Part II

Ultrafast dynamics from pump-probe spectra

The fs-pump-probe spectra are the center of this thesis. The experimental results are given first, ordered by the molecules examined, i.e. I_2 , ClF and Cl_2 . Their interpretation is discussed in chapter 7, ordered by topic. Some additional spectra are shown there that demonstrate a special effect in detail. The discussions include DIM-simulations from the cooperating theory group of Prof. Gerber on the closely related system F_2/Ar_{54} , and the good agreement of experimental and theoretical results support the interpretations. In some cases, as for the solvent induced spin-flip (chapter 7.6), the prediction by theory was given before the experimental evidence. In the case of depolarization and angular reorientation (chapter 2.3.5), it is evident that the process is contained in the simulations, but they have to be evaluated accordingly. The 2-D-quantum mechanical wave packet simulations by M. Korolkov are discussed in chapter 2.1.5 together with the energy loss. This shows the beautiful mutual stimulation of theory and experiment that emerges from the cooperation within the SfB 450. In the discussion (chapter 7), the input from the theoretical collaborations will be clearly delimited from the fruits of this thesis. In future, the experiments will be compared to a detailed theoretical study the system ClF/Ar itself. The DIM-simulations are currently pursued in the PhD thesis of Maike Schröder in the group of Prof. Manz.