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Attosecond dynamics of electrons in molecules and liquids[1]

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The ultrafast motion of electrons and holes following light-matter interaction is fundamental to a broad range of chemical and biophysical processes. In this lecture, I will discuss two recent experiments carried out in our group that measure the atomic-scale motion of charge with attosecond temporal resolution (1 as = 10^{-18} s). The first experiment is carried out on isolated, spatially oriented molecules in the gas phase. We advance high-harmonic spectroscopy to resolve spatially and temporally the migration of an electron hole immediately following ionization of iodoacetylene, while simultaneously demonstrating extensive control over the process. A multidimensional approach, based on the measurement of both even and odd harmonic orders, enables us to reconstruct both quantum amplitudes and phases of the electronic states with a resolution of ∼100 as. We separately reconstruct quasi-field-free and laser-controlled charge migration as a function of the spatial orientation of the molecule and determine the shape of the hole created by ionization [2]. The second experiment is carried out on a free-flowing microjet of liquid water. We use an attosecond pulse train synchronized with a near-infrared laser pulse to temporally resolve the process of photoemission from liquid water using the RABBIT technique. We measure a delay on the order of 50 as between electrons emitted from the HOMO of liquid water compared to that of gas-phase water and a substantially reduced modulation contrast of the corresponding sidebands. Since our measurements on solvated water molecules are referenced to isolated ones, the measured delays reflect (i) the photoionization delays caused by electron transport through the aqueous environment and (ii) the effect of solvation on the parent molecule. The relative modulation contrast, in turn, contains information on (iii) the modification of transition amplitudes and (iv) dephasing processes. These experiments make the liquid phase and its fascinating mechanisms accessible to attosecond science [3].

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[2]P. M. Kraus, B. Mignolet, D. Baykusheva, A. Rupenyan, L. Horny, E. F. Penka, G. Grassi, O. I. Tolstikhin, J. Schneider, F. Jensen, L. B. Madsen, A. D. Bandrauk, F. Remacle, and H. J. Wörner, *Science* **350**, 790 (2015)

[3]I. Jordan, M. Huppert, A. von Conta, L. Seiffert, M. Arbeiter, Th. Fennel and H.J. Wörner, *to be published*