANN^{2,1}, and FRANK NOÉ^{2,1,3} — ¹Freie Universität Berlin, Berlin, Germany — ²Microsoft Research AI4Science, Berlin, Germany — ³Rice University, Houston, USA

Obtaining accurate ground and low-lying excited states of electronic systems is crucial in a multitude of important applications. One ab initio method for solving the Schrödinger equation that scales favorably for large systems is variational quantum Monte Carlo (QMC). The recently introduced deep QMC approach uses ansatzes represented by deep neural networks and generates nearly exact ground-state solutions for molecules containing up to a few dozen electrons, with the potential to scale to much larger systems where other highly accurate methods are not feasible. Here, we extend one such ansatz (PauliNet) to compute electronic excited states. We demonstrate our method on various small atoms and molecules and consistently achieve high accuracy for low-lying states. To highlight the potential of our method, we compute the first excited state of the much larger benzene molecule, as well as the conical intersection of ethylene, with PauliNet matching results of more expensive high-level methods.

MO 7.5 Wed 12:15 F142

The performance of CCSD(T) for the calculation of dipole moments in diatomics — ●XIANGYUE LIU¹, LAURA MCKEMMISH², and JESÚS PÉREZ-RÍOS³ — ¹Fritz-Haber-Institut der Max-Planck-Gesellschaft, Faradayweg 4-6, 14195 Berlin, Germany — ²School of Chemistry, UNSW Sydney, Sydney, NSW 2052, Australia — ³Department of Physics and Astronomy, Stony Brook University, Stony Brook 11794, New York, USA

Electric dipole moment plays an important role in understanding intermolecular interactions. High-quality electric dipole moments are essential for accurate predictions of vibrational and rotational spectroscopy. In this work, the performance of CCSD(T) (coupled cluster with single, double, and perturbative triple excitations) has been evaluated with accurate experimental data of diatomic molecules. In particular, CCSD(T) accuracy for the equilibrium bond length, vibrational frequency, and dipole moments has been discussed. We find that CCSD(T) gives accurate predictions on dipole moments for most of the molecules in the test set. However, disagreements have been found for a few molecules, which can hardly be explained by relativistic or multi-reference effects. The impacts of basis set family and size have also been discussed.

MO 7.6 Wed 12:30 F142

Non-Local Polarizability Density as a Building Block for Dispersion Density Functionals — ●SZABOLCS GÖGER¹, PETER SZABO^{2,3}, DMITRY FEDOROV¹, and ALEXANDRE TKATCHENKO¹ — ¹University of Luxembourg, 1511 Luxembourg, Luxembourg — ²Katholieke Universiteit Leuven, 3000 Leuven, Belgium — ³Royal Belgian Institute for Space Aeronomy, 1180 Uccle, Belgium

Dispersion interactions, stemming from long-range electron correlations, are not properly captured by many electronic structure methods. A proper framework to tackle this problem requires determining the correlation energy via the adiabatic-connection dissipation-fluctuation theorem, but a robust unified formalism is yet to be developed [1]. In this work, we attempt to build such a general method based on the non-local polarizability, which is expressed as a polarization-polarization correlation function. This quantity is evaluated for a number of model systems and contrasted to real atoms and molecules. The model studies presented here, along with prior work on semi-local polarizability functionals [2], pave the way toward developing a unified non-local polarizability functional for molecules and materials, aimed to describe the short-range and long-range correlations on equal footing.

[1] Hermann and Tkatchenko, Phys. Rev. Lett. 124, 146401 (2020)

[2] Vydrov and Van Voorhis, Phys. Rev. Lett. 103, 063004 (2009)

MO 7.7 Wed 12:45 F142

Few-Body Physics of the Trapped Atoms: The Configuration Interaction Approach — ●MATEE UR REHMAN¹, ALEJANDRO SAENZ¹, FABIO REVUELTA PEÑA², PAUL WINTER¹, and SIMON SALA¹

— ¹Humboldt-Universität zu Berlin — ²Universidad Politécnica de Madrid

Two independent theoretical approaches are developed for the computational treatment of interacting ultracold atoms in versatile trap potentials. One approach considers the two-body system in centroid and relative coordinates have recently successfully demonstrated the inelastic confinement-induced resonances occurs due to the anharmonicity of the trap potentials, however an extension beyond two particles is not possible, but evidently of interest. This motivates to consider an alternative approach in absolute coordinates based on standard quantum-chemistry approaches, that uses cartesian Gaussians (GTOs) as ba-

sis functions and performs the configuration interaction calculations with a flexible choice of interaction potentials, hence allow considering arbitrary optical tweezer arrays. As the delta-pseudopotential does not work in beyond-mean field description, the Gaussian interaction potential using GTOs allows an efficient solution of multi-centered six-dimensional interaction integrals, but its validity has to be investigated. So this talk will demonstrate both theoretical models by focusing on their pros and cons for different interatomic interaction potentials, and then address the question that, Is the Gaussian interaction potential is sufficient (respectively in which parameter regime is it sufficient) to be used (or within which accuracy it can be used).

MO 8: Members' Assembly

Time: Wednesday 13:15–14:00

Location: F142

All members of the Molecular Physics Division are invited to participate.

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 BRANCHI¹, LINGFENG GE², FELIX SCHELL¹, KILIAN DICKSON³, MARK MERO¹, HORST ROTTKE¹, SERGUEI PATCHKOVSKII¹, MARC VRAKING¹, VARUN MAKHIJA³, and JOCHEN MIKOSCH² — ¹Max-Born-Institut, Berlin, Germany — ²Universität Kassel, Kassel, Germany — ³Univ. 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 LEE¹, SIMON RANECKY¹, SUDHEENDRAN VASUDEVAN¹, NICOLAS LADDA¹, TONIO ROSEN¹, SAGNIK DAS¹, JAYANTA GHOSH¹, HENDRIKE BRAUN¹, DANIEL REICH², ARNE SENFTLEBEN¹, and THOMAS BAUMERT¹ — ¹Institut für Physik, Universität Kassel, Heinrich-Plett-Str. 40, 34132 Kassel, Germany. — ²Dahlem 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 gen-

eration 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 superohmic 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} — ¹Physikalisches Institut, Albert-Ludwigs-Universität Freiburg, Hermann-Herder-Straße 3, D-79104, Freiburg, Germany — ²EUCOR Centre for Quantum Science and Quantum Computing, Albert-Ludwigs-Universität Freiburg, Hermann-Herder-Straße 3, D-79104, Freiburg, Germany — ³Max Planck Institute for the Structure and Dynamics of Matter, Luruper Chaussee 149, D-22761 Hamburg, Germany — ⁴The Hamburg Centre for Ultrafast Imaging, Luruper Chaussee 149, D-22761 Hamburg, Germany — ⁵Clarendon Laboratory, University of Oxford, Parks Road, Oxford OX1 3PU, United Kingdom

We investigate optimal states of photon pairs to excite a target transi-

tion 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 LINDEL¹, EDOARDO CARNIO¹, STEFAN YOSHI BUHMANN², and ANDREAS BUCHLEITNER¹ — ¹University of Freiburg, Germany — ²University 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.

MO 10: Collisions (joint session MO/Q)

Time: Wednesday 14:30–16:15

Location: F142

MO 10.1 Wed 14:30 F142

Gas phase investigations on dynamics of the reaction of tantalum cation Ta⁺ with carbon dioxide CO₂ — ●MARCEL META¹, MAXIMILIAN HUBER¹, MAURICE BIRK¹, ATILAY AYASLI², TIM MICHAELSEN², ROLAND WESTER², and JENNIFER MEYER¹ — ¹RPTU Kaiserslautern-Landau, Fachbereich Chemie, Kaiserslautern, Germany — ²Universität Innsbruck, Institut für Ionenphysik und Angewandte Physik, Innsbruck, Austria

The dynamics of the *oxygen atom transfer* (OAT) reaction Ta⁺ + CO₂ → TaO⁺ + CO in gas phase could be investigated under single collision conditions. The measured energy and angle differential cross sections allow us to probe the rearrangement of atoms during reaction, i.e. the atomistic dynamics [1]. The preset results were measured with our new 3D velocity map imaging setup in Kaiserslautern. The reaction is exothermic and spin forbidden in the ground state but takes place due to an efficient crossing from the quintet surface over to the triplet surface. Hence, it was found that the reaction almost proceeds with collision rate at room temperature [2-4]. The TaO⁺ velocity map images shows dominant indirect dynamics even at high collision energies with most of the additional collision energy partitioned into internal excitation.

[1] J. Meyer, R. Wester, Annu. Rev. Phys. Chem. 2017, 68, 333;

[2] R. Wesendrup, H. Schwarz, Angew. Chem. Int. Ed. 1995, 34, 2033; [3] G. K. Koyanagi, D. K. Bohme, J. Phys. Chem. A 2006, 110, 1232; [4] N. Levin, J. T. Margraf, J. Lengyel, K. Reuter, M. Tschurl, U. Heiz, Phys. Chem. Chem. Phys. 2022, 24, 2623

MO 10.2 Wed 14:45 F142

Dissociative recombination of ArH⁺ at the Cryogenic Storage Ring — ●ÁBEL KÁLOSI^{1,2}, MANFRED GRIESER², LEONARD W. ISBERNER^{3,2}, DANIEL PAUL^{1,2}, DANIEL W. SAVIN¹, STEFAN SCHIPPERS³, VIVIANE C. SCHMIDT², ANDREAS WOLF², and OLDŘICH NOVOTNÝ² — ¹Columbia Astrophysics Laboratory, Columbia University, New York, NY 10027, USA — ²Max-Planck-Institut für Kernphysik, 69117 Heidelberg, Germany — ³I. Physikalisches Institut, Justus-Liebig-Universität Gießen, 35392 Gießen, Germany

ArH⁺ is an important probe of the cosmic ray flux in interstellar space. Cosmic rays are the dominant ionization source for H and H₂ in the cold interstellar medium (ISM). This ionization initiates astrochemistry in the cold phases of the ISM. The cosmic ray ionization rate (CRIR) is thus an important parameter for both chemical and dynam-

ical models of the ISM. ArH⁺ forms via cosmic ray ionization of Ar, but can be destroyed via dissociative recombination (DR) with free electrons. Astronomical observations of ArH⁺, combined with chemical models, enable one to quantitatively estimate the CRIR. Such models require reliable rate coefficients that take into account the low internal excitation of the ArH⁺, as occurs in the cold ISM. To this end, we have performed merged-beams DR experiments with ArH⁺ in the Cryogenic Storage Ring where, at an ambient temperature of ~ 10 K, the ions relaxed to their lowest rotational states.

MO 10.3 Wed 15:00 F142

Laser induced forced evaporative cooling of molecular anions — ●ERIC ENDRES¹, JONAS TAUCH², SABA HASSAN², MARKUS NÖTZOLD¹, ROLAND WESTER¹, and MATTHIAS WEIDEMÜLLER² — ¹Institut für Ionenphysik und Angewandte Physik, Universität Innsbruck, Austria — ²Physikalisches Institut, Universität Heidelberg, Germany

Cooling molecular anions is key for the production of cold antihydrogen and the creation of anionic coulomb crystals and would open up new fundamental research areas in modern physics and chemistry. An established technique to store and cool anions are collisions with buffer gas in multipole radio frequency ion traps. However, the temperature is limited by the temperature of the used cryogenic cooling medium.

In this contribution we present forced evaporative cooling down to 2.2(8) K by means of photodetachment of an anionic OH⁻ ensemble, confined in a multipole wire trap. [1] This enables a phase space density approaching the near-strong Coulomb coupling regime. The anion cooling dynamics are described by a quantitative analysis of the experimental results with a full thermodynamic model [2] without any fitting parameters. In principle, this technique can be used for cooling basically any anionic species below the temperature of liquid helium.

[1] J. Tauch, et al. arXiv preprint arXiv:2211.11264 (2022).

[2] A. Crubellier. J. Phys. B, 23(20), 3585 (1990).

MO 10.4 Wed 15:15 F142

Product spin and binding energy propensities for three-body recombination of ultracold atoms — ●JINGLUN LI¹, SHINSUKE HAZE¹, JOSÉ P. D'INCAO^{1,2}, DOMINIK DORER¹, MARKUS DEISS¹, EBERHARD TIEMANN³, PAUL S. JULIENNE⁴, and JOHANNES HECKER DENSCHLAG¹ — ¹Institut für Quantenmaterie, Universität Ulm, Germany — ²JILA, University of Colorado, USA — ³Institut für Quantenoptik, Leibniz Universität Hannover, Germany — ⁴JQI, University

of Maryland, USA

Three-body recombination (TBR) is an elementary chemical reaction process, in which free atoms collide to form a molecule and release the binding energy E_b into the translational movement of the molecule and the third atom. Knowing favored molecular products in TBR is crucial for various fields such as astrophysics, atmospheric physics, and physical chemistry. In recent years we have been working experimentally and theoretically on TBR of ultracold atoms and have achieved great progress in identifying the molecular product distribution on a full quantum state resolution level. In particular, for ultracold Rb atoms we find that TBR intends to produce a molecule preserving the initial spins of two atoms that form it and that the state-to-state reaction rate follows roughly a power-law scaling $L_3 \propto 1/E_b$. Our numerical simulations predict that the $1/E_b$ propensity even holds, with a different prefactor, for two specific groups of molecular products disfavored by the spin propensity. We further elaborate a more comprehensive theoretical investigation on different alkali-metal species to explore the modifications and breakdowns of these propensities.

MO 10.5 Wed 15:30 F142

Two-photon optical shielding of collisions between ultracold polar molecules. — ●CHARBEL KARAM¹, MARA MEYER ZUM ALTEN BORGLÖH³, ROMAIN VEXIAU¹, MAXENCE LEPERS², SILKE OSPELKAUS³, NADIA BOULOUPA-MAAFA¹, LEON KARPA³, and OLIVIER DULIEU¹ — ¹Université Paris-Saclay, CNRS, Laboratoire Aimé Cotton, Orsay 91400, France — ²Laboratoire interdisciplinaire Carnot de Bourgogne, Cedex F-21075 Dijon, France — ³Institut für Quantenoptik, Leibniz Universität Hannover, 30167 Hannover, Germany

We propose a method to engineer repulsive long-range interactions between ultracold ground-state molecules using optical fields, thus preventing short-range collisional losses. It maps the microwave coupling recently used for collisional shielding onto a two-photon transition, and takes advantage of optical control techniques. In contrast to one-photon optical shielding [Phys. Rev. Lett. 125, 153202 (2020)], this scheme avoids heating of the molecular gas due to photon scattering. The proposed protocol, exemplified for $^{23}\text{Na}^{39}\text{K}$, should be applicable to a large class of polar diatomic molecules.

MO 10.6 Wed 15:45 F142

Light controlled engineering of long-range molecular states — ●PATRICK MISCHKE, JANA BENDER, TANITA KLAS, FLORIAN BINOTH,

THOMAS NIEDERPRÜM, and HERWIG OTT — Department of Physics and Research center OPTIMAS, RPTU Kaiserslautern-Landau

We experimentally engineer the deformation of the 5S-6P potential of Rubidium atoms at large interatomic distances on the order of several hundred Bohr radii. This deformation leads to a potential shape that supports bound molecular states.

To achieve this, we couple off-resonantly to an ultra-long range Rydberg molecule potential using strong laser driving. Properties that are commonly associated with Rydberg molecules, usually formed by an Rydberg atom and a ground state atom, are optically admixed to the 5S-6P pair state.

We spectroscopically observe the photoassociated 5S-6P molecules. The change in binding energy for different experimental coupling parameters is in qualitative agreement with a simple theoretical model.

MO 10.7 Wed 16:00 F142

Electric-field-controlled dipolar collisions between trapped polyatomic molecules — ●FLORIAN JUNG, MANUEL KOLLER, JINDARATSAMEE PHROMPAO, MARTIN ZEPPENFELD, ISABEL M. RABEY, and GERHARD REMPE — Max-Planck-Institut für Quantenoptik, Hans-Kopfermann-Strasse 1, 85748 Garching, Germany

Polar symmetric top molecules exhibit a permanent electric dipole moment which creates strong anisotropic interactions, and allows them to be manipulated with moderate electric fields. This, together with their multitude of internal states, renders them promising for applications in e.g. quantum information processing or cold chemistry. For these applications reaching high-density low-temperature ensembles is imperative. This requires the capability to control collisional losses, which is a challenging task.

By combining a cryogenic buffer-gas cell with a centrifuge decelerator and an electrostatic trap, we can now confine up to 2×10^7 CH_3F molecules at a temperature of ~ 350 mK for several seconds, achieving densities of up to $10^7/\text{cm}^3$, which allows for the observation of collisions [1]. We employ a homogeneous control field, covering a large fraction of our trap to mitigate collisional losses and obtain inelastic loss coefficients below $4 \times 10^{-8} \text{cm}^3/\text{s}$. An ab-initio theory shows excellent agreement with our experiment and highlights dipolar relaxation as the major loss mechanism. These findings are immediately relevant for cold molecular collision studies and an important step towards the observation of re-thermalisation between polyatomic molecules.

[1] M. Koller *et al.*, Phys. Rev. Lett. **128**, 203401 (2022).

MO 11: Quantum Technologies (joint session Q/MO/QI)

Time: Wednesday 14:30–16:30

Location: E214

Invited Talk

MO 11.1 Wed 14:30 E214

BMBF-Förderprogramm: Wissenschaftliche Vorprojekte — ●BERNHARD IHRIG and JOHANNES MUND — VDI Technologiezentrum GmbH

Die zweite Quantenrevolution und die schnell voranschreitenden Entwicklungen in der Photonik bieten großes Potenzial für Anwendungen in Ökonomie, Ökologie und Gesellschaft. Zugleich sind neue Erkenntnisse aus der Grundlagenforschung in einem frühen Stadium hinsichtlich der Herausforderungen und Risiken bei der Umsetzung oftmals kaum zu beurteilen. Daher müssen wissenschaftlich-technische Vorarbeiten eine Grundlage schaffen, die es ermöglicht, das Potenzial einer neuen Erfindung bzw. der neuen wissenschaftlichen Erkenntnis zu bewerten.

Das Bundesministerium für Bildung und Forschung (BMBF) beachtet daher, sogenannte Wissenschaftliche Vorprojekte (WiVoPro) im Bereich der Photonik und der Quantentechnologien auf Grundlage des Forschungsprogramms Quantensysteme zu fördern. Das Ziel dieser Vorprojekte besteht darin, wissenschaftliche Fragestellungen im Hinblick auf zukünftige industrielle Anwendungen in den Quantentechnologien und der Photonik zu untersuchen. Sie sollen die bestehende Forschungsförderung ergänzen und eine Brücke zwischen Grundlagenforschung und industriegeführter Verbundförderung schlagen.

Wir als Projektträger VDI Technologiezentrum GmbH möchten die Maßnahme in diesem Rahmen vorstellen, bewerben und Ihre Fragen für eine mögliche Förderung beantworten.

MO 11.2 Wed 15:00 E214

Mikrofabrikation von Ionenfallen für einen skalierbaren Quantencomputer — ●EIKE ISEKE^{1,2}, FRIEDERIKE GIEBEL^{1,2}, NILA KRISHNAKUMAR^{1,2}, KONSTANTIN THRONBERENS^{1,2}, JACOB STUPP^{1,2}, AMADO BAUTISTA-SALVADOR^{1,2} und CHRISTIAN OSPELKAUS^{1,2} — ¹Leibniz Universität Hannover, Hannover, Deutschland — ²Physikalisch Technische Bundesanstalt, Braunschweig, Deutschland

Die Ionenfallentechnologie ist eine vielversprechende Option auf dem Weg zur Entwicklung eines skalierbaren Quantencomputers. Eine mögliche Realisierung stellt die Multilagen-Ionenfalle dar [1]. Durch multiple Lagen wird die Integrationsdichte entscheidend erhöht und es können neuartige Ionenfallendesigns realisiert werden.

Die zunehmende Komplexität der Fallen stellt neue Anforderungen an die Mikrofabrikationsmethoden. Forschung und Entwicklung in diesem Feld fokussieren sich unter anderem auf die Interposer-Technologie, das Thermokompressionsbonds und die Substratdurchkontaktierung mittels TSVs (through silicon vias).

Diese fortschrittlichen Fabrikationsmethoden ermöglichen die Skalierung der Plattform sowohl durch die Möglichkeit die Anzahl der geführten Signale zu erhöhen, als auch durch die gesteigerte Zuverlässigkeit der Verbindungstechnologie.

MO 11.3 Wed 15:15 E214

Squeezed States of Light for Future Gravitational Wave Detectors at a Wavelength of 1550 nm — ●FABIAN MEYLAHN^{1,2}, BENNO WILLKE^{1,2}, and HENNING VAHLBRUCH^{1,2} — ¹Max Planck Institute for Gravitational Physics (Albert Einstein Institute), D-30167 Hannover, Germany — ²Leibniz Universität Hannover, D-30167 Hannover, Germany

The generation of strongly squeezed vacuum states of light is a key technology for future ground-based gravitational wave detectors (GWDs) to reach sensitivities beyond their quantum noise limit. For some proposed observatory designs, an operating laser wavelength of 1550 nm or around 2 μm is required to enable the use of cryogenically cooled silicon test masses for thermal noise reduction. Here, we present the first the direct measurement of up to 11.5 dB squeezing at 1550 nm over the complete detection bandwidth of future ground-based GWDs ranging from 10 kHz down to below 1 Hz. Furthermore, we directly observe a quantum shot-noise reduction of up to 13.5 dB at megahertz frequencies. This allows us to derive a precise constraint on the absolute quantum efficiency of the photodiode used for balanced homodyne detection. These results hold important insight regarding the quantum noise reduction efficiency in future GWDs, as well as for quantum information and cryptography, where low decoherence of nonclassical states of light is also of high relevance.

MO 11.4 Wed 15:30 E214

A single-photon source based on hot Rydberg atoms — ●JAN REUTER^{1,2}, MAX MÄUSEZAHN³, FELIX MOUNTSILIS³, TILMAN PFAU³, TOMMASO CALARCO^{1,2}, ROBERT LÖW³, and MATTHIAS MÜLLER¹ — ¹Forschungszentrum Jülich GmbH — ²Universität zu Köln — ³Universität Stuttgart

The leading effects of a single-photon source based on Rydberg atoms are the strong van-der-Waals interaction between the atoms as well as the collective decay of the atom ensemble. Our setup is a vapor cell filled with Rubidium atoms which we excite via three different laser pulses. The decay of this excitation will then lead to the emission of a single photon. To ensure robustness, we investigated the behavior of moving Rydberg atoms and optimized the laser pulse sequence. For that, we simulated the transitions of Rubidium atoms from the ground state over the Rydberg state up to the singly-excited collective states. We can show that the collective decay of the single excitations leads to a fast and directed photon emission, while double excitations show no or only weak collective properties.

MO 11.5 Wed 15:45 E214

Resolving photon numbers using ultra-high-resolution timing single-channel electronic readout of a conventional superconducting nanowire single photon detector — ●GREGOR SAUER^{1,2}, MIRCO KOLARCZIK³, RODRIGO GOMEZ^{1,2}, HELMUT FEDDER³, and FABIAN STEINLECHNER^{1,2} — ¹Institute of Applied Physics, Abbe Center of Photonics, Friedrich Schiller University, 07743 Jena, Germany — ²Fraunhofer Institute for Applied Optics and Precision Engineering IOF, 07745 Jena, Germany — ³Swabian Instruments GmbH, 70435 Stuttgart, Germany

Photon-number-resolving (PNR) detectors are indispensable building blocks for applications in quantum communications, computing, and sensing. PNR is commonly achieved by multiplexing onto several superconducting nanowire single-photon detectors (SNSPD) or using transition-edge sensors with energy- and photon-number resolution. This comes at the cost of resource overhead (for multiplexing) or long recovery times (for transition-edge sensors).

Here, we show how ultra-high-resolution timing measurements of the rising and falling edge of electrical pulses generated from the SNSPDs enable to distinguish photon numbers of up to 5 in a single-shot mea-

surement. This provides a practical and comparably low-cost PNR detector, offering high detection efficiency and operational repetition rate. We present the implementation of such a PNR detector system (in the telecom C-band) and its characterization by measuring the photon-number statistics of a 300fs-pulsed coherent input source with tunable average photon number and repetition rate.

MO 11.6 Wed 16:00 E214

N00N-states for super-resolving quantum imaging and sensing — ●GIL ZIMMERMANN¹ and FABIAN STEINLECHNER^{1,2} — ¹Institute of Applied Physics, Abbe Center of Photonics, Friedrich Schiller University, 07743 Jena, Germany — ²Fraunhofer Institute for Applied Optics and Precision Engineering IOF, 07745 Jena, Germany

Quantum measurement techniques can serve to improve precision imaging and sensing through entanglement. Employing N00N-states, i.e., maximally path-entangled photon-number states of two modes, the Heisenberg limit $1/N$ with N photons can be reached in precision phase measurements, thus overcoming the shot-noise limit. Furthermore, the Rayleigh diffraction limit can be overcome by a factor N . Therefore, the goal is to efficiently generate high N00N-states with $N > 2$ to improve current sensing schemes achieving super-resolution and super-sensitivity. High-N00N states with $N=5$ photons have already been generated experimentally with high fidelity, as shown by Afek et al. This talk will focus on schemes with relatively low complexity to generate high N00N-states. In addition, applications of high-N00N states, e.g., in the context of quantum-enhanced lidar systems or quantum microscopy, are discussed, taking into account their high fragility due to interactions with the environment.

MO 11.7 Wed 16:15 E214

Non-destructive measurement of phonon number states using the Autler-Townes effect — ●MARION MALLWEGER¹, MURILO DE OLIVEIRA², ROBIN THOMM¹, HARRY PARKE¹, NATALIA KUK¹, GERARD HIGGINS^{1,3}, ROMAIN BACHELARD^{2,4}, CELSO VILLAS-BOAS², and MARKUS HENNRICH¹ — ¹Department of Physics, Stockholm University, Sweden — ²Departamento de Física, Universidade Federal de São Carlos, Brazil — ³Department of Microtechnology and Nanoscience (MC2), Chalmers University of Technology, Sweden — ⁴Université Côte d'Azur, CNRS, Institut de Physique de Nice, France

Quantum technologies employing trapped ion qubits are currently some of the most advanced systems with regards to experimental methods in quantum computation, simulation and metrology. This is primarily due to the excellent control available over the ions' motional and electronic states. In this work we present a new method to measure the distribution of motional number states in a non-destructive manner. The technique can be applied to all platforms where a quantum harmonic oscillator is coupled to a three level system. We demonstrate the technique using a single trapped $^{88}\text{Sr}^+$ ion. The method relies on the Autler-Townes effect that arises when two levels are strongly coupled while being probed by a third level. If the two levels are coupled on a sideband transition, then the magnitude of the Autler-Townes splitting depends on the phonon number state. This new method provides a robust and efficient way of measuring motional states of quantum harmonic oscillators. It can even be applied to perform single shot measurements of phonon number states in a non-destructive way.

MO 12: Interaction with Strong or Short Laser Pulses II (joint session A/MO)

Time: Wednesday 14:30–16:15

Location: F107

Invited Talk

MO 12.1 Wed 14:30 F107

Adiabatic properties of the bicircular attoclock — ●PAUL WINTER and MANFRED LEIN — Leibniz University Hannover

If the right field-strength ratio between a circularly polarized laser pulse and its counter-rotating second harmonic is chosen, one can create a quasilinear electric field in the temporal vicinity of the maximal field. In contrast to conventional linear polarization, rescattering is avoided and a detailed study of direct ionization in strong fields is possible.

The well-defined direction of the field at the ionization time enables us to investigate orientation dependencies in the ionization of molecules in a controlled manner. As our main observables, the ionization yield and the orientation-dependent attoclock shift (i.e. the

potential-induced shift of the peak of the electron momentum distribution) are obtained by solving the two-dimensional time-dependent Schrödinger equation for HeH^+ and H_2 .

In the regime of small Keldysh parameter $\gamma = \sqrt{2I_p} \frac{\omega}{E} \ll 1$, ionization can be described by two-step models, in which the electron travels classically after tunneling out. A crucial factor in these adiabatic models (and hence for the predicted attoshift) is the location of the exit point, which is sensitive to molecular properties such as the dipole moment and the polarizability of the ionized orbital.

MO 12.2 Wed 15:00 F107

Towards strong-field XUV coherent control — ●F. RICHTER¹, C. MANZONI², A. NGAI¹, M. MICHELBAACH¹, D. UHL¹, F. LANDMESSER¹, N. RENDLER¹, S. D. GANESHAMANDIRAM¹, C.

CALLEGARI³, M. DI FRAIA³, N. PAL³, O. PLEKAN³, G. SANSONE¹, K. PRINCE³, T. LAARMANN⁴, M. MUDRICH⁵, P. REBERNIK³, R. FEIFEL⁶, R. SQUIBB⁶, M. WOLLENHAUPT⁷, S. HARTWEG¹, G. CERULLO², F. STIENKEMEIER¹, and L. BRUDER¹ — ¹Institute of Physics, University of Freiburg — ²Dipartimento di Fisica, Politecnico di Milano — ³Elettra - Sincrotrone Trieste S.C.p.A. — ⁴Department of Physics, University of Hamburg — ⁵Department of Physics and Astronomy, Aarhus University — ⁶Department of Physics, University of Gothenburg — ⁷Institute of Physics, University of Oldenburg

Within the NIR and VIS wavelength regime there are various coherent control schemes. However, for coherent control in the XUV regime two major challenges arise: (i) The technical challenge to manipulate the pulses. (ii) XUV radiation induces typically extremely fast relaxation dynamics, which compete with the coherent control scheme. Ultrafast control schemes are, hence, paramount which can be achieved by using intense pulses beyond the weak field regime. Intense optical fields are known to induce Rabi oscillations leading to Autler-Townes level splittings. We investigate the population control of the respective sub-levels as shown in the NIR [1]. We will present simulations of the expected Autler-Townes splitting as well as preliminary results from our beamtime at the free electron laser FERMI.

[1] M. Wollenhaupt et al., Phys. Rev. A 68, 015401 (2003).

MO 12.3 Wed 15:15 F107

The N-shaped partition method: A novel parallel implementation of the Crank Nicolson algorithm — ●FRANCISCO NAVARRETE and DIETER BAUER — Institute of Physics, University of Rostock

We develop an algorithm to solve tridiagonal systems of linear equations, which appear in implicit finite-difference schemes of partial differential equations (PDEs), being the time-dependent Schrödinger equation (TDSE) an ideal candidate to benefit from it. Our N-shaped partition method optimizes the implementation of the numerical calculation on parallel architectures, without memory size constraints. Specifically, we discuss the realization of our method on graphics processing units (GPUs) and the Message Passing Interface (MPI). In GPU implementations, our scheme is particularly advantageous for systems whose size exceeds the global memory of a single processor. Moreover, because of its lack of memory constraints and the generality of the algorithm, it is well-suited for mixed architectures, typically available in large high performance computing (HPC) centres. We also provide an analytical estimation of the optimal parameters to implement our algorithm, and test numerically the suitability of our formula in a GPU implementation. Our method will be helpful to tackle problems which require large spatial grids for which ab-initio studies might be otherwise prohibitive both because of large shared-memory requirements and computation times.

MO 12.4 Wed 15:30 F107

Dephasing effects in high-order harmonic generation from finite Su-Schrieffer-Heeger chains — ●CHRISTOPH JÜRSS and DIETER BAUER — Institute of Physics, University of Rostock, Germany

The Su-Schrieffer-Heeger (SSH) model describes a linear, one-dimensional chain that displays topological effects. Due to its simplicity, the SSH-model has been used to study numerous effects in

topological insulators. The most interesting feature of topologically non-trivial insulators are their topologically protected edge states. It was shown in previous studies that the generation of high-order harmonics can be influenced by the topological nature of the solid and even by just the edge states themselves. In order to obtain more realistic simulated harmonic spectra, relaxation and dephasing effects should be taken into account. This is usually done for the bulk, i.e., with no edge states present. In this work, we implement dephasing for the finite SSH-model and compare the results to those from the respective bulk.

MO 12.5 Wed 15:45 F107

Delay time and Non-Adiabatic Calibration of the Attoclock.

Multiphoton process versus tunneling in strong field interaction —

●OSSAMA KULLIE¹ and IGOR IVANOV² — ¹Institute for Physics, University of Kassel — ²Institute for Basic Science (IBS), Gwangju 61005, Republic of Korea

Recent measurement of the tunneling time in attosecond experiments (termed attoclock), triggered a hot debate about the tunneling time, the role of time in quantum mechanics and the separation of the interaction with the laser pulse into two regimes of a different character, the multiphoton and the tunneling (field-) ionization. In the adiabatic field calibration, we showed in earlier works (see e.g. [1]) that our real tunneling time model fits well to the experimental data. In the present work [2], we investigate the nonadiabatic case (see [3]) and combine it with a new result of a numerical integration of the TDSE (see [4]). Our model explains the experimental of Hofmann et al [3] with an excellent agreement. Our model is appealing because it offers a clear picture of the multiphoton and tunneling. In the nonadiabatic case, the barrier itself is mainly driven by multiphoton absorption and the number of the absorbed photons depends on the δ -value of the barrier height. Surprisingly, for a field strength $F < F_a$ (the atomic field strength) the model always indicates a time delay with respect to the lower quantum limit at $F = F_a$. Its saturation at the adiabatic limit explains the well-known Hartman effect or Hartman paradox. [1] O. kullie. PRA 92 052118 (2015). [2] O. kullie and I. Ivanov arxiv.2005.09938v4. [3] J. of Mod. Opt. 66, 1052, 2019. [4] Phys. Rev. A 89, 021402, 2014.

MO 12.6 Wed 16:00 F107

Writing waveguides in polymers with femtosecond laser. —

●DMITRII PEREVOZNIK^{1,2} and UWE MORGNER^{1,2,3} — ¹Institut für Quantenoptik, Welfengarten 1, 30167, Hannover — ²Cluster of Excellence PhoenixD (Photonics, Optics, and Engineering -Innovation Across Disciplines), Hannover, Germany — ³Laser Zentrum Hannover e.V., Hollerithalle 8, D-30419 Hannover, Germany

Writing waveguides with femtosecond laser is a very promising technique and has already proven its performance in glasses and crystals. Nevertheless, writing waveguides in polymers is a just developing field and polymer material can offer the potential to create low-cost and complex structures inside the volume of the material. Singlemode waveguides with propagation losses of 0.6 *m were achieved by putting modifications, done by femtosecond laser, around waveguide core forming different geometries. Also shown are various optical elements embedded in waveguides, such as waveguide splitters or Bragg gratings.

MO 13: Poster II

Time: Wednesday 16:30–19:00

Location: Empore Lichthof

MO 13.1 Wed 16:30 Empore Lichthof

Towards the production of groundstate RbYb — ●CHRISTIAN SILLUS, BASTIAN POLLKLESENER, ARNE KALLWEIT, CÉLINE CASTOR, and AXEL GÖRLITZ — Heinrich-Heine Universität

Ultracold dipolar molecules constitute a promising system for the investigation of topics like ultracold chemistry, novel interactions in quantum gases, precision measurements and quantum information. Here we report on first experiments in our apparatus for the production of ultracold RbYb molecules. This setup constitutes an improvement of our old apparatus, where the interactions in RbYb and possible routes to molecule production have already been studied extensively [1,2]. In the new setup a major goal is the efficient production of ground state RbYb molecules. We employ optical tweezers to transport individually cooled samples of Rb and Yb from their separate production chambers to a dedicated science chamber. Here we start

to study interspecies interactions of different isotopes by overlapping crossed optical dipole traps. To explore the pathways towards ground state molecules we start with photoassociation spectroscopy near the intercombination line of Yb.

[1] M. Borkowski et al., PRA 88, 052708 (2013)

[2] C. Bruni et al., PRA 94, 022503 (2016)

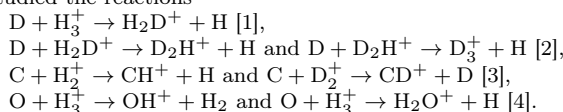
MO 13.2 Wed 16:30 Empore Lichthof

Merged-beams experiments on molecular ion-neutral reactions for astrochemistry — ●PIERRE-MICHEL HILLENBRAND¹,

XAVIER URBAIN², and DANIEL WOLF SAVIN³ — ¹Justus-Liebig Universität, Giessen, Germany — ²Université catholique de Louvain, Louvain-la-Neuve, Belgium — ³Columbia University, New York, USA

The gas-phase formation of complex molecules in the interstellar medium proceeds dominantly through barrierless ion-neutral reactions

at typical temperatures of 10–100 K. Our merged-beams apparatus at Columbia University in New York City enables us to measure energy-dependent absolute cross sections of molecular formation processes for reactions of singly-charged molecules with neutral atoms and derive temperature-dependent thermal rate coefficients for individual product channels. Focusing on key reactions implemented in astrochemical models as well as on systems of fundamental interest, we have recently studied the reactions



For example, the branching ratio for the two product channels of the $\text{O} + \text{H}_3^+$ reaction is relevant for accurately modeling the gas-phase formation of water in the diffuse and dense molecular clouds.

[1] *Astrophys. J.* **877**, 38 (2019). [2] *J. Chem. Phys.* **154**, 084307 (2021). [3] *Phys. Chem. Chem. Phys.* **22**, 27364 (2020). [4] *Astrophys. J.*, **927**, 47 (2022).

MO 13.3 Wed 16:30 Empore Lichthof

UV Photoexcitation-Photoemission Map of the Hydrogen Molecule — •GABRIEL KLASSEN¹, ANDREAS HANS¹, PHILIPP SCHMIDT², CATMARN KÜSTER-WETEKAM¹, JOHANNES VIEHMANN¹, and ARNO EHRESMANN¹ — ¹Institut für Physik und CINSaT, Universität Kassel, Heinrich-Plett-Str. 40, 34132 Kassel, Germany — ²European XFEL, Holzkoppel 4, 22869 Schenefeld, Germany

The hydrogen molecule, the simplest molecule we know, has been studied extensively in the past. Although most of the details of its electronic potentials are well understood, there is no complete set of experimentally determined absolute data with quantitative absorption and dispersed emission cross sections. These cross sections, however, are necessary to interpret spectra of gaseous clouds in space, where H₂ may be used as a probe molecule, once its cross sections are known.

On the road towards such a full data set for the hydrogen molecule we are performing experiments where H₂ is excited by small-bandwidth monochromatized synchrotron radiation and its emission is recorded dispersedly with high resolution. By scanning synchrotron radiation over parts of the UV spectrum and measuring the consecutive fluorescence we are able to construct a 2d-map with distinct features being representative for the H₂ rovibronic structure.

MO 13.4 Wed 16:30 Empore Lichthof

Ultrafast spectroscopy of intramolecular dynamics and photodissociation of a single trapped molecular ion — •ZHENLIN WU, BRANDON FUREY, STEFAN WALSER, GUANQUN MU, RENÉ NARDI, and PHILIPP SCHINDLER — Institut für Experimentalphysik, Universität Innsbruck, Innsbruck, Austria

Trapped atomic ions are one of the most promising platforms for quantum simulation and quantum computation. By co-trapping molecular ions with atomic ions, quantum logic spectroscopy can be performed to investigate the rovibrational structure of various molecular species, as demonstrated recently with diatomic molecular ions including CaH^+ and N_2^+ . In a similar system, we plan to explore ultrafast intramolecular dynamics in complicated polyatomic molecular ions with femtosecond laser pulses. In particular, we will measure the net momentum transfer of the pump and the delayed probe pulse. Such momentum change can be detected by preparing the common motion of the ion crystal in a non-classical state. With this scheme, we aim to study the evolution of vibrational excitations inside a single polyatomic molecule of chemical or biological importance.

We are currently investigating the interaction between ultrafast laser pulses and molecular ions by measuring the photodissociation threshold of CaOH^+ . The molecules are generated by chemical interactions between trapped Ca^+ ions and water molecules that can be introduced with a gate valve into the main experimental chamber. After the reaction, a Ca^+ ion turns “dark” and femtosecond laser pulses with variable wavelength are applied to dissociate it back to a “bright” Ca^+ ion.

MO 13.5 Wed 16:30 Empore Lichthof

Spectroscopic and collision studies of the laser-cooling candidate C_2^- — •CHRISTINE MARIA LOCHMANN, MARKUS NÖTZOLD, SRUTHI PURUSHU MELATH, ROBERT WILD, and ROLAND WESTER — Institut für Ionenphysik und Angewandte Physik, Universität Innsbruck, Austria

The precise control of the external and internal motion of negative ions has been of increasing interest in the past decades. One possible

method to reach this control is through laser cooling, which has never been achieved for a negatively charged particle, as only a few anions provide the strongly bound electronic excited and adequate internal energy states suitable for laser cooling schemes. One such candidate is the molecular carbon anion C_2^- . Here, we present our studies on the spectroscopy of C_2^- carried out in a cryogenic 16-pole RF wire trap at 6 K to determine precise laser cooling transition frequencies. We are able to resolve the spin-rotation splitting in C_2^- with an accuracy of 20 MHz and determine the rotational and translational temperature of the ions [1]. A useful method to repopulate the ground state of the molecule could be through vibrational quenching. We studied the quenching rate into the vibrational ground state of C_2^- in cold collisions with H₂. At high H₂ densities the collision also leads to a three-body reaction which forms C_2H^- . We measured the temperature dependent three-body rate between 10 K and 28 K to benchmark quantum calculations.

[1] M. Nötzold et al., *Phys. Rev. A* **106**, 023111 (2022)

MO 13.6 Wed 16:30 Empore Lichthof

State-to-state ion-molecule reaction dynamics — •TIM MICHAELSEN, DASARATH SWARAJ, ARNAB KHAN, ROBERT WILD, and ROLAND WESTER — Institut für Ionenphysik und Angewandte Physik, Universität Innsbruck, Technikerstraße 25/3 A-6020 Innsbruck, Austria

The study of reaction dynamics aims to gain a mechanistic understanding how chemical reactions occur on an atomistic level. The combination of crossed beam experiments with velocity map imaging [VMI], especially in comparison with trajectory calculations, are a powerful tool to observe these dynamics [1]. Product state detection for ion-molecule reactions were limited by energy resolution, at least for non di-atomic products [2].

Here we present a new setup, that aims push the required resolution towards product vibrational state detection in larger systems, which combined with prepared ground state reactants prior to the reactive collision, will let us obtain state-to-state differential cross sections. This is achieved by a newly designed laser-ionization source for H_2^+ and implementation of a Rydberg-tagging scheme to detect neutral hydrogen atom products in coincidence [3]. First reactions of interest are $\text{H}_2^+ + \text{Ne}$ and H_2 , where little is known experimentally about the energy partitioning and dynamics.

[1] J. Meyer, E. Carrascosa, et al. *Nat. Chem.* **13**, 977-981 (2021)

[2] T. Michaelsen, et al., *J. Chem. Phys.* **147**, 013940 (2017)

[3] M. Qiu, et al., *Science* **311** 5766, 1440-1443 (2006)

MO 13.7 Wed 16:30 Empore Lichthof

Towards state-to-state ion-molecule reaction dynamics — •TIM MICHAELSEN, DASARATH SWARAJ, FLORIAN TRUMMER, ARNAB KHAN, ROBERT WILD, and ROLAND WESTER — Institut für Ionenphysik und Angewandte Physik, Universität Innsbruck, Technikerstraße 25/3, A-6020 Innsbruck, Austria

The study of reaction dynamics aims to gain an understanding how chemical reactions occur on an atomistic level. Experiments combining crossed beams with velocity map imaging [VMI] are a powerful tool to observe these dynamics [1]. In the past product state detection for ion-molecule reactions was limited by energy resolution, at least for non-diatom products [2].

Here we present a new setup, that aims to push the required resolution towards product vibrational state detection in larger systems, which combined with prepared ground state reactants prior to the reactive collision, will let us obtain state-to-state differential cross sections. This is achieved by a newly designed laser-ionization source for H_2^+ and implementation of a Rydberg-tagging scheme to detect neutral hydrogen atom products in coincidence [3]. First reactions of interest are $\text{H}_2^+ + \text{Ne}$ and H_2 , where interesting quantum dynamics have been predicted and little is known experimentally about the energy partitioning.

[1] J. Meyer, E. Carrascosa, et al. *Nat. Chem.* **13**, 977-981 (2021)

[2] T. Michaelsen, et al., *J. Chem. Phys.* **147**, 013940 (2017)

[3] M. Qiu, et al., *Science* **311** 5766, 1440-1443 (2006)

MO 13.8 Wed 16:30 Empore Lichthof

Development of a Cryogenic Beam Source for Cold Calcium Monofluoride Molecules — •PHILLIP GROSS, OLE PROCHNOW, ALEXANDRA KÖPF, and TIM LANGEN — 5. Physikalisches Institut, Universität Stuttgart

The production of ultracold molecular quantum gases promises to add long-range dipolar interactions to the quantum simulation toolbox.

Here we first show theoretically that such dipolar interactions can lead to the formation of new phases of matter in bulk molecular Bose-Einstein condensates, such as droplet states and supersolids. Second, we present a new setup to realize these phases of matter in an experiment. In this setup we work with calcium monofluoride molecules, which are characterized by large electric dipole moments and well-established laser cooling strategies. In a first step to create a gas of these molecules with high phase space density, we present the design of a cryogenic buffer gas beam source, as well as extensive simulations of transversal and longitudinal laser cooling of the resulting molecular beam.

MO 13.9 Wed 16:30 Empore Lichthof
Imaging Photoelectron Circular Dichroism in Mass-Selected Chiral Anions — ●VIKTORIA KATHARINA BRANDT, JENNY TRIP-TOW, ANDRÉ FIELICKE, GERARD MEIJER, and MALLORY GREEN — Molecular Physics Department of the Fritz Haber Institute of the Max Planck Society

Photoelectron Circular Dichroism (PECD) is a chiral effect that manifests in the angle-dependent photoemission of an electron upon irradiation of a chiral molecule by circularly polarized light. The use of anions in this technique allows for mass-selectivity and eliminates the need for X-ray based ionization sources, thus leading to a potentially robust analytical tool for chiral discrimination of multicomponent gas-phase samples. PECD spectroscopy of neutral chiral species has flourished over the past decades. A PECD effect in anions, however, has only been observed recently. By coupling pre-photodetachment mass selection, tuneable detachment, and velocity-map imaging anion photoelectron spectroscopy, we provide an energy-resolved PECD signal for mass-selected anions. We have recently demonstrated this successfully in the study of the deprotonated 1-indanol anion, where we observed a PECD effect for several detachment channels and a maximum PECD effect of 11%. Current aims of the project are focused on improving the resolution for spectroscopic PECD measurements. Such improvements will pave a way to an understanding of the underlying cause in the difference in electron scattering dynamics of neutral and charged molecules, as well as work towards a robust analytical method for the chiral analysis of dilute, but complex, chemical samples.

MO 13.10 Wed 16:30 Empore Lichthof
Implementation of time-correlated single photon counting for studying photo-activated processes in aggregates of organic molecules attached to rare gas clusters — ●ALEKSANDR DEMIANENKO, MORITZ MICHELBACH, SEBASTIAN HARTWEG, and FRANK STIENKEMEIER — Institute of Physics, University of Freiburg, Germany

Collective effects in organic semiconductors affect excited state lifetimes, important for organic optoelectronic and photovoltaic applications. Previous studies addressed radiative and non-radiative decay mechanisms of the processes connected to the collective effects in aggregates of polyacenes attached to the surface of solid Ne and Ar clusters. Fluorescence quantum yield and excited singlet state's lifetime decreased upon increasing sample molecules surface density [1,2]. The limited temporal resolution of a few ns, due to the ns-laser excitation and PMT detection, made disambiguation of different contributions to the lifetime reduction challenging.

This project is aimed at enhancing the temporal resolution of fluorescence measurements by implementing time correlated single photon counting. This technique allows us to cover the sub-ns lifetime region currently not achievable in our cluster doping apparatus. We present the current status of the ongoing upgrade of our setup and discuss advantages and implementation challenges.

[1] M. Bohlen et al. *J. Chem. Phys.* 156, 034305 (2022).

[2] S. Izadnia et al. *J. Phys. Chem. Lett.* 8, 2068 (2017).

MO 13.11 Wed 16:30 Empore Lichthof
Learning potential energy surfaces for collisions of H₂+ molecules and rare gas atoms — ●KARL P. HORN¹, LUIS ITZA VAZQUEZ-SALAZAR², CHRISTIANE P. KOCH¹, and MARKUS MEUWLY² — ¹Theoretische Physik, Freie Universität Berlin, Germany — ²Department of Chemistry, Universität Basel, Switzerland

The latest developments in experimental techniques and coupled channel calculations have led to measurements and theoretical simulations of ever increasing accuracy, such as in the case of Helium/Neon and H₂ cold collisions. The utilisation of methods such as multi-reference configuration interaction (MRCI) and full configuration interaction (FCI) ab-initio calculations has already resulted in potential energy

surfaces (PESs) of very high quality [1]. Whilst experiment and theory already show good agreement using these existing potentials, high resolution measurements unlock the enticing possibility of exploiting experimental data for the purpose of improving existing PESs. In this work, we employ a closed loop parameter optimisation in order to morph [2] He-H₂⁺ PESs determined at several levels of theory, including second order Moller Plesset, MRCI and FCI by minimising the difference between collision cross sections from quantum wavepacket simulations and experiment. This yields a collection of PESs all consistent with experimental observables within certain error bounds and provides insight into which regions of the PES are sampled and which require further probing by new experiments.

[1] *Phys. Chem. Chem. Phys.*, 21, 24976-24983, 2019

[2] *J. Chem. Phys.*, 110, 8338, 1999

MO 13.12 Wed 16:30 Empore Lichthof
Reactive Scattering of the oxygen atom transfer reaction between Carbondioxide and group V cations — ●MAXIMILIAN HUBER¹, MARCEL META¹, ATILAY AYASLI², TIM MICHAELSEN², ROLAND WESTER², and JENNIFER MEYER¹ — ¹RPTU Kaiserslautern-Landau Fachbereich Chemie, Kaiserslautern, Germany — ²Universität Innsbruck Institut für Ionenphysik und angewandte Physik, Innsbruck, Austria

With our method we investigate the atomistic dynamics of elementary reactions like the oxygen atom transfer of CO₂ with transition metal cations under single collision conditions. We achieve this by recording energy and angle differential cross sections by means of product ion velocity distributions after a reactive collision. [1]

Here, we present our first results for the oxygen atom transfer reaction Nb⁺ + CO₂ → NbO⁺ + CO measured with our 3D crossed beam velocity map imaging experiment. The reaction is exothermic by 1.68 eV and is spin forbidden in its electronic ground state. Nevertheless the reaction was found to have relativ high reaction efficiencies at room temperature. [2,3] We recorded energy dependent velocity map images of the Niobium reaction at different energies. Also, we compared these results to the reaction of the heavier homologue tantalum Ta⁺ + CO₂ → TaO⁺ + CO.

[1] J. Meyer, R. Wester, *Annu. Rev. Phys. Chem.* 2017, 68, 333. [2] M.R. Sievers, P.B. Armentrout, *Int. J. Mass. Spec.*, 1998, 179/180, 103. [3] G. K. Koyanagi, D. K. Bohme, *J. Phys. Chem. A* 2006, 110, 1232.

MO 13.13 Wed 16:30 Empore Lichthof
Absorption spectroscopy and modeling laser cooling of barium monofluoride — ●FELIX KOGEL, MARIAN ROCKENHÄUSER, EINIUS PULFNEVICIUS, and TIM LANGEN — 5. Physikalisches Institut, Universität Stuttgart

Cold molecular gases are the starting point for studies of cold chemistry, quantum simulation and precision measurements of fundamental physics. Here, we report on our effort towards direct laser cooling of BaF molecules. We perform high-resolution absorption spectroscopy to derive an improved set of molecular constants for the bosonic isotopologues ¹³⁸BaF and ¹³⁶BaF and use these constants to model laser cooling using simulations based on the optical Bloch equations. We find effective Doppler, sub-Doppler, and coherent cooling strategies for the bosonic, as well as for the fermionic BaF isotopologues. This provides important guidance for the corresponding experiments ongoing in our group, and paves the way for tests of fundamental symmetries using these molecules.

MO 13.14 Wed 16:30 Empore Lichthof
Readout techniques for the implementation of a trace-gas sensor for nitric oxide — ●MAURICE SCHAMBER^{1,4}, YANNICK SCHELLANDER^{2,4}, FABIAN MUNKES^{1,4}, PATRICK KASPAR^{1,4}, PHILIPP NEUFELD^{1,4}, ALEXANDER TRACHTMANN^{1,4}, MARIUS WINTER^{2,4}, PATRICK SCHALBERGER^{2,4}, NORBERT FRÜHAUF^{2,4}, HASSAN HAYERI^{3,4}, PHILIPP HENGEL^{3,4}, LARS BAUMGÄRTNER^{3,4}, JENS ANDERS^{3,4}, ROBERT LÖW^{1,4}, TILMAN PFÄU^{1,4}, and HARALD KÜBLER^{1,4} — ¹5. Physikalisches Institut, Universität Stuttgart, Pfaffenwaldring 57, 70569 Stuttgart — ²Institut für Großflächige Mikroelektronik, Universität Stuttgart, Allmandring 3b, 70569 Stuttgart — ³Institut für Intelligente Sensorik und Theoretische Elektrotechnik, Universität Stuttgart, Pfaffenwaldring 47, 70569 Stuttgart — ⁴Center for Integrated Quantum Science and Technology (IQST), Universität Stuttgart

Two different readout techniques for nitric oxide spectroscopy are presented. The first approach is based on the detection of charged par-

ticles created by collisional ionization of Rydberg molecules in a low-pressure through-flow gas cell setup, whereas the second enables the detection of photons in the deep UV based on a-IGZO semiconductor technology. Both methods are presented within the scope of the development of a trace-gas sensor for nitric oxide.

MO 13.15 Wed 16:30 Empore Lichthof
Collisions in a quantum gas of bosonic $^{23}\text{Na}^{39}\text{K}$ molecules

— ●MARA MEYER ZUM ALTEN BORGLOH¹, PHILIPP GERSEMA¹, LEON KARPA¹, KAI KONRAD VOGES¹, JULE CAROLINE HEIER¹, SILKE OSPELKAUS-SCHWARZER¹, CHARBEL KARAM², and OLIVIER DULIEU² — ¹Institut für Quantenoptik, Leibniz Universität Hannover — ²Université Paris-Saclay, CNRS, Laboratoire Aimé Cotton

We report about our experiments with quantum gases of polar $^{23}\text{Na}^{39}\text{K}$ molecules. We discuss both atom-molecule and molecule-molecule collisions including the origin of loss processes in a cloud of chemically stable $^{23}\text{Na}^{39}\text{K}$ molecules.

Furthermore, we discuss a method for suppressing molecular loss using two optical photons at Raman resonance, leading to a potential barrier that protects the colliding molecules from reaching the short range.

MO 13.16 Wed 16:30 Empore Lichthof
Surface charge removal by UV illumination in a microstructured electrostatic trap for cold molecule research

— ●JINDARATSAMEE PHROMPAO, MICHAEL ZIEMBA, YVAN BRIARD, FLORIAN JUNG, MARTIN ZEPPENFELD, ISABEL RABEY, and GERHARD REMPE — Max-Planck-Institut für Quantenoptik, Hans-Kopfermann-Straße 1, 85748 Garching, Germany

Polar molecules are an excellent platform to conduct fascinating research ranging from cold chemistry to tests of fundamental physics. Precise determination of molecular constants is essential for these applications. Toward this end, long observation time [1] is beneficial, which can be achieved by trapping the cold molecules. In our experiment, we employ a microstructured electrostatic trap [2] to confine the molecules for several seconds. The trap potential is created by applying high voltages to the trap electrodes. This, however, induces surface charges that can broaden the electric-field distribution, which renders precise addressing and controlling of the trapped molecules difficult. Thus, surface charge removal is crucial to overcome these problems.

Here, we will highlight the working principles of the electrostatic trap. The characteristics of high-voltage-induced surface charge build up will be described. Furthermore, surface charge removal by UV illumination will be presented. We found that the surface charge removal efficiency depends on the applied UV intensity and wavelength in a nontrivial way.

[1] A. Prehn et al., Phys. Rev. Lett. 127, 173602 (2021).

[2] B.G.U. Englert et al., Phys. Rev. Lett. 107, 263003 (2011).

MO 13.17 Wed 16:30 Empore Lichthof
Challenges and solutions in dispersed fluorescence measurements and determining their absolute cross sections

— ●ROBIN ISE¹, JOHANNES VIEHMANN¹, PHILIPP SCHMIDT², ARNE SCHRÖDER¹, ANDREAS HANS¹, and ARNO EHRESMANN¹ — ¹University of Kassel, Kassel, Germany — ²European XFEL, Hamburg, Germany

Measurements of the absorption and emission of electromagnetic radiation have always been a key contribution to the understanding of the fundamental structure of atoms and molecules. While the ex-

perimental setup used for these measurements, called PIFS (Photon Induced Fluorescence Spectroscopy) and developed by our group, has seen great success in the past, in obtaining large-scale emission maps of H_2 in the near to vacuum ultraviolet spectral range, the improvement of the experimental methods may allow to determine absolute cross sections of the measured process. This ongoing optimization process of the experimental setup shall be illustrated in this work, including the reconfiguration of the setup to accommodate for measurements in the magic angle and an improved method of acquiring absolute pressure data of the medium in question.

MO 13.18 Wed 16:30 Empore Lichthof
A Reaction Microscope for the Cryogenic Storage Ring CSR

— ●FELIX HERRMANN, DAVID V. CHICHARRO, ROBERT MOSHAMMER, CLAUS DIETER SCHRÖTER, and THOMAS PFEIFER — Max-Planck-Institut für Kernphysik, Saupfercheckweg 1, 69117 Heidelberg, Germany

For studying molecular break-up reactions on slow and cold molecular ions and clusters at the cryogenic storage ring CSR, we have designed a dedicated in-ring reaction microscope (ReMi). The ReMi is a combined electron and ion spectrometer for energy and angular resolved particle detection created in an elementary collision process. This spectrometer will deliver kinematically complete data on the reaction dynamics. The implementation into the storage ring of this first cryogenic ReMi worldwide started in May 2022. Its finalization is expected for spring 2023. Subsequently, first fundamental in-ring experiments, reactions like electron transfer, molecular break-up or proton transfer at room temperature conditions, followed by measurements at cryogenic conditions ($< 10\text{ K}$) in a future beamtime period are envisaged. The setup of the complex apparatus, in detail one of the MCP detectors with time and position sensitive delay-line read-out along with the design adaptation for operation at cryogenic temperatures, the supersonic gas jet and laser incouplings, as well as first test measurements will be shown.

MO 13.19 Wed 16:30 Empore Lichthof
New apparatus for synchrotron-based photoelectron spectroscopy of cold and mass-selected ions in the gas phase

— STÖCKS PHILLIP, BÄR FABIAN, WEISE LUKAS, and ●VON ISSENDORFF BERND — Institute of Physics at the University of Freiburg, Freiburg i.Br., Germany

We present the development and construction of a new apparatus for cluster research at the BESSY II synchrotron at the Helmholtz Centre in Berlin (HZB). A magnetron sputter source provides the clusters for the apparatus. The setup allows mass selection with a Quadrupole Mass Spectrometer (QMS) before the clusters are stored. The trap is a linear Radio Frequency (RF) trap. Photoelectrons are emitted through interaction with the synchrotron beam. These are forced out of the trap with a magnetic field using the principle of the magnetic bottle on parallel trajectories in one direction. In this way, sufficient intensities can be achieved. The photoelectrons then pass through a magnetic guiding field and experience a deflection from the synchrotron axis. Subsequently, the photoelectrons are directed and focused by electric ion optics so that the photoelectrons can be guided into the Hemispherical Spectrum Analyser (HSA) as magnetic field-free as possible. At the current state of the project we will especially focus on the simulated photoelectron paths guided and focused by our newly developed photoelectron extractor. Furthermore the detailed design of the whole experiment will be introduced.

MO 14: Ultrafast Dynamics II (joint session MO/A)

Time: Thursday 11:00–13:00

Location: F102

MO 14.1 Thu 11:00 F102
Isosteric molecules in the time-domain

— ●MAXIMILIAN POL-LANKA, CHRISTIAN SCHRÖDER, and REINHARD KIENBERGER — Chair for Laser and X-ray Physics, E11, Technische Universität München, Germany

We report on photoemission timing measurements performed on isosteric molecules in the gas phase on attosecond timescales. Comparing the photoemission time delay between the respective σ and π orbitals in the inner and outer valence states of CO_2 and N_2O leads to a deeper insight into the characteristics of isosterism in the time-domain. Additionally, the isoelectronicity of CO and N_2 is investigated in detail as

a complementary study. Due to the similarities in molecular structure (isostericity) and electronic configurations (isoelectronicity) the pure effect of the specific molecular/orbital characteristics is expected to be probed. We are not only able to experimentally assess the relative photoemission delay between respective outer and inner valence states, but also performing absolute photoemission timing via attosecond streaking spectroscopy using iodomethane (I_4d state) as a reference. The experimental data show great similar tendencies but also differences between the compared molecular orbitals, which are determined but not completely understood up to now. Therefore, further theoretical considerations and accurate modelling of the process of laser-dressed photoionization and the information encoded in photoemission timing

measurements on molecular targets are necessary. This will help us gaining a greater understanding of the correlations between molecular geometry and photoemission time and therefore the isosteric influence.

MO 14.2 Thu 11:15 F102

RABBITT experiments in a vibrationally active ammonia molecule — ●LISA-MARIE KOLL¹, DAVID SORRIBES ORTIZ², IGNACIO MARTÍNEZ CASASÚS², TOBIAS WITTING¹, LORENZ DRESCHER^{1,4}, OLEG KORNILOV¹, MARC JJ VRAKING¹, FERNANDO MARTIN³, and LUIS BAÑARES² — ¹Max Born Institute, Berlin — ²Universidad Complutense de Madrid — ³Universidad Autónoma de Madrid and IMDEA Nanociencia — ⁴University of California, Berkeley

Many RABBITT (Reconstruction of Attosecond Beatings By Interferences of Two-photon Transitions) experiments have been carried out so far for (rare gas) atoms to disentangle the evolution of an electronic wave packet by measuring the photoemission time delays. In molecules, the experiments are more complicated due to the nuclear degrees of freedom. Previously, RABBITT experiments could resolve vibrations in the photoelectron spectrum of N₂ [1], and N₂O [2]. Here, we present RABBITT experiments on NH₃ using the velocity map imaging (VMI) technique. NH₃ belongs to the C_{3v} symmetry group and, as a result of the photoionization process, symmetry is shifted to the D_{3h} conformation in NH₃⁺. The ionization of ammonia is accompanied by rich vibrational structure, such as the long vibrational progression in the v₂ umbrella inversion mode of the X₂A₂ state. These RABBITT experiments present an interesting and challenging case for full-dimensional theoretical calculations and help to demonstrate the capability of the RABBITT method to study in depth vibronic dynamics in polyatomic molecules. [1] S. Haessler et al., Phys. Rev. A 80, 01140R (2009) [2] L. Cattaneo et al., Nature Phys. 14, 733 (2018)

MO 14.3 Thu 11:30 F102

Control of ion+photoelectron entanglement in an attosecond pump-probe experiment — ●LISA-MARIE KOLL, TOBIAS WITTING, and MARC JJ VRAKING — Max Born Institute, Berlin, Germany

Quantum mechanical entanglement is a vibrant topic, culminating in this year's Nobel price award. In attosecond science, it is common to use radiation in the extreme ultra-violet (XUV) regime to study atomic and electronic dynamics. Due to their high photon energy any sample (solid, liquid or gaseous) placed in their path is ionized, creating a bipartite quantum system, i.e. an ion+photoelectron. Entanglement between those sub-systems can have measurable consequences for any attosecond experiment [1]. To illustrate the role of entanglement in photoionization we designed an experimental protocol, which utilizes a pair of phase-locked XUV pulses [2] and an IR pulse to ionize hydrogen molecules. The initially entangled ion+photoelectron system created by the XUV photoionization process is converted by the IR pulse into a coherent superposition of the gerade and ungerade electronic state of the ionic molecule leading to the observation of electronic localization [3]. By changing the time delay between the two XUV pulses the degree of ion+photoelectron entanglement is controlled and as a consequence the degree of electronic coherence. This can lead to the suppression of the observable electronic localization for certain time delays.

[1] L.-M. Koll et. al., Physical Review Letters 128, 043201 (2022)

[2] L.-M. Koll et al., Optics Express 30, 7082-7095 (2022)

[3] G. Sansone et al., Nature 465, 763-766 (2010)

MO 14.4 Thu 11:45 F102

High-order spectroscopy at 100 kHz repetition rate — ●KATJA MAYERSHOFER¹, SIMON BÜTTNER¹, TIM SCHEMBRI^{2,3}, MATTHIAS STOLTE^{2,3}, FRANK WÜRTHNER^{2,3}, and TOBIAS BRIXNER^{1,3} — ¹Institut für Physikalische und Theoretische Chemie, Universität Würzburg, Am Hubland, 97074 Würzburg — ²Institut für Organische Chemie, Universität Würzburg, Am Hubland, 97074 Würzburg — ³Center for Nanosystems Chemistry (CNC), Universität Würzburg, Theodor-Boveri-Weg, 97074 Würzburg

With our new laser setup we can carry out shot-to-shot femtosecond spectroscopy experiments at 100 kHz repetition rate, which decreases the measurement time to a few seconds for conventional coherent two-dimensional spectroscopy experiments. For the shot-to-shot measurements an acousto-optical modulator pulse shaper was implemented, which can arbitrarily shape pulses at 100 kHz repetition rate, and we detect full spectra with a line camera at 100 kHz. The increase in repetition rate and the ability to measure shot-to-shot also leads to a better signal-to noise ratio as a larger number of averages can be

measured in the same time frame as previous measurements. As an exemplary measurement, we show data taken using the new development of fifth-order transient absorption spectroscopy. This method can be used to analyze the exciton dynamics in J-type coupled merocyanine dye films, which are interesting in view of their optoelectronic properties.[1]

[1] A. Liess, et al., Adv. Funct. Mater. 2019, 29, 1805058.

MO 14.5 Thu 12:00 F102

High-resolution rapid-scanning two-dimensional electronic spectroscopy — ●NICOLAI GÖLZ, FRIEDEMANN LANDMESSER, DANIEL UHL, FRANK STIENKEMEIER, and LUKAS BRUDER — Institute of Physics, University of Freiburg, Hermann-Herder-Str. 3, 79104 Freiburg, Germany

Two-dimensional electronic spectroscopy (2DES) is an effective ultrafast spectroscopic technique to study dynamics of matter with a high spectro-temporal resolution. Extending the method to weakly perturbed molecular and cluster species in the gas phase permits very high spectral resolution [1,2]. However, in this case, the attainable resolution is limited by the acquisition time of corresponding long delay scans. To solve this problem, we have implemented a rapid-scanning method developed by the Ogilvie group [3] which reduces the acquisition time by up to 2 orders of magnitude. First results will be presented.

[1] L. Bruder et al., Nat Commun 9, 4823 (2018)

[2] U. Bangert et al., Nature Communications 13:1, 3350 (2022)

[3] D. Agathangelou et al., J. Chem. Phys.155, 094201 (2021)

MO 14.6 Thu 12:15 F102

Coherent multidimensional spectroscopy of molecular and cluster beam samples — ●YILIN LI, ARNE MORLOK, ULRICH BANGERT, DANIEL UHL, FRANK STIENKEMEIER, and LUKAS BRUDER — Institute of Physics, University of Freiburg, Germany

Coherent multidimensional spectroscopy is a versatile technique enabling further insights into intra- and inter-molecular couplings on ultrashort time scales. We have extended the method to molecular nanosystems prepared in the gas phase [1,2]. Recently we started analysing 2D beating maps to obtain information about the electronic and vibrational coherences in the systems, which are otherwise covered by linebroadening mechanisms. First results will be presented.

[1] L. Bruder et al., Nat Commun 9, 4823 (2018)

[2] U. Bangert, F. Stienkemeier, L. Bruder, Nat Commun 13, 3350 (2022)

MO 14.7 Thu 12:30 F102

Simplifying the Analysis of Transient Absorption Data by Polarization Control — ●YI XU¹, LARS MEWES¹, ERLING THYRHAUG¹, HEINZ LANGHALS², and JÜRGEN HAUER¹ — ¹Dynamical Spectroscopy, Department of Chemistry, Technical University of Munich, 85748 Garching, Germany — ²Department of Chemistry, Ludwig-Maximilians-Universität München, 81377 Munich, Germany

Ultrafast energy transfer in donor-acceptor (D-A) systems with orthogonal transition dipole moments (TDMs) is of fundamental interest due to its incompatibility with Förster theory. An in-depth theoretical treatment calls for specific experimental tools, which we provide by polarization-controlled transient absorption (TA) spectroscopy with broadband detection. We isolate pure donor and acceptor parts of the total signal. This provides a strategy to greatly reduce spectral congestion in complex systems. The polarization-associated spectra can be isolated to the contributions either parallel (S_z) or orthogonal (S_y) to the excitation TDM. The derived expressions read $S_z = 3 \cdot S_{MA} \cdot V_z(r(\lambda, t), \beta)$ and $S_y = 3 \cdot S_{MA} \cdot V_y(r(\lambda, t), \beta)$, where S_{MA} represents the magic angle spectra. The corresponding unit vectors $V_z(r(\lambda, t), \beta)$ and $V_y(r(\lambda, t), \beta)$ are both functions of the time resolved anisotropy $r(\lambda, t)$ and the angle β stands for differences between the TDMs defining the first and last light-matter interaction in a TA-sequence. We find that $\beta \approx 30^\circ$ is an optimal choice to disentangle the D and A-signals. This proves the non-orthogonality within the "orthogonal" D-A system.

MO 14.8 Thu 12:45 F102

Probing Nonadiabatic Dynamics with Attosecond Pulse Trains and soft X-ray Raman Spectroscopy — ●LORENZO RESTAINO, DEEPENDRA JADOUN, and MARKUS KOWALEWSKI — Department of Physics, Stockholm University, Albanova University Centre, SE-106 91 Stockholm, Sweden

Ultrafast electronic coherences, generated by a photoexcited wave

packet passing through a conical intersection, can be detected by linear off-resonant X-ray Raman probes. A hybrid femtosecond or attosecond probe pulse can be employed to generate a Raman spectrum that maps the energy difference between the involved electronic states. We investigate how attosecond pulse trains perform as probe pulses in the framework of this spectroscopic technique, instead of single Gaussian

pulses. We explore different schemes for the probe pulse, as well as the impact of parameters of the pulse trains on the signals. We use two different model systems, representing molecules of different symmetry, and quantum dynamics simulations to study the difference in the spectra. The results suggest that such pulse trains are well suited to capture the key features associated with the electronic coherence.

MO 15: Rotational- and Vibrational-resolution Spectroscopy

Time: Thursday 11:00–13:00

Location: F142

Invited Talk

MO 15.1 Thu 11:00 F142

Excited state dipole moments from rotationally resolved Stark spectroscopy — ●MICHAEL SCHMITT, MATTHIAS ZAJONZ, and MARIE-LUISE HEBESTREIT — Heinrich-Heine-Universität Düsseldorf, Institut für Physikalische Chemie I, Universitätsstraße 26.43.02

Rotationally resolved electronic Stark spectroscopy is a versatile tool for the accurate determination of rotational constants of molecules in both electronic states, connected by the electronic transition, centrifugal distortion constants, barriers to hindered internal motions, transition dipole moments, and the permanent dipole moments of both states. The latter provide an easy access to the electronic nature of the excited state, because they differ in size and in direction for different electronic states. The most reliable values for dipole moments of ground and electronically excited states are obtained from gas phase electronic Stark experiments, since the dependence of the frequency shift of individual rovibronic lines from the electric field strength yields immediately the dipole moment in ground and excited state. A variation of the solvatochromic effect will be presented, namely the thermochromic effect, which is able to yield dipole moment changes upon electronic excitation of molecules in solution that are largely independent of the solvent used, to close the gap between gas phase dipole moment changes and those obtained in solutions. Although used for many years in organic chemistry, the Lippert-Mataga theory has severe deficiencies, which will be highlighted in the contribution. We will present a thorough analysis of the photo-physics of the six different positional isomers of cyanoindole in the gas phase and in solution.

MO 15.2 Thu 11:30 F142

High resolution continuous wave spectroscopy on the $A^2\Sigma^+ \leftarrow X^2\Pi_{3/2}$ transition in nitric oxide — ●PHILIPP NEUFELD^{1,3}, PATRICK KASPAR^{1,3}, FABIAN MUNKES^{1,3}, ALEXANDER TRACHTMANN^{1,3}, YANNICK SCHELLANDER^{2,3}, ROBERT LÖW^{1,3}, TILMAN PFAU^{1,3}, and HARALD KÜBLER^{1,3} — ¹Physikalisches Institut, Universität Stuttgart, Pfaffenwaldring 57, 70569 Stuttgart — ²Institut für Großflächige Mikroelektronik, Universität Stuttgart, Allmandring 3b, 70569 Stuttgart — ³Center for Integrated Quantum Science and Technology (IQST), Universität Stuttgart

Within the scope of the development of a new kind of gas sensor [1,2], we employ Doppler-free saturated absorption spectroscopy on the $A^2\Sigma^+ \leftarrow X^2\Pi_{3/2}$ transition in nitric oxide (NO) for different total angular momenta J on the P_{12} branch. Spectroscopy is performed in continuous wave operation at 226 nm in a 50 cm long through-flow cell. Via phase sensitive detection by a lock-in amplifier the hyperfine structure of the $X^2\Pi_{3/2}$ state of NO is partially resolved. The data is compared to previous measurements, showing good agreement. Investigation of the dependence of the spectroscopic feature on power and pressure, should yield hyperfine constants, natural transition linewidth and the collisional cross-section between NO molecules. [1] P. Kaspar et. al., OSA Optical Sensors and Sensing, 19-23 July, 2021 [2] J. Schmidt et. al., Appl. Phys. Lett. 113, 011113 (2018)

MO 15.3 Thu 11:45 F142

Determination of the ortho-to-para ratio of H_3^+ in a cryogenic ion trap — PHILIPP SCHMID, CARSTEN CZAPCZYK, THOMAS SALOMON, OSKAR ASVANY, and ●STEPHAN SCHLEMMER — I. Physikalisches Institut der Universität zu Köln, Köln, Germany

A clean sample of ortho (o) and para (p) H_3^+ is prepared in a cryogenic 22-pole ion trap by removing one nuclear spin species. This isolation is reached on time scales below one second by a selective excitation of the ν_2 vibrational mode of H_3^+ addressing a rotational state associated with one of the two species. In subsequent collisions of the excited H_3^+ with He the vibration-to-translation (V-T) energy transfer produces fast H_3^+ of the addressed spin configuration which then leaves the trap

via a small electrostatic barrier. Following this protocol the o/p-ratio of H_3^+ coming from the ion source is determined to be 1:1, as expected. In the presence of normal hydrogen, n- H_2 , in the cryogenic ion trap the o/p-ratio of H_3^+ reaches a stationary value close to 1:3 which is rationalized by the ordering of the lowest energy states of o- H_3^+ and p- H_3^+ . This work shows that nuclear spin specific and also structural isomer specific preparation and analysis is now possible in cryogenic ion traps. With this aid action spectra of isomer mixtures can be disentangled and it will become possible to determine isomer branching ratios of chemical reactions, as well as to study state-specific reactions like for the fundamental $H_3^+ + H_2$ collision system considered in this work.

MO 15.4 Thu 12:00 F142

Leak-out spectroscopy: a new, universal action spectroscopy method — ●PHILIPP C. SCHMID, THOMAS SALOMON, SVEN THORWIRTH, OSKAR ASVANY, and STEPHAN SCHLEMMER — I. Physikalisches Institut, Universität zu Köln, Köln, Germany

Rotational-resolved vibrational spectra of molecular ions are recorded using action spectroscopy techniques in cryogenic ion traps [1]. Thereby the spectrum is identified as a change of the ion mass by fragmentation upon photon absorption, pre-dissociation of tagged ions or via laser induced chemical reactions. Although all these techniques have advanced significantly, action spectroscopy cannot be applied to many ions when the aforementioned methods fail to work. Here, we present a new method of action spectroscopy, called leak-out spectroscopy (LOS) [2]. LOS does not rely on a change in the mass of the ion, but on the transfer of internal energy of an excited ion to its translation energy upon the collision with a neutral partner. This allows the fast ions to escape the trap potentials and thus the spectrum can be recorded by detecting the ions leaving the trap. Based on this principle, LOS is intrinsically background free and suitable for virtually any ion. In recent measurements we used LOS to record the ro-vibrational resolved ν_1 C-H stretching vibration spectrum of l- C_3H^+ . [1] B.A. McGuire et al., Nature Review Physics 2(2020)402. [2] P.C. Schmid et al., J. Phys. Chem. A 126(2022)8111.

MO 15.5 Thu 12:15 F142

Spectroscopic studies of acylium- and thioacylium ions — ●SVEN THORWIRTH¹, OSKAR ASVANY¹, THOMAS SALOMON¹, MARCEL BAST¹, PHILIPP C. SCHMID¹, MICHAEL E. HARDING², IGOR SAVIC³, JOSÉ L. DOMÉNECH⁴, and STEPHAN SCHLEMMER¹ — ¹I. Physikalisches Institut, Universität zu Köln, Köln Germany — ²Institut für Nanotechnologie, Karlsruher Institut für Technologie (KIT), Karlsruhe, Germany — ³Department of Physics, Faculty of Sciences, University of Novi Sad, Novi Sad, Serbia — ⁴Instituto de Estructura de la Materia (IEM-CSIC), Madrid, Spain

Acylium- and thioacylium ions, R-CO⁺ and R-CS⁺, are classes of astrochemically relevant molecular ions that have received relatively little attention from molecular spectroscopy so far. Triggered by recent laboratory and astronomical observations of the polyatomic HC₃O⁺, HC₃S⁺, and CH₃CO⁺ species (see, Thorwirth et al. 2020, Cernicharo et al. 2021 and references therein) our laboratory investigations have now been extended towards other selected (thio)acylium species. Here, we would like to present a status report on our studies performed using various action spectroscopy schemes in combination with state-of-the-art ion traps.

Thorwirth, S., Harding, M. E., Asvany, O. et al. 2020, Mol. Phys. 118, e1776409
Cernicharo, J., Cabezas, C., Bailleux, S. et al. 2021, Astron. Astrophys. 646, L7

MO 15.6 Thu 12:30 F142

Luminescent and excited state properties of bimetallic coinage metal NHC-complexes — •DANIEL MARHÖFER¹, PIT BODEN¹, SOPHIE STEIGER¹, CHRISTOPH KAUB², PETER ROESKY², and GEREON NIEDNER-SCHATTEBURG¹ — ¹Dept. of Chemistry, TUK, Erwin-Schrödinger-Str. 52-54, 67663 Kaiserslautern — ²Institute of Inorganic Chemistry, KIT, Engesserstr. 15, 76131 Karlsruhe

A series of bimetallic coinage metal complexes containing a specific, bipyridyl substituted, N-heterocyclic carbene ligand was investigated via luminescence spectroscopy as well as step-scan FTIR spectroscopy. The series contains the monometallic and the heterobimetallic gold complexes as well as the homobimetallic silver and copper compounds. The luminescence measurements were performed both in potassium bromide matrix and in (conditionally frozen) ethanoic solution. The emission lifetimes were determined via time-correlated single photon counting in the temperature range between 5 K and 290 K. The excited state structures were studied by electronic excitation by a pulsed UV laser followed by step-scan FTIR probing, allowing the determination of the IR absorption of the electronically excited molecules. The obtained excited state spectra were then compared to both the ground state vibrational spectrum as well as the quantum-chemically calculated lowest energy singlet and triplet states' vibrational spectra. A pronounced dependence of the excited state IR absorption, the emis-

sion colour and the excited state lifetimes on the metal centers could be observed and finally assigned to cooperative effects in specific metal combinations.

MO 15.7 Thu 12:45 F142

2DIR Spectroscopy of an Organic Azide Model — •CLAUDIA GRÄVE, JÖRG LINDNER, and PETER VÖHRINGER — Clausius-Institut für Physikalische und Theoretische Chemie, Rheinische Friedrich-Wilhelms-Universität Bonn, Wegelerstraße 12, 53115 Bonn, Germany

The photoinduced dynamics of azides is of great importance in organic chemistry but not yet understood, even at a qualitative level. We are currently utilizing ultrafast spectroscopy to address these dynamics.

Here, we report on first results from femtosecond two-dimensional infrared (2DIR) spectroscopy of 1-cyano-2-(4-azidophenyl)-acetylene to understand vibrational energy transfer (VET) to the solvent and intramolecular vibrational energy redistribution (IVR). Our 2DIR data show that IVR from the azide moiety through the phenyl ring onto the para-positioned cyano-acetylene group occurs on a time scale of 3.5 ps. In contrast, VET in chloroform solution requires 20 ps. Finally, a coherent vibrational quantum beat can be detected, which dephases within roughly 1 ps.

MO 16: Atomic Clusters (joint session A/MO)

Time: Thursday 11:00–13:00

Location: F107

Invited Talk

MO 16.1 Thu 11:00 F107

Efficient and accurate simulation of wide-angle single-shot scattering — •PAUL TUEMMLER, BJÖRN KRUSE, CHRISTIAN PELTZ, and THOMAS FENNEL — Institute for Physics, University of Rostock, Albert-Einstein-Str. 23-24, D-18059 Rostock, Germany

In recent years coherent diffractive imaging has been established as a powerful method for the structural investigation of unsupported nanoparticles. A large number of studies have been successfully performed in the small angle regime, where the recorded scattering image is directly connected to the target's density projection along the optical axis. An established technique to invert the scattering image is the well-known phase retrieval algorithm. Single-shot 3d information only becomes available when scattering signal can be recorded at wide scattering angles, which typically requires wavelengths of several object diameters. However, in this scattering regime a direct inversion via phase retrieval is no longer possible and iterative forward fitting schemes have to be applied. These schemes require many iterations and therefore heavily rely on an efficient method to calculate scattering images. Unfortunately, optical parameters in the long wavelength regime are typically quite far from vacuum parameters, such that absorption and multiple scattering effects become important. So far, available methods either lack the necessary accuracy (e.g. MSFT methods) or the numerical efficiency (e.g. FDTD).

Here we present a rigorous split step method that retains the efficiency of multislice methods, while yielding accuracy comparable to Mie and FDTD methods.

MO 16.2 Thu 11:30 F107

3D Femtosecond Snapshots of Silver Nanoclusters — •ALESSANDRO COLOMBO for the FLASH-SilverClusters-Collaboration — Laboratory for Solid State Physics, ETH Zurich, 8093 Zurich, Switzerland

Thanks to X-ray Free-Electron Lasers, Coherent Diffraction Imaging (CDI) allows femtosecond snapshots of matter at the nanoscale. When the diffracted light is recorded up to a sufficiently wide scattering angle, a single two-dimensional diffraction pattern carries 3D structural information on the sample. However, the non-trivial mathematical link between the sample's 3D shape and the 2D diffraction pattern renders 3D single-shot CDI a scientific challenge. Here we present a reconstruction method [1] that unveils the intriguing three-dimensional architectures of free-flying silver nanoclusters, retrieved from single wide-angle scattering images acquired at the soft X-ray Free-Electron Laser FLASH in Hamburg. The retrieved shapes of the silver clusters show satisfactory reliability and consistency, also revealing new structural motifs. Thanks to its great versatility, the method is then further extended to nanocrystals agglomerates, allowing for the first time a direct 3D insight into their growth process and surprising structures. This work

represents a strong proof of concept for this imaging approach, raising the bar of the capabilities of 3D coherent diffraction imaging from single shot.

[1] Colombo, A., et al. arXiv:2208.04044 (2022).

MO 16.3 Thu 11:45 F107

Quenching of photon emission in heterogeneous noble gas clusters — •LUTZ MARDER, CATMARNA KÜSTNER-WETEKAM, NILS KIEFER, DANA BLOSS, ANDRÉ KNIE, ARNO EHRESMANN, and ANDREAS HANS — Institut für Physik und CINSaT, Universität Kassel, Heinrich-Plett-Str. 40, 34132 Kassel, Germany

Noble gas clusters represent prototype systems suited for the investigation of fundamental atomic and molecular processes. The Van-der-Waals bonds enable new relaxation pathways not available in isolated systems. Many of these have been studied during the recent years, often using coincidence measurement techniques.

We present our state-of-the-art experiment where both electrons and photons were detected in coincidence, which allows for investigation of multi-particle decay pathways after ionization with synchrotron radiation. The results show that the addition of a heavier noble gas to clusters of a lighter noble gas strongly alters the emission by the opening of faster ionizing decay channels compared to the radiative decay.

MO 16.4 Thu 12:00 F107

Electron-Photon Coincidence Measurements at Synchrotron Facilities with Arbitrary Filling Pattern — •JOHANNES VIEHMANN, ANDREAS HANS, CHRISTIAN OZGA, and ARNO EHRESMANN — Institut für Physik und CINSaT, Universität Kassel, Heinrich-Plett-Str. 40, 34132 Kassel, Germany

Coincidence measurements are an important experimental tool in atomic or molecular physics. Our group has used electron-photon coincidence measurements to investigate rare gas clusters after synchrotron irradiation. The clusters exhibit a plethora of local and non-local electronic relaxation processes after core hole excitation. Most of these pathways produce free electrons and/ or photons. In order to distinguish signals of certain pathways from the general background, coincidence measurements are very useful.

So far, the combination of coincidence techniques with synchrotron radiation has mainly been restricted to the single bunch operation mode of the synchrotron facility due to difficulties in data acquisition. Here, we present a solution to combine coincidence measurements with multi-bunch operation modes and an example of using such technique to study rare gas clusters.

MO 16.5 Thu 12:15 F107

Quantum nanofriction in trapped ion chains with a topological defect — •LARS TIMM¹, LUCA A. RÜFFERT², HENDRIK WEIMER^{1,3}, LUIS SANTOS¹, and TANJA E. MEHLSTÄUBLER^{2,4} —

¹Institut für Theoretische Physik, Appelstr. 2, 30167 Hannover — ²Physikalisch-Technische Bundesanstalt, Bundesallee 100, 38116 Braunschweig — ³Institut für Theoretische Physik, Hardenbergstr. 36, 10623 Berlin — ⁴Institut für Quantenoptik, Welfengarten 1, 30167 Hannover

After an introduction into the fundamental properties of the Frenkel-Kontorova model, one of the paradigmatic models of nanofriction, I will present the observation of a sliding to pinned transition with the help of a topological defect inside a two-dimensional self-assembled ion crystal. Subsequently, I shortly introduce one major consequence of this so-called Aubry transition, i.e. the localization of energy in the pinned phase of the defect. In the main part of my talk I discuss the quantized version of the Frenkel-Kontorova model and the consequences for the Aubry transition inside an ion crystal. As for that matter, we make use of a simple single particle formalism treating the defect as a quasiparticle, which captures the important dynamics of the defect close to the Aubry transition. This convenient approach gives access to its quantum properties revealing its quantum tunneling on a micron length-scale in a range of trap configurations and lets us identify a transition into a quasi-classical regime away from the transition point. Lastly, we give estimates for the temperature requirements and strategies to observe these effects in an experiment.

MO 16.6 Thu 12:30 F107

Experimental studies on Interatomic Coulombic Decay after inner-shell ionization of heterogeneous rare gas clusters — ●CATMARN KÜSTNER-WETEKAM¹, LUTZ MARDER¹, DANA BLOSS¹, NILS KIEFER¹, ÜWE HERGENHAHN², ARNO EHRESMANN¹, PRÉMYSL KOLOREŇČ³, and ANDREAS HANS¹ — ¹Institut für Physik und CIN-SaT, Universität Kassel, Heinrich-Plett-Str. 40, 34132 Kassel, Germany — ²Fritz-Haber-Institut der Max-Planck-Gesellschaft, Faradayweg 4-6, 14195 Berlin, Germany — ³Institute of Theoretical Physics, Charles University, V Holesovickach 2, 180 00 Prague, Czech Republic

Non-local decay mechanisms play an important role in the relaxation of electronic vacancies in dense media such as biological samples. To explore these mechanisms in a less complex environment, rare gas clusters can be used as a prototype system for experiments. The use of multi-coincidence spectroscopy enables the detection of core-level Interatomic Coulombic Decay (ICD), which is a comparatively weak process in relation to the local Auger decay followed by Radiative Charge Transfer (RCT). Here, we present the observation of changes in ICD efficiency when going from homogeneous Ar and Kr clusters to heterogeneous ArKr clusters and thereby introducing a different environment to the excited atom in the respective cluster.

MO 16.7 Thu 12:45 F107

Electron-photon-coincidence investigations on neighbor induced photoelectron recapture in argon clusters — ●NILS KIEFER, CAROLIN HONISCH, CATMARN KÜSTNER-WETEKAM, NIKLAS GOLCHERT, ARNO EHRESMANN, and ANDREAS HANS — Institute of Physics, University of Kassel, Kassel, Germany

Noble gas clusters are an ideal prototype system for fundamental research on atomic and molecular processes. The Van-der-Waals-bound atoms create an environment, which enables further decay pathways and scattering effects. These have been studied already with high resolution electron spectroscopy and multi-electron-coincidence spectroscopy. With use of a state-of-the-art experimental set-up, which allows coincident electron and photon detection, radiative and electronic processes after excitation of clusters with synchrotron radiation can be directly observed. Here, we present the results of electron-photon-coincidence measurements of a recent experiment on argon clusters. Here a slow photoelectron after 2p ionization is expected to be scattered on neighboring atoms in a "Bremsstrahlung"-like process. Thus, the scattered electron can be recaptured to the Ion and further decay in a resonant Auger-like process.

MO 17: Quantum Optics and Quantum Information with Rigid Rotors (joint session MO/Q/QI)

Time: Thursday 14:30–16:30

Location: F102

MO 17.1 Thu 14:30 F102

Cooling and control of the translational and rotational motion of a nano rotor — ●PETER BARKER¹, ANTONIO PONTIN¹, MARKO TOROS², HAYDEN FU¹, TANIA MONTEIRO¹, JONATHAN GOSLING¹, and MARKUS RADEMACHER¹ — ¹University College London, UK — ²University of Glasgow, Glasgow, UK

There has been significant interest in controlling the motional degrees of isolated, single nanoparticles, trapped within optical fields in high vacuum. They are seen as ideal candidates for exploring the limits of quantum mechanics in a new mass regime while they are also massive enough to be considered for future laboratory tests of the quantum nature of gravity. In this talk I will report on the control and cooling of all translational and rotational degrees of freedom of a nanoparticle trapped in an optical tweezer using cooling via coherent elliptic scattering where translational temperatures in the 100 μ K range were reached, while temperatures as low as 5 μ mK were attained in the librational degrees of freedom. I will also outline nanoparticle characterisation techniques based on the control and measurement of the librational and translational motion. This work opens up future applications in quantum science and the characterisation of single isolated nanoparticles free of interference from a substrate.

MO 17.2 Thu 14:45 F102

Polarization control of optically levitated nanoparticles — ●YANHUI HU, JAMES SABIN, MUDASSAR RASHID, and JAMES MILLEN — Department of Physics, King's College London, Strand, London

The optical control of anisotropic particles opens up applications in torque sensing and the study of rotational quantum mechanics. The angular modes of a levitated particle are markedly different from the linear modes, and new tools are required to achieve full control. In the Levitated Nanophysics Group at King's College London we work with nanofabricated silicon nanorods, which allow enhanced control over all degrees-of-freedom. We control the rotation of the nanorods through a recently discovered method for generating transverse optical vortices, which can be used to exert a large torque on an array of levitated nanoparticles.

We also present a method to simultaneously cool all of the linear and angular modes of levitated, anisotropic particles, without the necessity for a delicate optical cavity.

MO 17.3 Thu 15:00 F102

Surface-induced decoherence and heating of charged rigid rotors — ●LUKAS MARTINETZ, KLAUS HORNBERGER, and BENJAMIN A. STICKLER — University of Duisburg-Essen

Levitating charged particles in ultrahigh vacuum provides a preeminent platform for quantum information processing, for quantum-enhanced force and torque sensing, for probing physics beyond the standard model, and for high-mass tests of the quantum superposition principle. Existing setups, ranging from single atomic ions to ion chains and crystals to charged molecules and nanoparticles, are crucially impacted by fluctuating electric fields emanating from nearby electrodes used to control the motion. In this article, we provide a theoretical toolbox for describing the rotational and translational quantum dynamics of charged nano- to microscale objects near metallic and dielectric surfaces, as characterized by macroscopic dielectric response functions. The resulting quantum master equations describe the coherent surface-particle interaction due to image charges and Casimir-Polder potentials as well as surface-induced decoherence and heating with the experimentally observed frequency and distance scaling. We explicitly evaluate the master equations for relevant setups, thereby providing the framework for describing and mitigating surface-induced decoherence as required in future quantum technological applications.

MO 17.4 Thu 15:15 F102

Decoherence-Free Rotational Degrees of Freedom for Quantum Applications — ●JULEN S. PEDERNALES, FRANCESCO COSCO, and MARTIN B. PLENIO — Institut für Theoretische Physik und IQST, Albert-Einstein-Allee 11, Universität Ulm, D-89081 Ulm, Germany

I will describe the use of spherical t-designs for the systematic construction of solids whose rotational degrees of freedom can be made robust to decoherence due to external fluctuating fields while simultaneously

retaining their sensitivity to signals of interest. Specifically, the ratio of the signal phase accumulation rate from a nearby source to the de-coherence rate caused by fluctuating fields from more distant sources can be incremented to any desired level by using increasingly complex shapes. This allows for the generation of long-lived macroscopic quantum superpositions of rotational degrees of freedom and the robust generation of entanglement between two or more such solids with applications in robust quantum sensing and precision metrology as well as quantum registers.

[1] J. S. Pedernales, F. Cosco, and M. B. Plenio, Phys. Rev. Lett. 125, 090501 (2020).

MO 17.5 Thu 15:30 F102

Group report: Precision spectroscopy and quantum information with trapped molecules — ●BRANDON FUREY, STEFAN WALSER, ZHENLIN WU, GUANQUN MU, RENE NARDI, and PHILIPP SCHINDLER — Institut für Experimentalphysik, Universität Innsbruck, Österreich

The quantum molecules group at the University of Innsbruck utilizes a range of innovative advances in molecular spectroscopy and quantum logic spectroscopy (QLS) to study molecular rovibrational structure and explore quantum information processing with trapped molecules. The efforts of our group are divided into three projects. The first is pump-probe recoil spectroscopy, where we measure the rovibrational population dynamics excited by a pump pulse by mapping them to the electronic state of an atomic ion via QLS. The second project investigates state-dependent force spectroscopy, where an optical tweezer generates a state-dependent force on a trapped molecule. Our third project is demonstrating superpositions of rotational states in a diatomic molecular ion built using stimulated Raman transitions driven by two beams from an optical frequency comb. This could pave the way for using quantum error correction to realize the use of trapped molecules for quantum information or memory. We are interested in creating the rotational superposition states that form the codewords of a truncated $Z_3 \subset Z_6$ linear rotor code. In order to demonstrate ultrafast light-matter interaction in our system, we have measured the photodissociation spectrum of CaOH^+ using an optical parametric amplifier.

MO 17.6 Thu 15:45 F102

From the rotation of a planar rigid rotor in electric fields to the semifinite-gap structure of an optical superlattice — ●MARJAN MIRAHMADI¹, BRETSLAV FRIEDRICH¹, BURKHARD SCHMIDT², and JESÚS PÉREZ-RÍOS^{1,3,4} — ¹Fritz-Haber-Institut der Max-Planck-Gesellschaft, Berlin, Germany — ²Weierstraß-Institut, Berlin, Germany — ³Department of Physics, Stony Brook University, NY, USA — ⁴Institute for Advanced Computational Science, Stony Brook University, NY, USA

We show that two seemingly unrelated problems – the trapping of an atom in a one-dimensional optical superlattice (OSL) formed by the interference of optical lattices whose spatial periods differ by a factor of two, and the libration of a polarizable planar rotor (PR) in combined electric and optical fields – have isomorphic Hamiltonians. It is possible to establish a map between the translations of atoms in the former system and the rotations of the rotor due to the

coupling of its permanent and induced electric dipole moments to the external fields. The latter system belongs to the class of conditionally quasi-exactly solvable problems in quantum mechanics and exhibits intriguing spectral properties. We make use of our findings to explain the *semifinite-gap* band structure of the OSL. This band structure follows from the eigenenergies obtained as solutions of the Whittaker-Hill equation and their genuine and avoided crossings. Furthermore, the mapping makes it possible to establish correspondence between concepts, such as localization on the one hand and orientation/alignment on the other.

MO 17.7 Thu 16:00 F102

Experimental advances in the quest for perfect enantiomer-specific state control of cold molecules — ●JUHYEON LEE¹, JOHANNES BISCHOFF¹, ALICIA. O. HERNANDEZ-CASTILLO², BORIS SARTAKOV¹, GERARD MEIJER¹, and SANDRA EIBENBERGER-ARIAS¹ — ¹Fritz Haber Institute of the Max Planck Society, Berlin, Germany — ²Harvey Mudd College, Claremont, California, USA

Enantiomer-specific state transfer (ESST) was recently developed using tailored microwave fields [1]. This technique enables the population or depopulation of a rotational state of a chosen enantiomer, providing a way of quantum-controlled chiral separation. Recently, we have explored spectroscopic schemes to overcome previous limitations in the transfer efficiency of ESST: thermal population of the rotational levels and M_J degeneracy [2]. We improved the transfer efficiency up to 50%, and quantitatively studied ESST for the first time [3]. The experimental ESST efficiency was $\sim 20\%$ lower than theoretically expected. We attribute this partially to imperfections in the microwave polarizations and their respective orthogonality. We show a method to experimentally determine the polarization of microwave fields in-situ by quantitative analysis of molecular Rabi oscillations.

[1] S. Eibenberger, et al., Phys. Rev. Lett. 118, 123002 (2017)

[2] M. Leibscher, et al., Commun. Phys. 5, 1 (2022).

[3] J. H. Lee, et al., Phys. Rev. Lett. 128, 173001 (2022)

MO 17.8 Thu 16:15 F102

Photoelectron circular dichroism in rotationally excited mixtures — ●ALEXANDER BLECH¹, LOREN GREENMAN², REINHARD DÖRNER³, and CHRISTIANE P. KOCH¹ — ¹Fachbereich Physik, Freie Universität Berlin, Berlin, Germany — ²Department of Physics, Kansas State University, Manhattan, KS, USA — ³Institut für Kernphysik, Goethe-Universität, Frankfurt am Main, Germany

Gas phase experiments with chiral molecules may be carried out with randomly oriented molecules because there exist enantiomer-sensitive observables that survive orientational averaging. The strength of these observables is directly related to the enantiomeric excess and vanishes in the limit of a racemic mixture. Here we turn the perspective around and investigate whether it is possible to detect chiral signatures from racemic, but rotationally excited mixtures. We focus on photoelectron circular dichroism (PECD), which is the forward-backward asymmetry in the photoelectron angular distributions of chiral molecules upon ionization with circularly polarized light. Based on an analysis of the electric dipole response in rotationally excited molecular ensembles, we show that PECD can be observed in racemic mixtures by breaking the isotropy of the orientational distribution.

MO 18: Cluster and Experimental Techniques (joint session MO/A)

Time: Thursday 14:30–16:30

Location: F142

MO 18.1 Thu 14:30 F142

Characterization of a simple supersonic expansion source for small molecular ions — ●LUKAS BERGER¹, AIGARS ZNOTINS¹, FLORIAN GRUSSIE¹, DAMIAN MÜLL¹, FELIX NUSSLIN¹, ARNAUD DOCHAIN², JOFFREY FRÉREUX², XAVIER URBAIN², and HOLGER KRECKEL¹ — ¹Max-Planck-Institut für Kernphysik, Saupfercheckweg 1, 69117 Heidelberg, Germany — ²Institute of Condensed Matter and Nanosciences, Université Catholique de Louvain, Louvain-la-Neuve, B-1348 Belgium

The Cryogenic Storage Ring (CSR) [1] at the MPI for Nuclear Physics allows for the storage of molecular ions of almost arbitrary mass at extreme vacuum (residual gas densities on the order of 1000 cm⁻³) and at low temperature ($T < 5\text{K}$). In this environment, small infrared-active molecular ions will cool to their lowest rotational states within

minutes. However, some astrophysically relevant molecular ions lack a permanent dipole moment (e.g.: H_2^+ , H_3^+ , H_3O^+) and have to be produced in cold ion sources prior to injection, as they do not cool on experimentally accessible time scales. Here we present the design of a simple supersonic expansion source based on a commercial pulsed valve and an electric discharge. It allows for the production of intense pulses of small molecular ions. We use high-resolution photodissociation spectroscopy of N_2O^+ , employing the STARGATE setup [2] at the Université Catholique de Louvain, to characterize the internal excitation of the molecular ions and extract their rotational temperature.

[1] Von Hahn et al., Rev. Sci. Instrum. 87, 063115 (2016)

[2] Bejjani et al., Rev. Sci. Instrum. 92, 033307 (2021)

MO 18.2 Thu 14:45 F142

OH^+He as simple example to illustrate fundamental con-

cepts of intermolecular interactions. — ●NIMA-NOAH NAHVI, DAVID MÜLLER, and OTTO DOPFER — Institut für Optik und Atomare Physik, Technische Universität Berlin, Hardenbergstr. 36, 10623 Berlin, Germany

The OH⁺He dimer is a simple cluster for which many concepts of intermolecular interactions can be demonstrated. Nowadays, OH⁺He can be understood quite well, which makes it an interesting and encompassing example for educational purposes.

OH⁺He_n ($n \leq 6$) clusters are grown inside a cryogenic 22-pole ion trap and detected using a reflectron mass spectrometer. The resulting mass spectra are explained with cluster structures determined by CCSD(T) calculations using CFOUR. We investigate and explain certain features of OH⁺He_n to illustrate concepts like solvation shells, potential energy functions, binding energies and vibrational frequencies, charge- and dipole-induced interactions, dispersion contribution, complete basis set limits and BSSE corrections. The results will be compared to those of H₂O⁺He_n and H₃O⁺He_n [1] to establish the effect of the number of protons on the interaction potential and cluster growth.

[1] Müller and Dopfer, Phys. Chem. Chem. Phys., 2022, 24, 11222-11233. <https://doi.org/10.1039/D2CP01192A>

MO 18.3 Thu 15:00 F142

Investigation of the homogeneous line width of organic molecules attached to rare-gas clusters — ●ARNE MORLOK, ULRICH BANGERT, LUKAS BRUDER, YLIN LI, and FRANK STIENKEMEIER — Institute of Physics, University of Freiburg, Hermann-Herder-Str. 3, 79104 Freiburg, Germany

Two-dimensional spectroscopy (2DES) is a powerful method to study dynamics of atoms and molecules with a high spectro-temporal resolution. In our group, we apply the technique to doped cluster beams, which act as miniature cryostats for different organic molecules [1,2]. Previous 2DES experiments on phthalocyanine (Pc) on neon clusters allowed resolution of the homogeneous linewidth of Pc and distinguished fluorescence from the C12 and C13 isotope [3]. We extended this investigation in varying the species of the spectroscopic matrix, hence the clusters, and the probed organic molecules. First results are presented, which suggest differences in the homogeneous linewidth and coherence times depending on the coupling between dopant and cluster environment.

- [1] L. Bruder et al., Nat. Commun. 9 4823 (2018).
 [2] L. Bruder et al., J. Phys. B: At. Mol. Opt. Phys. 52 183501 (2019).
 [3] U. Bangert et al., Nat. Commun. 13 3350 (2022).

MO 18.4 Thu 15:15 F142

Experimental cross sections for the uptake of alkyl alcohols by binary HNO₃/H₂O clusters — YIHUI YAN¹, ANDRIY PYSANENKO², KAROLINA FARNIKOVA², EVA PLUHAROVA², MICHAL FARNIK², and ●JOZEF LENGYEL¹ — ¹TUM School of Natural Sciences, Technical University of Munich, Garching, Germany — ²Heyrovsky Institute, Czech Academy of Sciences, Prague, Czech Republic

The uptake of oxidized organic compounds by hydrated acid clusters accounts for a substantial portion of atmospheric aerosol particles. Through joint experimental and computational studies, we investigate the uptake of alkyl alcohols on pre-existing hydrated HNO₃ clusters. In our experiments, the HNO₃/H₂O clusters pass through a chamber filled with a particular gas, the molecules collide with the clusters and can stick to the surface. The efficiency of this process is given by the uptake cross section, which is determined experimentally by a combination of mass spectrometry and velocity measurements in a molecular beam. Our previous experiments have shown that the uptake probabilities for the oxidized molecules are significantly larger than for the corresponding volatile organic compounds (VOCs). This increase is attributed to hydrogen bonds between the molecules and clusters, whereas the interactions of the parent VOCs are weaker and nonspecific. In this study, we examine the effect of different O-H bond positions in the oxidized molecule and different carbon chain length on the pickup probability. To learn the details of the pickup process, all experimental measurements are supplemented by MD simulations.

MO 18.5 Thu 15:30 F142

Femtosecond pump-probe spectroscopy of tetracene in the gas phase and in helium nanodroplets — AUDREY SCOGNAMIGLIO, NICOLAS RENDLER, ●SEBASTIAN HARTWEG, and FRANK STIENKEMEIER — Institute of Physics, University of Freiburg, Germany

Acenes, specifically tetracene and pentacene, are promising candidates for applications in organic photovoltaics since ensembles of these molecules can undergo singlet fission to produce two triplet excitations from a single singlet excitation. This process may thus allow to produce multiple charge carriers from a single absorbed photon. The potential applications motivate the fundamental study of the ultrafast excitation dynamics of these molecules and their aggregates to provide a fundamental understanding of the underlying processes and energetics. We will present a study of femtosecond pump-probe photoelectron spectroscopy and mass spectrometry of the excited state dynamics of tetracene excited to its brightest singlet state using UV photons. We will compare results for effusive tetracene with preliminary data for tetracene and its clusters in superfluid helium nanodroplets. The doping of organic molecules into superfluid helium nanodroplets offers a promising path to extend the study from isolated molecules to clusters of acenes, but also imposes additional challenges.

MO 18.6 Thu 15:45 F142

Plasmon quenching of a single gold nanoparticle in the gas phase — ●BJÖRN BASTIAN, BENJAMIN HOFFMANN, SOPHIA LEIPPE, and KNUT R. ASMIS — Universität Leipzig, Wilhelm-Ostwald-Institut, Linnéstraße 2, D-04103 Leipzig

A split-ring electrode trap design has been optimized to quasi-continuously monitor the mass of single nanoparticles for action spectroscopy, fluorescence spectroscopy and temperature-programmed desorption experiments. The aim is to investigate inherent properties of individual particles and their relation to parameters such as size, shape, temperature, charge state or surface functionalization. One example is a collective electron oscillation of metallic nanoparticles called *localized surface plasmon resonance*. New results are presented that show electronic action spectra of the plasmon resonance of a single 50 nm diameter gold particle and its stepwise quenching by radiative heating.

The particle mass is proportional to its secular frequency in the trap, which is typically monitored by resonant excitation. The resonance frequency is observed as a dip in the intensity of light scattered from the particle when sweeping an excitation frequency. Cryogenic cooling allows to control the adsorption of a messenger gas. Action spectra are obtained by observing the mass loss from desorption of the messenger due to absorption of light in the visible or infrared range.

We present results on plasmon quenching and current progress in implementing infrared action spectroscopy and to better characterize the particle temperature, adsorption and desorption dynamics.

MO 18.7 Thu 16:00 F142

Setup Of A Spectrometer To Detect Raman Optical Activity — ●KLAUS HOFMANN and INGO FISCHER — Universität Würzburg, 97074 Würzburg, Germany

Raman Optical Activity (ROA) is a type of vibrational circular dichroism: chiral samples show different Raman intensities when utilizing circular polarized light. The ROA signal is very sensitive to the molecular geometry and environment of the sample, which can be analyzed by comparing the spectrum with quantum chemical calculations. ROA spectra exhibit high levels of noise and are prone to false signals, since the intensity difference is roughly 0.1% of the corresponding Raman peak.[1]

For this project, a Raman spectrometer was custom-built and modified to detect ROA. A modulation scheme repeatedly converts linear to right and left circular polarized light for excitation. Python was used to automate the experiment, data acquisition and post-processing. The beam path of the spectrometer and its implemented error reduction schemes[2] are shown. Post-processing is used to evaluate the signal. Literature known samples were measured to validate the spectrometer.

- [1] V. Parchaňský, J. Kapitán, P. Bouř, RSC Adv. 2014, 4, 57125.
 [2] W. Hug, Appl. Spectrosc. 2003, 57, 1.

MO 18.8 Thu 16:15 F142

Salt effects on the translocation dynamics of polycationic peptide nucleic acids through a protein nanopore — ●IOANA CEZARA BUCATARU¹, ALINA ASANDEI², LOREDANA MEREUȚA¹, and TUDOR LUCHIAN¹ — ¹Department of Physics, *Alexandru I. Cuza* University, Iasi, Romania. — ²Sciences Department, Interdisciplinary Research Institute, *Alexandru I. Cuza* University, Iasi, Romania

Peptide nucleic acids (PNAs) are synthetic molecular constructs that mimic DNA in structure, but with an uncharged pseudopeptide backbone made of N-(2-aminoethyl)-glycine, having the ability to form Watson-Crick complementary duplexes with regular DNA. Due to its distinctive properties, PNAs displayed considerable potential for appli-

cation in molecular diagnostics and antisense therapies. The addition of different charged sidechains to the neutral PNA structure plays an essential part in addressing solubility-related issues that are linked with the use of these molecules. The single-molecule investigations used here focus on the interactions of different length polyarginine-conjugated PNAs (poly(Arg)-PNAs) with the α -hemolysin (α -HL) nanopore, under an applied transmembrane voltage. The effect of ionic strength

on the translocation kinetics is demonstrated by using different salt concentrations in the recording buffer. Our results indicate that low ionic strength increases the electrophoretic mobility of poly(Arg)-PNA probes as they pass through the nanopore and reduces their volume. The current findings highlight the intricate interplay between conformation and ion environment that influences the inherent flexibility and function of poly(Arg)-functionalized PNAs.

MO 19: Interaction with Strong or Short Laser Pulses III (joint session A/MO)

Time: Thursday 14:30–16:00

Location: F107

Invited Talk

MO 19.1 Thu 14:30 F107

Intra-cavity photoelectron tomography with an intra-cavity velocity-map imaging spectrometer at 100 MHz repetition rate — •JAN-HENDRIK OELMANN¹, TOBIAS HELDT¹, LENNART GUTH¹, JANKO NAUTA^{1,2}, NICK LACKMANN¹, VALENTIN WÖSSNER¹, STEPAN KOKH¹, THOMAS PFEIFER¹, and JOSÉ R. CRESPO LÓPEZ-URRUTIA¹ — ¹Max-Planck-Institut für Kernphysik, Heidelberg, Germany — ²Current address: Department of Physics, Swansea University, Singleton Park, SA2, United Kingdom

In a first experiment, we used intra-cavity velocity-map imaging (VMI) at 100 MHz repetition rate to investigate multi-photon ionization (MPI) events of xenon with high count rates, even at very low intensities [1]. For that, ultrashort pulses from a near-infrared frequency comb laser were amplified in a passive femtosecond enhancement cavity that we now use for extreme-ultraviolet frequency comb generation [2].

For tomographic reconstruction of photoelectron angular distributions (PADs) [3], we developed a compact VMI spectrometer and a polarization-insensitive enhancement cavity [4]. We will present our new setup that collects electron-energy spectra at high rates and allows to tomographically reconstruct 3D PADs from intra-cavity xenon MPI.

[1] J. Nauta et al., *Opt. Lett.* 45(8), 2156 (2020). [2] J. Nauta et al., *Opt. Exp.* 29(2), 2624 (2021). [3] M. Wollenhaupt et al., *Appl. Phys. B* 95(4), 647-651 (2009). [4] J.-H. Oelmann et al., *Rev. Sci. Instrum.*, accepted (2022).

MO 19.2 Thu 15:00 F107

Free electron vortices in multiphoton ionization of molecules — •DARIUS KÖHNKE, CORNELIA OPP, TIM BAYER, and MATTHIAS WOLLENHAUPT — Carl von Ossietzky university Oldenburg, Institute of Physics, Germany

Since their theoretical proposal [1] and their first experimental demonstration [2], free electron vortices have attracted significant attention. So far, most of the theoretical and all of the experimental investigations were performed on atoms. Here, we present the first experimental demonstration of free electron vortices by multiphoton ionization (MPI) of molecules. Specifically, we study the creation of molecular vortices on C₆₀ fullerenes using counter rotating circularly polarized femtosecond laser pulse sequences generated from a white-light supercontinuum. Since the discovery of the C₆₀ molecule it has served as a benchmark system to study photo-induced dynamics in complex systems. Due to its high symmetry, the C₆₀ molecule, is an ideal system to bridge the gap between atoms and more complex systems such as polyatomic molecules and clusters. It has been shown that C₆₀ exhibits distinct atom-like electronic orbitals, termed superatomic molecular orbitals (SAMOS) [3], which play an important role in the MPI of fullerenes. By tomographic reconstruction of the three-dimensional photoelectron momentum distribution, we show that ionization from a SAMO with the polarization-tailored laser field creates a six-armed free electron vortex. [1] J. M. Ngoko Djioke et. al, *Phys. Rev. Lett.*, 115(11), 2015 [2] D. Pengel et. al, *Phys. Rev. Lett.*, 118(5), 2017 [3] M. Feng et. al, *Science*, 320(5874), 2008

MO 19.3 Thu 15:15 F107

Intra-cavity multiphoton processes in a standing wave frequency comb — •TOBIAS HELDT¹, JAN-HENDRIK OELMANN¹, LENNART GUTH¹, JANKO NAUTA¹, PRACHI NAGPAL¹, NICK LACKMANN¹, NELE GRIESBACH¹, CHRISTOPH DÜLLMANN², THOMAS PFEIFER¹, and JOSÉ R. CRESPO LÓPEZ-URRUTIA¹ — ¹Max-Planck-Institut für Kernphysik, 69117 Heidelberg, Germany — ²Johannes Gutenberg-Universität, 55099 Mainz, Germany

The coherence of frequency combs gives rise to a wide field of power-

ful spectroscopic techniques. Additionally, the high repetition rate of a comb leads to experimentally manageable count rates even for processes with low cross-sections. We use these properties to study the nonlinear regime of atomic and solid-state light-matter interaction. To reach the necessary field strengths, we enhance a frequency comb at 100 MHz in a cavity to intensities of over 10¹³ W/cm². The polarization insensitive bow-tie ring cavity supports counterpropagating pulses which collide in the focus, where they generate a standing wave for the time of the pulse overlap. This geometry is promising because not only does it reduce the interaction volume, enhancing the resolution of our velocity map imaging (VMI) spectrometer [1], but it also allows Doppler-free excitation of atoms. The polarization of both pulses can be controlled independently and an additional third pulse renders versatile pump-probe experiments possible. Further, we plan to probe the field with nanometric tips and we investigate how plasmons could lead to an excitation of the nuclear isomeric state of thorium-229 on such tips. [1] J.-H. Oelmann et al., *Rev. Sci. Instrum.*, accepted (2022)

MO 19.4 Thu 15:30 F107

Time-resolved three-body fragmentation of the CH₂I₂ molecule upon XUV irradiation — •FLORIAN TROST¹, SEVERIN MEISTER¹, HANNES LINDENBLATT¹, KIRSTEN SCHNORR¹, SVEN AUGUSTIN¹, YIFAN LIU¹, FARZAD HOSSEINI², MUSTAFA ZMERLI², MARKUS BRAUNE⁵, MARION KUHLMANN⁵, SERGIO DÍAZ-TENDERO⁴, FERNANDO MARTÍN⁴, RENAUD GUILLEMIN², MARIA NOVELLA PIANCASTELLI³, MARC SIMON², THOMAS PFEIFER¹, and ROBERT MOSHAMMER¹ — ¹Max-Planck-Institut für Kernphysik, Heidelberg — ²Sorbonne Université, Paris — ³Uppsala Universitet — ⁴Universidad Autónoma de Madrid — ⁵DESY, Hamburg

Knowledge of de-excitation, charge redistribution and fragmentation of molecules upon XUV irradiation is essential for our understanding of light-matter interaction. Here I present the sequential three-body fragmentation of diiodomethane (CH₂I₂) following 4d inner-shell ionisation of one iodine atom. The data was obtained by time-resolved XUV-XUV pump-probe measurements using the reaction microscope endstation at the free-electron laser FLASH2 at DESY. In the two-step dissociation of the CH₂I₂ molecule a rotating intermediary state is identified through time-resolved 3D momentum correlation of the fragments. These results are supported by classical as well as quantum-mechanical simulations.

MO 19.5 Thu 15:45 F107

Time operator, real tunneling time in strong field interaction and the attoclock. — •OSSAMA KULLIE — Institute for Physics, University of Kassel.

In the present work [1], we show that our real tunneling time relation derived in earlier works [2] can be derived from an observable or a time operator, which obeys an ordinary commutation relation. Moreover, we show that our real tunneling time can also be constructed from the well-known Aharonov-Bohm time operator. This shows that the specific form of the time operator is not decisive, and dynamical time operators relate identically to the intrinsic time of the system. It contrasts the famous Pauli theorem, and confirms the fact that time is an observable, i.e. the existence of time operator and that the time is not a parameter in quantum mechanics. Furthermore, we discuss the relations with different types of tunneling times such as Eisenbud-Wigner time, dwell time and the statistically defined tunneling time. We conclude with the hotly debated interpretation of the attoclock measurement and the advantage of the real tunneling time picture versus the imaginary one. [1] O. Kullie, *Phys. Rep.* 2020,2, 233. [2] O. K. Phys. Rev. A. **92**, 052118 (2015), O. K. Ann. of Phys. **389**, 333 (2018), [4] O. K. Mathematics **6**, 192 (2018).

MO 20: Poster III

Time: Thursday 16:30–19:00

Location: Empore Lichthof

MO 20.1 Thu 16:30 Empore Lichthof

Photoelectron Circular Dichroism of fenchone induced by coherent broadband laser pulses — ●ERIC KUTSCHER, ANTON ARTEMYEV, and PHILIPP DEMEKHIN — Institut für Physik und CIN-SaT, Universität Kassel, Heinrich-Plett-Str. 40, 34132 Kassel, Germany

Angular distributions of photoelectrons ionized by circularly polarized light from randomly oriented chiral molecules are asymmetric with respect to the propagation direction of the light [1]. This Photoelectron Circular Dichroism (PECD) appears also in the multiphoton ionization regime [2], which opens up a possibility for the coherent control of the effect. So far, the quantum control of PECD has only been discussed in a pump-probe excitation scheme with narrowband pulses [3]. Broadband (bb) laser pulses support photons in a large energy interval [4] and can be tailored in their amplitude and phase with pulse-shaping techniques. Here, the multiphoton PECD induced by coherent bb pulses with a 1-3 eV energy spectrum is studied theoretically for R(-)-fenchone molecules with different chirps and intensities of the pulses by utilizing the time-dependent single-center method [5]. Our results confirm a possibility to control the multiphoton PECD by tailored coherent bb pulses.

- [1] B. Ritchie, *Phys. Rev. A* **13**, 1411 (1976).
- [2] C. Lux *et al.*, *Angew. Chem. Int. Ed.* **51**, 5001 (2012).
- [3] R. E. Goetz *et al.*, *Phys. Rev. Lett.* **122**, 013204 (2019).
- [4] H. Hundertmark *et al.*, *Opt. Express* **17**, 1919 (2009).
- [5] A.N. Artemyev *et al.*, *J. Chem. Phys.* **142**, 244105 (2015).

MO 20.2 Thu 16:30 Empore Lichthof

Nondipole-Induced Asymmetry in the photoelectron emission distribution from fixed-in-space CO molecules — ●DMITRII REZVAN¹, KIM KLYSSEK², SVEN GRUNDMANN², ANDREAS PIER², NIKOLAY NOVIKOVSKIY¹, NICO STRENGER², DIMITRIOS TSITSONIS², MAX KIRCHER², ISABEL VELA-PEREZ², KILIAN FEHRE², FLORIAN TRINTER², MARKUS SCHÖPFLE², TILL JAHNKE², REINHARD DÖRNER², and PHILIPP DEMEKHIN¹ — ¹Institut für Physik und CINSaT, Universität Kassel, Heinrich-Plett-Str. 40, 34132 Kassel, Germany — ²Institut für Kernphysik, J.W. Goethe-Universität, Max-von-Laue-Str. 1, 60438 Frankfurt am Main, Germany

A nondipole-induced asymmetry in the angular distributions of C and O 1s-photoelectrons of fixed-in-space CO molecules is studied experimentally and theoretically at a photon energy of 905 eV. We demonstrate how scattering and nondipole effects determine the final emission distributions. The calculations were carried out in the frozen-core Hartree-Fock approximation using the stationary single center method [1], while the experiment was carried out using the COLTRIMS reaction microscope [2] available at beamline P04 of PETRA III, DESY. The emission distributions possess a strong scattering peak in the direction of the neighboring atom. This peak is either enhanced or suppressed by the nondipole contributions, respectively, if the scatterer points to the forward or backward direction with respect to the light propagation.

- [1] Ph. V. Demekhin *et al.*, *J. Chem. Phys.* **134**, 024113 (2011).
- [2] R. Dörner *et al.*, *Phys. Rep.* **330**, 95 (2000).

MO 20.3 Thu 16:30 Empore Lichthof

Measuring discriminatory optical forces on chiral molecules — ●TIANYU FANG, NICK VOGLEY, KILIAN SINGER, and DAQING WANG — Institute of Physics, University of Kassel, Heinrich-Plett-Straße 40, 34132 Kassel, Germany

We aim at a proof-of-principle experimental demonstration of enantiomer separation using optical forces created by a spatially tailored optical field. Chiral molecules with opposite handedness scatter polarized light differently. Consequently, the scattering of a photon exerts a force on the scatterer. This force can be tailored to be enantiomer specific, such that molecules of opposite handedness are pushed to opposite directions. In this poster, we outline an experiment, in which such discriminatory forces will spatially separate enantiomers in a propagating molecular beam.

MO 20.4 Thu 16:30 Empore Lichthof

Information theoretical approach to coupled electron-nuclear wave packet dynamics: Time-dependent differential Shannon

entropies — ●PETER SCHÜRGER and VOLKER ENGEL — Universität Würzburg, Institut für Physikalische und Theoretische Chemie, Emil-Fischer-Str. 42, 97074 Würzburg, Germany

We study differential Shannon entropies determined from position space quantum probability densities in a coupled electron-nuclear system. In calculating electronic and nuclear entropies, one gains information about the localization of the respective particles and also about the correlation between them. For a Born-Oppenheimer (BO) dynamics the correlation decreases at times when the wave packet reaches the classical turning points of its motion. If a strong non-adiabatic coupling is present, leading to a large population transfer between different electronic states, the electronic entropy is approximately constant. Then, the time-dependence of the entropy reflects the information on the nucleus alone, and the correlation is absent. A decomposition of the entropy into contributions from the participating electronic states reveals insight into the state-specific population and nuclear wave packet localization.

MO 20.5 Thu 16:30 Empore Lichthof

Spectroscopy of the (2)¹Σ_u state in Rb₂ — ●DOMINIK DORER¹, SHINSUKE HAZE¹, MARKUS DEISS¹, EBERHARD TIEMANN², and JOHANNES HECKER DENSCHLAG¹ — ¹Institut für Quantenmaterie, Universität Ulm — ²Institut für Quantenoptik, Leipzig Universität Hannover

We report a systematic study of the (2)¹Σ_u electronic state of the rubidium dimer. For this we carried out single-photon photoassociation spectroscopy with an ultracold sample of ground state atoms. Using either a pulsed or a more narrow cw laser we investigated the vibrational and rotational structure for both isotopologues, ⁸⁷Rb and ⁸⁵Rb. From a theoretical analysis including also previously recorded data from other experiments, we got new insights of spin-orbit and rotational interactions for the (2)¹Σ_u state. These results are also of interest for state-selective molecule detection schemes on the basis of resonance-enhanced multiphoton ionization (REMPI) as the (2)¹Σ_u can serve as an appropriate intermediate state.

MO 20.6 Thu 16:30 Empore Lichthof

Spectroscopic Characterization of Aluminum Monofluoride — ●NICOLE WALTER^{2,1}, JOHANNES SEIFERT¹, BOSIS SARTAKOV¹, and GERARD MEIJER¹ — ¹Fritz Haber Institute of the Max Planck Society, Berlin — ²Humboldt University of Berlin

Aluminum monofluoride (AlF) possesses highly favorable properties for laser cooling, both via the A¹Π and a³Π states. Determining efficient pathways between the singlet and the triplet manifold of electronic states will be advantageous for future experiments at ultralow temperatures. The lowest rotational levels of the A¹Π, *v* = 6 and b³Σ⁺, *v* = 5 states of AlF are nearly iso-energetic and interact via spin-orbit coupling. These levels thus have a strongly mixed spin-character and provide a singlet-triplet doorway. We here present a hyperfine resolved spectroscopic study of the A¹Π, *v* = 6 // b³Σ⁺, *v* = 5 perturbed system in a jet-cooled, pulsed molecular beam. From a fit to the observed energies of the hyperfine levels, the fine and hyperfine structure parameters of the coupled states, their relative energies as well as the spin-orbit interaction parameter are determined. The standard deviation of the fit is about 15 MHz. We experimentally determine the radiative lifetimes of selected hyperfine levels by time-delayed ionization, Lamb dip spectroscopy and accurate measurements of the transition lineshapes. The measured lifetimes range between 2 ns and 200 ns, determined by the degree of singlet-triplet mixing for each level.

MO 20.7 Thu 16:30 Empore Lichthof

Photoelectron Spectroscopy of Simple Diamondoids — ●SIMONE STAHL, PARKER CRANDALL, MARKO FÖRSTEL, and OTTO DÖFFER — Institut für Optik und Atomare Physik, Technische Universität Berlin, Germany

Diamondoids are a class of aliphatic molecules with cage-like structures and are a bridge between small hydrocarbons and large nanodiamond macromolecules. Extrasolar nanodiamonds have been discovered in carbonaceous chondrites and could account for more than 5% of cosmic and 40% of tertiary carbon in the interstellar medium.¹ Due to similarities between their infrared spectra and the unidentified infrared emission bands of young stars, diamondoids could be

present in these environments.² However, laboratory measurements of the optical properties of diamondoids, particularly of their cations and functionalized variants, remains incomplete and are necessary for astronomical observations. Here, we present the first photoelectron spectra of cyanoadamantane and amantadine. These are compared to newly measured spectra of adamantane, diamantane, and urotropine, with improved resolution and a more thorough assignment of spectral features with the aid of TD-DFT calculations.^{3,4} The effect of functionalization on the ionization potential, as well as the size and shape of the cage structure on the PES spectrum will be discussed.

1. Henning, Th. & Salama, F. *Science* 282, 2204-2210 (1998).
2. Pirali, O. et al. *ApJ* 661, 919-925 (2007).
3. Schmidt, W. *Tetrahedron* 29, 2129-2134 (1973).
4. Rander, T. et al. *J. Chem. Phys.* 138, 024310 (2013).

MO 20.8 Thu 16:30 Empore Lichthof

Analysis of statistical moments – a new technique to find Legendre coefficients in photoelectron distributions with increased precision — ●SIMON RANECKY, SAGNIK DAS, SUDHEENDRAN VASUDEVAN, TONIO ROSEN, HAN-GYEOL LEE, JAYANTA GHOSH, HENDRIKE BRAUN, ARNE SENFTLEBEN, and THOMAS BAUMERT — Uni Kassel, Experimentalphysik III

The ionisation of isotropically aligned molecules with linear or circular polarized light leads to a characteristic distribution of the photoelectrons. The radial part of the distribution relates to the energy of the photons and the ionisation potential of the molecules. The angular part can be described by Legendre polynomials of the order up to twice the number of photons involved in the ionisation [1].

In velocity map imaging, which we use to detect photoelectrons, the 3d electron distribution is projected electrostatically on a phosphorous screen. The right orientation of the spectrometer leads to no information loss in the Abel-projection, but an Abel-inversion like rbases [2] is necessary to find out the distribution of the Legendre coefficients.

As an alternative way to retrieve the Legendre coefficients, the centroid and higher moments of the photoelectron projection can be used. With the calculated moments of single Legendre polynomials, a linear system of equations can be compiled that makes the Legendre coefficients accessible. For simple photoelectron distributions, we will demonstrate smaller uncertainties as compared to rbases.

- [1] Yang, *Phys. Rev.* 74, 764; Dixit et al., *J. Chem. Phys.* 82, 3546
- [2] Hickstein et al., *Rev. Sci. Instrum.* 90, 065115

MO 20.9 Thu 16:30 Empore Lichthof

Photoelectron circular dichroism on fenchone: from multiphoton to the tunnel ionization regime using near-infrared femtosecond laser pulses. — ●SUDHEENDRAN VASUDEVAN, HAN-GYEOL LEE, SIMON T. RANECKY, NICOLAS LADDA, TONIO ROSEN, SAGNIK DAS, JAYANTA GHOSH, HENDRIKE BRAUN, ARNE SENFTLEBEN, and THOMAS BAUMERT — Institut für Physik, Universität Kassel, Heinrich-Plett-Str. 40, 34132 Kassel

Photoelectron circular dichroism (PECD) is a pronounced forward/backward asymmetry in the ionization of chiral molecules by circularly polarized light. PECD is dominated by the electric-dipole effect, which gives rise to a larger magnitude of asymmetry as compared to circular dichroism in absorption. In this contribution, we use nearly Fourier-limited near-infrared pulses directly from a titanium sapphire femtosecond laser to excite electronic intermediate states in fenchone molecule via a (3+1+1+1) resonance-enhanced multiphoton ionization (REMPI) scheme. We scanned the intensity of the laser and measured the angular distribution of the photoelectrons from the multiphoton almost to the tunnel ionization regime. Herein we present our measured photoelectron spectra, PECD, and mass spectra during the intensity scan.

MO 20.10 Thu 16:30 Empore Lichthof

Temperature-dependant Luminescence and Step-scan FTIR Investigations of Tetranuclear Cu(I) Complexes — ●SOPHIE STEIGER¹, PIT BODEN¹, JASMIN BUSCH², DANIEL MARHÖFER¹, STEFAN BRÄSE², and GEREON NIEDNER-SCHATTEBURG¹ — ¹Department of Chemistry, TUK, Germany — ²IOC, KIT, Germany

In this contribution, we present the investigation of the temperature-dependent luminescence and the luminescent states of tetranuclear copper(I) complexes. The investigations were performed by (time-resolved) luminescence and static as well as step-scan FTIR spectroscopy. The measurements were performed in a temperature range of 290 K to 10 or 5 K in KBr matrix or pure powder. The complexes contain a tetranuclear Cu₄X₄ (X=Cl, Br, I) core and two 2-

(diphenylphosphino)-pyridine ligands with or without a methyl substituent in 4-position of the pyridine ring units. As known from previous work^[1], the same type of complexes with ligands with a methyl group in 6-position shows a pronounced thermochromism, which was not detected for the Cu₄I₄ complex with methyl group in 4-position in KBr matrix. The Cu₄I₄ complex without methyl group shows the thermochromism in powder measurements but it is less resolved in KBr matrix. All other investigated complexes have only slight temperature-dependent shifts in the luminescence. With step-scan FTIR spectroscopy in comparison with quantum chemical calculation, the excited states were assigned to the different excited state structures.

[1] *Chem. Eur. J.* 2021, 27, 5439-5452.

MO 20.11 Thu 16:30 Empore Lichthof

Calculation of the Stark effect of Rydberg states in nitric oxide — ●ALEXANDER TRACHTMANN^{1,4}, FABIAN MUNKES^{1,4}, PATRICK KASPAR^{1,4}, PHILIPP NEUFELD^{1,4}, YANNICK SCHELLANDER^{2,4}, LARS BAUMGÄRTNER^{3,4}, ROBERT LÖW^{1,4}, TILMAN PFAU^{1,4}, and HARALD KÜBLER^{1,4} — ¹⁵. Physikalisches Institut, Universität Stuttgart, Pfaffenwaldring 57, 70569 Stuttgart — ²Institut für Großflächige Mikroelektronik, Universität Stuttgart, Allmandring 3b, 70569 Stuttgart — ³Institut für Intelligente Sensorik und Theoretische Elektrotechnik, Universität Stuttgart, Pfaffenwaldring 47, 70569 Stuttgart — ⁴Center for Integrated Quantum Science and Technology (IQST), Universität Stuttgart

We demonstrate Stark effect calculations of high-lying Rydberg states in nitric oxide and compare them to measurements at room temperature. These states are generated using a three-photon continuous-wave excitation scheme. The readout is based on the detection of charged particles created by collisional ionization of Rydberg molecules. Details of the calculation techniques are given.

MO 20.12 Thu 16:30 Empore Lichthof

Decoherence estimates for spin-boson systems from semiclassical Gaussian wavepacket propagation — ●SREEJA LOHO CHOUDHURY¹, CAROLINE LASSER², ROCCO MARTINAZZO^{3,4}, and IRENE BURGHARDT¹ — ¹Institute of Physical and Theoretical Chemistry, Goethe University Frankfurt — ²Technische Universität München, Zentrum Mathematik, Deutschland — ³Department of Chemistry, Università degli Studi di Milano, Italy — ⁴Instituto di Scienze e Tecnologie Molecolari, CNR, Milano, Italy

We investigate the time scale of decoherence in spin-boson type systems, by calculating the subsystem purity as a measure of decoherence. For a general system-bath setting, the decoherence time can be expressed in terms of a universal Gaussian decay of the purity [O. Prezhdo and P. Rossky, *Phys. Rev. Lett.* **81**, 5294 (1998)], but exponential decay is often predicted from Markovian master equations. The transition between these forms of decoherence has recently been discussed [B. Yan and W. H. Zurek, *N. J. Phys.* **24**, 113029 (2022)]. Against this background, we investigate the pure dephasing case in a spin-boson system using semiclassical Gaussian wavepackets, notably relying on the variational multiconfigurational Gaussians (vMCG) [G. A. Worth and I. Burghardt, *Chem. Phys. Lett.* **368**, 502 (2003)] and multiconfiguration time-dependent Hartree (MCTDH) [M. H. Beck, A. Jäckle, G. A. Worth, H.-D. Meyer, *Phys. Rep.* **324**, 1 (2000)] formalism for wavefunction propagation. We further consider quantum-semiclassical hybrid wavefunctions and examine the effect of semiclassical scaling on the subsystem purity.

MO 20.13 Thu 16:30 Empore Lichthof

Photoelectron spectroscopy study of anthracene anions — ●KEVIN SCHWARZ, AGHIGH JALEHDOOST, and BERND V. ISSENDORFF — Institute of Physics, University of Freiburg, Hermann-Herder-Str. 3, 79104 Freiburg, Germany

As semiconducting organic materials generate more and more interest, there are still properties of said materials left to be investigated more in depth. After the development of a high temperature-suited pulsed valve to produce clusters of organic material such as anthracene has been concluded recently, first results of energy-resolved photoelectron spectroscopy (PES) studies on anthracene anions (C₁₄H₁₀⁻) are shown.

MO 20.14 Thu 16:30 Empore Lichthof

Action Spectroscopic Infrared Detection of NCCO⁺ — ●MARCEL BAST¹, THOMAS SALOMON¹, OSKAR ASVANY¹, IGOR SAVIĆ², SANDRA BRÜNKEN³, MATHIAS SCHÄFER⁴, STEPHAN SCHLEMMER¹, and SVEN THORWIRTH¹ — ¹I. Physikalisches Institut,

Universität zu Köln, Köln, Germany — ²Department of Physics, Faculty of Sciences, University of Novi Sad, Serbia — ³FELIX Laboratory, Institute for Molecules and Materials, Radboud University, Nijmegen, the Netherlands — ⁴Institute of Organic Chemistry, Department of Chemistry, University of Cologne, Köln, Germany

The linear NCCO⁺ ion has been studied spectroscopically for the first time using the Free Electron Laser for Infrared eXperiments (FELIX) in combination with the 4K 22-pole ion trap facility FELion. The vibrational spectrum of NCCO⁺ was observed in the range from 500 to 1400 and 2000 to 2500 cm⁻¹ using resonant photodissociation of the corresponding Ne-complex. Spectroscopic assignments are in very good agreement with high-level quantum-chemical calculations.

In a consecutive study, the rotationally resolved ν_2 band of the bare NCCO⁺ ion was measured around 2150 cm⁻¹ using the COLTRAP2 instrument in Cologne. Here, the recently developed action spectroscopic method Leak-Out Spectroscopy (LOS)^a was used, that traces the bare ion rather than a weakly bound complex. In this manner, as will be shown on this poster, a nearly background-free absorption spectrum with a large signal-to-noise ratio was obtained.

^a Schmid et al. 2022, J. Phys. Chem. A 126, 43, 8111-8117

MO 20.15 Thu 16:30 Empore Lichthof

Towards probing vibrational dynamics in methyl p-tolyl sulfoxide via time-resolved PECD — ●NICOLAS LADDA, SUDHEENDRAN VASUDEVAN, TONIO ROSEN, HAN-GYEOL LEE, SAGNIK DAS, JAYANTA GHOSH, HENDRIKE BRAUN, ARNE SENFLEBEN, and THOMAS BAUMERT — Institut für Physik, Universität Kassel, Heinrich-Plett-Strasse 40, 34132 Kassel, Germany

The goal is to investigate the dynamic change of chiral character upon laser-induced vibrational motion in methyl p-tolyl sulfoxide (MTSO). To this end, we aim to measure the forward/backward asymmetry of the photoelectron angular distribution with respect to the propagation direction of the ionizing circularly polarized light of the randomly oriented chiral molecule, known as photoelectron circular dichroism (PECD) [1]. The vibrational motion - the umbrella motion of the sulfoxide molecule - changes the chiral character of the molecule, which can be investigated by studying the time-resolved PECD with UV femtosecond laser pulses. For this purpose, a two-color pump-probe setup consisting of the 3rd and 4th harmonics of a Ti:Sa laser system is used. The experimental setup has already been tested with static PECD measurements on fenchone and MTSO.

[1] N. Böwering, T. Lischke, B. Schmidtke, N. Müller, T. Khalil, and U. Heinzmann Phys. Rev. Lett. 2001, 86, 1187

MO 20.16 Thu 16:30 Empore Lichthof

Two-photon excitation spectroscopy of metal-organic frameworks — ●HONGXING HAO¹, YANG CUI¹, FERDINAND BERGMEIER², SIMON N. DEGER³, ALEXANDER PÖTHIG³, EBERHARD RIEDLE², ROLAND A. FISCHER³, and JÜRGEN HAUER¹ — ¹Dynamical Spectroscopy, Department of Chemistry, Technical University of Munich, 85748 Garching, Germany — ²Lehrstuhl für BioMolekulare Optik, Ludwig-Maximilians-Universität München, 80538 München, Germany — ³Chair of Inorganic and Metal-Organic Chemistry, Department of Chemistry, Technical University of Munich, 85748 Garching, Germany

Metal-organic frameworks (MOFs) or coordination polymers (CPs) constitute a novel class of materials and have several interesting photo-physical properties such as a high multiphoton absorption cross section. Here, we introduce a spectroscopic experiment to measure two-photon excitation spectra from the visible to the near infrared based on a non-collinear optical parametric amplifier and a phase-stable common-path birefringent interferometer via nonlinear Fourier-transform spectroscopy. We present two-photon excitation spectra in the range between 380 to 470 nm with a data acquisition time of under one minute per spectrum.

We demonstrate the feasibility of this approach by using standard dyes as test samples. After incorporating two-photon absorption active organic linker molecules in crystalline CPs or MOFs, the spectra show a shift of oscillator strength. We suggest excitonic coupling between the linkers as the mechanism behind this targeted modification of two-photon absorption property.

MO 20.17 Thu 16:30 Empore Lichthof

Control of (bio-) nanoparticles with external fields — ●JINGXUAN HE^{1,2,3}, LENA WORBS^{1,2}, JANNIK LÜBKE^{1,2,3}, SURYA KIRAN PERAVALI^{1,4}, ARMANDO D. ESTILLORE¹, AMIT K. SAMANTA^{1,3}, and JOCHEN KÜPPER^{1,2,3} — ¹Center for Free-Electron Laser Science, Deutsches Elektronen-Synchrotron DESY, Hamburg, Germany — ²Department of Physics, Universität Hamburg, Germany — ³Center for Ultrafast Imaging, Universität Hamburg, Germany — ⁴Fakultät für Maschinenbau, Helmut-Schmidt-Universität, Germany

Unraveling the elementary steps of biological processes and chemical reactions is a long-time goal of the scientific community. By using x-ray single-particle diffractive imaging, we can investigate the three-dimensional molecular structure of individual nanoparticles at atomic resolution through reconstructing a series of two-dimensional diffraction patterns [1]. However, because of the typically low signal-to-noise ratio, this requires the collection of a large amount of individual diffraction patterns. Since every intercepted particle is destroyed by the intense x-ray pulse, a new and preferably identical sample particle has to be delivered to every pulse. Here, we present an approach to prepare dense beams of cold and controlled nanoparticles and macromolecules with buffer-gas-cell-cooling and aerodynamic-focusing techniques. Besides, we demonstrated several control mechanisms with external fields that can help to realize a stream of pure, identical particles. We also showed that the particles' arbitrary orientation in space can be controlled by applying laser alignment.

[1] M. M. Seibert, *et al.*, *Nature* **470**, 78 (2011)

MO 20.18 Thu 16:30 Empore Lichthof

TrapREMI: A reaction microscope inside an electrostatic ion beam trap — FRANS SCHOTSCH¹, ●ILJA ZEBERGS¹, SVEN AUGUSTIN^{1,2}, HANNES LINDENBLATT¹, LUDWIG HOIBL³, DENIS DJENDJUR³, CLAUDIUS DIETER SCHRÖTER¹, THOMAS PFEIFER¹, and ROBERT MOSHAMMER¹ — ¹Max-Planck-Institute for Nuclear Physics, Saupfercheckweg 1, 69117 Heidelberg, Baden-Württemberg, Germany — ²Paul Scherrer Institut, Forschungsstrasse 111, 5232 Villigen, Switzerland — ³Department of Physics and Astronomy, Ruprecht-Karls University, 69120 Heidelberg, Baden-Württemberg, Germany

A novel setup has been developed to investigate the reaction dynamics of (molecular) ions with a variety of projectiles in kinematically complete crossed-beam experiments. The setup combines an electrostatic ion beam trap (EIBT) with a reaction microscope (REMI).

The EIBT stores ions in oscillatory motion at several keV and allows further preparation e.g. cooling, relaxation, pulsing of the beam, mass selection. Reactions are induced by crossing the ion beam with another beam in the field-free region of the EIBT, where the REMI is located. The REMI, oriented along the target beam axis, allows for detection of all fragments in coincidence, including neutral particles, and reconstruction of their 3D-momenta.

During first REMI tests a laser pulse was used as the projectile to induce photodissociation in molecular ions. Currently a gas jet was assembled to provide an atomic/molecular beam and first simple ion-atom/molecule collision experiments are conducted.

MO 20.19 Thu 16:30 Empore Lichthof

Collisional excitation and dissociation of CO molecules by ion impacts and applications to astrophysics — ●MASATO NAKAMURA — College of Science and Technology, Nihon University, Funabashi, Japan

The CO molecule is one of the most abundant molecules in interstellar space and a major component of the stellar (solar) winds is proton. Thus, the collisional excitation and collision-induced dissociation (CID) by proton impacts are expected to play important roles in the chemical evolution of interstellar molecules. Here we present a theoretical work on the energy transfer and fragmentation of CO molecule by proton impact at hyperthermal energies. The classical trajectory calculation and the sudden-limit model are applied to estimate the energy transfer from translational to internal degrees of freedom. The threshold energy for the dissociation is calculated by for the impact of singly charged ions. For comparison, the calculation is performed also for nitrogen molecule. The nitrogen molecule is a major component of atmosphere of Titan.

MO 21: Molecular Physics with X-rays

Time: Friday 11:00–13:00

Location: F102

MO 21.1 Fri 11:00 F102

X-ray induced luminescence for DNA molecules in aqueous solution via nitrogen core ionization — ●YUSAKU TERAO¹, YOSHIKI KUMAGAI¹, ISSEI SUZUKI¹, TAKAHIRO TSUCHIYA¹, MASATOSHI UKAI¹, AKINARI YOKOYA², KENTARO FUJII², YOSHIHIRO FUKUDA³, and YUJI SAITOH⁴ — ¹Dept. of Applied Physics, Tokyo University of Agriculture and Technology, Koganei, Tokyo 184-8588, Japan — ²Institute for Quantum Life Science, Naka-gun, 319-1195, Ibaraki, Japan — ³SPRING-8 Service Co. Ltd., Tatsuno-shi, Hyogo 679-5165, Japan — ⁴Synchrotron Radiation Research Center, Hyogo 679-5148, Japan

Radiation effect to biological matter can lead to various kinds of damages on a molecular level. However, a protective environmental effect of surrounding water is expected to play an important role to suppress damage inductions. To get to know about the protective environmental effect, we studied intermediate species along the reaction pathway of core ionized DNA molecules in aqueous solution. We conducted X-ray induced luminescence spectroscopy for nucleotides in aqueous solution using monochromatic soft X-rays in the nitrogen K-shell region at SPRING-8 BL23SU. We obtained the excitation spectra of total luminescence yields for four nucleotides samples and the filtered luminescence spectra as well. Those results indicated the luminescence from parent base species which are maintained even after the Auger ionization of core-hole state, implying significant emission processes of their excess charge and energy can take place in the relaxation pathway.

MO 21.2 Fri 11:15 F102

Experimental investigation of solvated metal ions after X-ray-induced ionization — ●DANA BLOSS¹, FLORIAN TRINTER^{2,3}, UWE HERGENHAHN², ARNO EHRESMANN¹, and ANDREAS HANS¹ — ¹Institut für Physik und Center for Interdisciplinary Nanostructure Science and Technology (CINSaT), Universität Kassel, Heinrich-Platt-Straße 40, 34132 Kassel — ²Fritz-Haber-Institut der Max-Planck-Gesellschaft, Faradayweg 4-6, 14195 Berlin — ³Institut für Kernphysik, Goethe-Universität, Max-von-Laue-Str.1, 60438 Frankfurt am Main

Exploration of the microscopic response of biological systems to ionizing radiation is a key to understanding radiation damage on a molecular level. Of special interest in this context are low-energy electrons (LEEs) with energies below 30 eV, which are known to be genotoxic. One source of these LEEs upon X-ray irradiation are secondary processes like the interatomic Coulombic decay (ICD) - an energy-transfer process - or the electron-transfer-mediated decay (ETMD) - a charge-transfer process. Both have attracted attention as a source of efficient localized emission of LEEs close to the site of ionization and been studied intensely throughout the last decades in clusters and solutions. A multi-step cascade of ICD and ETMD processes upon X-ray ionization of microsolvated metal ions, e.g., Mg²⁺, was predicted. We experimentally investigate this theoretically predicted LEE emission for Mg²⁺, Ca²⁺ and Al³⁺ solutions at different synchrotron facilities with a combination of liquid microjets and coincident electron detection.

MO 21.3 Fri 11:30 F102

Tracking the birth of the solvated electron with x-rays — ●ARTURO SOPENA MOROS¹, LUDGER INHETER¹, and ROBIN SANTRA^{1,2} — ¹Center for Free-Electron Laser Science, DESY — ²Department of Physics, Universität Hamburg

The attention that the solvated electron, e⁻(aq), has received over the past half century, both from theory and experiment, is well justified due to its fundamental role in the chemistry of water. Its high reactivity and its role in radiation damage of DNA [1] have important implications in physics, chemistry, and biology. Albeit the apparent simplicity of this system, a unified picture of the solvation process remains elusive. In this theoretical study, we explore the capabilities of time-resolved x-ray absorption spectroscopy (XAS) as a new tool to investigate the ultrafast dynamics of the formation of the solvated electron.

We used classical and ring-polymer molecular dynamics together with a recently published neural network-based force field trained on MP2 electronic structure calculations [2] to simulate the evolution of an excess electron placed in bulk water. The simulations reveal that

e⁻(aq) localizes in a cavity with a radius of 2.4 Å within 1.6 ps. For snapshots of the simulations, we have calculated the XAS using the ab-initio electronic structure toolkit XMOLECULE [3]. The calculations reveal that it is possible to trace the solvation dynamics of the electron in liquid water by inspecting distinct features in the time-resolved XAS.

[1] Chem. Rev. 112, 5553 (2012). [2] Nat. Commun. 12, 766 (2021). [3] Struct. Dyn. 2, 041707 (2015)

MO 21.4 Fri 11:45 F102

Chemical effects on the dynamics of organic molecules irradiated with high intensity x rays — ●SOURAV BANERJEE^{1,2}, ZOLTAN JUREK^{1,2,3}, MALIK MUHAMMAD ABDULLAH², and ROBIN SANTRA^{1,2,3,4} — ¹Center for Free-Electron Laser Science (CFEL), Notkestr. 85, 22607 Hamburg, Germany — ²Deutsches Elektronen-Synchrotron (DESY), Notkestr. 85, 22607 Hamburg, Germany — ³The Hamburg Centre for Ultrafast Imaging, Luruper Chaussee 149, 22761 Hamburg, Germany — ⁴Department of Physics, Universität Hamburg, Notkestr. 9-11, 22607 Hamburg, Germany

High intensity ultra-short x-ray pulses, produced by X-ray free-electron lasers (XFEL), induce complex dynamics in matter. Over the past several years, numerical simulations for various molecular systems and clusters have successfully been performed with the classical molecular dynamics (MD) based hybrid code XMDYN.

The present study [1] extends the XMDYN toolkit to capture the effect of charge transfer and chemical bonds. The chemical bonds are approximated by the reactive force field (ReaxFF) description. The system under examination is a small organic molecule, glycine, which, being an amino acid, has relevance to general biological systems. Effects on key quantities, e.g., time dependent charge states, etc. are analyzed. The present study suggests that the chemical effects are of importance for realistically achievable fluences, whereas bondless simulation is accurate for high fluence.

[1] S. Banerjee et al., Struct. Dyn. 9, 054101 (2022) and references therein.

MO 21.5 Fri 12:00 F102

Probing the UV-induced ultrafast dynamics of thionucleobases with time-resolved x-ray absorption at the Free Electron Laser FLASH — ●F LEVER^{1,2}, D MAYER^{1,2}, D PICCONI⁶, S ALISAUSKAS², F CALEGARI², S DUESTERER², C EHLERT⁴, R FEIFEL³, M KUHLMANN², T MAZZA⁷, J METJE¹, M ROBINSON^{1,2}, R SQUIBB³, A TRABATTONI^{2,8}, M WARE⁵, P SAALFRANK¹, T WOLF⁵, and M GHUER^{1,2} — ¹Universität Potsdam, Germany — ²DESY, Germany — ³University of Gothenburg, Sweden — ⁴Heidelberg Institute for Theoretical Studies, Germany — ⁵SLAC, USA — ⁶University of Groningen, Netherlands — ⁷XFEL, Germany — ⁸IQO, Germany

Light-induced ultrafast reactions play a fundamental role in the photochemistry of DNA, as they can lead to the formation of lesions in the genetic code. Thionucleobases are sulfur-substituted nucleobases that show an increased rate of UV-induced lesion formation. In a previous work by this group, we have investigated the photoinduced dynamics of 2-thiouracil (2-tUra) via time resolved x-ray photoemission [1,2]. In this work, we study the ultrafast dynamic 2-thiouracil in a UV-pump, x-ray probe experiment at the Free Electron Laser FLASH. The molecule is excited by the pump pulse in the S2 state ($\pi\pi^*$ character), and its dynamic is probed by a delayed x-ray pulse that is resonant with 2p and 2s excitations of the sulfur atom. The localized probe on the sulfur allows us to image the excitation of the dark S1 ($n\pi$) state within a 100fs time scale, and to follow oscillations in its population fraction with a period of 200fs. [1] Mayer et al. Nat Comm 13, 198 (2022) [2] Lever et al. J. Phys. B 54 014002 (2021)

MO 21.6 Fri 12:15 F102

Creating Electronic Molecular Movies Using Time-Resolved X-Ray Photoelectron Spectroscopy at Free-Electron Lasers — ●DENNIS MAYER¹, FABIANO LEVER¹, DAVID PICCONI², SKIRMANTAS ALISAUSKAS¹, AGATA AZZOLINI³, FRANCESCA CALEGARI^{3,4,5}, GIOVANNI CIRMI^{1,4}, STEFAN DÜSTERER¹, ULRIKE FRÜHLING¹, ALICE GREEN^{6,7}, INGMAR HARTL¹, MARION KUHLMANN¹, TOMMASO MAZZA⁶, STEFFEN PALUTKE¹, SEBASTIAN SCHULZ¹, ANDREA TRABATTONI^{3,8}, ATIA TUL NOOR¹, and MARKUS GÜHR¹ — ¹DESY, Hamburg, Germany — ²University of Groningen, The Netherlands —

³CFEL, DESY, Hamburg, Germany — ⁴The Hamburg Centre for Ultrafast Imaging, Germany — ⁵University of Hamburg, Germany — ⁶European XFEL, Schenefeld, Germany — ⁷Stanford PULSE Institute, Menlo Park, USA — ⁸University of Hannover, Germany

Recently, we were able to extend the chemical shift concept to excited state molecular dynamics [1]. Here, we present a follow-up experiment performed at FLASH, showing C 1s time-resolved photoelectron spectra of 2-thiouracil after UV excitation. The photoelectron spectra show a strong shift towards lower binding energies suggesting a charge movement towards the pyrimidine ring. These measurements verify the concept of the excited-state chemical shift [1] and pave the way to create electronic molecular movies for the UV-induced relaxation using time-resolved x-ray photoelectron spectroscopy.

[1] Mayer et al., Nature Communications 13, 198 (2022)

MO 21.7 Fri 12:30 F102

Tracking Conical Intersections with Nonlinear X-ray Raman Spectroscopy — ●DEEPENDRA JADOUN and MARKUS KOWALEWSKI — Department of Physics, Stockholm University, Albanova University Centre, SE-106 91 Stockholm, Sweden

Conical intersections (CIs) play a vital role in processes such as the event of vision, and DNA damage from sunlight. A CI appears in a molecule when electronic and nuclear motions are strongly coupled, and it acts as a funnel that guides a molecule from one electronic state to another. Substantial efforts have been made toward understanding such non-adiabatic phenomena. X-ray Raman techniques have been proposed in the past to investigate the presence of a CI in molecules.

We propose a two-dimensional Raman probe scheme that uses a visible/infrared pulse and an ultra-short X-ray Raman probe pulse. The visible/infrared pulse interaction creates a coherent superposition of electronic states in the molecule. Probing the coherent superposi-

tion using ultra-short X-ray pulses allows to visualize the dynamic energy separation between electronic states throughout the photochemical process. The lifetime of the coherent superposition created using the infrared pulse can be read directly from two-dimensional Raman spectra. Therefore, the method aids to the observation of multiple indicators of a conical intersection and may allow for a more detailed study of non-adiabatic dynamics in molecules.

MO 21.8 Fri 12:45 F102

X-FEL induced Coulomb explosion: Advanced analysis of coincident ion momenta — ●BENOÎT RICHARD^{1,2,3}, ROBIN SANTRA^{1,2,3}, and LUDGER INHETER^{1,2,3} — ¹CFEL, DESY, Hamburg, Germany — ²CUI, Hamburg, Germany — ³Universität Hamburg, Hamburg, Germany

Intense x-ray pulses from x-ray free-electron lasers (X-FEL) ionize small molecular systems leading to their violent dissociation into several atomic fragments on a femtosecond time scale. Recent experiments with the 2-iodopyridine molecule have demonstrated that measuring the momenta of only 3 fragments in coincidence is sufficient to distinguish individual ion contributions. [1] However, combining all the different n-particle coincidence data, where in each a different subset of ion fragments is detected, into a joint distribution is not trivial.

We present a general method to perform such analysis. We demonstrate its application using simulation data of the X-FEL-induced Coulomb explosion of 2-iodopyridine and use it to extract correlations between ion momenta. We show that such correlations contain fingerprints of collisions during the explosion. [2] Moreover, we demonstrate that this data is related to the structure of the molecule just before being hit by the x-ray pulse that destroyed it.

[1] R. Boll, J. Schäfer, B. Richard et al. 2022. Nat. Phys. 18, 423-428

[2] B. Richard et al. 2021. J. Phys. B: At. Mol. Opt. Phys. 54 194001

MO 22: Theoretical and Computational Molecular Physics

Time: Friday 11:00–13:15

Location: F142

Invited Talk MO 22.1 Fri 11:00 F142
A QED Theory of Mediated RET Between a Pair of Chiral Molecules — ●AKBAR SALAM — Department of Chemistry, Wake Forest University, Winston-Salem, NC 27109, USA

A QED [1] theory of relayed resonance energy transfer (RET) [2] between a pair of identical chiral molecules by one or two neutral, passive, electric dipole polarizable bodies, is developed. Migration of excitation energy occurs via the exchange of a single virtual photon between any two interacting particles. Isotropic Fermi golden rule transfer rates are evaluated for direct and bridge-mediated pathways. Rates from both routes depend on the chirality of the donor and acceptor moieties [3-5]. For conveyance of energy to take place indirectly, a multi-level model must be adopted for each mediator polarizability. Furthermore, the rate is found to be a maximum when the three or four particles are collinear. Useful understanding is achieved of propagation of energy between optically active entities in a medium with uniform dielectric constant [6]. [1]*A. Salam, Molecular Quantum Electrodynamics, John Wiley & Sons, Inc., 2010. [2]*A. Salam, Atoms 6, 56 (2018). [3]*D. P. Craig and T. Thirunamachandran, J. Chem. Phys. 109, 1258 (1998). [4]*A. Salam, J. Phys. Chem. A 125, 3549 (2021). [5]*A. Salam, J. Chem. Phys. 157, 104110 (2022). [6]*J. Franz, S. Y. Buhmann and A. Salam, arXiv:2209.15400 [quant-ph].

MO 22.2 Fri 11:30 F142

Excitation transport in molecular aggregates with thermal motion — ●RITESH PANT and SEBASTIAN WÜSTER — Indian institute of science education and research, Bhopal, India

Molecular aggregates can under certain conditions transport electronic excitation energy over large distances due to the long range dipole-dipole interactions. These interactions are also the characteristics of Rydberg aggregates which have been proved as the quantum simulators for molecular aggregates. An idea that naturally arises in Rydberg aggregates, is adiabatic excitation transport through atomic motion, where slow motion of the atoms combined with excitation transport can result in efficient and guided transport of the excitation from one end of an atomic chain to the other. Based on the analogy between

Rydberg- and Molecular aggregates, in ref. [1] we explore whether the adiabatic excitation transport can play a functional role in molecular aggregates in the absence of intra-molecular vibrations. But because the transport is partially adiabatic and because it involves transitions between non-eigenstates, it is challenging to estimate the adiabaticity of transport in molecular aggregates. Hence, in ref [2] we established a measure to quantify the adiabatic character of quantum transitions in general. Next, the effect of intramolecular vibrations is included by extending our calculation for excitation transport to an open-quantum-system technique [3].

[1] R. Pant and S. Wüster, Physical Chemistry Chemical Physics 22, 21169 (2020). [2] R. Pant, et al., <https://arxiv.org/abs/2007.10707>. [3] R. Pant, et al., (Manuscript in preparation)

MO 22.3 Fri 11:45 F142

Rotational and vibrational decay of diatomic molecules near a surface — ●NICOLAS SCHÜLER, OMAR JESÚS FRANCA SANTIAGO, and STEFAN YOSHI BUHMANN — Institute of Physics, University of Kassel, Germany

We study the rotational and vibrational motion of chiral diatomic molecules near a dielectric surface [1]. We use macroscopic quantum electrodynamics [2] to obtain the decay rate of the molecule. We determine the critical distances at which retarded (nonretarded) interactions with the surface become dominant, which happens when the distances are much larger (smaller) than the atomic transition wavelengths, respectively. We explore potential links of the rotational and vibrational dynamics with quantum friction.

References:

[1] Stefan Yoshi Buhmann, M. R. Tarbutt, Stefan Scheel, and E. A. Hinds, Phys. Rev. A 78, 052901 (2008).

[2] D.T. Butcher, S.Y. Buhmann, and S. Scheel, New J. Phys. 14, 113013 (2012).

MO 22.4 Fri 12:00 F142

Medium-assisted chiral discrimination via resonance energy transfer — ●JANINE CHRISTINE FRANZ¹, STEFAN YOSHI BUHMANN², and AKBAR SALAM³ — ¹University of Freiburg, Ger-

many — ²University of Kassel, Germany — ³Wake Forest University, Winston-Salem, USA

Resonance energy transfer between two chiral molecules can be used in principle to discriminate enantiomers [1]. Using macroscopic quantum electrodynamics, we study how to enhance and control this effect by means of an intervening medium and propose a distinct, but related way to discriminate between enantiomers by using a medium with known chirality [2].

When embedding molecules in a macroscopic medium, the microscopic structure of the medium close to the embedded molecule needs to be taken into account; we achieve this by using the so-called local-field correction. We present the influence of a magneto-(di)electric medium surrounding the energy-exchanging molecules, where exotic media have a non-trivial impact on the degree of discrimination [3]. When considering a chiral medium that takes an active part in the chiral discrimination, we find that considering local-field effects results in a surprising prediction for the discrimination.

- [1] D. P. Craig et al., *J. Chem. Phys.* 109, 1258 (1998)
- [2] S. Y. Buhmann et al., *New J. Phys.* 14 083034 (2012)
- [3] J. Franz et al., arXiv:2209.15400 [quant-ph]

MO 22.5 Fri 12:15 F142

Analysis of the dopamine D2 receptors interaction with three radiopharmaceuticals using molecular dynamics techniques — NASTASIA-SANDA MOLDOVEAN-CIOROIANU, •DIANA-GABRIELA TIMARU, and VASILE CHIS — Babes-Bolyai University, Faculty of Physics, Cluj-Napoca, Romania

It is widely known that the dopamine neurotransmitter and its receptors play a pivotal role in neuronal signal transduction and other metabolic processes. Using molecular dynamics (MD) techniques, this study seeks to find and describe the optimal binding positions between the D2 dopamine receptor (D2DR) and three [11C]-labeled synthetic compounds: [11C]-FLB 457, raclopride ([11C]-RACL), and halobenzazepine ([11C]-SCH).

Previous research has demonstrated that the FLB 457 molecule has a high affinity for the D2 dopamine subunit. In addition, [11C]-tagged RACL is already employed as a PET scan radiotracer.

Our analysis showed, in good agreement with experimental results, that RACL ligand bound at the upper portion of the D2 dopamine receptor had the highest interaction energy patterns. In addition, these discoveries pave the path for future research. Perspectives include more MD studies as well as thorough production time and free energy estimates. The same set of radiopharmaceuticals will be docked in the receptor's top pocket, with the D2DR structure encased in a DOPC/phospholipid bilayer membrane.

MO 22.6 Fri 12:30 F142

Molecules for photodynamic therapy. Photophysical properties of 1O2 and 1,3-diphenylbenzofuran — •STEFAN STAN and VASILE CHIS — Babes-Bolyai University, Faculty of Physics, Cluj-Napoca, Romania

The first part of the study focuses on the calculation of the excitation

energies of the O2 molecule in the first and second excited states, using the [8,6]-CASSCF method and the complete basis set extrapolation technique. The calculated energies are 0.983 and 1.671 eV, in excellent agreement with the available experimental data (0.98 and 1.63 eV). As a result of the transition to the first excited state, the DPBF molecule is significantly planarized, and the bond lengths undergo significant changes only in the benzofuran group.

Several density functionals were tested for the reproduction of the photophysical parameters of the molecule, and the results clearly show that the cam-B3LYP functional provides the best agreement with the experimental data, for all photophysical parameters.

The first excited state of the molecule is due to the π - π^* transition between the HOMO-LUMO orbitals. The transition was described based on the natural transition orbitals and the difference between the electronic densities corresponding to the excited state and the fundamental one. An excellent agreement has been obtained between the experimental radiative lifetime (5.52 ns) and the calculated counterpart (5.34 ns) at PCM(DMSO-cam-B3LYP/6-311G(2d,p)//APFD/6-311+G(2d,p) level of theory.

MO 22.7 Fri 12:45 F142

Gas phase sugar synthesis: the formation of glycolaldehyde — •WEIQI WANG¹, XIANGYUE LIU¹, and JÉSUS PÉREZ-RÍOS² — ¹Fritz-Haber Institute, Max-Planck Society, Berlin — ²Stony Brook University, Stony Brook, New York

Gas-phase sugar synthesis is one of the most important reactions for the understanding of prebiotic chemistry in space. In this work, ab initio molecular dynamics has been employed to study the formation of glycolaldehyde from protonated formaldehyde, which is the first step of the carbon chain growth reaction. In particular, the isomerization of neutral formaldehyde molecule has been discussed. The reaction networks between neutral formaldehyde and protonated formaldehyde has been constructed to understand the formation of glycolaldehyde.

MO 22.8 Fri 13:00 F142

Kinetic and thermodynamic theory study of sequential reaction of Koop type hydroamination and [3 and 2] Meisenheimer rearrangement — •MORTEZA FARAHANI — Department of Physical Chemistry, Islamic Azad university, Malayer Branch, Malayer, Iran Farahanijokar@yahoo.com

Koop Norborne type hydroamination using N-allyl-N-methylhydroxylamine at 120 °C and tetrahydrofuran solvent leads to the production of unstable dipolar N-oxide intermediate which turns into a neutral product with higher stability after [3 and 2] Meisenheimer rearrangement. The kinetics and thermodynamics of this sequential reaction have been studied at the computational level of MN15L/aug-cc-pVTZ. The first stage of the reaction is exothermic and non-spontaneous, while the second stage is exothermic and spontaneous. The adverse effects of entropy and high temperature make the rate constant of the first stage, despite the lower activation energy, smaller than the second stage, and the balance is not towards the production of the dipolar N-oxide intermediate.

MO 23: Ultrafast Dynamics III (joint session MO/A)

Time: Friday 14:30–16:30

Location: F102

MO 23.1 Fri 14:30 F102

Photodissociation dynamics of CH₂Br — •LILITH WOHLFART, CHRISTIAN MATTHAEI, and INGO FISCHER — Julius-Maximilians-Universität Würzburg, 97074 Würzburg, Germany

Bromomethyl belongs to the class of organic halogen radicals. Therefore, it can potentially influence the atmosphere by reacting with the ozone layer and causing its depletion similar to HCFCs. The photoionization of bromomethyl was already investigated by several groups, including Steinbauer and coworkers. They determined the ionization energy and structure with VUV synchrotron radiation and investigated the dissociative photoionization. To obtain further insights into the dissociation of bromomethyl, we analyzed the fragments of the radical using velocity map imaging (VMI).

CH₂Br-NO₂ was used as a precursor for the halogenated methyl radical, because the weaker C-NO₂ bond can be cleaved through pyrolysis. Subsequently, laser light with 235 nm was deployed to dissociate the

formed CH₂Br radical. The major dissociation pathway gave the methylene and bromine fragments which were detected with SPI at 118 nm and [1+1']-REMPI at 356 nm respectively. With velocity map ion imaging, the translational kinetic energy distribution of the photofragments was determined. The recorded images of the bromine and methylene photofragments showed an anisotropic distribution, implying a direct dissociation.

MO 23.2 Fri 14:45 F102

Fragmentation of fulminic acid, HCNO, following core excitation and ionization — •DOROTHEE SCHAFFNER¹, MARIUS GERLACH¹, TOBIAS PREITSCHOPF¹, EMIL KARAEV¹, FABIAN HOLZMEIER², JOHN BOZEK³, and INGO FISCHER¹ — ¹Universität Würzburg, 97074 Würzburg — ²imec, 3001 Leuven, Belgium — ³Synchrotron SOLEIL, 91190 Saint-Aubin, France

In 2008 fulminic acid, HCNO, was first detected in space in the starless cores B1, L1544 and L183.^[1] Its isomer HNCO is also ubiquitous in

interstellar systems.^[2] Due to their composition of biogenic elements, the CHNO isomers have been proposed to play a prebiotic role as intermediates for organic life. Investigating the molecules' interaction with X-ray radiation is critical to understand their fate in space.

Here we report a study of the fragmentation of fulminic acid in the gas phase after resonant core excitation and core ionization on the K-edge of carbon, oxygen and nitrogen. The ionic fragmentation products formed after the Auger decay were investigated at the PLEIADES beamline at the synchrotron SOLEIL using Auger-electron/photoion coincidence spectroscopy. Branching ratios were determined which show a site-selective fragmentation upon ionization and excitation. Different fragmentation tendencies could be related to differences in the occupation of the Auger final states.

[1] N. Marcelino, J. Cernicharo, B. Tercero, E. Roueff, *Astrophys. J.* **2008**, 690, L27-L30.

[2] Nguyen-Q-Rieu, C. Henkel, J. M. Jackson, R. Mauersberger, *Astron. Astrophys.* **1991**, 241, L33-L36.

MO 23.3 Fri 15:00 F102

Ultrafast dynamics of OCS — ●WUWEI JIN¹, SEBASTIAN TRIPPEL^{1,3}, HUBERTUS BROMBERGER^{1,3}, TOBIAS RÖHLING¹, KAROL DLUGOLECKI¹, SERGEY RYABCHUK¹, ERIK MÄNSSON¹, ANDREA TRABATTONI¹, VINCENT WANIE¹, IVO VINKLÁREK⁴, FRANCESCA CALEGARI¹, and JOCHEN KÜPPER^{1,2,3} — ¹Center for Free-Electron Laser Science CFEL, Deutsches Elektronen-Synchrotron DESY, Hamburg — ²Department of Physics, Universität Hamburg — ³Center for Ultrafast Imaging, Universität Hamburg — ⁴Department of Chemical Physics and Optics, Faculty of Mathematics and Physics, Charles University, Prague, Czech Republic

Imaging ultrafast photochemical reactions with atomic-spatial and femtosecond-temporal resolution is one of the ultimate goals of physical chemistry and the molecular sciences [1]. We will discuss our ultrafast (sub 10 fs) time-resolved study of the photodissociation dynamics of carbonyl sulfide (OCS) after UV-photoexcitation at $\lambda = 267$ nm. OCS was purified and separated from the helium seed gas using the electrostatic deflector [2]. The UV-induced dynamics was studied through strong field ionization using a velocity map imaging spectrometer in combination with a Timepix3 camera [3].

[1] J. Onvlee, S. Trippel, and J. Küpper, *doi:10.1038/s41467-022-33901-w*, arXiv:2103.07171 [physics]

[2] YP Chang, D. Horke, S. Trippel, and J. Küpper, *Int. Rev. Phys. Chem.* **34**, 557 (2015), arXiv:1505.05632 [physics]

[3] H. Bromberger, *et al.*, *J. Phys. B Atomic Mol. Opt. Phys.* **55**, 144001 (2022), arXiv:2111.14407 [physics]

MO 23.4 Fri 15:15 F102

Ultrafast dynamics in iodomethane induced by few-fs ultraviolet pulses — ●SERGEY RYABCHUK^{1,2}, LORENZO COLAIZZI^{2,3}, ERIK P. MÄNSSON³, KRISHNA SARASWATHULA³, JESÚS GONZÁLEZ-VÁZQUEZ⁴, VINCENT WANIE³, ANDREA TRABATTONI^{3,5}, FERNANDO MARTÍN^{4,6,7}, and FRANCESCA CALEGARI^{1,2,3} — ¹The Hamburg Centre for Ultrafast Imaging, Hamburg, Germany — ²Universität Hamburg, Hamburg, Germany — ³DESY, Hamburg, Germany — ⁴Universidad Autónoma de Madrid, Madrid, Spain — ⁵Leibniz Universität Hannover, Hannover, Germany — ⁶IMDEA-Nanoscience, Madrid, Spain — ⁷IFIMAC, Madrid, Spain

Iodomethane has been extensively used as a model system to study photodissociation dynamics by ultraviolet (UV) light excitation. The molecule is prompted to rapid fragmentation along the C-I bond due to the repulsive character of the neutral states accessed by single UV photon absorption in the energy range of 4.1-5.4 eV. In this work, we used 4 fs UV pulses with central energy of 4.9 eV as a pump with delayed few-cycle infrared pulses, probing the dynamics via multi-photon ionization. The dynamics of methyl and iodine fragments allow us to track the field-free wavepacket evolution on the neutral states with unprecedented time resolution. Moreover, the experimental observations combined with a theoretical study revealed that the implementation of such ultrashort pulses allows for the production of an intact parent ion with a 5-fs lifetime, preventing molecular cleavage. This becomes possible only in a narrow time window close to the Frank-Condon regime before the dissociation takes place.

MO 23.5 Fri 15:30 F102

Photoemission chronoscopy of the Iodoalkanes — ●CHRISTIAN SCHRÖDER, MAXIMILIAN POLLANKA, PASCAL SCIGALLA, ANDREAS DUENSING, MICHAEL MITTERMAIR, MAXIMILIAN FORSTER, MATTHIAS OSTNER, and REINHARD KIENBERGER — Physik Department, Tech-

nische Universität München, James-Frank-Str. 1, 85748 Garching, Germany

Photoemission timing measurements on primary and secondary iodoalkanes up to 2-iodobutane are performed in the gas phase and reveal an unexpected and yet unexplained dependency of the I4d photoemission time delay τ_{I4d} on the molecular species.

The experiment is carried out at photon energies around the giant resonance in the I4d $\rightarrow \epsilon f$ channel, which is expected to be largely indifferent to its chemical environment. If true, observable differences in the I4d photoemission time between molecules should be a direct consequence of differences experienced by the leaving photoelectron during its propagation alone, and not due to a distortion of its initial bound state.

We find a strong variation of τ_{I4d} between different molecules, but no clear correlation with the attached functional group's size as it has been suggested by previous theoretical studies (S. Biswas *et al.*, *Nature Physics* **16** (2020)).

MO 23.6 Fri 15:45 F102

X-ray diffractive imaging of UV-induced ultrafast dynamics in CF₂I₂ — ●NIDIN VADASSERY^{1,3}, SEBASTIAN TRIPPEL^{1,2}, and JOCHEN KÜPPER^{1,2,3} — ¹Center for Free-Electron Laser Science CFEL, Deutsches Elektronen-Synchrotron DESY, Hamburg — ²Department of Physics, Universität Hamburg — ³Department of Chemistry, Universität Hamburg

Disentangling chemical dynamics, including the traversal of transition states, provides important insight into (bio)chemical processes. Roaming for example, a proposed ultrafast mechanism occurring in unimolecular photodissociation, follows an unusual reorientation motion after the excitation near bond dissociation energies [1]. Difluorodiodomethane (CF₂I₂), has shown roaming mechanism following excitation with 350 nm light.

X-ray pulses generated at XFELs provide the opportunity to study such ultrafast rearrangements on the atomic scale with femtosecond resolution by diffractive imaging. Here, we present a computational proposal of the time-resolved coherent x-ray diffractive imaging of the photodissociation of CF₂I₂ using CMI diffract, an in-house python software to predict and analyze molecular-ensemble diffraction patterns[2]. We also detail efforts to produce a pure sample of CF₂I₂ using the electric deflector in the eCOMO end-station, in preparation for UV-pump x-ray-probe studies at EuXFEL.

[1] D. Townsend, *et al.*, *Science* **306**, 1158, (2004)

[2] J. Küpper, *et al.*, *Phys. Rev. Lett.* **112**, 083002, (2014), arXiv:1307.4577

MO 23.7 Fri 16:00 F102

Dynamics of H₂-roaming processes, H₃⁺ formation, and cationic fragmentation in ethanol and aminoethanol initiated above and below the double-ionization threshold — ●AARON NGAI¹, JAKOB ASMUSSEN², BJÖRN BASTIAN², MATTEO BONANOMI^{3,4}, CARLO CALLEGARI⁵, MICHELE DI FRAIA⁵, KATRIN DULITZ^{1,6}, RAIMUND FEIFEL⁷, SARANG GANESHAMANDIRAM¹, SEBASTIAN HARTWEG¹, SIVARAMA KRISHNAN⁸, AARON LAForge⁹, FRIEDEMANN LANDMESSER¹, BEN LTAIEF LTAIEF², MORITZ MICHELBAACH¹, NITISH PAL⁵, OKSANA PLEKAN⁵, NICOLAS RENDLER¹, FABIAN RICHTER¹, AUDREY SCOGNAMIGLIO¹, TOBIAS SIXT¹, RICHARD SQUIBB⁷, AKGASH SUNDARALINGAM², FRANK STIENKEMEIER¹, and MARCEL MUDRICH² — ¹Physikalisches Institut, Albert-Ludwigs-Universität Freiburg, Germany — ²Department of Physics and Astronomy, Aarhus University, Denmark — ³Dipartimento di Fisica and CIMaInA, Università degli Studi di Milano, Italy — ⁴Istituto di Fotonica e Nanotecnologie, CNR-IFN, Milano, Italy — ⁵Elettra - Sincrotrone Trieste S.C.p.A., Basovizza, Trieste, Italy — ⁶Institut für Ionenphysik und Angewandte Physik, Universität Innsbruck, Austria — ⁷Department of Physics, University of Gothenburg, Sweden — ⁸Department of Physics, Indian Institute of Technology Madras, Chennai, India — ⁹Department of Physics, University of Connecticut, US

The trihydrogen cation (H₃⁺) is the simplest and one of the most abundant triatomic cation in the universe. It plays a crucial role in interstellar gas-phase chemistry as it facilitates molecule-forming chemical reactions. Building upon the work of Ekanayake [1], we further investigated the competition between pathways leading to H₃⁺ formation in doubly ionized ethanol and 2-aminoethanol molecules and their respective clusters using time-resolved XUV-UV pump-probe spectroscopy. While formation of H₃⁺ in doubly-ionized alcohol molecules is due to intramolecular H₂-roaming, H₃⁺ formation in clusters likely occurs

via more complicated intermolecular pathways involving fragmentation and recombination of excited ionic fragments e.g. in nanoplasmas [2]. We compare results between XUV-photoionization below and above the double-ionization threshold, including the lifetimes of intermediate states. Notably, we report the absence of H_3^+ -formation in aminoethanol, and the suppression of H_2 -roaming in ethanol clusters.

[1] Ekanayake, N. *et al. Nature Comm.* **9**, 5186 (2018).

[2] Michiels, R. *et al. Phys. Chem. Chem. Phys.* **22**, 7828 (2020).

MO 23.8 Fri 16:15 F102

Probing well aligned molecular environments on surfaces in the time-domain — ●PASCAL SCIGALLA¹, CHRISTIAN SCHRÖDER¹, SVEN PAUL¹, PETER FEULNER², and REINHARD KIENBERGER¹ — ¹Chair for laser and x-ray physics, E11, Technische Universität München, Germany — ²Surface and Interface Physics, E20, Technische Universität München, Germany

We report on the photoemission timing measurements of well-aligned

iodomethane and -ethane molecules on a Pt111 surface. In this set of experiments we clock the $I4d$ photoemission of iodine against the Platinum valence photoemission using the attosecond streak camera technique, allowing the extraction of a relative photoemission delay. As the $I4d$ photoemission in the selected energy range is dominated by a giant resonance in the $I4d \rightarrow ef$ channel its photoemission time is mostly unaffected by its chemical environment. Thus any observed change in the photoemission delay can be attributed to the traversed potential landscape of the molecule. By carefully selecting the detection angle and crystal surface coverage we can reliably choose whether only parts of the molecule or its entirety was traversed by the detected photoelectron wavepackets. It is furthermore possible to investigate the influence of slight coverage variations onto the observed photoemission delay. Planned, complementary scattering simulations will be used to gain deeper insight into the observations with the goal to establish photoemission timing experiments as an efficient and accurate means to study molecular environments on surfaces.