

UV Dual-Comb Spectroscopy with Femtosecond Temporal Resolution

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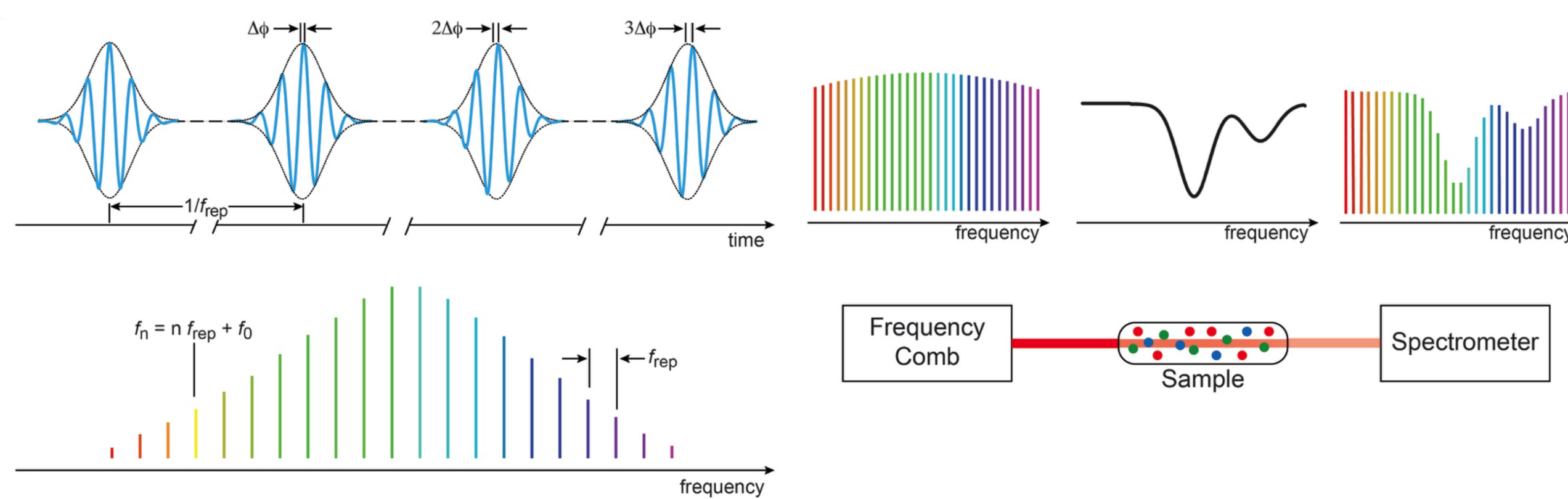
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Motivation

The project aims at combining the unique advantages of ultraviolet (UV) Dual-Comb Spectroscopy (DCS) concerning its fast acquisition time and unprecedented spectral resolution in the UV with a state-of-the-art temporal resolution in the femtosecond (fs) regime. This unique combination of ultrahigh spectral and temporal resolution paves the way for the observation of subtle effects as for example wave packet dynamics in gases. The observation of which has so far been impeded by the insufficient resolution of existing UV grating spectrographs.

Frequency Combs

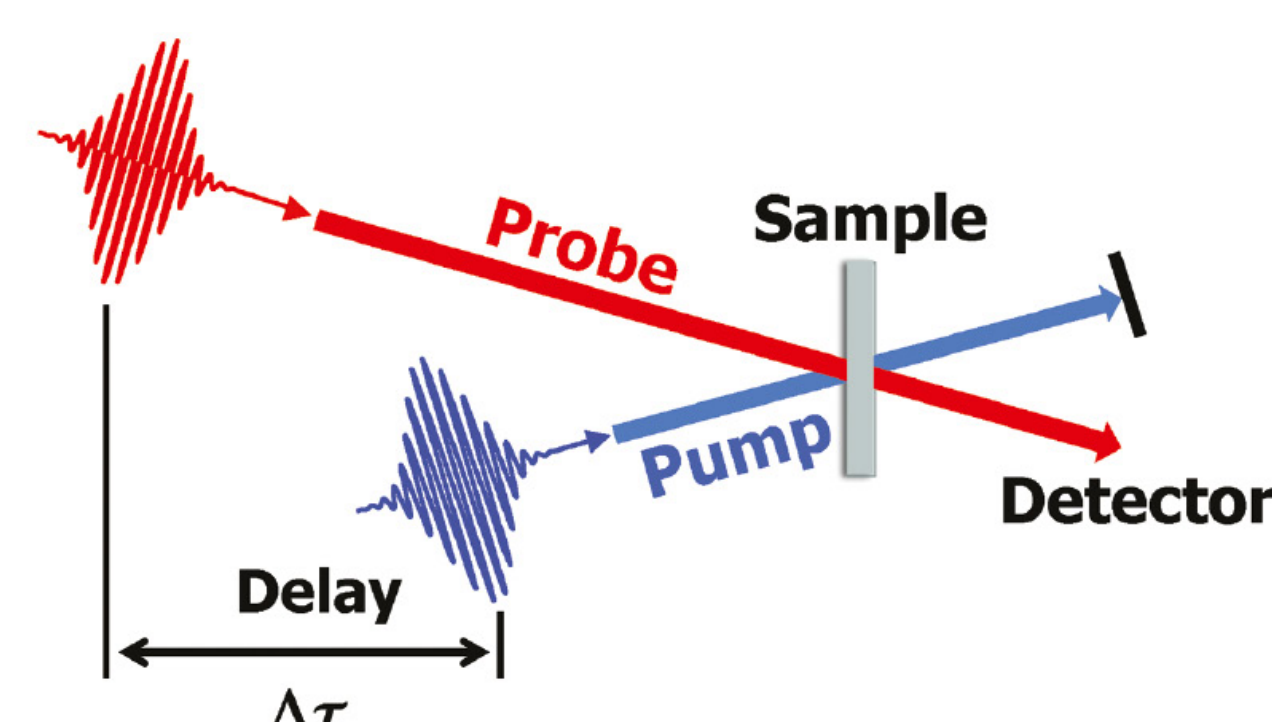
- **Description**
 - Consists of equidistant lines in the frequency domain
 - Is not a continuum
 - Pulsed or cw seed laser
- **Properties**
 - Spacing surpasses spectrometer resolution
 - Discrete in frequency domain, train of pulses in time domain



Usually appearing quite ordinary when viewed from a spectrometer, frequency combs actually consist of a vast amount of narrowly spaced laser lines [3].

Pump-Probe Spectroscopy

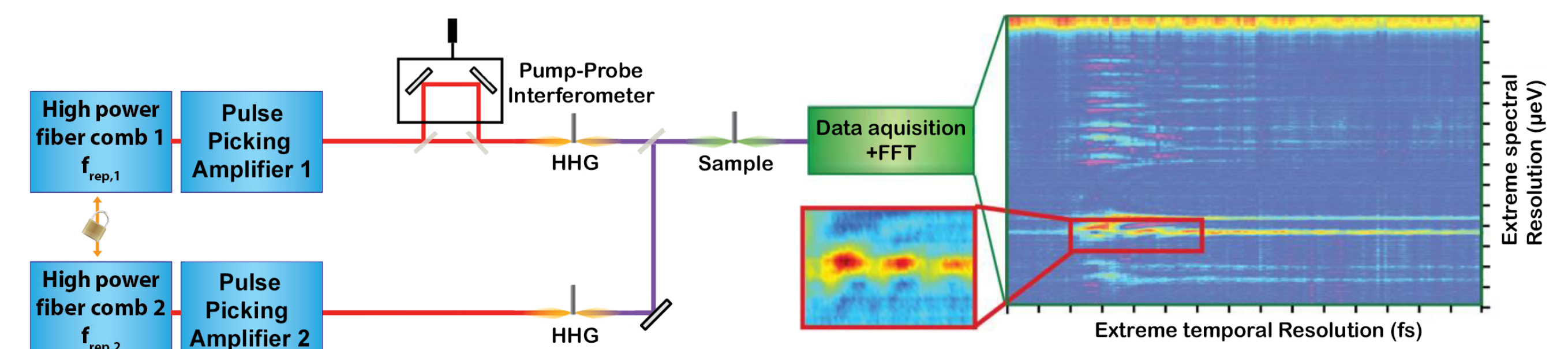
- **Methodology**
 - Illumination of a sample with two individual laser pulses
 - High energy, narrowband pump beam
 - Lower energy, broadband probe beam
 - Femto- to nanosecond temporal delay between pulses
 - First, one induces change in a sample, then measures it



Via this experimental method fundamental physical processes, such as the lifetimes of excited states or ultrafast electron processes, become accessible [5].

References

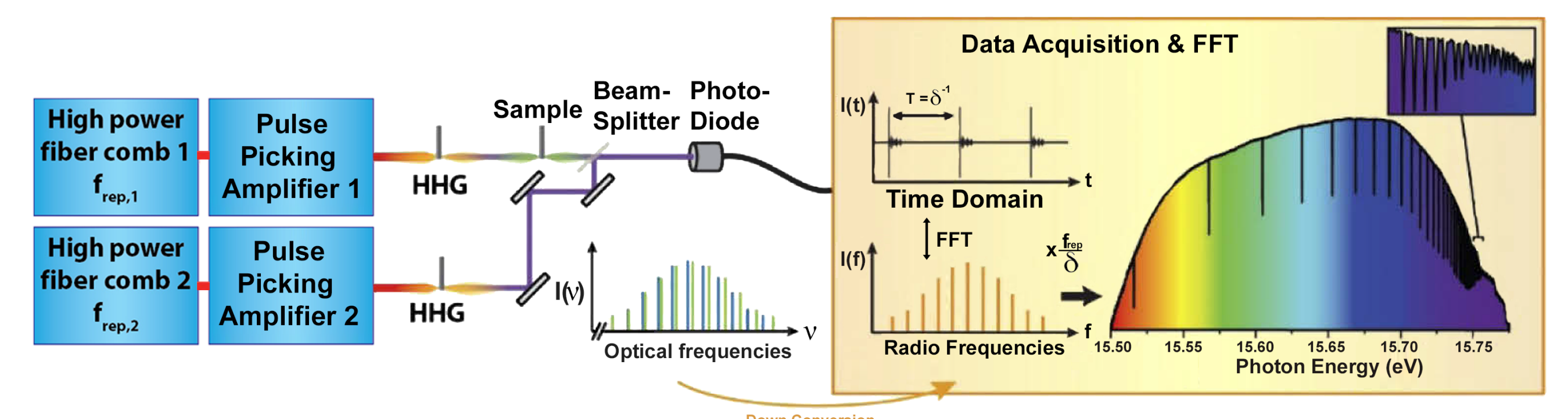
- [1] Bernhardt, B., private communication. Figure adapted.
- [2] Beck, A.R., et al. "Attosecond transient absorption probing of electronic superpositions of bound states in neon: detection of quantum beats", *New J. Phys.* 16 113016 (2014). <https://doi.org/10.1088/1367-2630/16/11/113016>
- [3] Picqué, N., Hänsch, T.W. "Frequency comb spectroscopy", *Nature Photon* 13, 146–157 (2019). Figure adapted. <https://doi.org/10.1038/s41566-018-0347-5>
- [4] Schuster, V., Liu, C., Klas, R., Dominguez, P., Rothhardt, J., Limpert, J., and Bernhardt, B. "Ultraviolet dual comb spectroscopy: a roadmap", *Opt. Express* 29, 21859-21875 (2021). Figure adapted. <https://doi.org/10.1364/OE.424940>
- [5] Lauth, J., Kinge, S. and Siebbeles, L.D.A.. "Ultrafast Transient Absorption and Terahertz Spectroscopy as Tools to Probe Photoexcited States and Dynamics in Colloidal 2D Nanostructures", *Zeitschrift für Physikalische Chemie*, vol. 231, no. 1, 2017, pp. 107-119. <https://doi.org/10.1515/zpch-2016-0911>
- [6] Gallmann, L. et al. "Attosecond science: recent highlights and future trends", *Annual review of physical chemistry* 63 (2012): 447-69. Figure adapted. <https://doi.org/10.1146/annurev-physchem-032511-143702>



Overview of UV Dual-Comb Spectroscopy with femtosecond temporal resolution. Results are a 2D plot covering the time and frequency domain simultaneously [1]. The data shown in the spectrogram are taken from [2].

Dual-Comb Spectroscopy

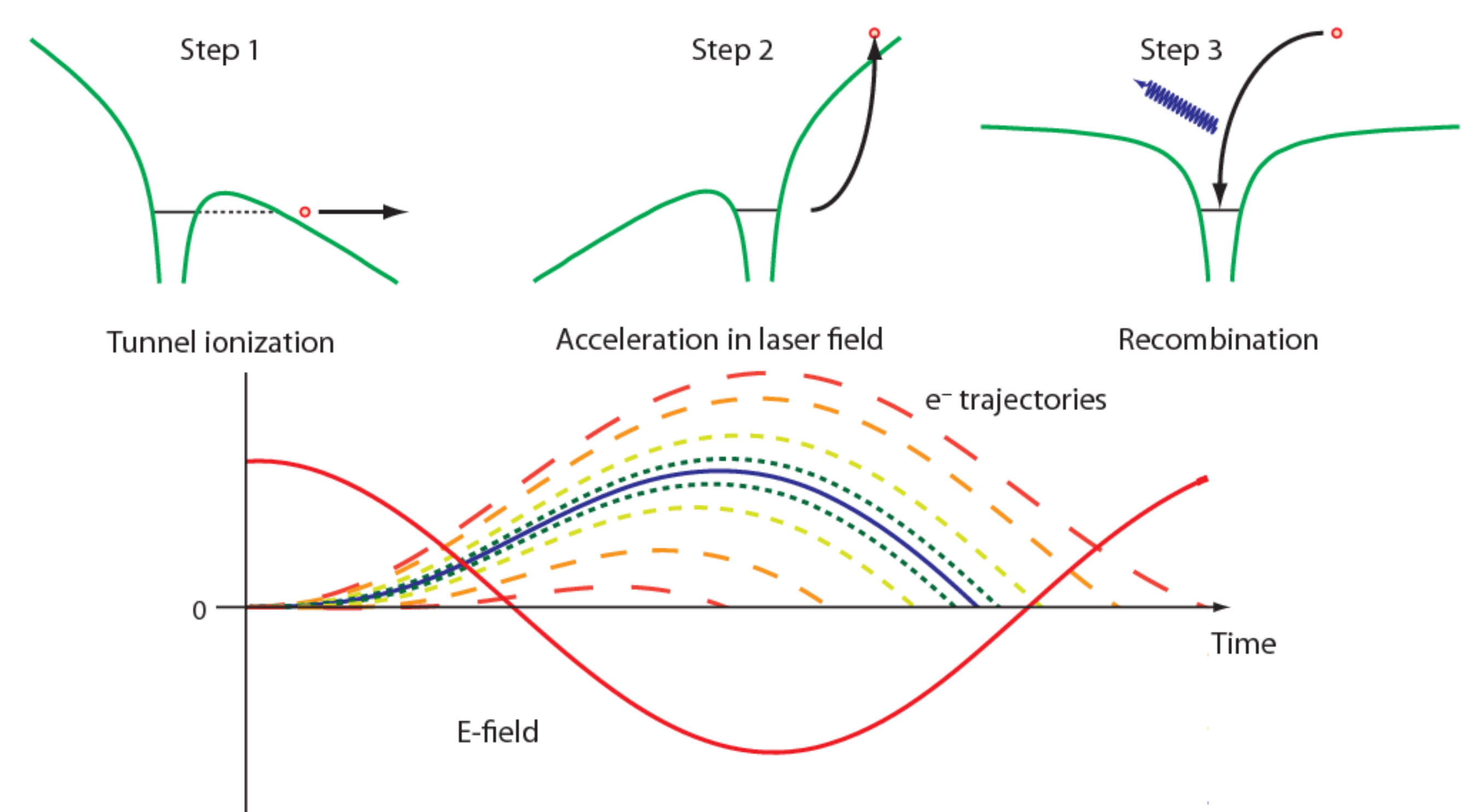
- **Superimposed frequency combs**
 - Ytterbium fiber lasers (IR)
 - Detuned repetition rates
 - Spectral broadening & HHG
 - Via fibers & gas target
- **Fourier Transformation**
 - Signal in the radio frequency domain
 - Interferogram via oscilloscope
 - Time to frequency domain



The signal measured in DCS is Fourier-transformed from time to frequency domain [4].

High Harmonic Generation

- **Conversion of laser light**
 - No viable UV laser sources
 - Access to 3rd & 5th harmonic
 - 3th: 1030 nm → 310 nm
 - 5th: 1030 nm → 206 nm
- **IR laser source → UV laser**
 - High power source laser
 - 2 x 100 W Ytterbium (IR)
 - Low conversion efficiency
 - mW to μW in the UV



The three step model of high harmonic generation (HHG) including electron trajectories [6].

