Multiscale pump-probe spectroscopy in Fatty Acid Photodecarboxylase

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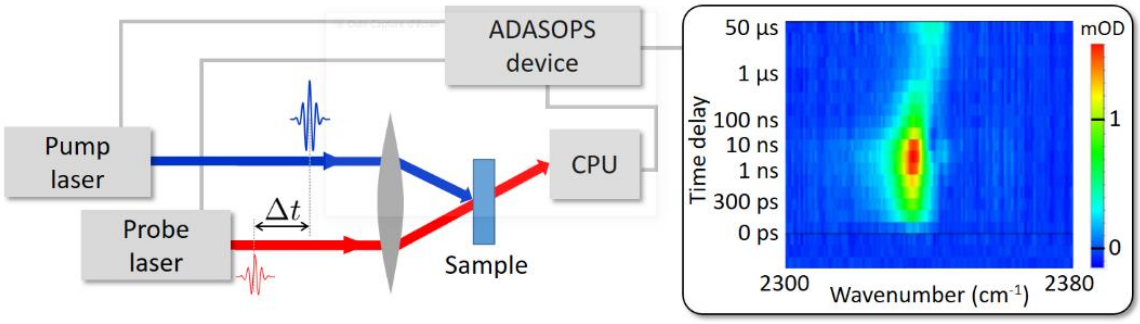
Femtosecond pump-probe spectroscopy is one of the most widespread techniques for investigating the ultrafast dynamics of numerous systems in physics, chemistry and biology. However, the standard method based on an optical delay line for controlling the pump-probe delay is not suitable to address the multiscale distribution of time constants, from picoseconds to milliseconds, often encountered in complex biomolecules. Indeed, multiscale control of the time delay in a single experiment requires the use of two distinct amplifiers for delivering pump and probe pulses [1]. Using Arbitrary Detuning Asynchronous Optical Sampling (ADASOPS), we have shown that this approach can be readily implemented using two pre-existing amplifiers, seeded by independent free-running oscillators [2-4]. A specially designed electronics (the ADASOPS device in the Figure below) triggers the two amplifiers while selecting appropriate oscillator pulses that are closest to desired time delays. Besides its simplicity, this scheme offers a great agility as the delay can be controlled for each individual pulse pair and characterized with a sub-picosecond resolution [3].



Using ADASOPS, we have performed multiscale time-resolved vibrational spectroscopy in Fatty Acid Photodecarboxylase (FAP), a recently-discovered photoenzyme [5,6]. As shown in the above Figure, the differential pump-probe spectrum clearly shows the release and subsequent decay of CO_2 from the fatty acid upon photo-excitation of the flavin cofactor. These findings contributed to the recent elucidation of the mechanisms and dynamics of this new photoenzyme [6].

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