TIME RESOLVED ALIGNMENT: FROM PARA-HYDROGEN CRYSTALS TO AMBIENT NITROGEN

Nina Owschimikow[1], Falk Königsmann[1], Phillip Giese[1], Jochen Mauer[1], Mizuho Fushitani[1], David Anderson[2], Nikolaus Schwentner[1]

[1]*Department of Experimental Physics, Free University Berlin, 14195 Berlin, Germany;*
[2]*Department of Chemistry, University of Wyoming, Laramie, WY 82071, USA;*

Strong, linearly polarized laser pulses are widely used to induce alignment in molecules possessing an anisotropic polarizability. A short nonresonant pulse prepares a coherent superposition of rotational eigenstates, as a result of which $\langle \cos^2\theta \rangle$ for the angle θ between polarization vector and molecular axis becomes time-dependent. The alignment induces birefringence and recording it between crossed polarizers (Optical Kerr Effect) provides a sensitive and convenient tool to detect the alignment dynamics. We demonstrate this in a collinear detection scheme using the fundamental of a Ti:sapphire laser for alignment and its second harmonic for probing [1].
At a field strength of about 10 TW/cm^2 very large phase shifts between ordinary and extraordinary beam can be achieved due to the large interaction length. In a sample of nitrogen gas under ambient conditions they lead to ellipticity angles α of the order of 10 degrees. Good polarizers allow to resolve α down to 10^{-5}, thus a huge range is available to study quantitatively and in detail the dynamics. The revival period of N_2 of about 8 ps provides an optimal clock to follow collisions, which occur for ambient pressure on a 100 ps time scale. We demonstrate that the phase dependent and the phase independent parts in θ enable to record the dephasing and the depopulation cross sections independently and we compare them with theoretical predictions and results from alternative experimental methods.
For pH_2-crystals at a temperature below the rotational energies the same technique reveals coherent oscillations as stimulated Raman-sidebands in the 94 fs range, which last for about 1000 periods. They originate from delocalized rotons and the subtle splitting into the three |M| sub-states leading in time frame to three components at 93.7, 94.2 and 94.8 fs, which can be resolved and are also visible in a beat pattern of 17 ps. These rotons appear together with a single-frequency oscillation with 900fs period, originating from a delocalized lattice phonon. It strictly follows the Raman-selection rule and is long lived, lasting for several ten ps.

References

[1]. F. Königsmann, M. Fushitani, N. Owschimikow, D. Anderson, N. Schwentner, Chem. Phys. Lett. **2008**, 458, 303.