Coherent multidimensional femtosecond spectroscopy – from basics to applications

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Abstract: At the core of our perception of chemical transformation are 'molecular movies': we understand a reaction when there is a satisfactory picture of bonds breaking and forming and when the sequence of such events is known. The appropriate timescale to observe and interpret chemical reactions is given by the period of molecular vibrations, i.e. in the femtosecond (fs) range. A carbon-carbon double bond for instance vibrates with a period of roughly 20 fs. Accordingly, femtosecond spectroscopy has established itself as a fundamental tool in physical-chemical sciences. Since its advent nearly three decades ago, femtosecond spectroscopy has diversified greatly, with transient absorption as the most wide spread technique. The recently emerged two-dimensional electronic spectroscopy (2D-ES) reports on the same non-linear signal as transient absorption, but with a maximally enriched information content; in 2D-ES, the signal is resolved both in excitation and emission frequencies as well as in its time-dependence. In my talk, I will introduce 2D-ES and discuss several of its applications on molecular dynamics in biologically relevant systems as well as in molecular J-aggregates.