SEVENTH FRAMEWORK PROGRAMME
Nano-scale Devices – New Functionalities

Molecular Logic Circuits – MOLOC

WP2 – KLK: Optical addressing of molecules with femtosecond MID IR pulses

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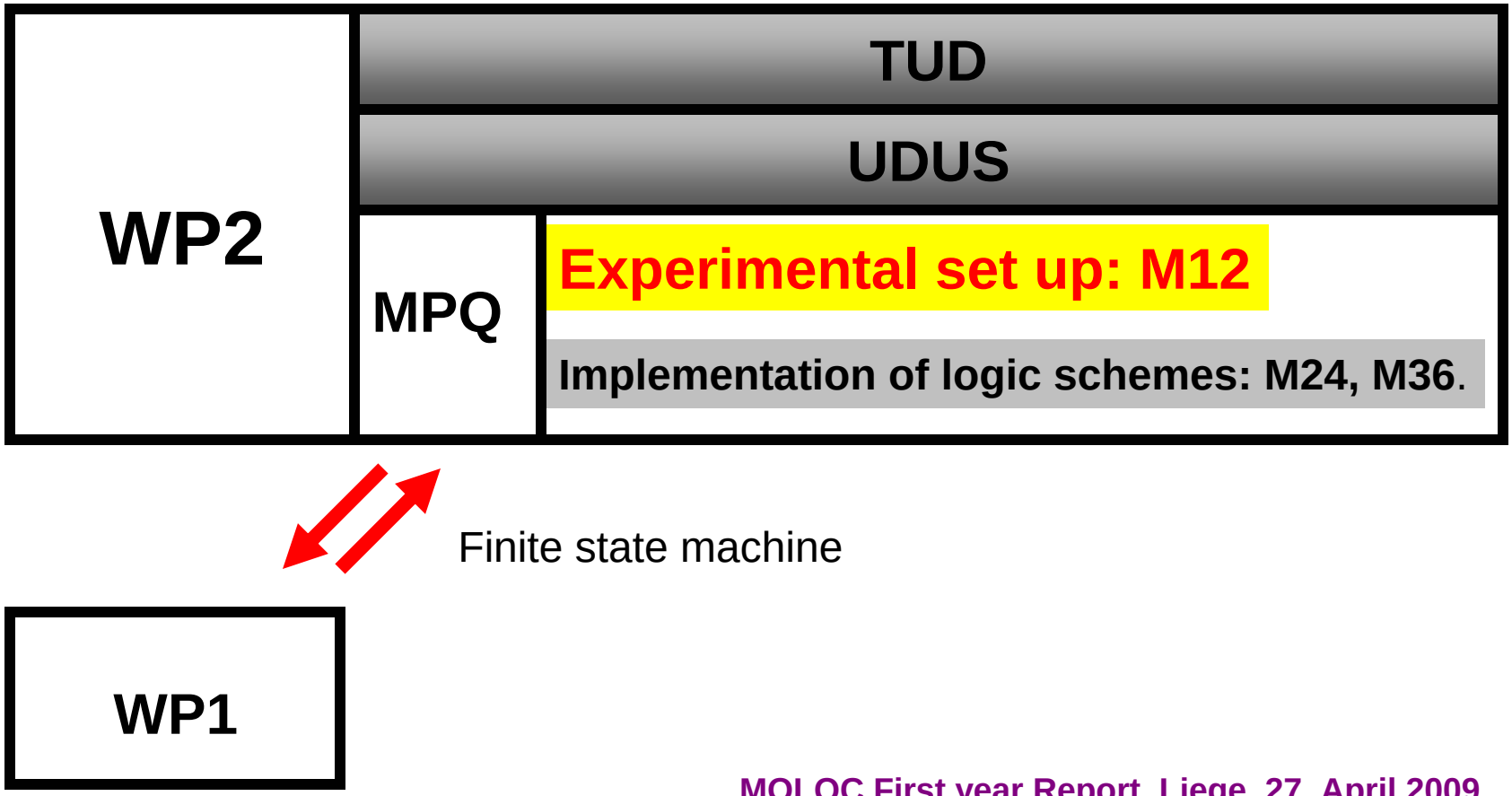
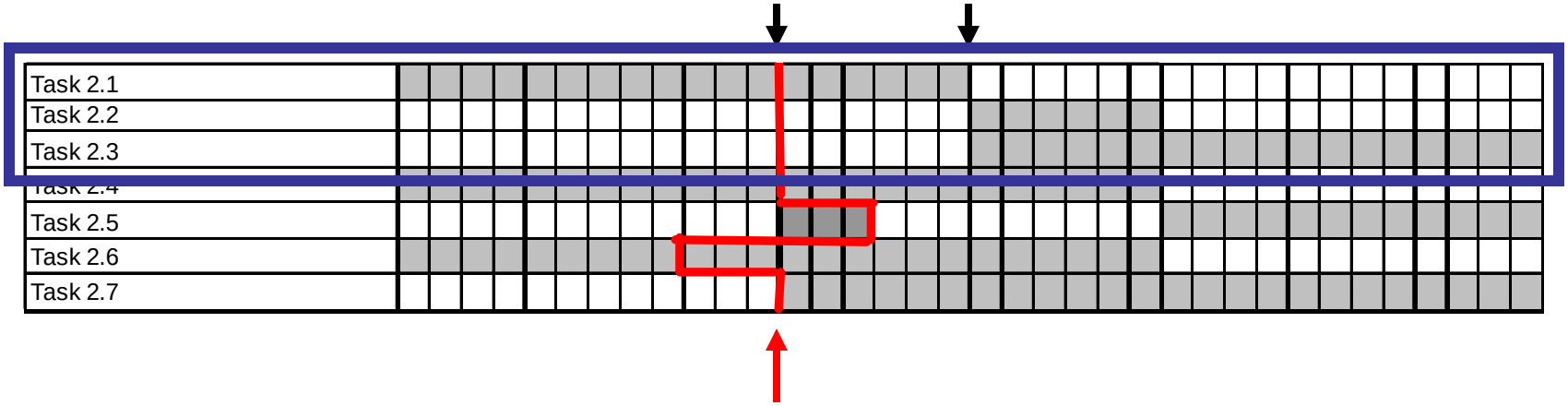
WP2: Coherent and incoherent multiphoton optical addressing

Milestones and collaborations



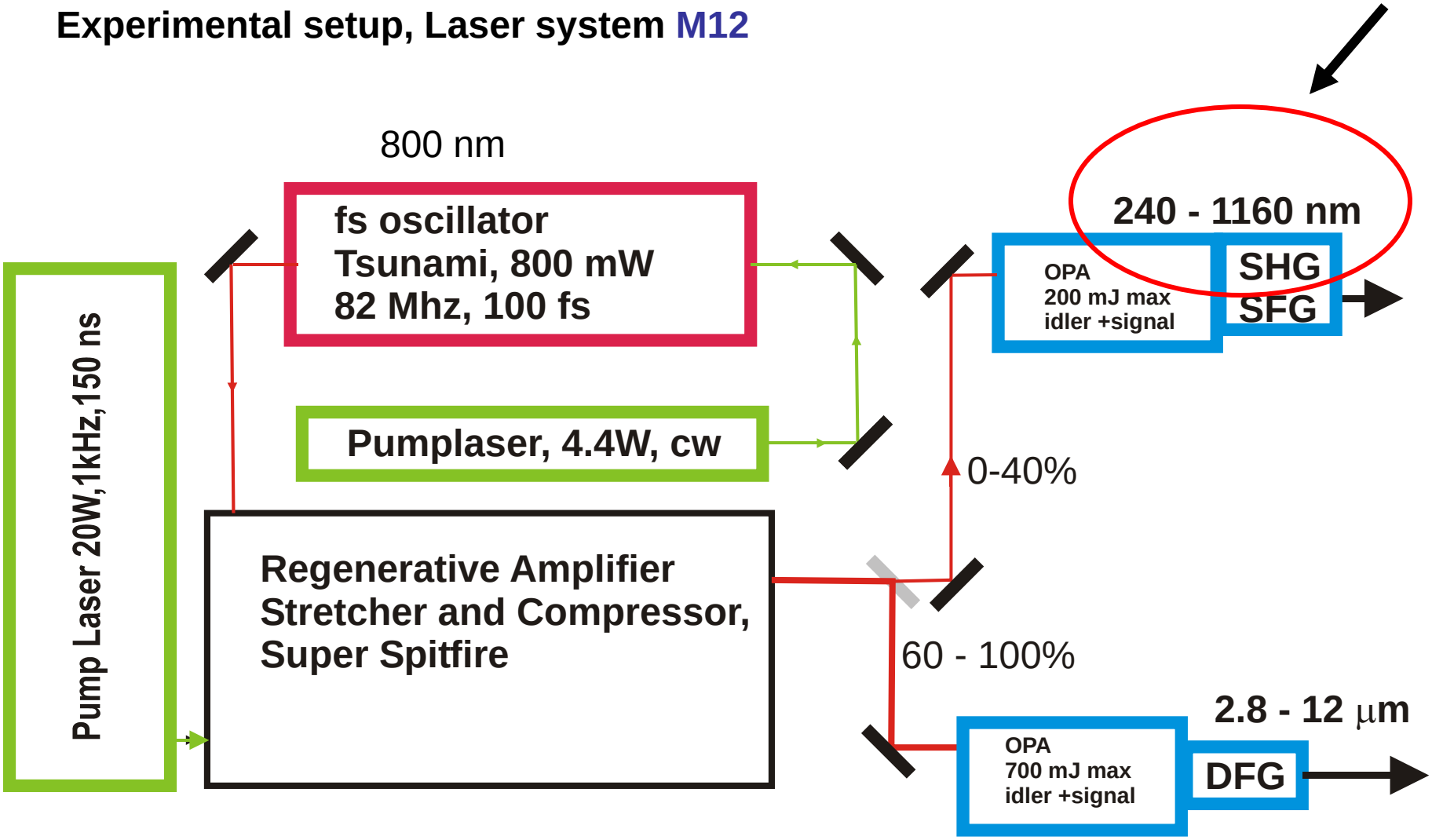
Project objectives, MPQ group

Task 2.1: Development, setup and characterization of a basic component in the experimental setup, i. e. Mid-infrared femtosecond pulse shaper for phase and amplitude control of of optical pulses, **M18**

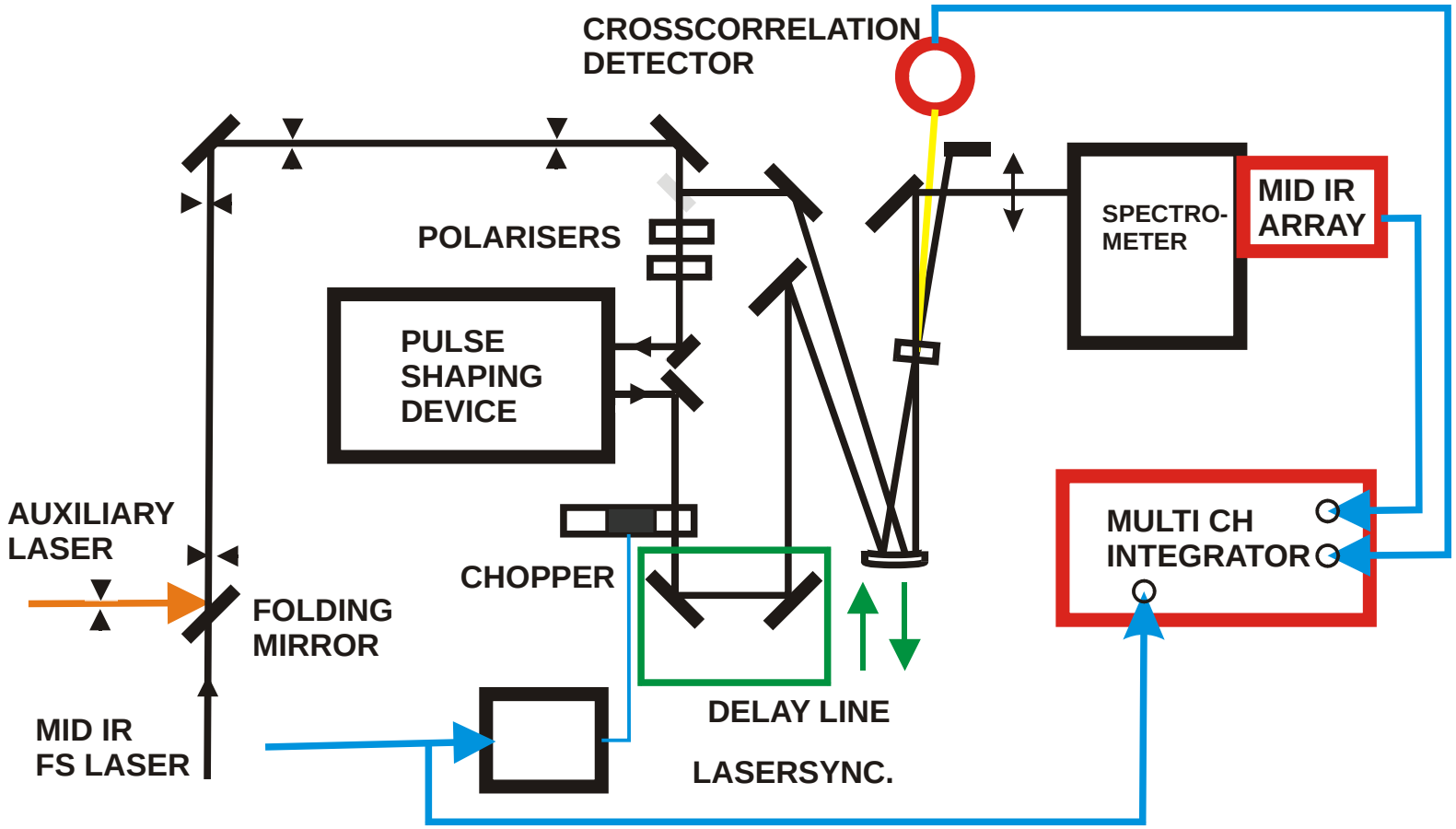


✓**Deliverable 2.1:** Experimental setup **M12**

Experimental setup, Laser system M12



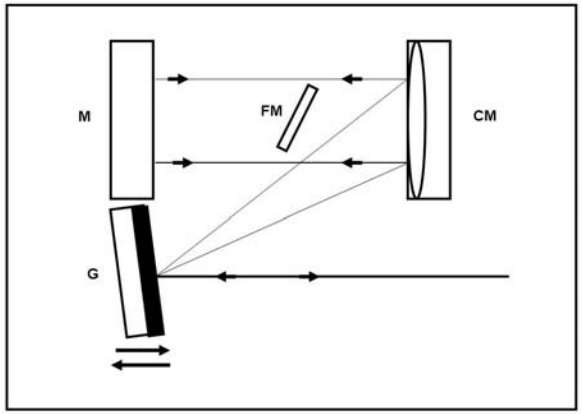
Experimental setup, Transient absorption setup M12



Experimental setup, Pulse shaper M18

PULSE SHAPING DEVICE

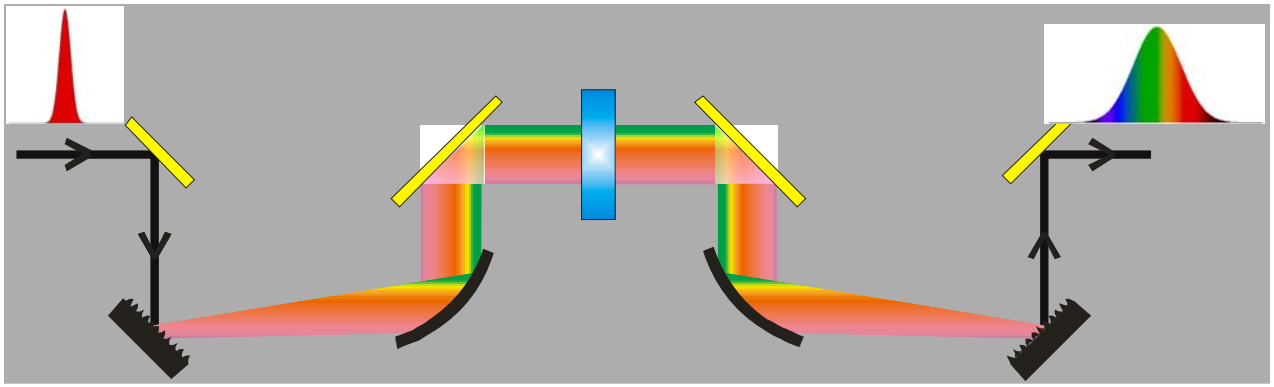
In use

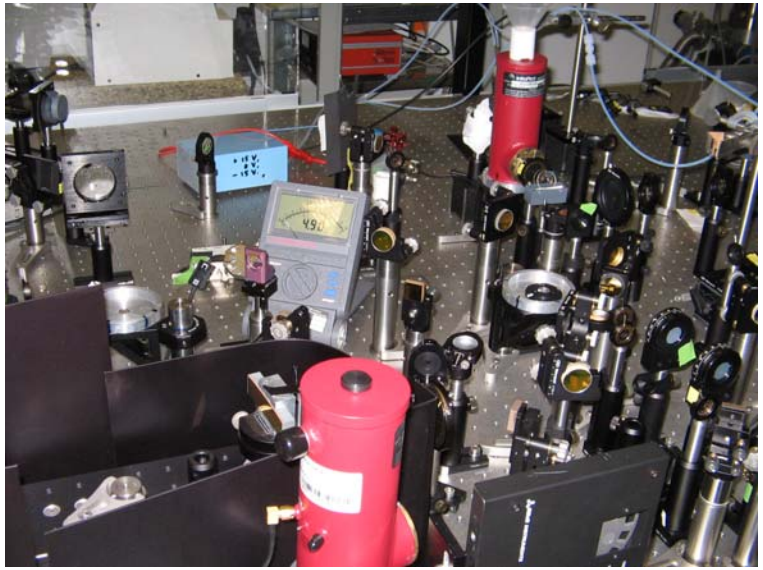


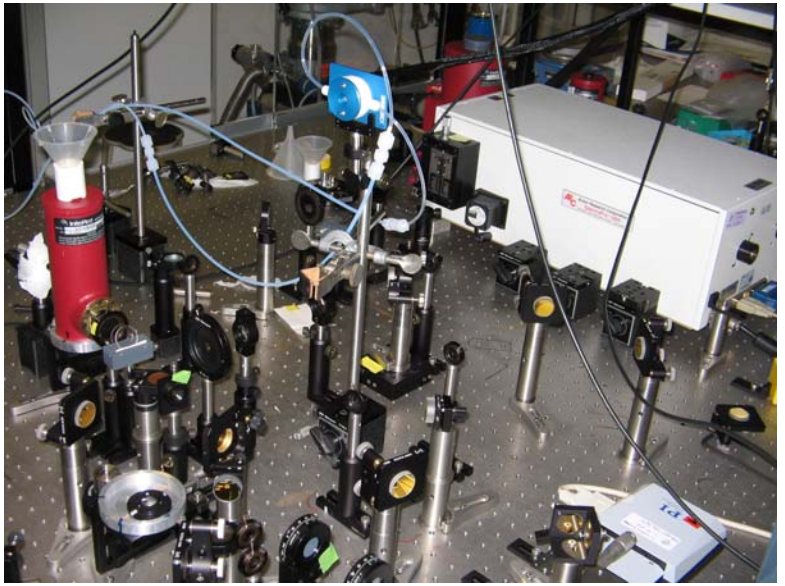
Chirping

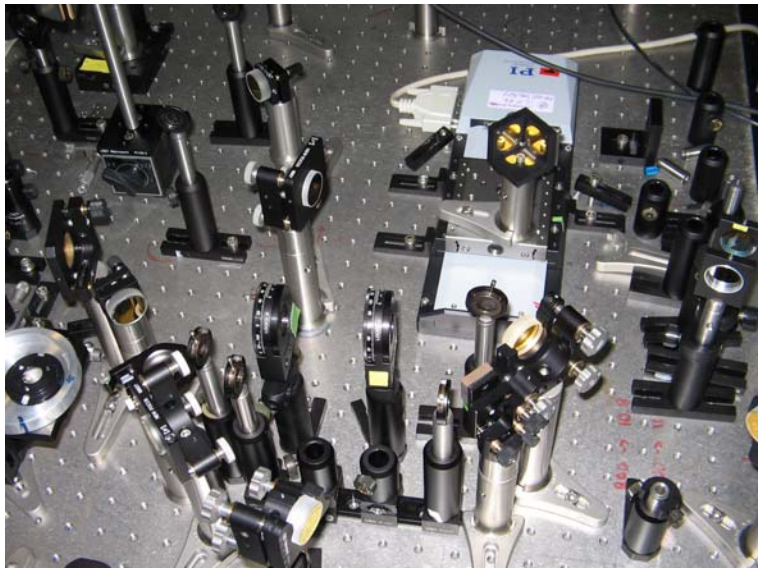
In progress....

Phase and amplitude shaping with acousto-optic modulator









Molecular internal degrees of freedom as a resource for computation

Spectroscopy transleted to logic:

MOLECULE	LOGIC
Vibrational state vectors	Logical observables
Molecular dynamics take place in ultrafast time scales	Fast processing
Fs spectroscopy allows simultaneous excitation of several molecular states	Paralell processing

Molecular internal degrees of freedom as a resource for computation

Features:

- Direct excitation of the vibrational levels in MID IR
- Transient absorption spectroscopy for monitoring dynamics
- IR active CO-stretching modes of tungsten-hexacarbonyl $W(CO)_6$.
- Aiming to excite higher population levels – higher degree of parallelism
- Aiming to excite population in controlable manner – shaping device

Why pulse shaping? (M18)

The phase and amplitude shaped pulses can produce input state in more controlable manner, i. e. with control of the input we control the output.

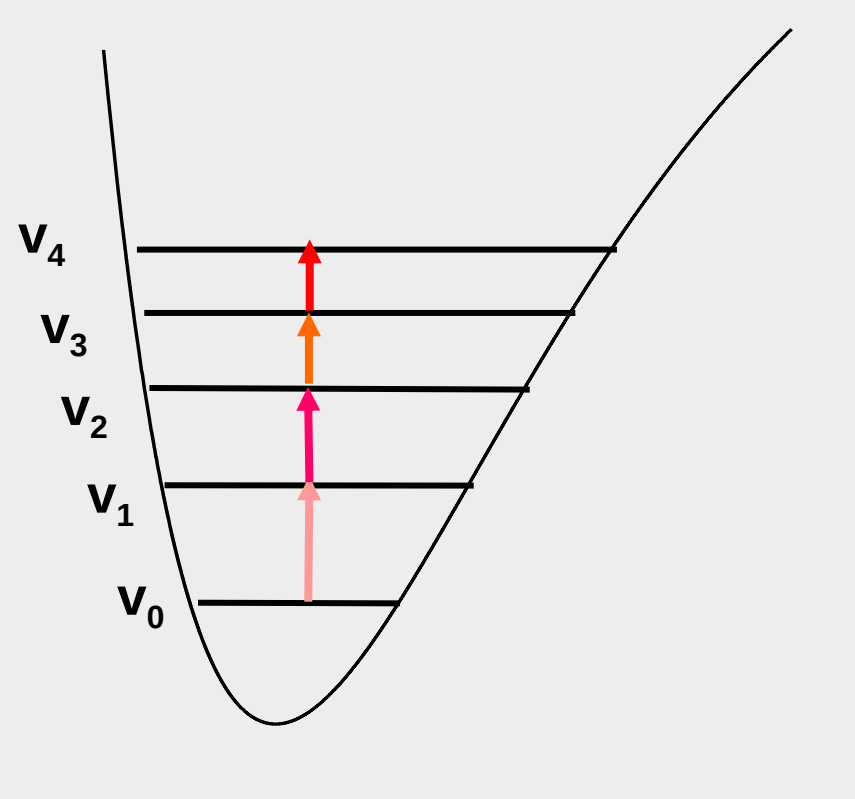
From technical point of view, shaping in MID IR is difficult.

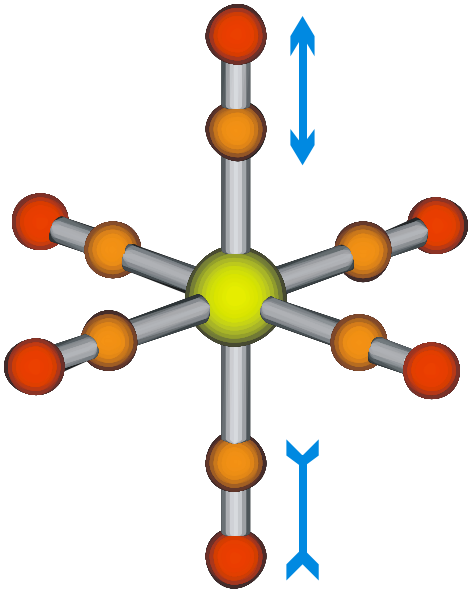
LCDs that are widely used in VIS/NIR spectroscopy are not transparent in MID IR. One has to use AOMs.

CO stretch in metal carbonyls:

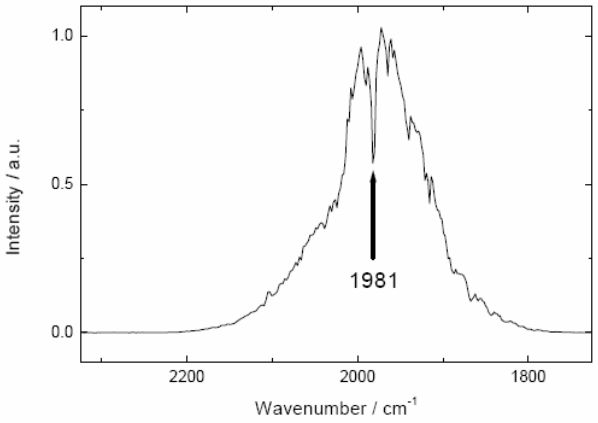
→ Large absorption cross section ($8.3\ 10^{-17}\ cm^2$)

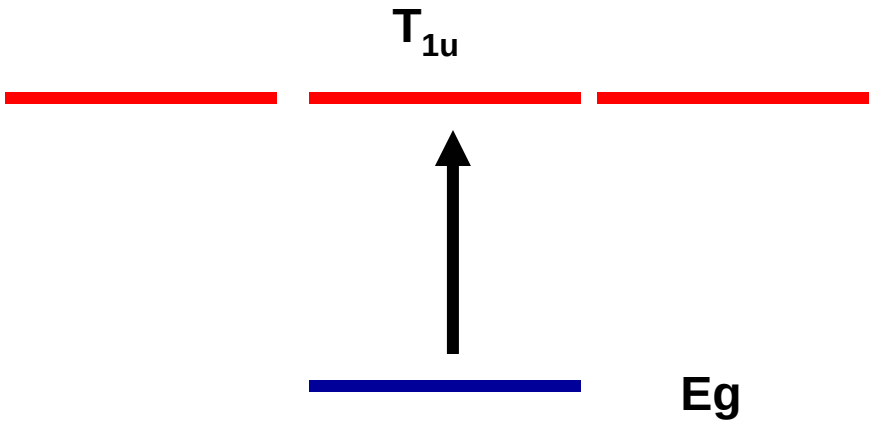
→ Molecular potential anharmonicity





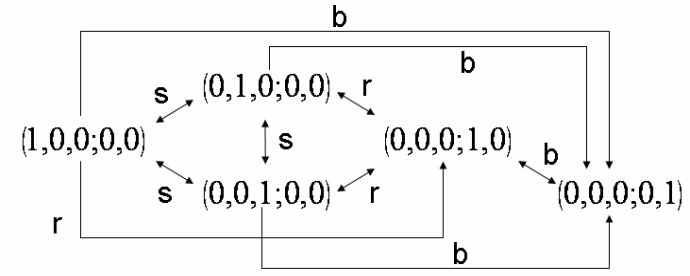
Case study: tungsten-hexacarbonyl $W(CO)_6$





One quantum excitation

Schematic representation of the relaxation of vibrationally excited $W(CO)_6$ molecule, represented by (T_{1ux} , T_{1uy}, T_{1uz} ; E_g , bath) where one quantum is initially placed in the infrared active T_{1u} state:

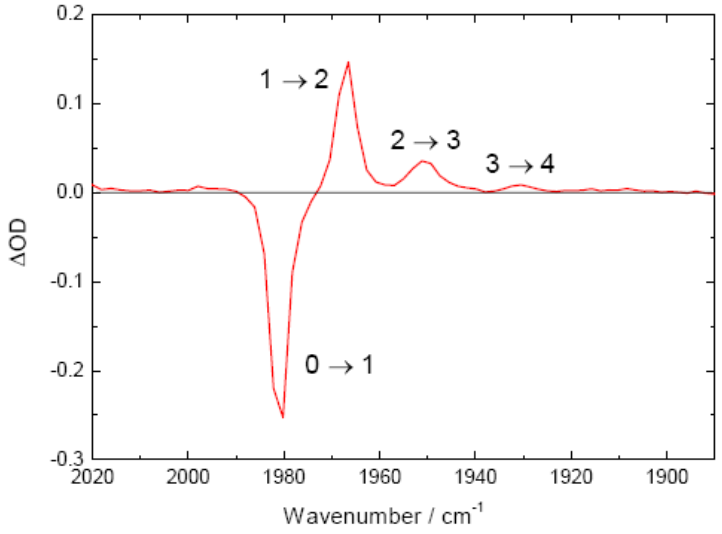


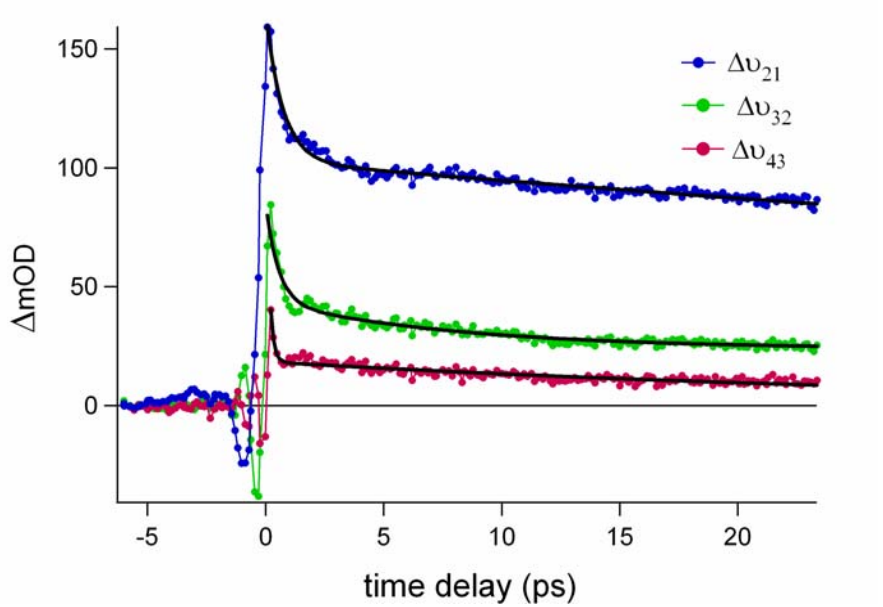
- E.A.Torres, K.L. Kompa, F. Remacle and R.D.Levine, *Chem Phys*., 347 (2008) 531

Parallelism increases with number of the excited quanta:

q	*No. of states*
1	*5*
2	*15*
3	*35*

Excitation up to the 4th level results:





Extracted life times for $W(CO)_6$ vibrational relaxation in room temperature CCl_4 ($1X10^{-3}$ M).
The lifetimes were extracted with double exponential decay curves.

State of the work

- Excited levels up to v=4 are observed
-The excitation with chirped pulses did not produce better signal to noise ratio.

Plans for the future

- Setting up the AOM shaper
- Trying some new molecules
- Introducing an UV/VIS beam as logical control