MAR11-2010-002752

Abstract for an Invited Paper
for the MAR11 Meeting of
the American Physical Society

Generation, characterization and spectroscopic use of ultrashort pulses fully tunable from the deep UV to the MIR

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The impressive work of Ian Walmsley has brought us invaluable new possibilities for the full characterization of ultrashort pulses. Spectroscopy of physical, chemical and biological relevance does, however, need pulses far from the 800 nm Ti:sapphire wavelength used for testing SPIDER and its advanced versions. Fortunately, optical parametric amplification (OPA) allows for easy generation of fully tunable pulses. I will review our efforts, highlighting noncollinear OPA, i.e. NOPA, for visible pulses shorter than 10 fs, mixing into the UV down to below 200 nm at 20 fs duration and novel hybrid schemes to efficiently reach the middle IR. I will show that these schemes can be used equally well from kHz to MHz repetition rates. The tunable ultrafast pulses in turn also demand improvements in characterization. The UV range led us to use difference frequency generation instead of the sum frequency mixing employed in the original SPIDER. The lack of proper beam splitters and auto-referencing led us to the use of two auxiliary pulses and the avoidance of any additional chirp added to the test pulse. We termed this zero-additional-phase SPIDER, i.e. ZAP-SPIDER. Lately, with increased use of UV pulses, we came to the conclusion, that the ubiquitous two-photon-absorption can well serve as nonlinearity, at least in UV autocorrelation measurement. How do we use this for full characterization? Hopefully, Ian will tell us! Since the proof is known to be in the eating, I will demonstrate the success of our technical efforts with examples taken from ultrafast molecular dynamics. Highly pronounced vibronic wavepackets in the product of ultrafast excited state proton transfer and the very primary processes leading to homolytic and heterolytic bond cleavage will serve as easy to comprehend illustrations.