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Development of an ultrafast pump and probe platform and its application in the characterization of photoisomerizing push-pull azobenzenes

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Ultrafast time-resolved spectroscopy is an essential field for characterization of materials, because it allows one to measure the dynamics of excited states and also probe different characteristics of materials on an ultrafast time scale (< 1 ns) depending of the type of light-matter interaction being employed. (1) In this work, a pump and probe setup with 150 - 220 fs time resolution and broad spectral coverage was developed. This kind of time-resolved technique relies, first, on a pump pulse to optically excite the sample. Then, a probe pulse is used to measure changes in the sample caused by the pump pulse as a function of the relative time delay between both pulses. The new setup can have pump pulses ranging from 210 nm to 3000 nm. Additionally, the probe pulses can be either produced by two OPAs at the 210 nm - 16 μm range or by White-Light Continuum generation with a 500-1030 nm bandwidth. The first kind of probe is used together with lock-in detection, to measure signals with resolution up to around 1 μOD. The second kind of probe is detected by a spectrometer because of its broadband nature. In this manner, the minimum resolution is worse, on the order of 1 mOD. Additionally, a new Non-collinear Optical Parametric Amplifier (NOPA) has been developed in the lab. Its output currently ranges from 640 to 1000 nm and it can possibly be second-harmonic duplicated. This new tool will allow measurements with pulses up to 18 fs to be performed. Thus, in the future, not only broad spectral coverage will be available in the setup, but also better time coverage. Furthermore, this setup was used to study four push-pull azobenzene derivatives, which are molecules that undergo photoisomerization on the excited state. (2-3) With the use of pump and probe spectroscopy, the isomerization dynamics for both isomers was shown to occur on a sub-10 ps time for both isomers. Additionally, by comparison of the four molecules, it was possible to propose two different isomerization pathways which are available for the *trans*.isomer, in contrast with only one for the *cis*.isomer. Transient absorption anisotropy measurements also show the geometrical distortions that occur during isomerization and allows the identification of a barrier in the ground state of the molecules which retards full vibrational relaxation. This was a very important finding, because it has shown that some structures on the Potential Energy Surface of the molecular states may be invisible to the most commonly used probes and the detection depends on the charge symmetry properties of the molecule. In addition, spectroscopic characterization of the *cis*.isomer was performed, which is not usually done for push-pull azobenzenes. Part of the study was developed in collaboration with Tiago Buckup, in the University of Heidelberg.

Palavras-chave: Espectroscopia ultrarrápida. Espectroscopia resolvida no tempo. Absorção transiente.

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