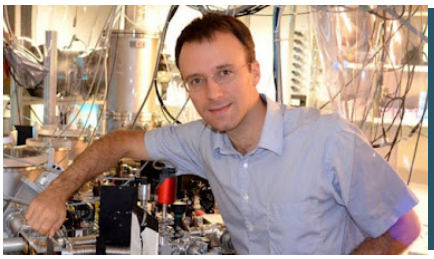
New Horizons Solvay Lectures in Chemistry



Prof. Hans Jakob Wörner
(ETH Zürich, Switzerland)

Hans Jakob Wörner's main research focus is the ultra-fast spectroscopy of molecules with an attosecond time resolution (1as = 10-18 s) and the development of new experimental methods to characterize the structure and dynamics of the valence shell of molecules. He has been an Assistant Professor at the Laboratory of Physical Chemistry in the Department of Chemistry and Applied Biosciences since 2010. He studied chemistry at ETH Zurich, graduating in 2003 and obtaining a doctorate in physical chemistry in 2007. From 2007, he worked as a postdoc at the Laboratoire Aimé Cotton of the CNRS (Centre national de la recherche scientifique) in Orsay, France, and the National Research Council in Ottawa, Canada.

Tuesday 5 October 2021 at 4.00 pm. (Online)

Attosecond charge migration and its interaction with nuclear motion: towards attochemistry

Charge migration is a periodic rearrangement of the charge distribution in molecules, driven by a coherent superposition of electronic states. It can be prepared in a variety of ways, including temporally confined ionization, excitation or through electron correlation. In this lecture, I will discuss the experimental measurement and reconstruction of attosecond charge migration in spatially oriented molecules [1]. These measurements revealed the essentially complete migration of an electron hole from one side of the iodoacetylene cation to the other in less than one femtosecond. Turning from high-harmonic spectroscopy to attosecond transient-absorption spectroscopy, I will discuss the experimental observation of decoherence and revival of attosecond charge migration driven by nuclear motion in the neutral silane molecule [2]. These results demonstrate a broadly applicable approach to inducing and probing charge migration in molecules, opening the door to controlling molecular dynamics on the electronic time scale. An outlook on the prospects of attochemistry will be given.

[1] P. M. Kraus, B. Mignolet, D. Baykusheva, A. Rupenyan, L. Horný, E. F. Penka, G. Grassi, O. I. Tolstikhin, J. Schneider, F. Jensen, L. B. Madsen, A. D. Bandrauk, F. Remacle, H. J. Wörner, Science 350, 790 (2015).
[2] D. Matselyukh, V. Despré, N. V. Golubev, A. I. Kuleff, H. J. Wörner, submitted (2021)

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Tuesday 12 October 2021 at 4.00 pm. (Online)

Attosecond time delays in molecules, clusters and liquids: towards electronic dynamics in solutions

Attosecond spectroscopy has enabled the observation of ionization dynamics on their natural time scale. These delays convey rich information on the scattering potential of the ionized system, as well as electronic correlations. In this lecture, I will discuss the experimental measurement of time delays in molecules [1]. Turning from gases to liquids, I will discuss the measurement of photoionization delays from liquid water [2] and the development of theoretical methods for their interpretation. Extending attosecond spectroscopy to size-resolved water clusters provided a bridge between the gas and liquid phases and yielded a molecular-level understanding of the mechanisms governing photoionization dynamics in the condensed phase [3]. A systematic correlation between the ionization delay and the spatial extension of the electron hole has been found, which suggests the possibility of studying electron-hole dynamics in weakly bound clusters and liquids with attosecond temporal resolution. These methods establish an experimental pathway to time-resolved studies of electronic dynamics in (micro-)solvated molecules.

[1]M. Huppert, I. Jordan, D. Baykusheva, A. von Conta, H. J. Wörner, Phys. Rev. Lett. 117, 093001 (2016)
[2] I. Jordan, M. Huppert, D. Rattenbacher, M. Peper, D. Jelovina, C. Perry, A. von Conta, A. Schild, H. J. Wörner, Science 369, 974 (2020)
[3] X. Gong, S. Heck, D. Jelovina, C. Perry, K. Zinchenko, H. J. Wörner, submitted (2021), arxiv.org/abs/2106.09459

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Tuesday 19 October 2021 at 4.00 pm. (Online)

Attosecond soft-X-ray and high-harmonic spectroscopies: bridging the complexity gap

One of the remaining challenges for attosecond spectroscopy is its extension to complex systems, such as large molecules, molecular aggregates or nanoparticles in solution. X-ray spectroscopy offers an attractive approach to this goal, owing to its element specificity and site sensitivity. In this lecture, I will discuss the development of table-top soft-X-ray spectroscopy and its application to observing the rearrangement of unoccupied molecular states during chemical reactions [1]. The generation of isolated attosecond soft-X-ray pulses with a duration of only 43 attoseconds has established the current world record of the shortest light pulse ever measured [2]. The spectroscopic application of such pulses has revealed the fastest conical-intersection dynamics observed to date, i.e. the sub-7-femtosecond electronic relaxation from the A to the X state in the ethylene cation [3]. Turning from the gas phase to the liquid phase, I will discuss recent results including the observation of suppressed dissociation of ionized pyridine in solution, as well as the ultrafast proton transfer of ionized urea dimers in aqueous solution. These results demonstrate the potential of attosecond soft-X-ray spectroscopy in addressing complex systems.

[1] Y. Pertot, C. Schmidt, M. Matthews, A. Chauvet, M. Huppert, V. Svoboda, A. von Conta, A. Tehlar, D. Baykusheva, J.-P. Wolf, H. J. Wörner, Science 355(6322), 264-267 (2017)
[2] T. Gaumnitz, A. Jain, Y. Pertot, M. Huppert, I. Jordan, F. Ardana-Lamas, H. J. Wörner, Opt. Exp. 25(22), 27506 (2017)
[3] K. S. Zinchenko, F. Ardana Lamas, I. Seidu, S. P. Neville, J. van der Veen, V. Utrio Lanfaloni, M. S. Schuurman, H. J. Wörner, Science 371 (6528), 489 (2021)

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