Max-Born-Institut

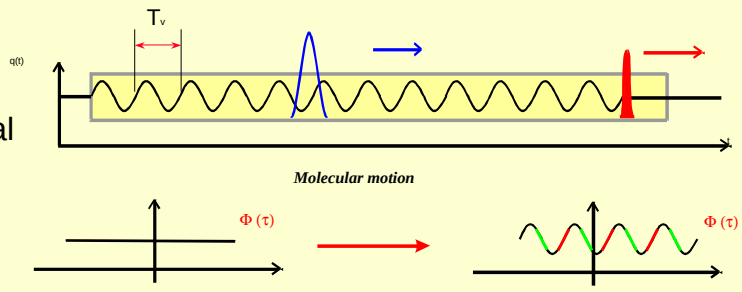
UP 2: Non-Linear Optical Excitation and Alignment Processes of Molecules in the Gas Phase

Results (2001-2003): Test of Pump-Control Technique by Mode Specific Excitation and Dissociation of Small Molecules

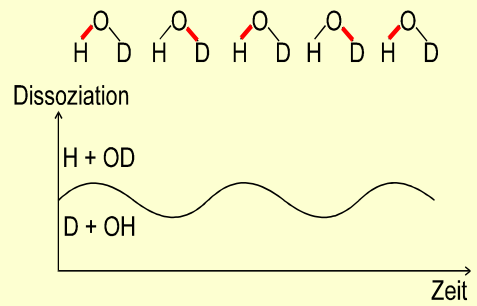
Motivation

Study of non-linear excitation mechanisms in a long, well defined interaction region

- new type of absorption spectroscopy
- formation of rotational and/or vibrational wave packets
- creation of new frequency components

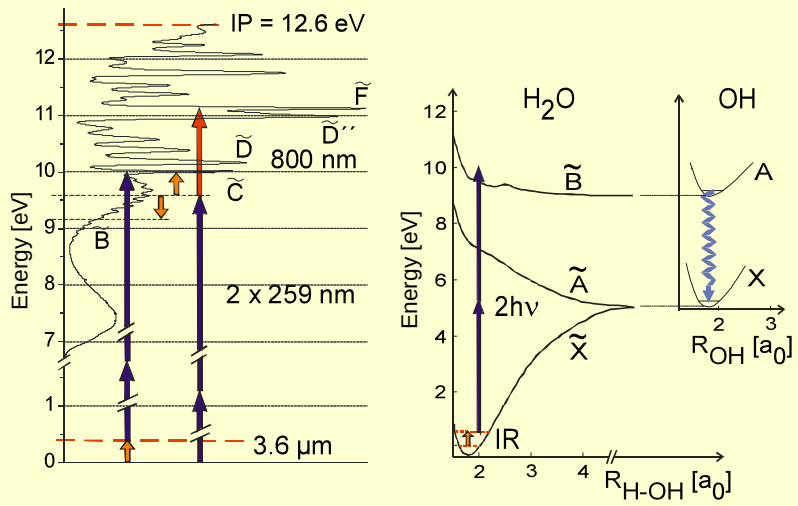


Mode specific IR excitation to control the dissociation pathway



Experimental

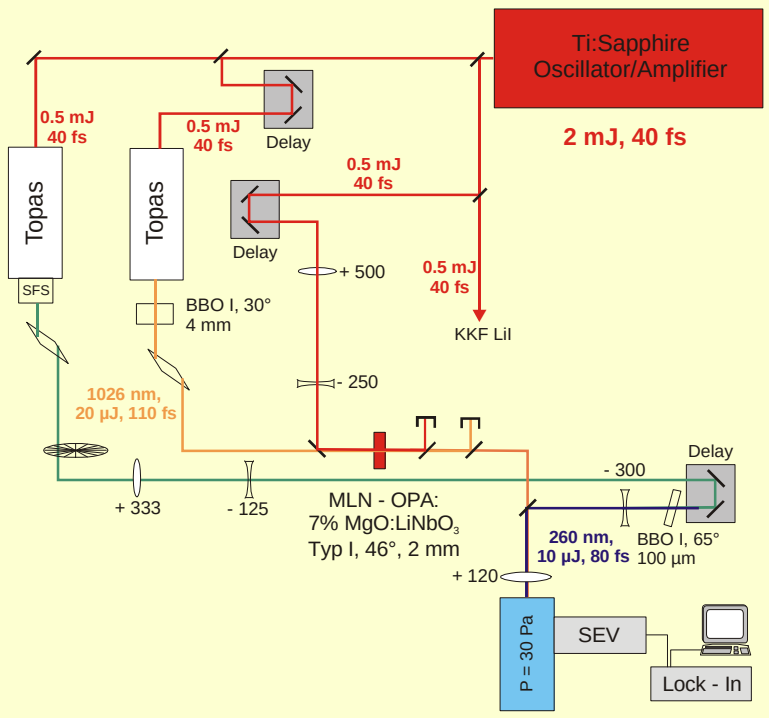
Multiphoton Excitation – Fluorescence Detection



- two-photon excitation at 244 nm – 264 nm
- OH(A) fluorescence yield dependent on vibrational excitation of the B̃ state

➡ control by pulses in IR or MIR

Setup Fluorescence-Experiment

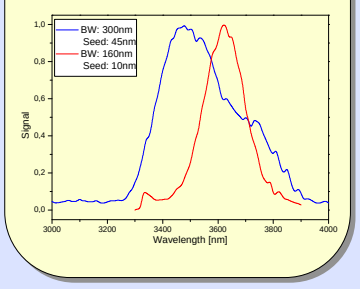


MIR - Generation

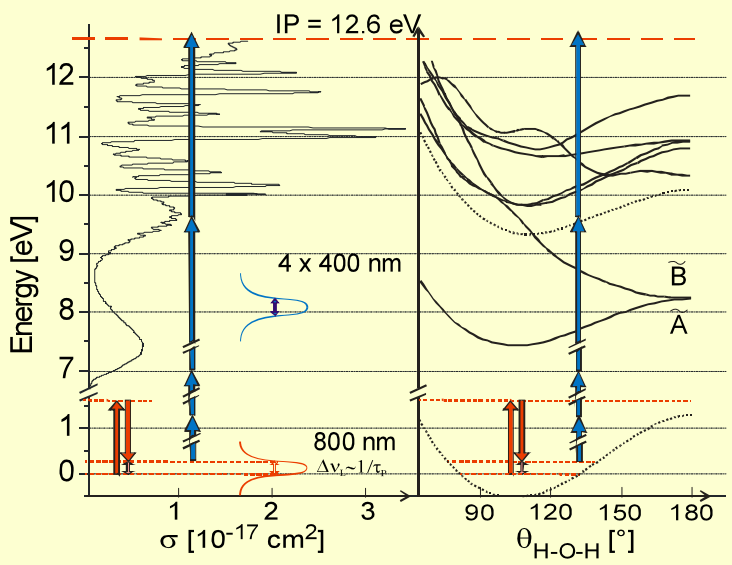
Femtosecond traveling-wave optical parametric amplification in $MgO:LiNbO_3$ (7% MgO)

- 0.35 µm - 5.5 µm
- GVM: $|1/v_I - 1/v_P|$, $|1/v_S - 1/v_P|$ £ 50 fs/mm @ 3 -4 µm

➡ narrow-band seeding

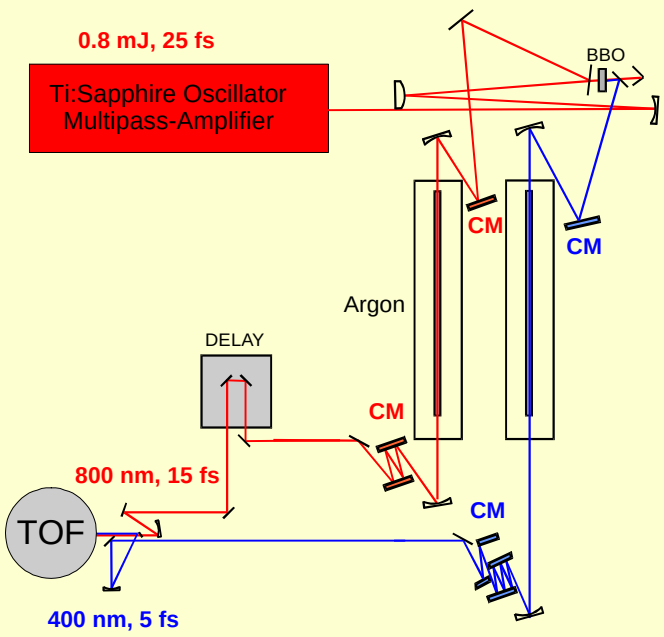


Impulsive Raman Excitation



Impulsive excitation at 800 nm, multiphoton ionisation with 400 nm (t_L £ 10 fs)

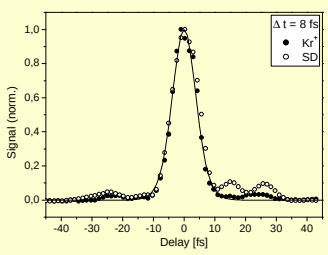
Setup Raman-Experiment



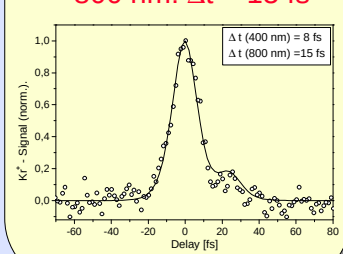
Short Pulse Generation financially supported by Deutsche Forschungsgemeinschaft

Laser Properties

Pulses @ 400 nm: Δt = 8 fs

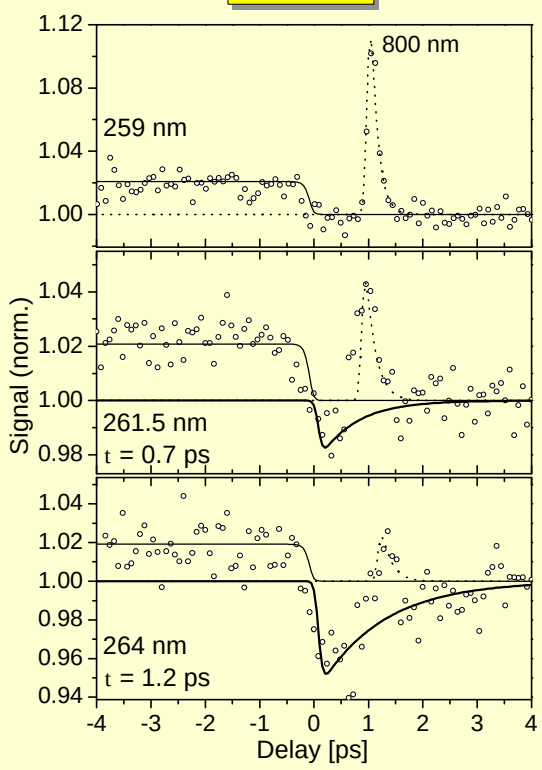


Pulses @ 800 nm: Δt = 15 fs



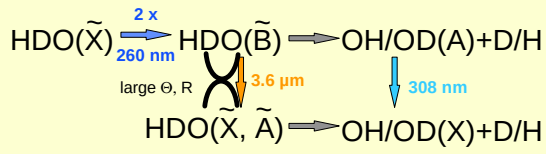
Results

B̃ state

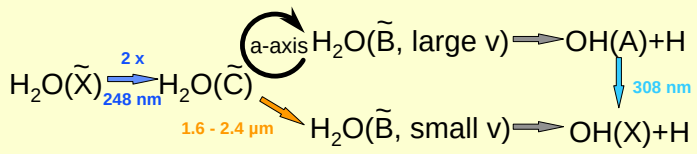


Analysis of Reaction Pathways

B̃ state: transit through a conical intersection



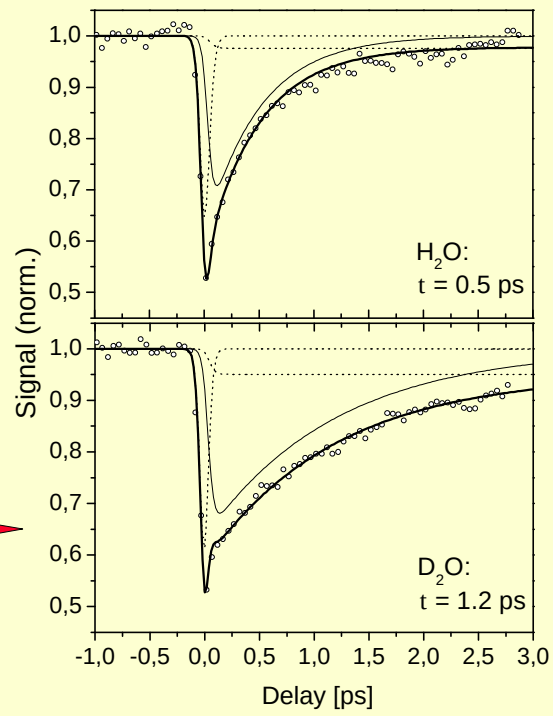
C̃ state: predissociation dynamics



D̃ state: predissociation mechanism

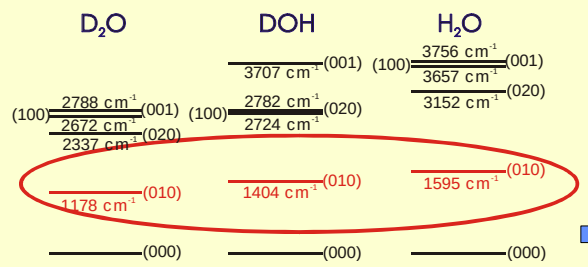
homogeneously electronic

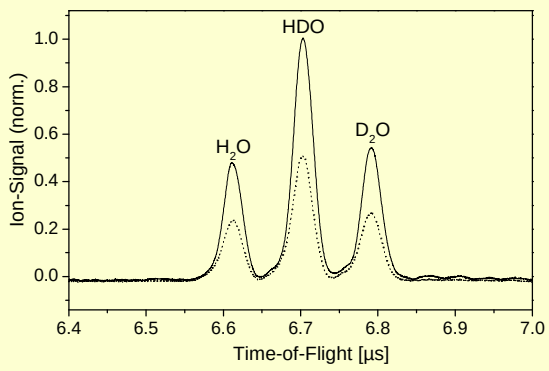
C̃ state



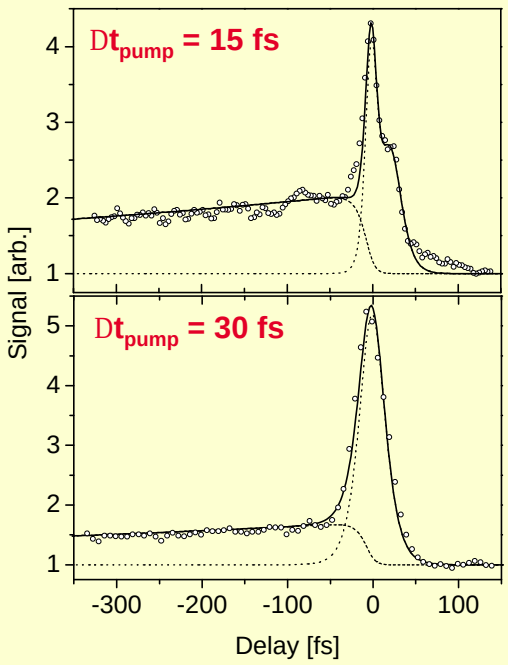
Impulsive Raman Excitation

Clear dependence of ionisation rate at H_2O, HDO and D_2O on pre-excitation



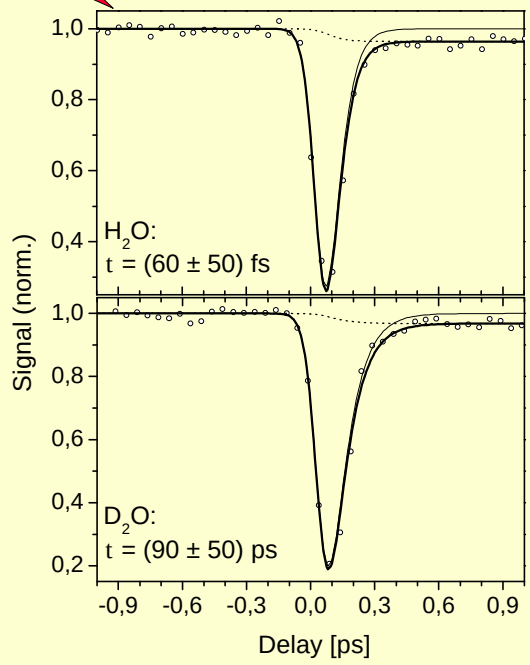


Effect of Pump Pulse Duration



Steinkellner et al., Appl. Phys. B, **73**(3), 279 (2001)

D̃ state

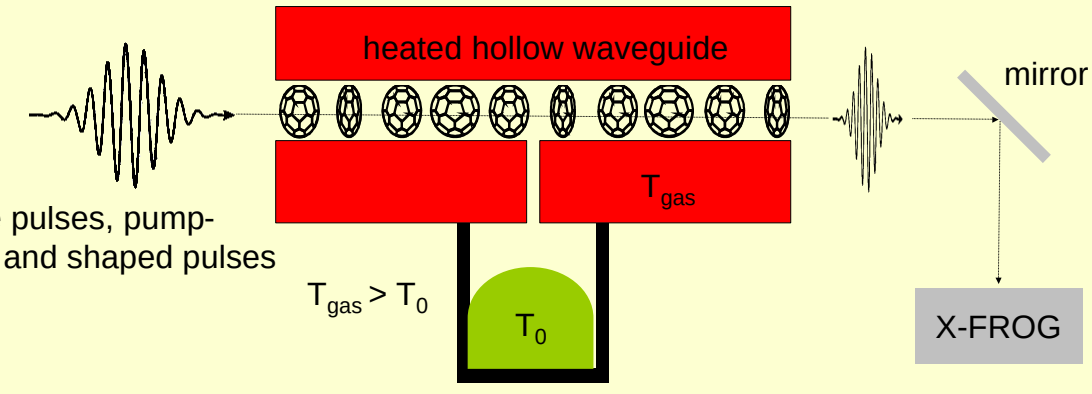


Steinkellner et al., submitted to JCP

Program for 2004-2007
Non-Linear Optical Excitation and Alignment Processes of Molecules in the Gas Phase

Hollow wave guides: also a new method for molecular spectroscopy for weakly absorbing species?

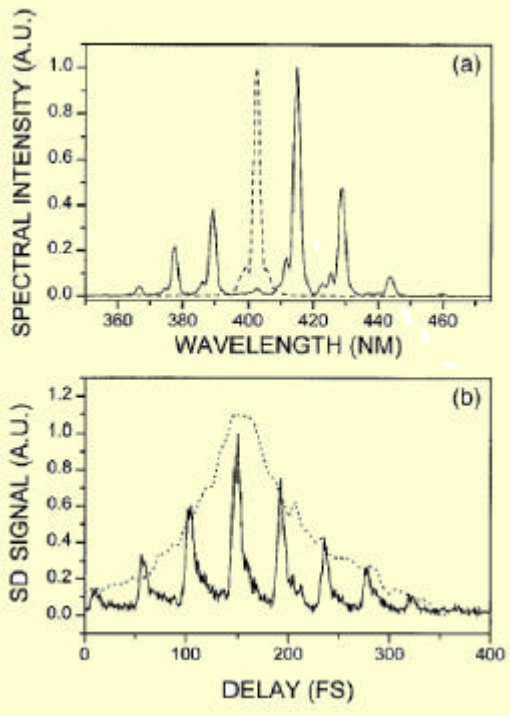
Long and well defined interaction region



- multielectron and impulsive excitation of coherent vibrational and/or rotational motion

  C_{60} - A_{1g}, T = 67 fs, Ω = 496 cm^{-1}

  C_{60} - H_{1g}, T = 123 fs, Ω = 271 cm^{-1}
- modulation of the refractive index
- scattered light gives insight in the induced electron and nuclear dynamics in the neutral system (complementary to UP1)
- studies of H_2O and different aromatic molecules (benzene -> tetrazene -> C_{60})



M. Wittmann et al.,
Opt. Lett. 26, 298 (2001)
SF_6-A_{1g} mode, T = 43 fs, Ω = 775 cm^{-1}
Pump pulse: 0,2 mJ, 24 fs, 790 nm
Probe pulse: 10 µJ, 100 fs, 400 nm