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Yet another minimum in high harmonic generation

K.I.T.P.
9th September 2010

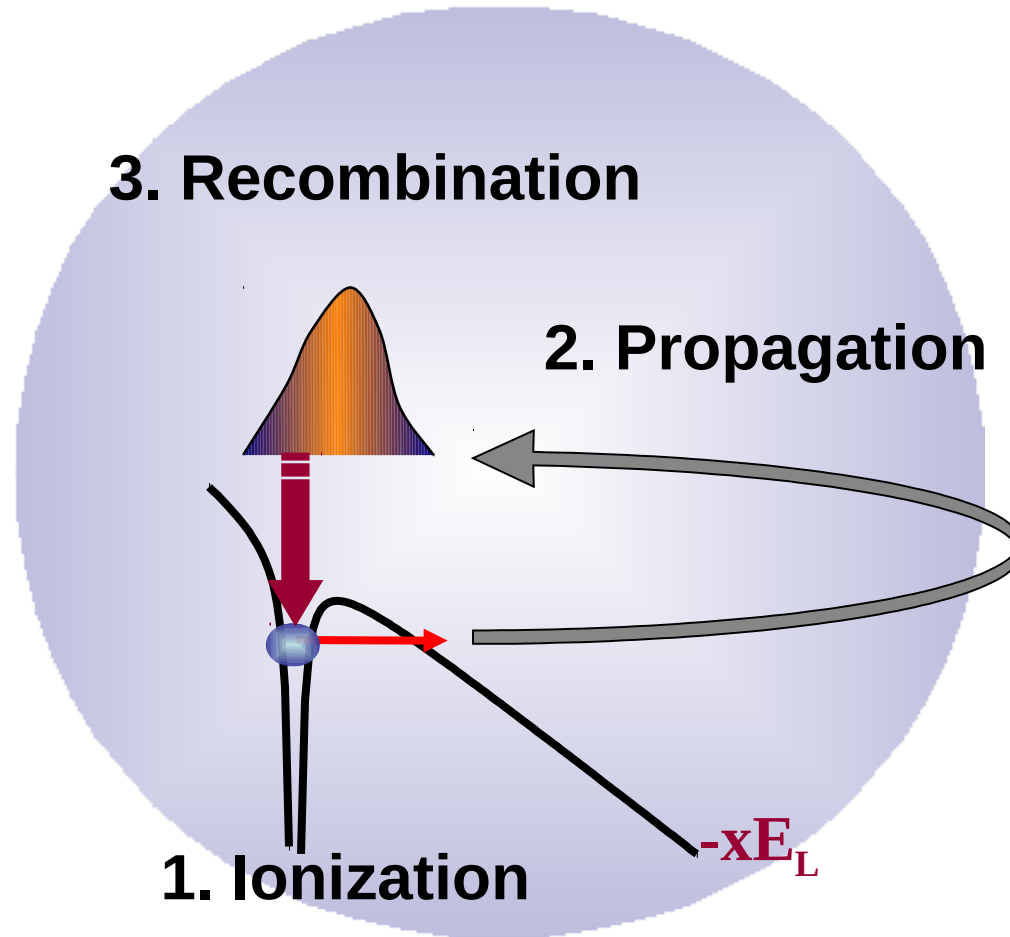
Alex Harvey



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High harmonic spectroscopy



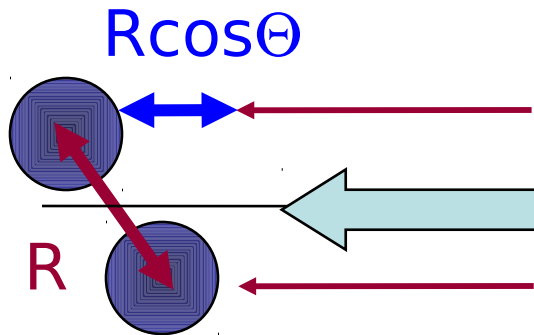
Potentially high temporal and spatial resolution

HH spectroscopy: spatial resolution

Spatial resolution comes from A scale de Broglie wavelength of the electron

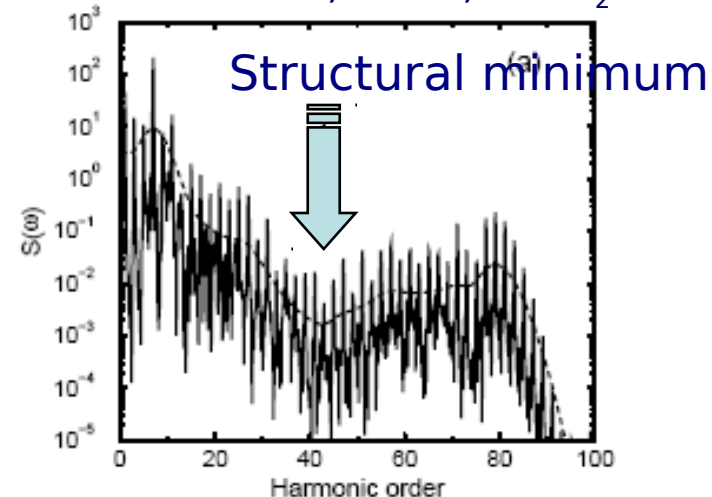
$$N\omega - I_p = \frac{p^2(N\omega)}{2}$$

$$pR \cos \theta = \begin{cases} \pi(2n+1) & - \text{ gerade} \\ \pi 2n & - \text{ ungerade} \end{cases}$$



M. Lein et al,
2002

M. Lein et al, 2002, 2D H_2^+

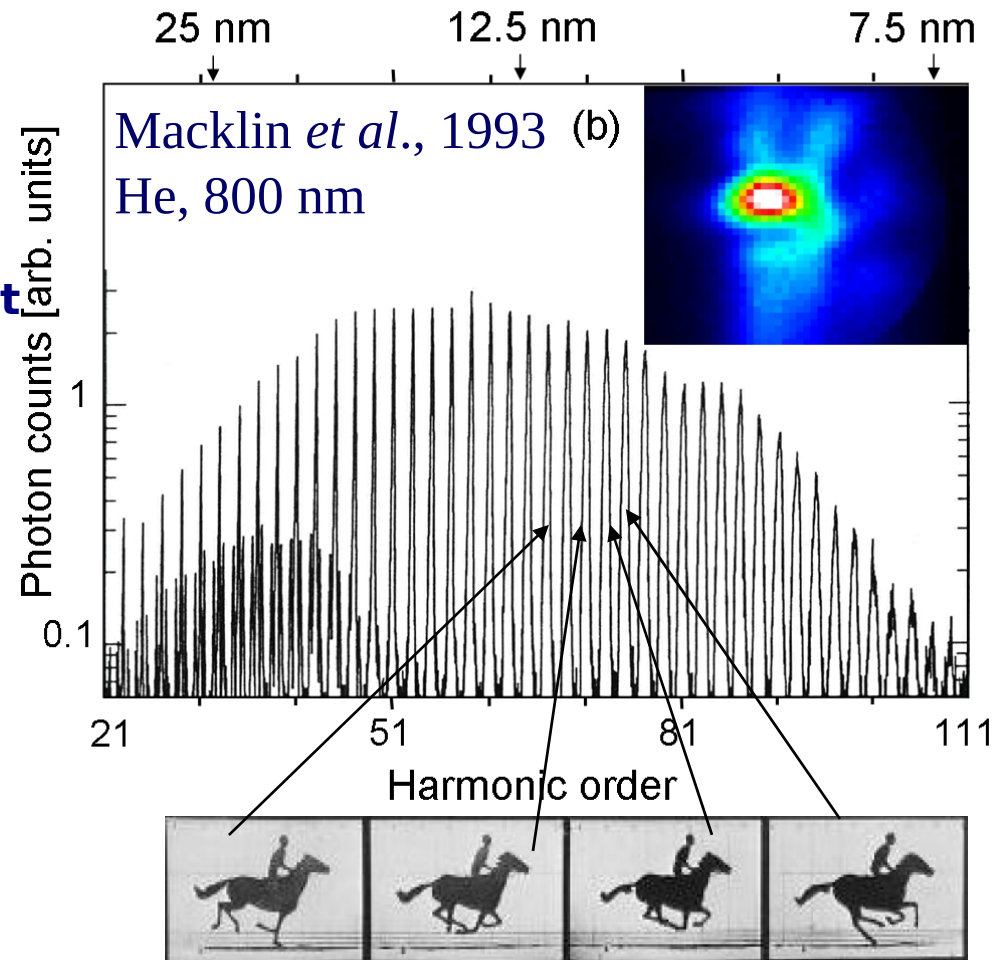


Potential to obtain structural information

HH spectroscopy: Temporal resolution

Temporal resolution comes from attosecond duration of the recollision event

- Emission happens in the half cycle
- Snapshot of recombining system at different times



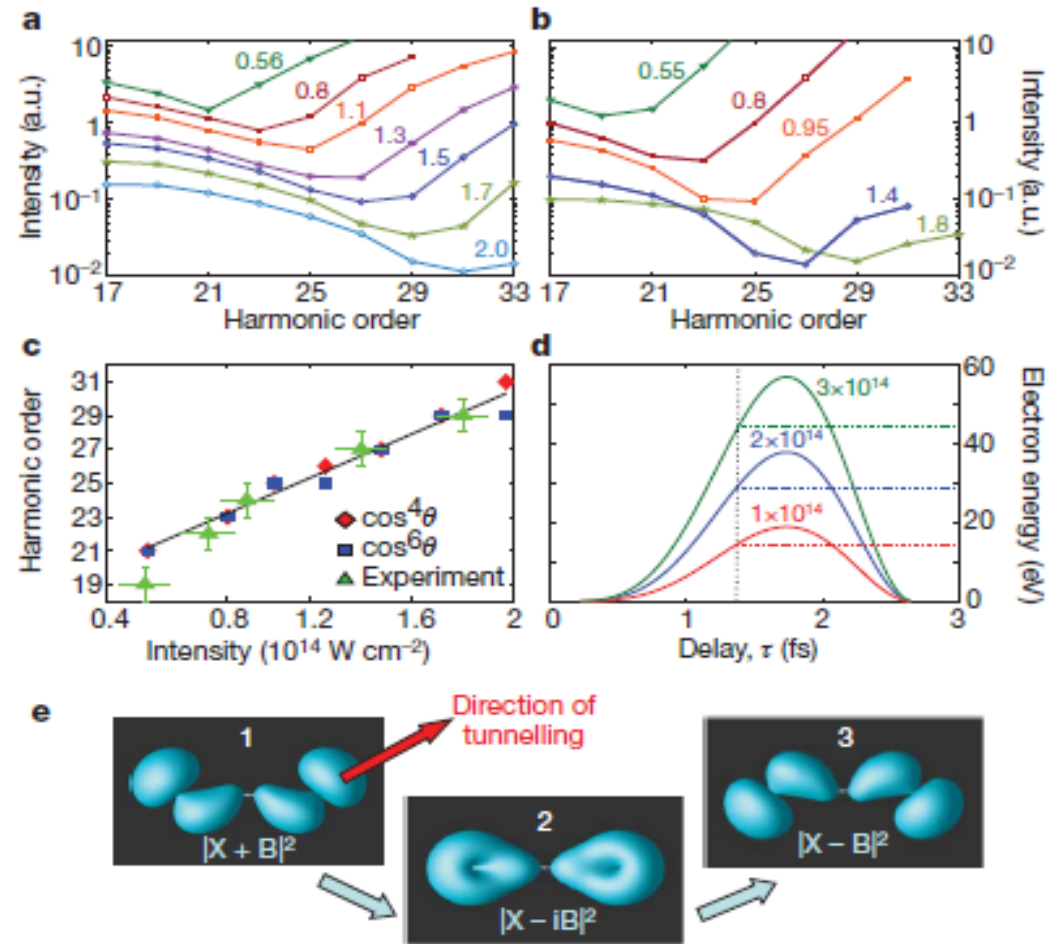
Atto-second temporal resolution

HH spectroscopy: Dynamical Minima

Temporal resolution comes from attosecond duration of the recollision event

- Minimum position depends on intensity.
- Interference from multiple channels.
- Fixed delay time → different return energies for different intensities.

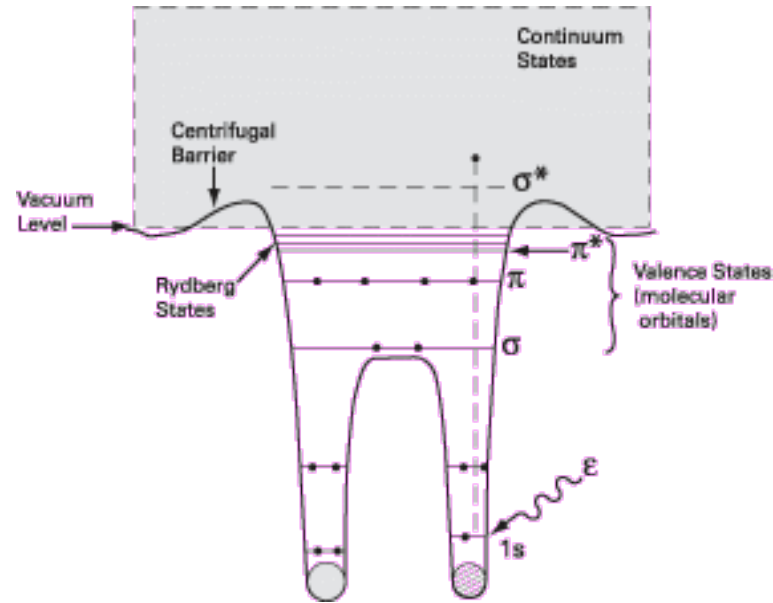
Longer recombination events exist



Carbon Dioxide

Smirnova *et al.*, Nature 2009

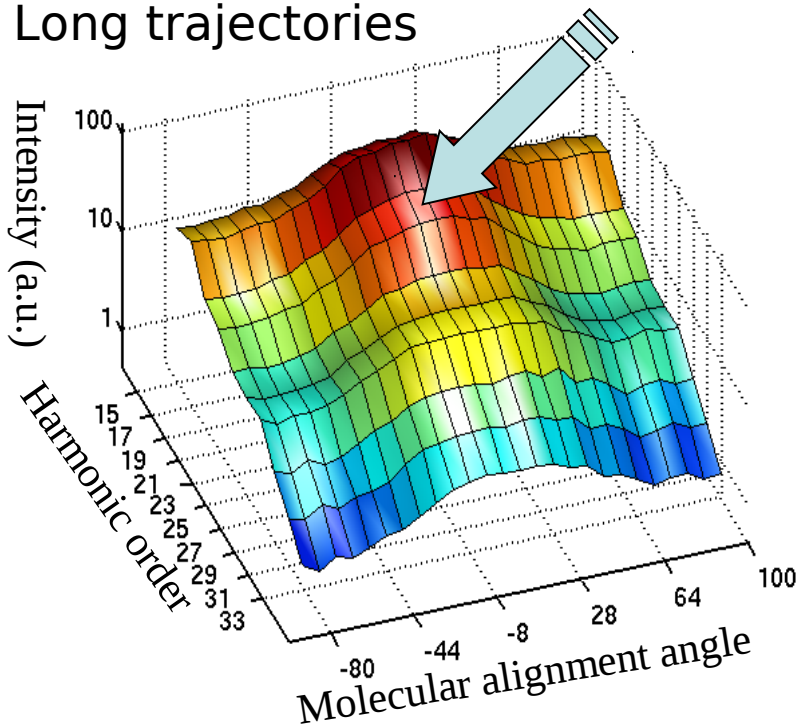
What is a shape resonance?



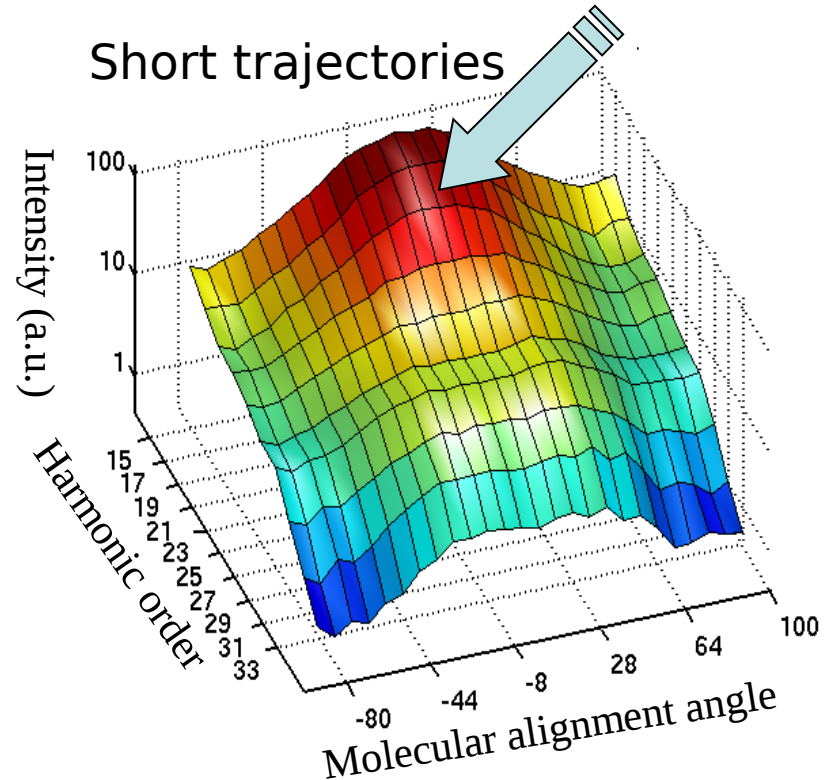
- Metastable state.
- Electron trapped by 'centrifugal barrier' in effective potential.
- Tunnels out

The shape resonance in N₂

Long trajectories



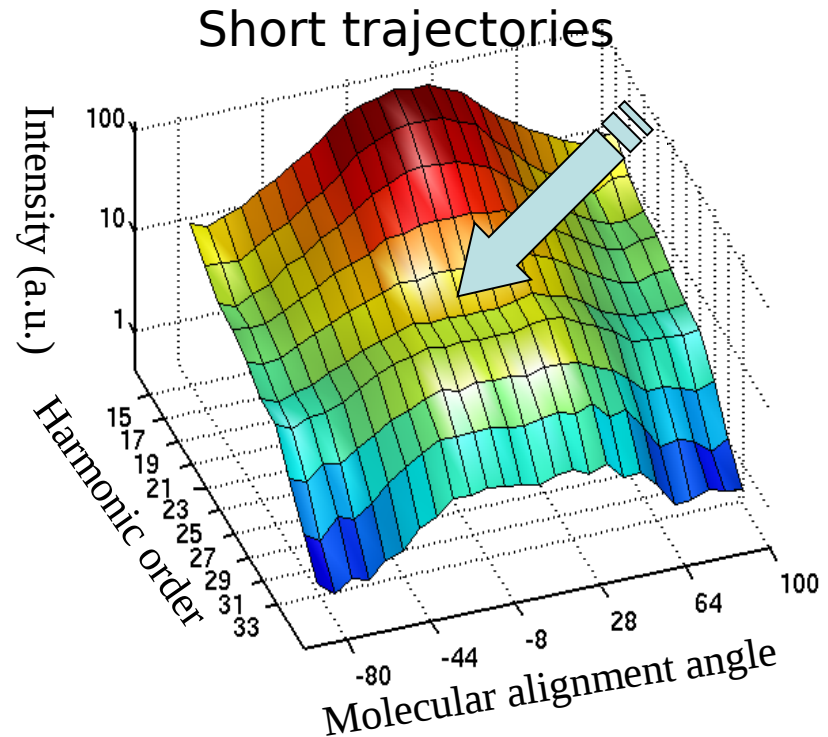
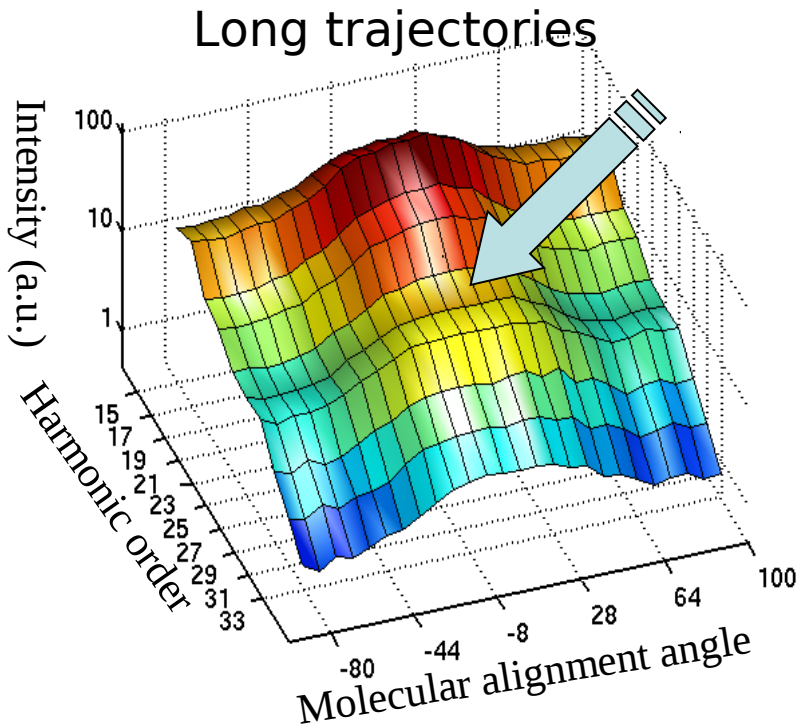
Short trajectories



Taken by Hartmut, Yann, Batiste, Misha and Olga in Bordeaux, March 2010

- N₂ has pronounced shape resonance in recombination
- Probably responsible for the big bump.

The mysterious minimum in N₂ : H₂3-25 @ 800 nm

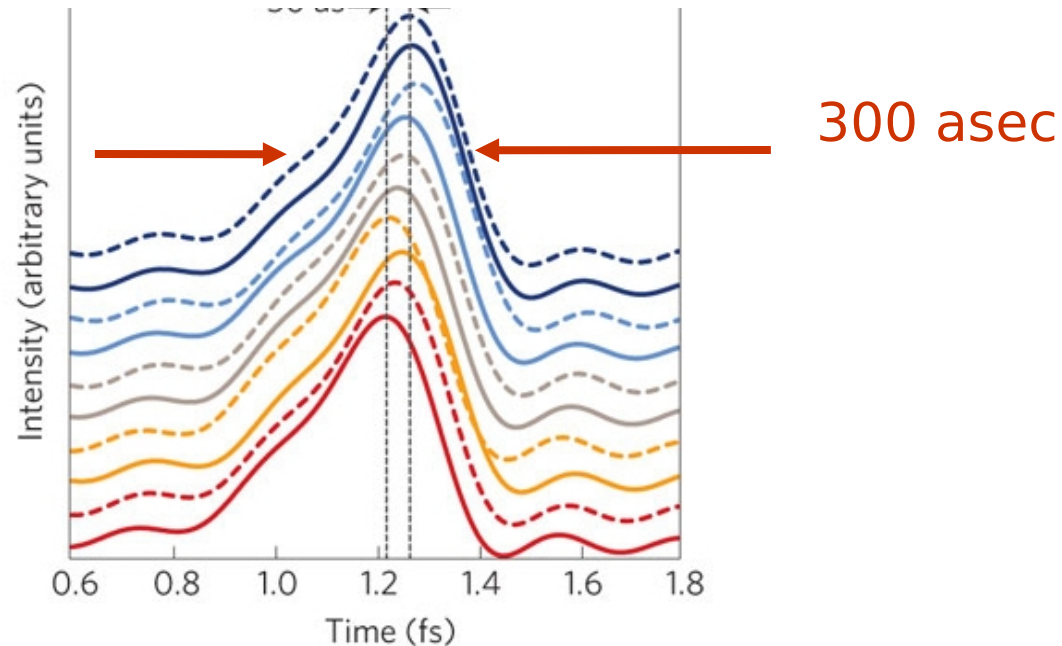


Taken by Hartmut, Yann, Batiste, Misha and Olga in Bordeaux, March 2010

- Same place for long and short trajectories – not dynamics
- Same place for all angles – not 2-center interference
- Where does it come from?

Shape resonance vs short time-scales

Haessler, S. *et al. Nature Phys.* **6**, 200–206 (2010)



Shape resonances trap electrons and should yield long emission tails

But the Paris experiment does not seem to see it!

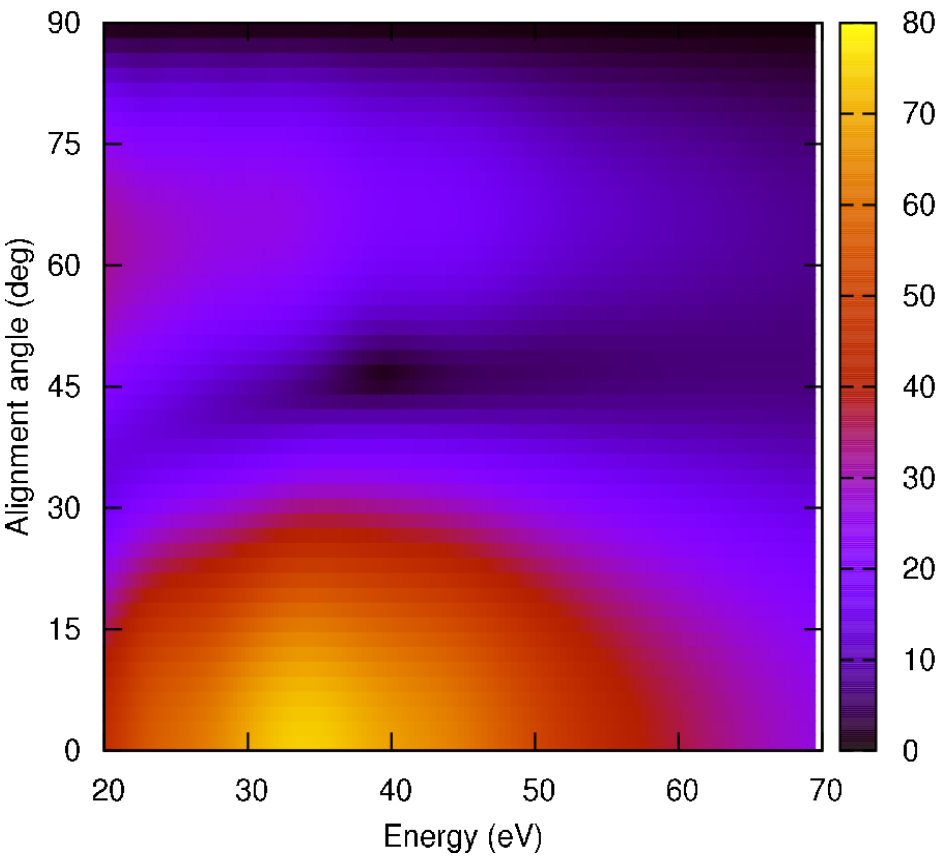
Why don't we filter **prompt attosecond emission out** of the stationary recombination matrix elements?

Dipole matrix elements

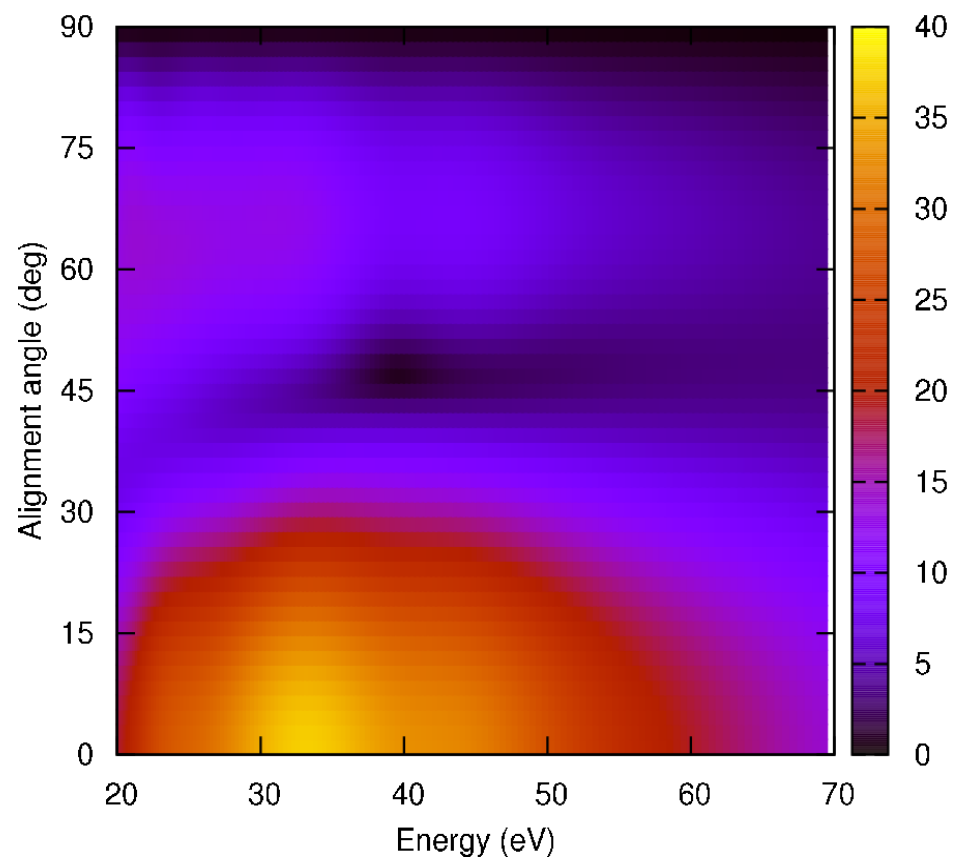
- **Created with FERM3D (Tonzani Comp. Phys. Comm. 2007)**
- **One-electron R-matrix code**
- **HF orbitals, exchange treated at LDA level, correlation potential.**
- **In future will use UK R-matrix codes**
- **Full multi-electron, multi-channel code with full treatment of exchange, polarisation and correlation.**

Time-resolving the shape resonance

5 fsec window



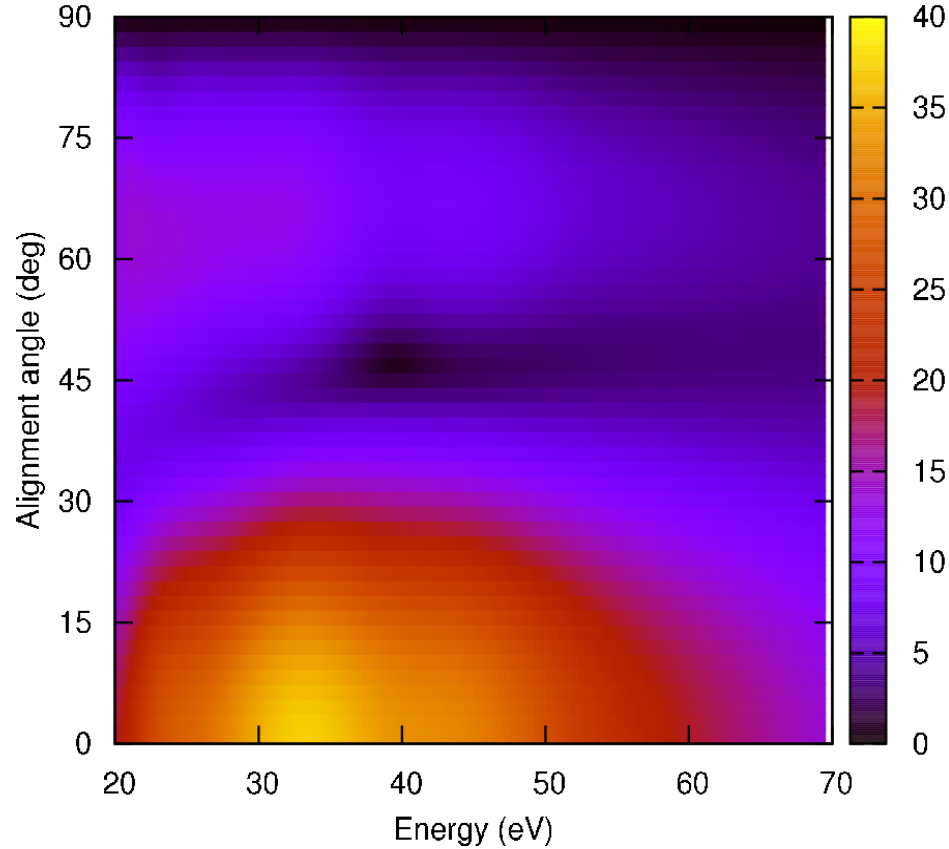
1.9 fsec window



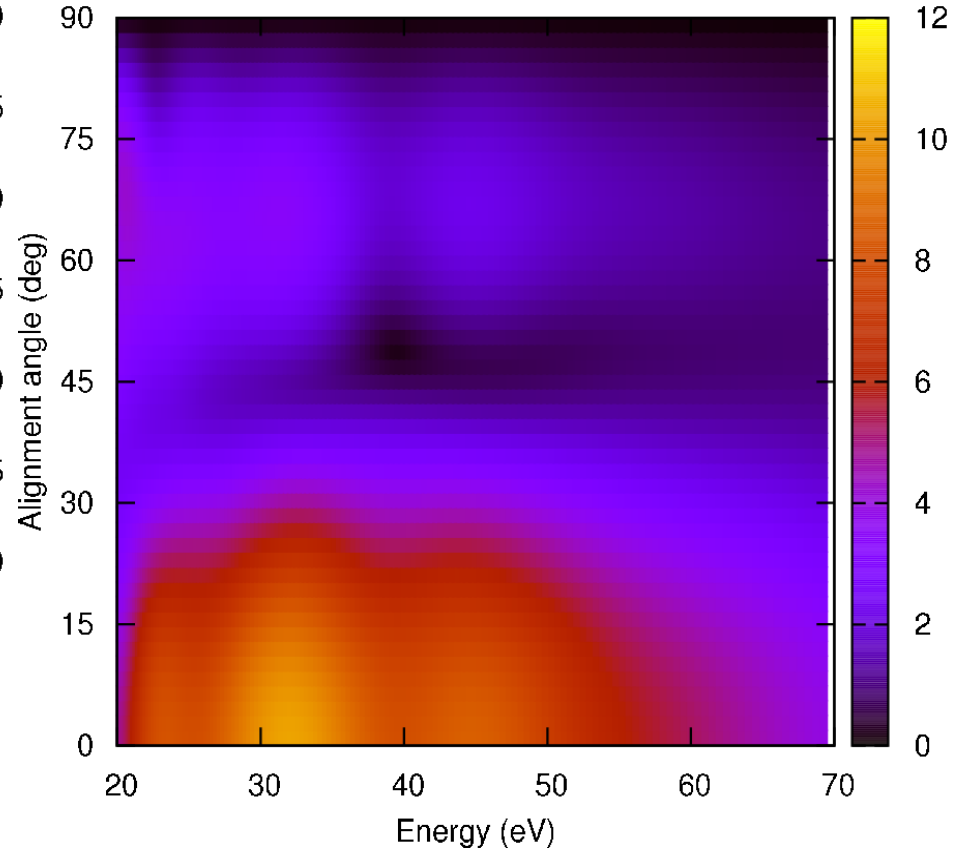
HOMO

Shape resonance vs short time-scales

1.9 fsec window

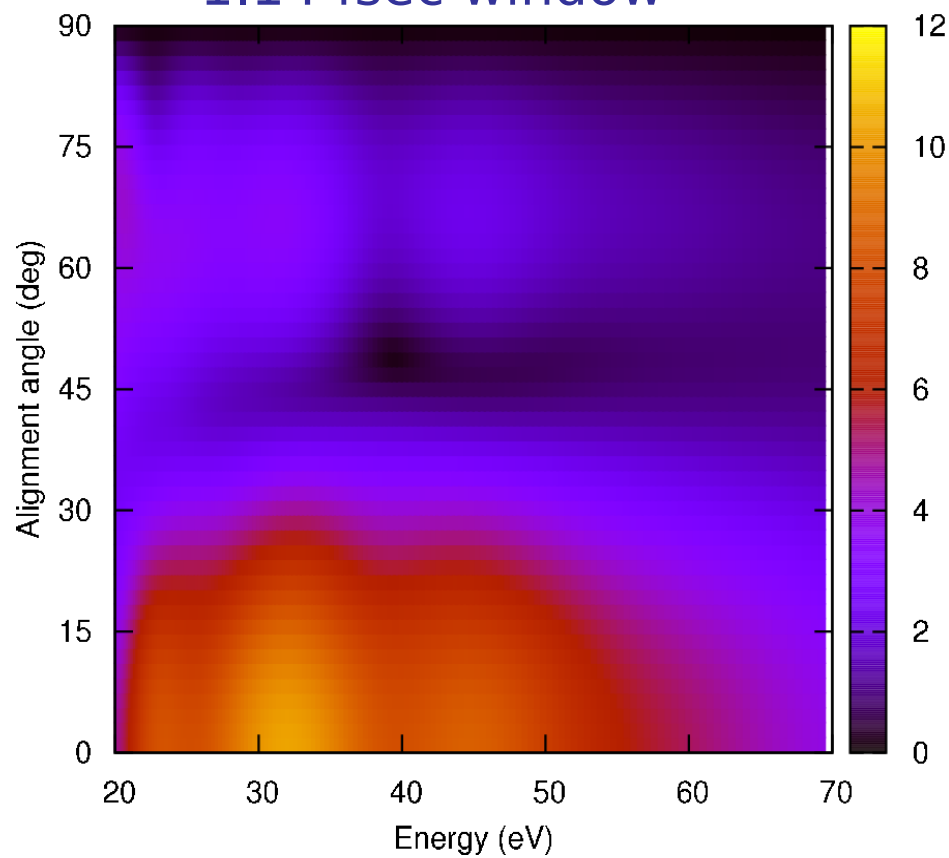


1.14 fsec window

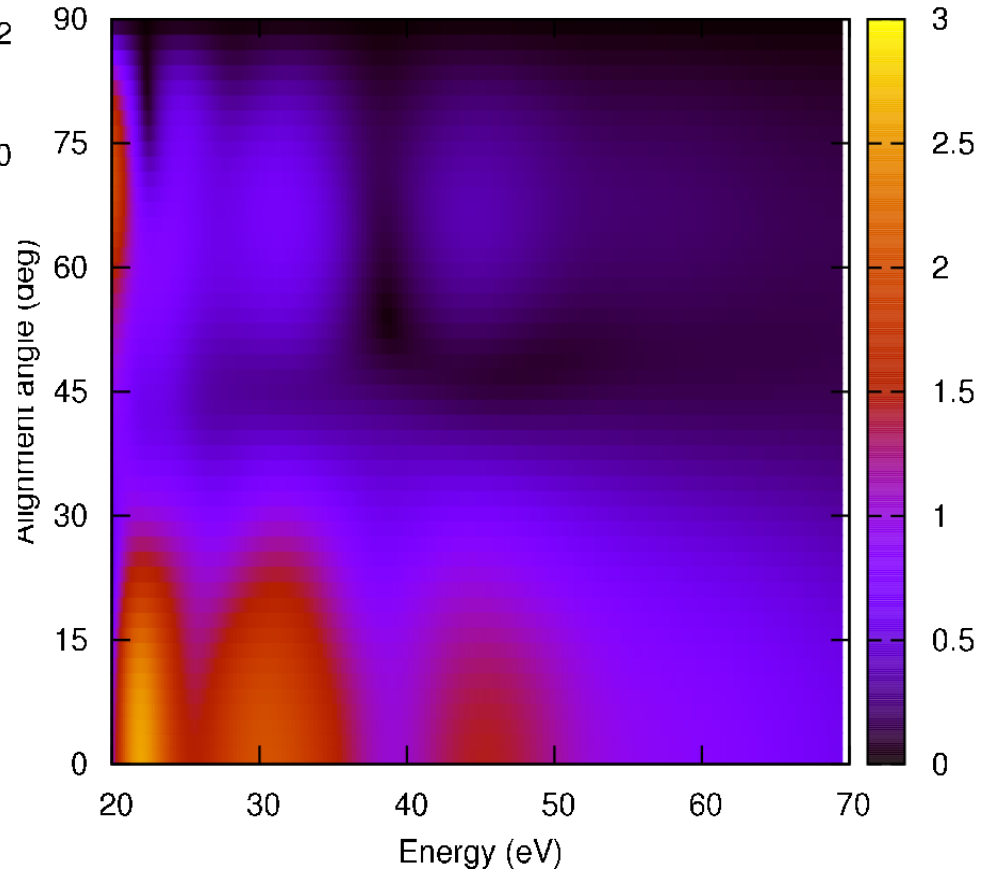


Shape resonance vs short time-scales

1.14 fsec window

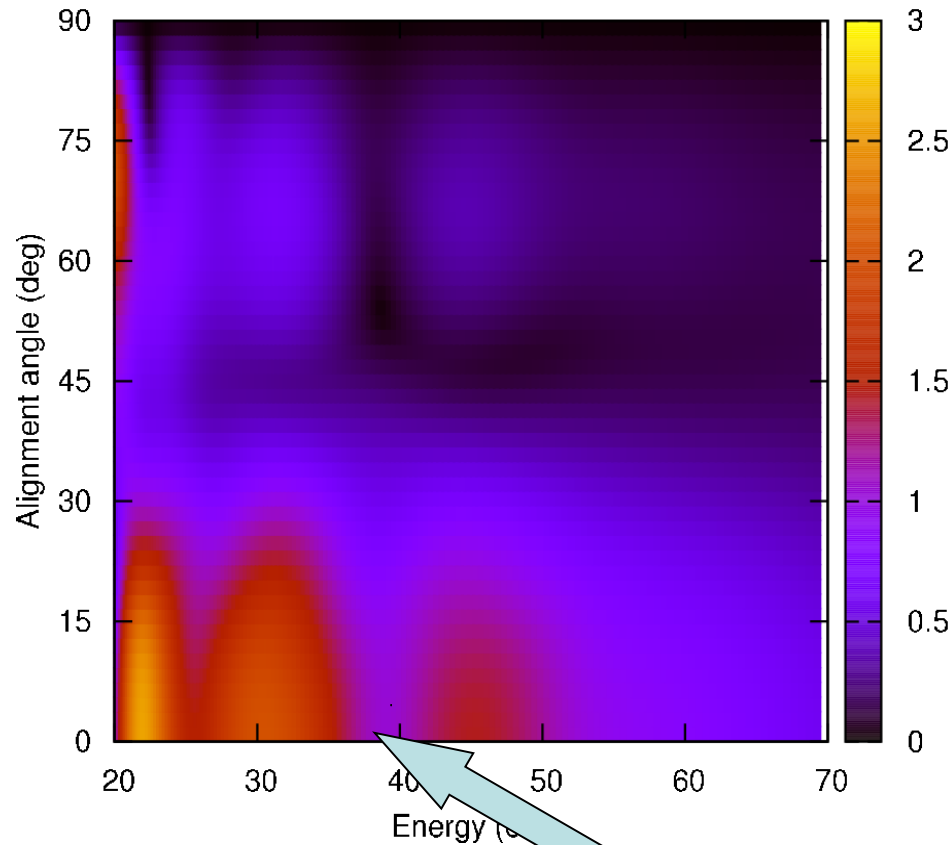


0.8 fsec window

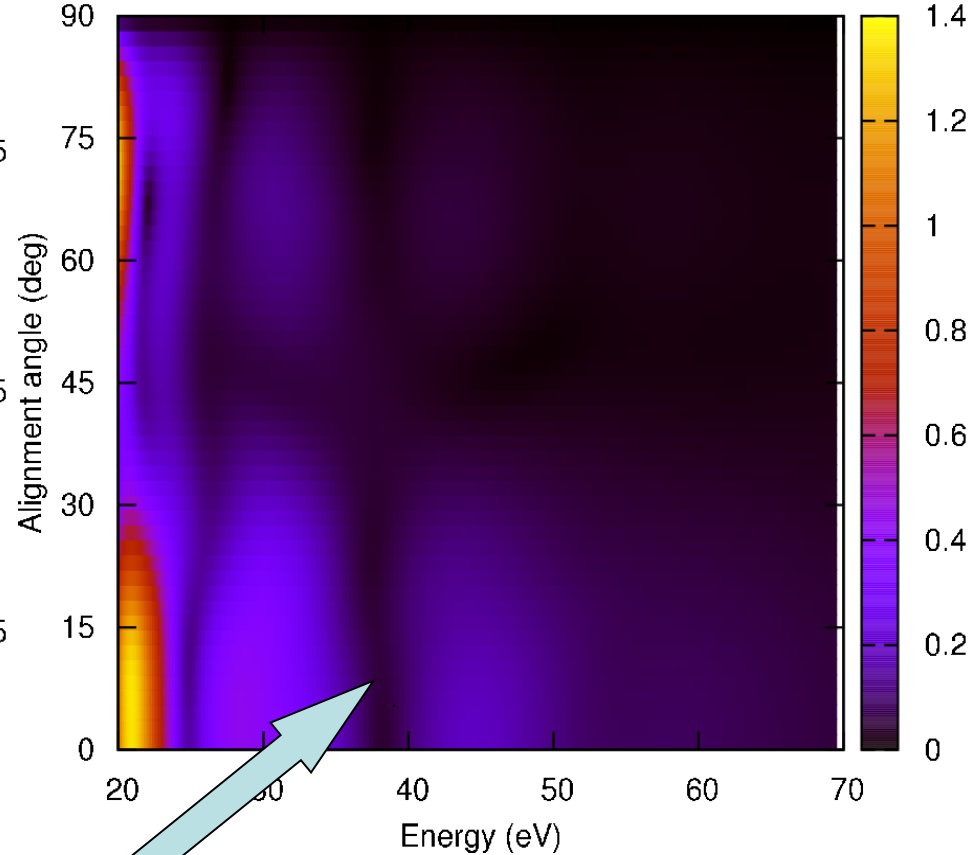


Shape resonance vs short time-scales

0.8 fsec window

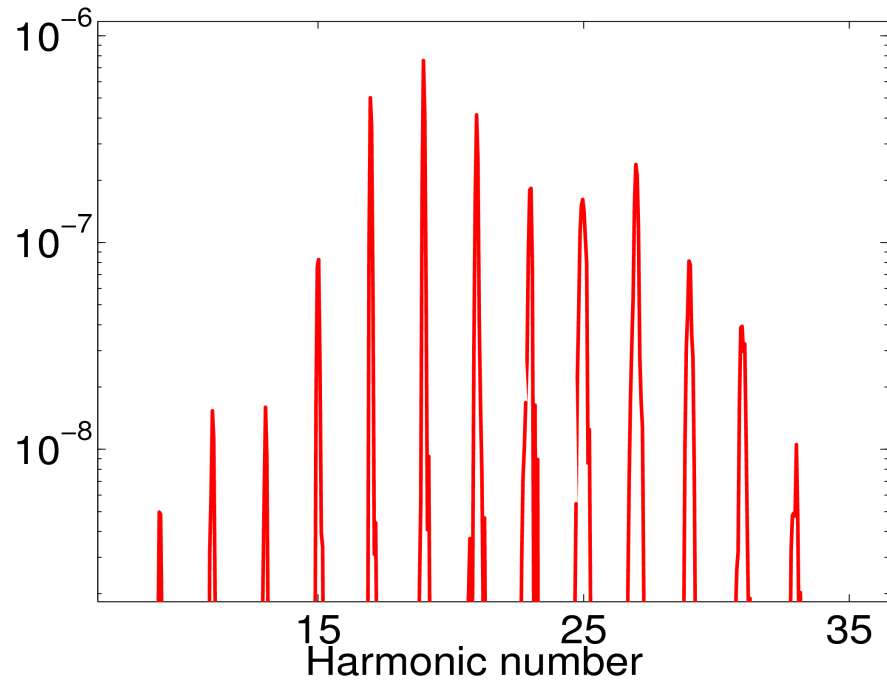


0.6 fsec window

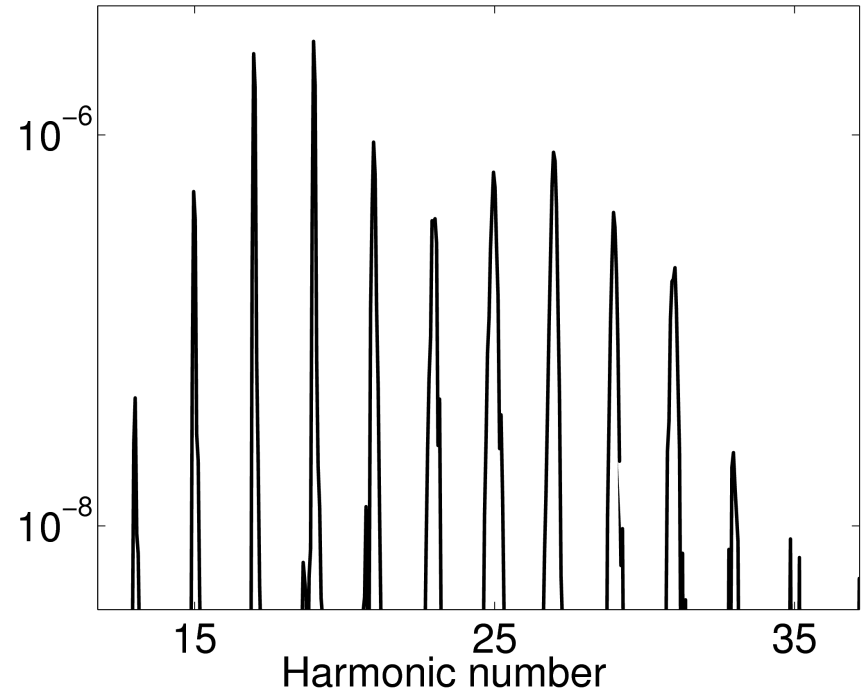


Minimum @ H25 !

Application to HHG



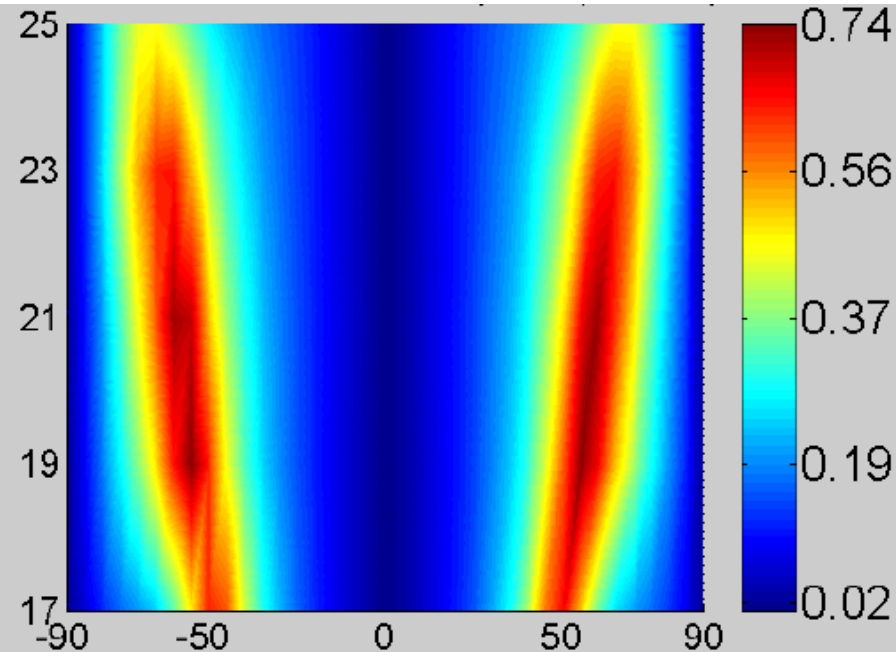
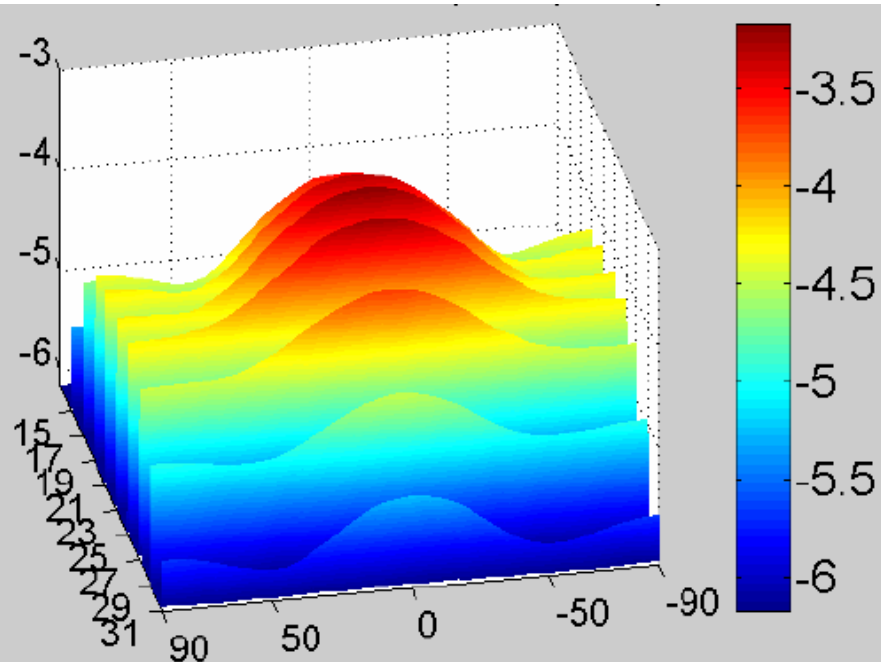
- 800 nm, $0.8 \text{ e}14 \text{ W/cm}^2$
- || alignment
- HOMO only



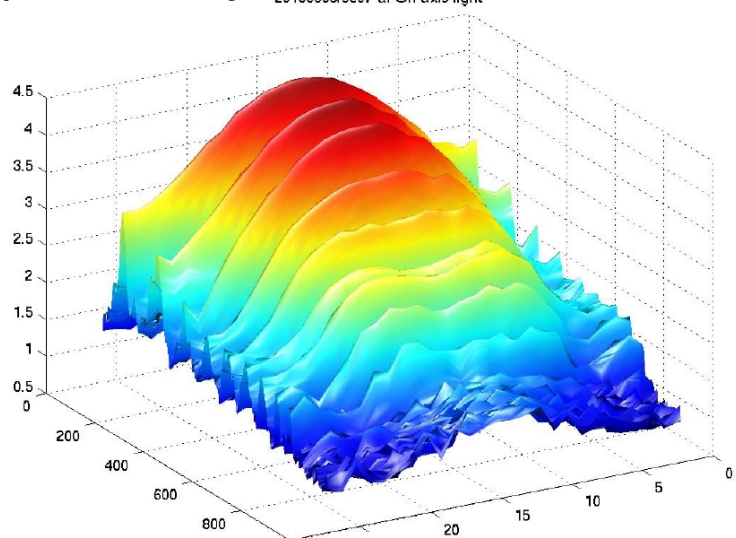
- 800 nm, $0.8 \text{ e}14 \text{ W/cm}^2$
- || alignment
- all channels

HHG no filtering (averaged over alignment distribution $\sim \cos^6$)

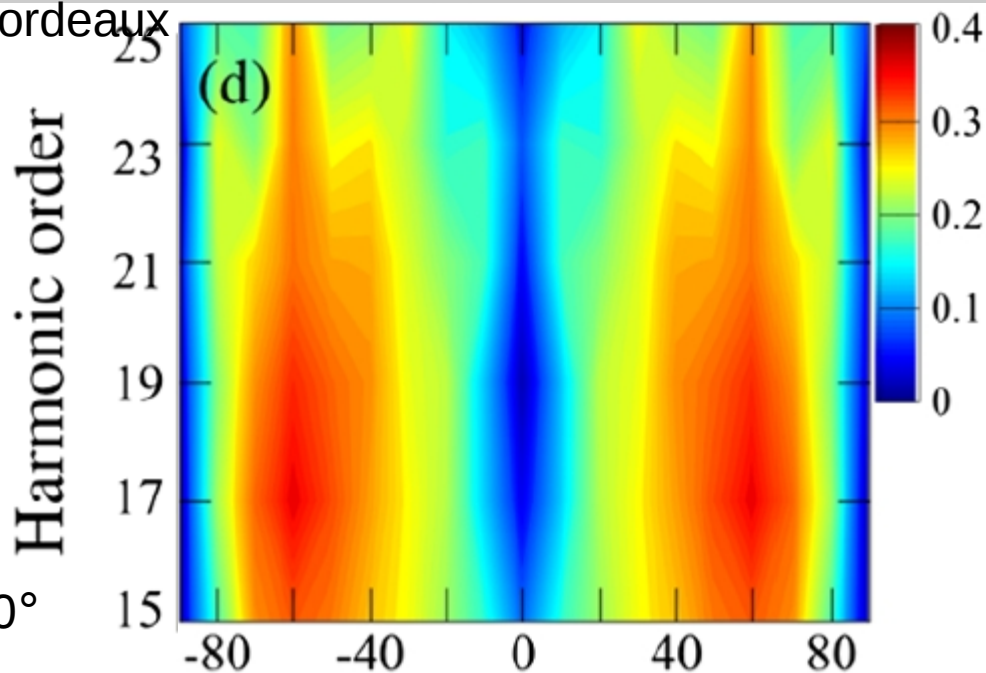
polarization



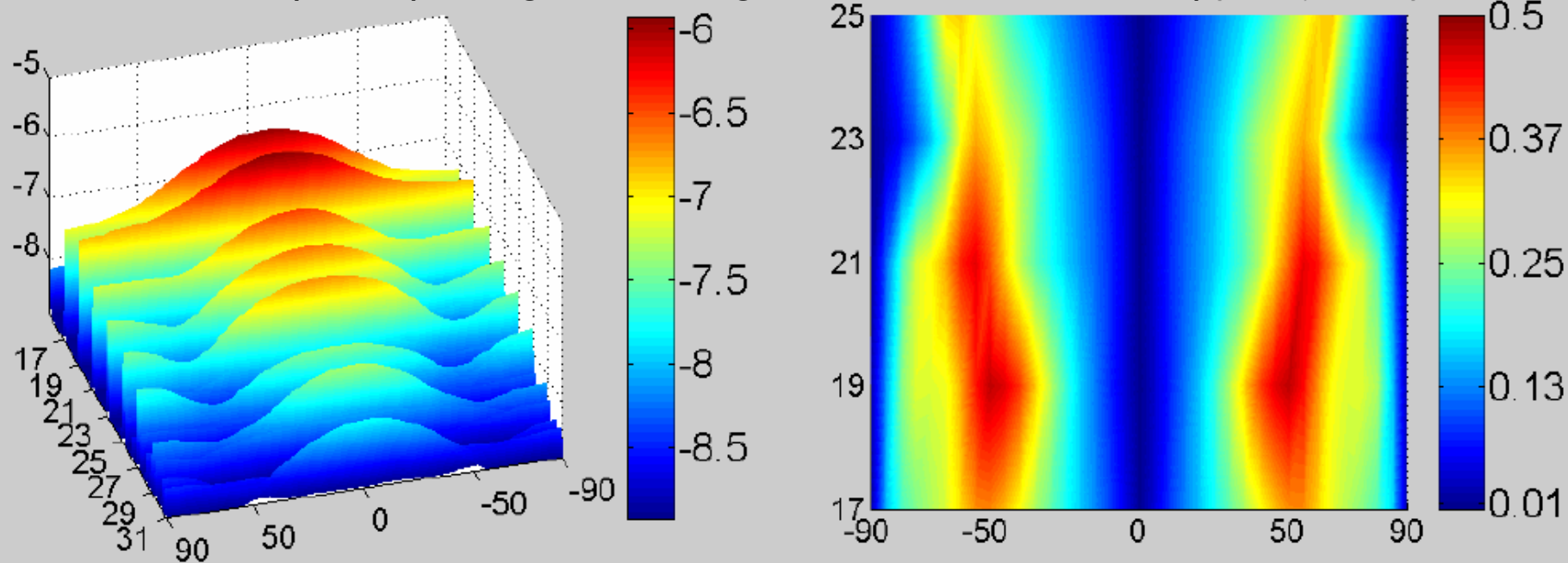
Experiment by Mairesse et al, CELIA, Bordeaux



10^{14} W/cm², starts at H17, -90° to +90°

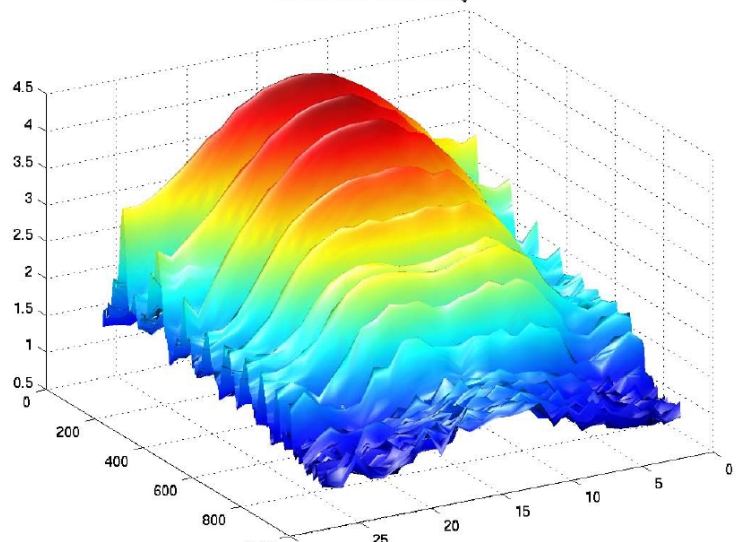


HHG for Filtered dipoles (averaged over alignment distribution $\sim \cos^6$) polarization

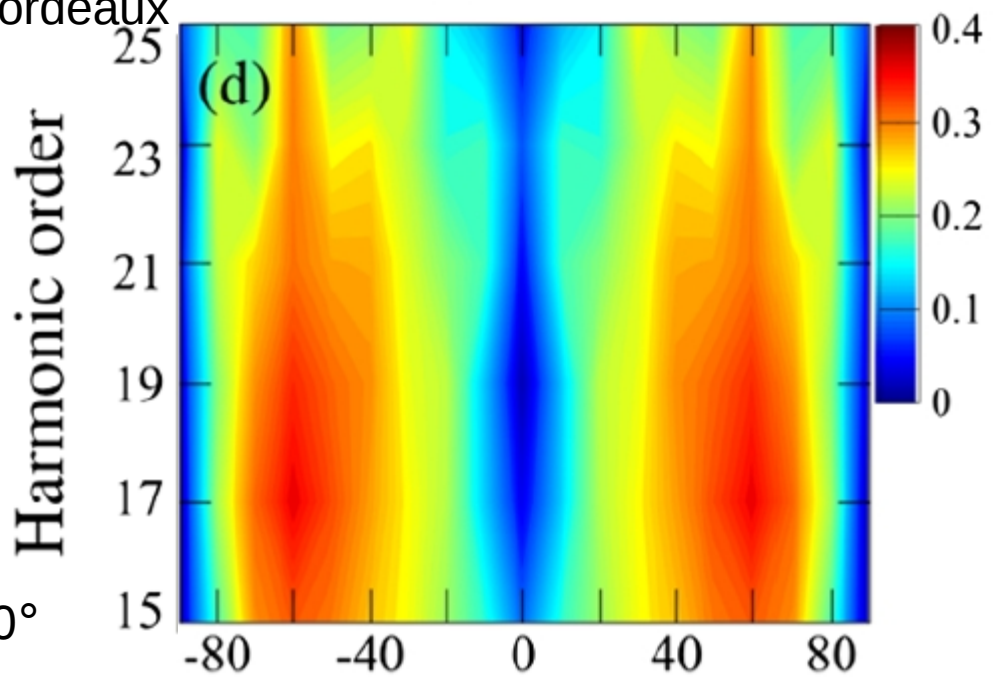


Experiment by Mairesse et al, CELIA, Bordeaux

20100303/sc07-el On-axis light



10^{14} W/cm², starts at H17, -90° to +90°



Conclusions

Shape-resonance at short times

- At short times, the resonance can create a minimum in emission near to a maximum
- The mysterious minimum in HHG from N₂ may be related to this very early dynamics of a shape resonance