

MO 28: Ultrafast Dynamics IV

Time: Friday 11:00–12:45

Location: HS XVI

MO 28.1 Fri 11:00 HS XVI

Generation of atomic coherence by ultrafast molecular photodissociation probed by heterodyned detected attosecond four-wave-mixing spectroscopy — ●FRANCESCO MONTORSI^{1,2}, PATRICK RUPPRECHT³, LEI XU⁴, NIRI GOVIND⁴, SHAUL MUKAMEL⁵, MARCO GARAVELLI², DANIEL M. NEUMARK³, STEPHEN R. LEONE³, and DANIEL KEEFER¹ — ¹Max-Planck-Institut für Polymerforschung, 55128 Mainz, Germany — ²Dipartimento di Chimica industriale Toso Montanari, Università di Bologna, 40136 Bologna, Italy — ³Department of Chemistry, University of California, Berkeley, California 94720, USA — ⁴Physical and Computational Sciences Directorate, Pacific Northwest National Laboratory, Richland, Washington 99352, United States — ⁵Department of Chemistry, Physics and Astronomy, University of California, Irvine, California 92697, USA

Electronic coherences (EC) in molecules can naturally form, after photoexcitation, in the vicinity of so-called conical intersections (CIs). Here, we theoretically investigate the evolution of EC generated during a photochemical process namely the bond cleavage in methyl iodine. Quantum dynamics simulations show that the coherence spawned in the molecule by a CI is rapidly transferred to the atomic iodine upon dissociation. This allows the preparation of a long-lived EC, making it interesting for quantum technology applications. Finally, we propose a spectroscopic scheme based on the interference of two attosecond four-wave mixing signals which enables a background-free tomography of such EC providing crucial information about its generation at the molecular CI

MO 28.2 Fri 11:15 HS XVI

Absolute Photoemission Timing of Surface-Oriented Iodoalkanes on Pt(111) — PASCAL FREISINGER¹, ●SVEN-JOACHIM PAUL¹, CHRISTIAN SCHRÖDER², KONSTANTIN SEIDENFUS¹, PETER FEULNER³, and REINHARD KIENBERGER¹ — ¹Chair for laser and x-ray physics, E11, Technische Universität München, Germany — ²University of California, Berkeley, USA — ³Surface and Interface Physics, E20, Technische Universität München, Germany

We report on attosecond streaking measurements of the electron photoemission process from the platinum (111) surface covered by surface-oriented iodoalkanes. Attosecond streaking enables measuring relative time delays in photoemission from two energetically different bound electronic states. This experiment investigated how the photoemission delay between the platinum valence band and the iodine 4d state in iodomethane, iodoethane, and atomic iodine depends on surface coverage.

Both Iodoalkanes align horizontally or vertically to the Pt(111) surface, depending on coverage. Therefore, changing the surface coverage changes the surface orientation of adsorbed iodomethane or iodoethane, thus enabling measuring photoemission delays under a fixed emission angle. This overcomes common attosecond streaking experiments in the gas phase, where the photoemission delay is averaged over the full solid angle.

Streaking atomic iodine enables referencing an established timing scheme in the gas phase, making retrieved photoemission delays comparable to gas phase measurements.

MO 28.3 Fri 11:30 HS XVI

Time resolved attosecond photoemission of isosteric molecules — ●MAXIMILIAN POLLANKA¹, CHRISTIAN SCHRÖDER¹, MAXIMILIAN FORSTER¹, PASCAL FREISINGER¹, SVEN-JOACHIM PAUL¹, ZDENEK MASIN², JAKUB BENDA², and REINHARD KIENBERGER¹ — ¹Physik-Department, Technische Universität München, Garching, Germany — ²Institute of Theoretical Physics, Faculty of Mathematics and Physics, Charles University, Prague, Czech Republic

In this work we report on the absolute photoemission timing of the outer valence states in the isosteric molecules CO₂ and N₂O utilizing attosecond streaking spectroscopy. We investigated on the photon energy dependant emission times using helium and iodomethane as a timing reference. Employing the time-dependant R-matrix approach, the channel-coupling effect turned out to be a significant delay contribution in CO₂ due to the resonant coupling of the NIR field with the B and C states separated by 1.3 eV. Based on the theoretical calculations we could disentangle the respective delay contribution of single states in an experimentally accessible mixed state, where in both mod-

ecules the Π orbital turned out to be the dominant electron supplier. By comparing the absolute photoemission times retrieved with both chronoscopes He and CH₃I, we could proof the validity of using HeI_s as well as I4d as complementary reference photolines. The great agreement of the absolute delays obtained with both timing references make us confident for further exploiting the I4d in prospective absolute timing experiments of outer valence states in molecules.

MO 28.4 Fri 11:45 HS XVI

Asymmetry parameter of the iodoalkanes — ●MAXIMILIAN FORSTER¹, MAXIMILIAN POLLANKA¹, SVEN-JOACHIM PAUL¹, PASCAL FREISINGER¹, CHRISTIAN SCHRÖDER¹, MICHELE ALAGIA², ALESSIO BRUNO³, ROBERT RICHTER², STEFANO STRANGES³, and REINHARD KIENBERGER¹ — ¹Technische Universität München — ²Elettra Synchrotron Trieste — ³Sapienza University of Rome

We measured the asymmetry parameter of photoemission from the Iodine 4d state in four different Iodoalkanes across an energy range of 60 to 160 eV.

We previously conducted photoemission delay measurements of the Iodoalkanes in our group using attosecond streaking. There we see a dependence on the species, the molecular surrounding of the iodine atom influences its photoemission delay. Since photoemission delay is interpreted as scattering delay, intramolecular scattering of the outgoing photoelectron is a logical explanation. It has been shown experimentally that the molecular environment can affect the asymmetry parameter, most likely also through intramolecular scattering. By measuring the asymmetry parameter we can compare the behavior to our previous measurements and clearly identify intramolecular scattering as the underlying cause for the behavior both in the time and the spectral domain. While we saw an effect, it is not the same as in our measurements, therefore the picture is not as intuitive as we had hoped.

The measurements were conducted at the Elettra synchrotron using the gas phase endstation.

MO 28.5 Fri 12:00 HS XVI

Time- and energy-resolved fluorescence measurements of collective effects in organic aggregates attached to rare gas clusters — ●ALEKSANDR DEMIANENKO, MORITZ MICHELBAACH, 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. A complete understanding of the energy level structure, and decay mechanisms require high spectral and temporal resolution. We present a setup combining conventional laser-induced fluorescence (LIF) spectroscopy with time-correlated single photon counting (TC-SPC) and discuss advantages and implementation challenges. Using wavelength-tunable nanosecond dye laser pulses allows us to measure high-resolution LIF spectra of transitions to highly excited states of tetracene embedded in superfluid helium nanodroplets, or deposited on solid rare-gas clusters. The newly implemented TCSPC detection in combination with a femtosecond laser system is aimed at studying radiative and non-radiative decay mechanisms connected to collective effects in aggregates of polyacenes. This technique allows us to cover the sub-ns lifetime region not previously reachable in our fluorescence measurements.

MO 28.6 Fri 12:15 HS XVI

A systematic study of vibrational decoherence of PTCDA in different environments using two-dimensional electronic spectroscopy — ●YILIN LI, ARNE MORLOK, ULRICH BANGERT, JAKOB GERLACH, BRENDAN WOUTERLOOD, FRANK STIENKEMEIER, and LUKAS BRUDER — Institute of Physics, University of Freiburg, Germany

We present a systematic study of the coupling and energy dissipation of vibrational modes in different environments, ranging from cold isolated molecules in the gas phase, to molecules embedded in different rare-gas cluster species and molecules dissolved in a solution at room temperature. As test molecule we chose 3,4,9,10-perylene tetracarboxylic dianhydride (PTCDA) and apply femtosecond pump probe and two-dimensional electronic spectroscopy (2DES) [1,2]. This

approach provides insight into the various aspects of vibrational decoherence, e.g. pure dephasing processes and energy dissipation into the environment, and can be applied in all environments, allowing for a direct comparison of the experimental results.

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

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

MO 28.7 Fri 12:30 HS XVI

Anomalous diffusion of free-base phthalocyanine on rare-gas clusters — ●ARNE MORLOK, ULRICH BANGERT, PHILIPP ELSÄSSER, YILIN LI, FELIX RIEDEL, TANJA SCHILLING, LUKAS BRUDER, and FRANK STIENKEMEIER — University of Freiburg, Institute of Physics, Hermann-Herder-Straße 3, 79104 Freiburg, Germany

Precise knowledge and control of the environment are essential in

diffusion studies, as interactions between a diffusing particle and its surroundings can lead to anomalous diffusion, deviating from simple Brownian motion. To address this, we employ cluster-isolation spectroscopy, which provides well-studied and weakly interacting rare-gas clusters as the environment. Experiments are conducted with solid rare-gas clusters of argon and neon doped with free-base phthalocyanine (H₂Pc) and interrogated with action-based two-dimensional electronic spectroscopy. The high spectro-temporal resolution of this technique allows us to resolve subtle changes in binding configurations with femtosecond precision. We report an anomalous diffusion behavior of H₂Pc in both of the dopant-host nanosystems on a picosecond timescale. Molecular dynamics simulations confirm these findings and further reveal that movement of H₂Pc is confined to rotations on a single facet of the icosahedral host cluster.