Ultrafast Dynamics - Tracking motion on fs-timescales and below

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Molecular dynamics take place on fs-timescales, electronic dynamics even reach down to attosecond levels. To study such ultrafast processes a variety of techniques have been developed. In my talk I will give an introduction to two such techniques, High Harmonic Generation and Free Electron Lasers, highlight select experiments and give an outlook on future developments.

Purpose: Write-Up to the author's talk in the physics master seminar module at Heidelberg University.

For more than a century people have been interested in capturing faster and faster processes on picture or video. In 1878 Eadweard Muybridge published his famous cabinet series "The Horse in Motion", for the first time resolving dynamics faster than what the human eye can follow[1]. Nowadays specialized camera systems are able to resolve processes ocurring at the ns-timescale and at the very high-end even at the ps-timescale[2,3].

In contrast to movies of single events taken by typical camera systems, a different imaging approach has to be employed to access the fs- and as-timescales, so-called pump-probe schemes (see Fig.1). Not a single-event is imaged frame after frame but instead a repeatable(!) process is triggered many times and snapshots are taken at varying time delays. For detection either spectrometers or momentum space imaging techniques can be employed.

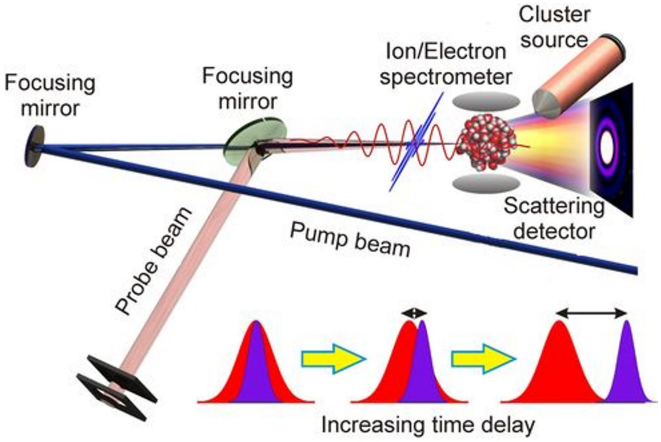

FIG. 1. Setup of a typical pump-probe experiment. A pump beam excites the system under study, triggering a repeatable process. A probe beam serves to take a snapshot of the system at a given time delay, which can be adjusted using a delay line for example. Copyright AG Krikunova and AG Möller, TU Berlin.

Using pump-probe schemes it is possible to study among others the dissociation of molecules, charge migration upon photoionization, molecular dynamics or movements of electronic wavepackets[4–11]. To that end a source of high intensity coherent light pulses of (sub-)fs duration in the UV/XUV or the X-ray regime is needed. Producing such light pulses is far from easy but there are two widespread methods for doing so: high harmonic generation (HHG), which is limited to the UV/XUV regime, and free electron lasers (FELs).

At intensities of $10^{14} - 10^{15}$ W/cm^2 the potential of an oscillating laser field has comparable magnitude to the Coulomb potential that binds electrons to the atom. By tunnel ionization, subsequent acceleration due to the force exerted by the laser beam on the electron and ultimately recollision of the electron into a bound state, a plateau of high harmonics of the fundamental laser frequency can be generated[4,5,12]. By employing different filtering and phase-matching techniques the resulting pulse can be compressed down to attosecond duration[13].

The motion of electrons after tunnel ionization has been beautifully visualized by recording diffraction images of electrons scattering back with the emitter atom[11]. One example of momentum space imaging is the measurement of electron-hole wavepacket dynamics in Ar^+ using a COLTRIMS (Cold Target Recoil Ion Momentum Spectroscopy) setup[7]. A further intriguing imaging technique is self-probing spectroscopy, wherein the high harmonics' spectrum is used to infer the dipole moments, in which e.g. the dissociation dynamics of molecules are encoded[8].

The highest-energy harmonic is limited to the cut-off energy $E_{max} = I_p + 3.17U_p$, where I_p is the ionization potential and U_p is the average kinetic energy of the electron in the laser field[12]. Furthermore the intensity of light produced by HHG is limited by the process' low efficiency[4]. To generate coherent light pulses at higher energies and intensities, large-scale facilities such as FELs are needed.

An FEL accelerates electrons from a particle accelerator in a wiggling fashion, thereby producing X-ray light. Using self-amplified spontaneous emission in undulators and different seeding techniques coherent light can be created[14,15]. With FELs events such as photochemical ring openings in 1,3-cyclohexadiene can be studied with sub-Ångström spatial and fs-temporal resolution - bringing us closer to capturing "molecular movies"[10].

Many fascinating dynamics are currently being investigated using ultrafast imaging techniques. Newly constructed large-scale facilities such as SLAC's LCLS-II and novel concepts, e.g. single-cycle attosecond pulses, promise an exciting future[16].

[1] E. Muybridge, The Horse in motion. "Abe Edgington," owned by Leland Stanford; driven by C. Marvin, trotting at a 2:24 gait over the Palo Alto track, 15th June 1878 / Muybridge, *Library of Congress, Washington, D.C. 20540 USA*

[2] Specialised Imaging Ltd., Pushing ultra-highspeed photography to the limit., `https://www.specialised-imaging.com/about-us/news/Pushing-ultra-high-speed-photography-to-the-limit`, Retrieved on 5th July 2021

[3] J. Liang, L. Zhu & L.V. Wang, Single-shot real-time femtosecond imaging of temporal focusing, *Light Sci Appl* **7**, 42 (2018).

[4] M.J.J. Vrakking, Attosecond Imaging, *Phys. Chem. Chem. Phys.* **16**, 2775-2789 (2014)

[5] P. Salières, A. Maquet, S. Haessler *et al.*, Imaging orbitals with attosecond and Ångström resolutions: toward attochemistry?, *Rep. Prog. Phys.* **75**, 062401 (2012)

[6] E. Goulielmakis, ZH. Loh, A. Wirth *et al.*, Real-time observation of valence electron motion, *Nature* **466**, 739–743 (2010)

[7] M. Kübel, Z. Dube, A.Yu. Naumov *et al.*, Spatiotemporal imaging of valence electron motion, *Nat Commun* **10**, 1042 (2019)

[8] S. Baker, J.S. Robinson, C.A. Haworth *et al.*, Probing Proton Dynamics in Molecules on an Attosecond Time Scale, *Science* **312**, 424-427 (2006)

[9] L. Belshaw, F. Calegari, M.J. Duffy *et al.*, Observation of Ultrafast Charge Migration in an Amino Acid, *J. Phys. Chem. Lett.* **3**, 24, 3751–3754 (2012)

[10] T.J.A. Wolf, D.M. Sanchez, J. Yang *et al.*, The photochemical ring-opening of 1,3-cyclohexadiene imaged by ultrafast electron diffraction, *Nat. Chem.* **11**, 504–509 (2019)

[11] J. Mauritsson, P. Johnsson, E. Mansten *et al.*, Coherent Electron Scattering Captured by an Attosecond Quantum Stroboscope, *Phys. Rev. Lett.* **100**, 073003 (2008)

[12] P.B. Corkum, Plasma perspective on strong field multiphoton ionization, *Phys. Rev. Lett.* **71**, 13, 1994–1997 (1993)

[13] P.M. Paul, E.S. Toma, P. Breger *et al.*, Observation of a Train of Attosecond Pulses from High Harmonic Generation, *Science* **292**, 1689-1692 (2001)

[14] J. Ullrich, A. Rudenko & R. Moshammer, Free-Electron Lasers: New Avenues in Molecular Physics and Photochemistry, *Ann. Rev. Phys. Chem.* **63**, 635 (2012)

[15] K. Zhukovsky, Undulators for Short Pulse X-Ray Self-Amplified Spontaneous Emission-Free Electron Lasers, High Energy and Short Pulse Lasers, R. Viskup, *IntechOpen* (2016)

[16] A. Mak, G. Shamuilov, P. Salén *et al.*, Attosecond single-cycle undulator light: a review, *Rep. Prog. Phys.* **82**, 025901 (2019)