Coherent Control with Shaped Ultrafast Laser Pulses

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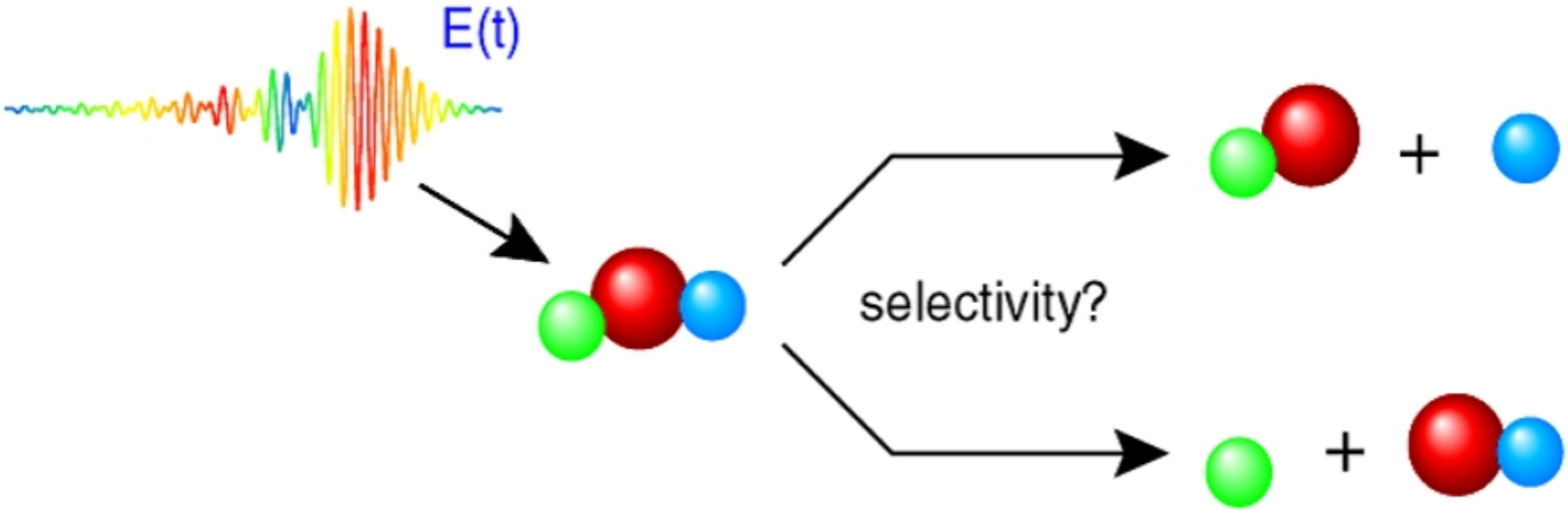
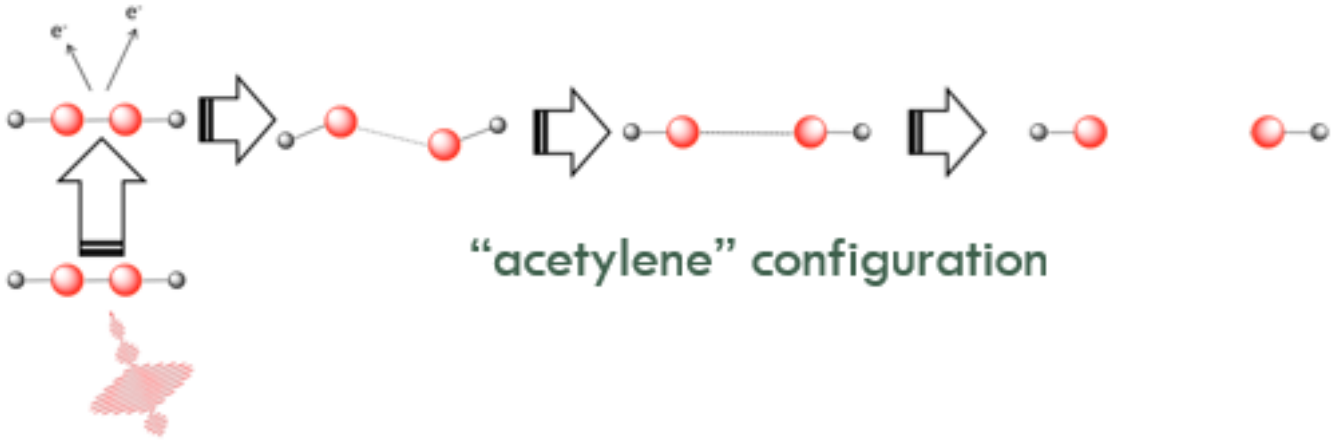
Coherent Control

Control of molecular dynamics (e.g. chemical reactions) with shaped ultrafast laser pulse

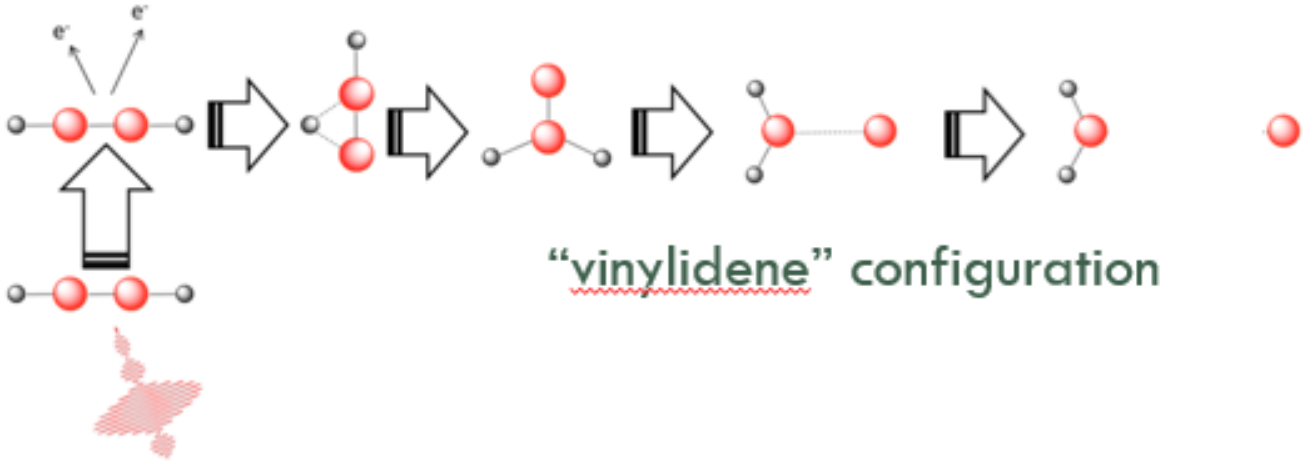


Example: Two possible dynamics of acetylene

Symmetric breakup $n\omega + C_2H_2 \rightarrow C_2H_2^{2+} \rightarrow CH^+ + CH^+$



Isomerization $n\omega + C_2H_2 \rightarrow C_2H_2^{2+} \rightarrow CH_2^+ + C^+$



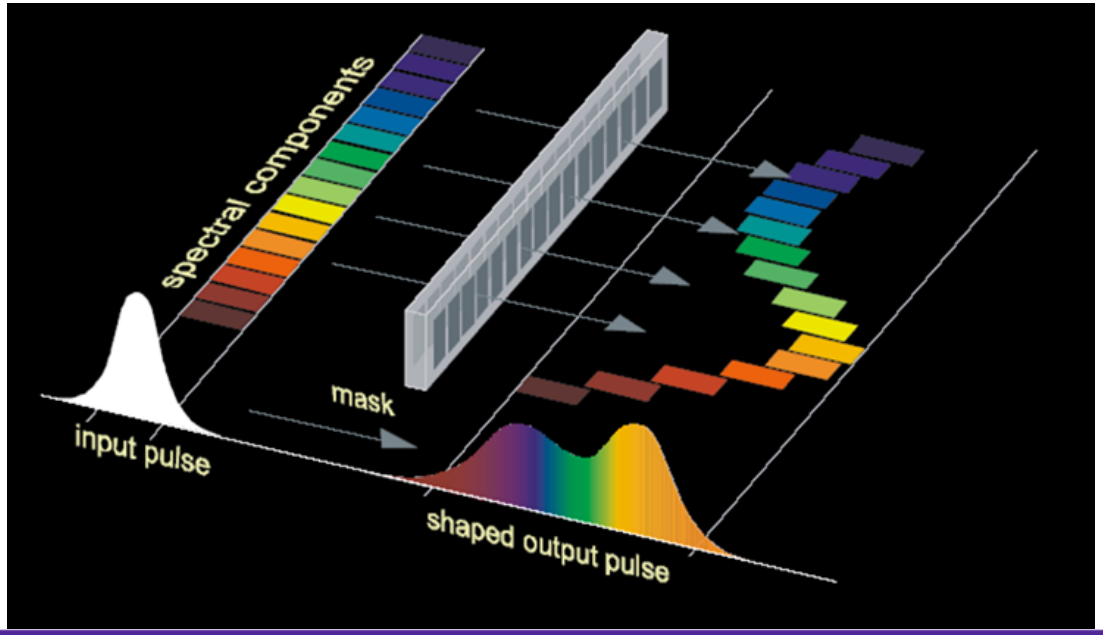
E. Wells *et al.*, *Nat. Comm.* **4** (2895), 2013.

How to Shape a Pulse

Use a pulse shaper (Spatial Light Modulator, Acousto-Optic Modulator)

Diffract the light into different spectral components (wavelengths)

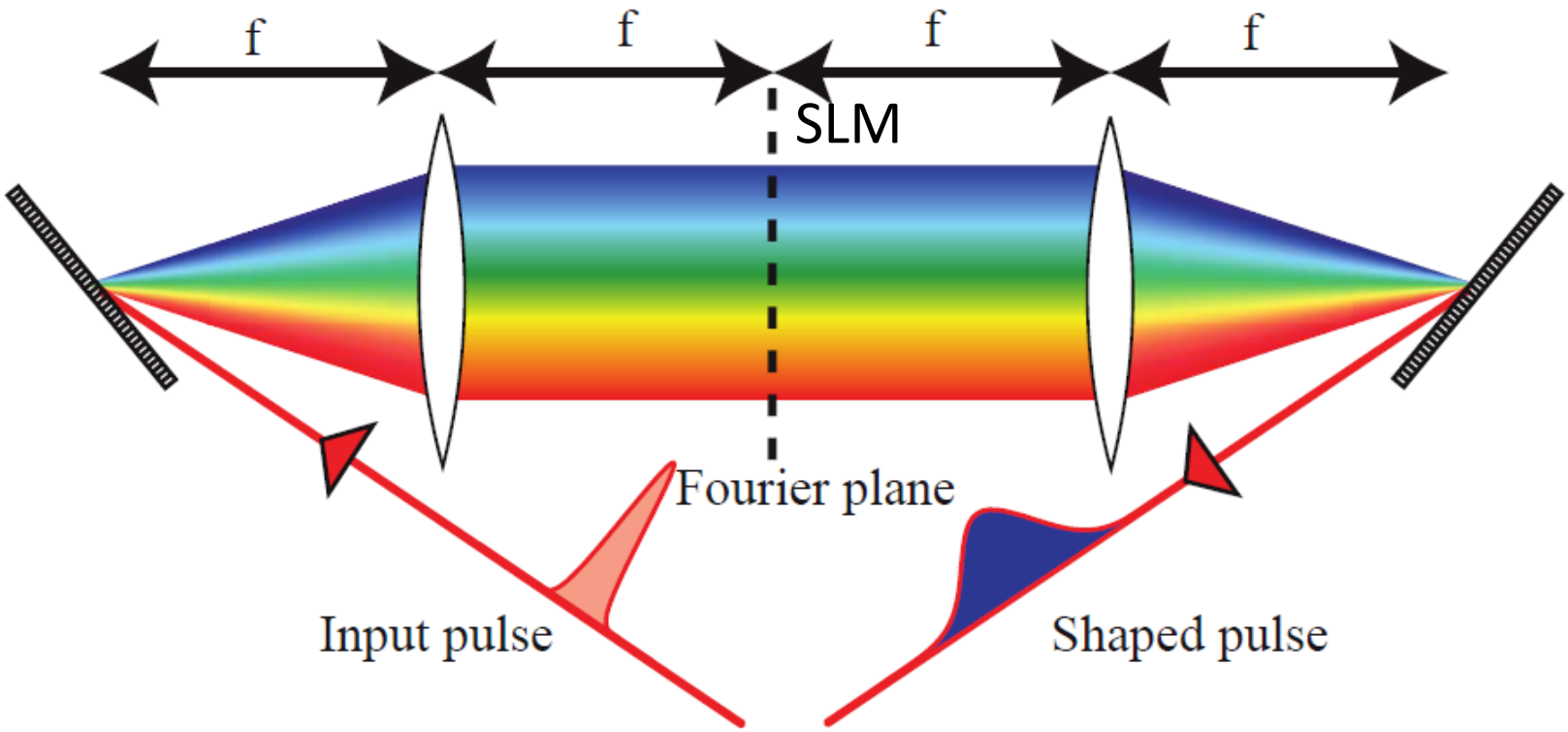
Each “pixel” of the pulse shaper acts differently on each component



Spatial Light Modulator (SLM)

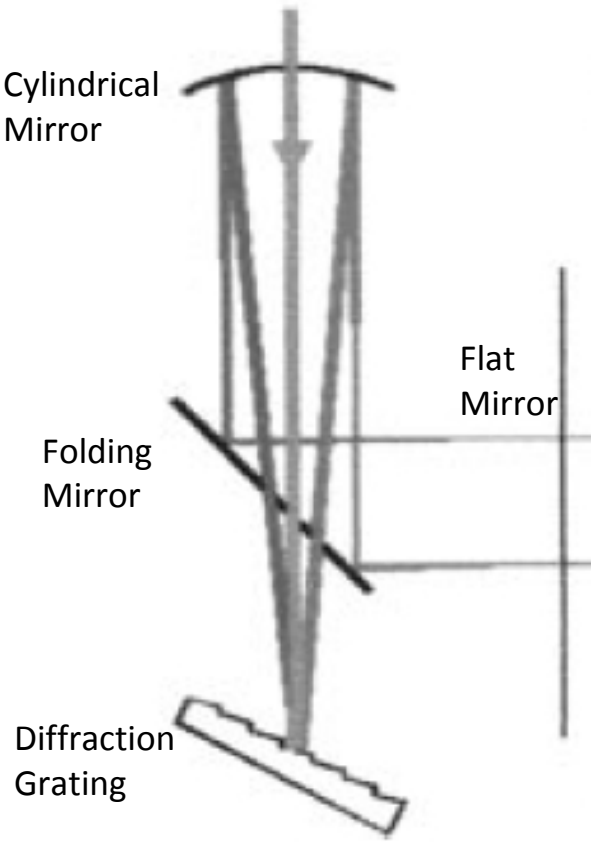
SLM is one type of pulse shaper

Goal of summer project: assemble SLM setup and test it



Ayhan Tajalli, *Génération et caractérisation d'impulsions façonnées - Application au contrôle spatio-temporel de la lumière diffusée* (2012).

SLM 4f Folded Geometry



Cylindrical Mirror

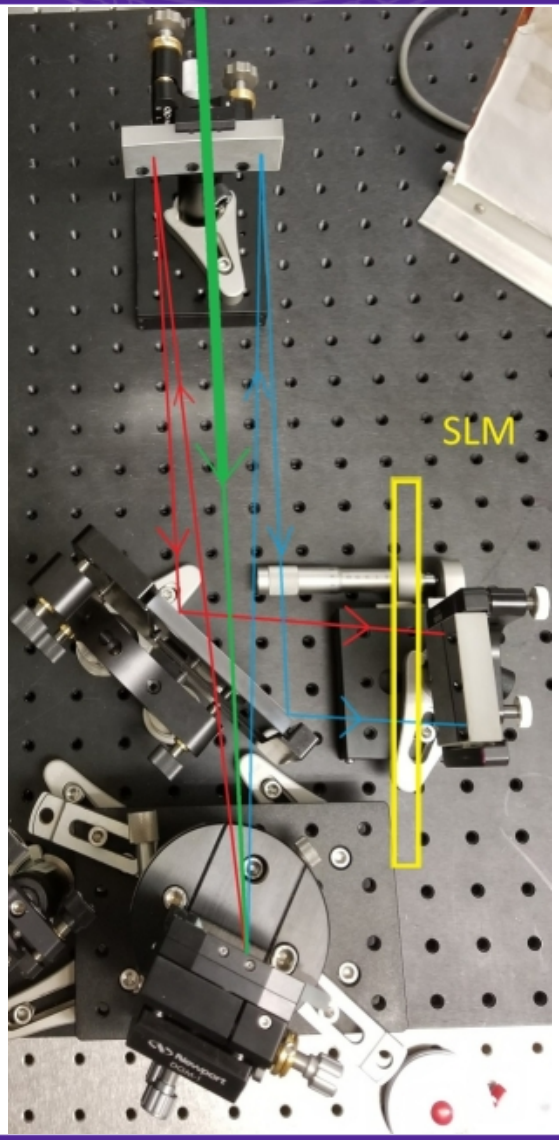
Input beam

Two boundaries for spectra

SLM

Folding Mirror

Diffraction Grating



Flat Mirror

T. Baumert *et al.*, Rev. Sci. Instrum., **74**, 4950 (2003).

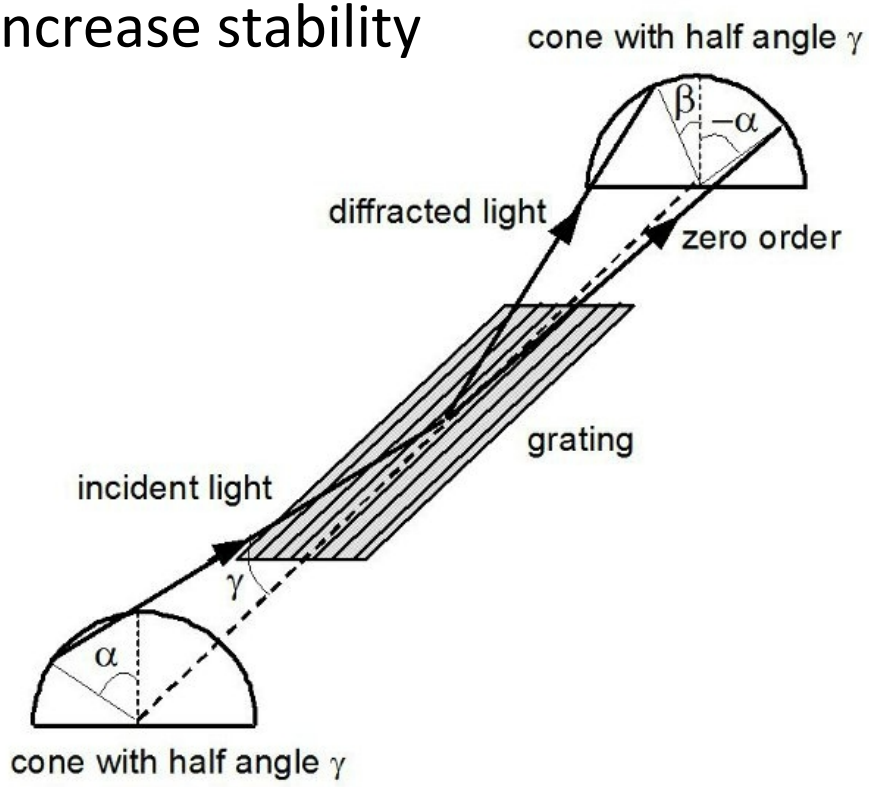
Advantages & Challenges

Advantages:

- Focusing mirror: reduce aberrations
- Fewer optics to deal with: increase stability

Challenges:

- 4f
- Conical diffractions



Luca Poletto, Paolo Villoresi and Fabio Frassetto, *Diffraction Gratings for the Selection of Ultrashort Pulses in the Extreme-Ultraviolet* (2010).

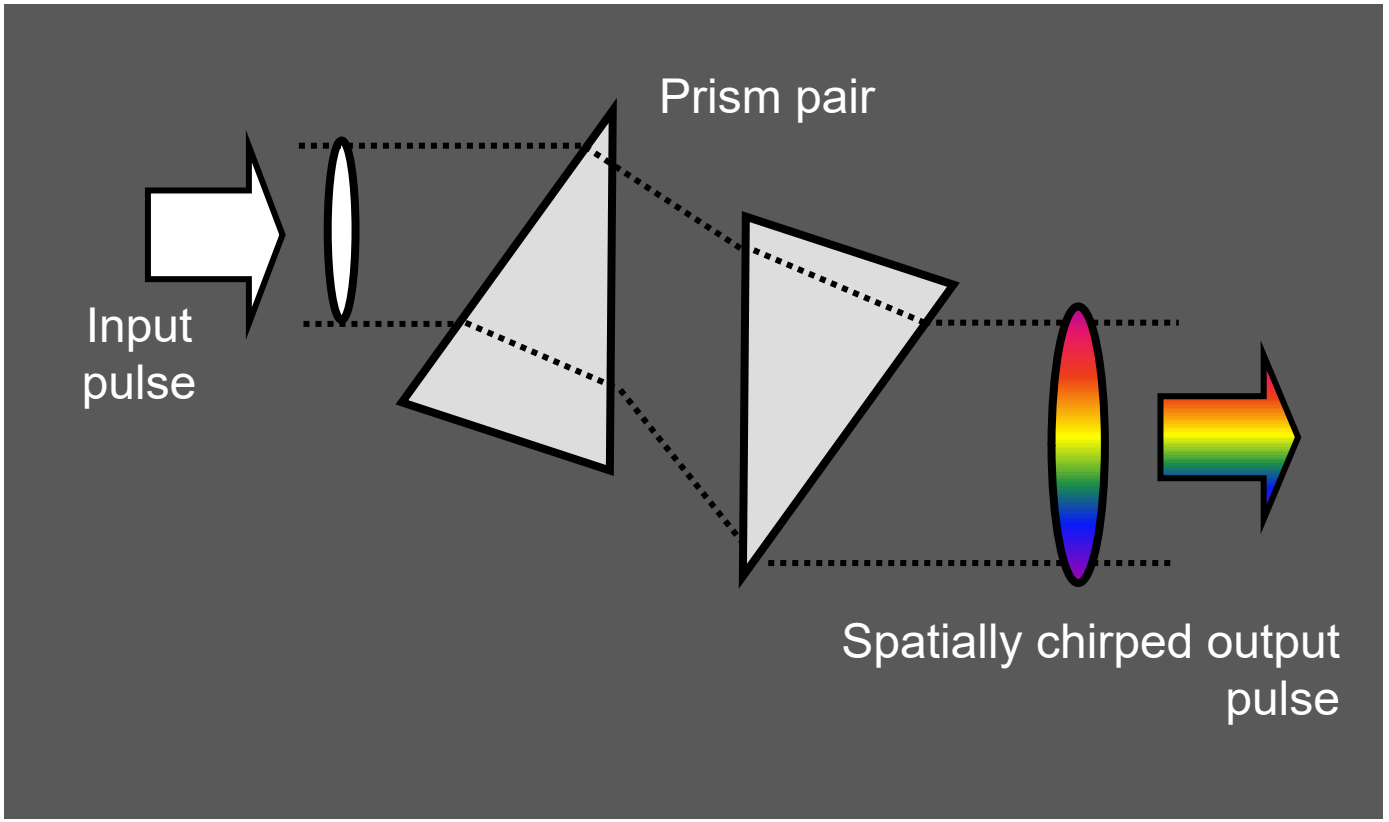
Experience/Progress

Got 4 days of beam time (with spatial chirp on the beam), so we need extra work to get a pulse shaping system (explanation of spatial chirp next slide)

Never assume the quality of laser is perfect

Spatial Chirp

Ultrafast laser pulse consists of a broad frequency range (many different colors) uniformly distributed on the beam spot. In a spatially chirped pulse, different frequencies appear at different positions on the beam spot.

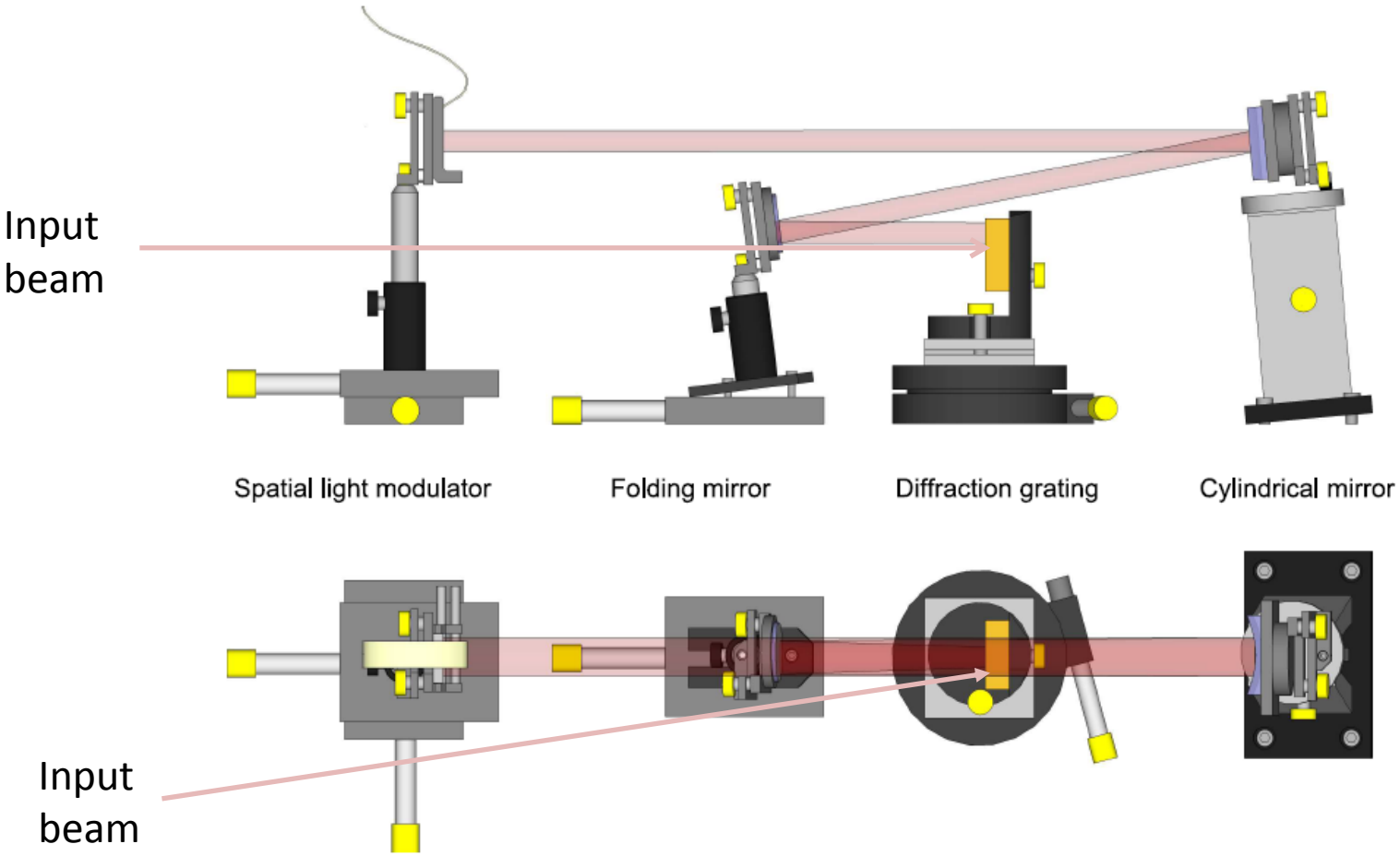


Experience/Progress (Cont'd)

Changed to an alternative geometry that caused less conical diffraction (figure next slide)

Finished geometric alignment of SLM setup

Alternative SLM Folded Geometry



Dann Sprunken, *a 2D Spatial Light Modulator for spatial-temporal shaping* (2008)

Future Work

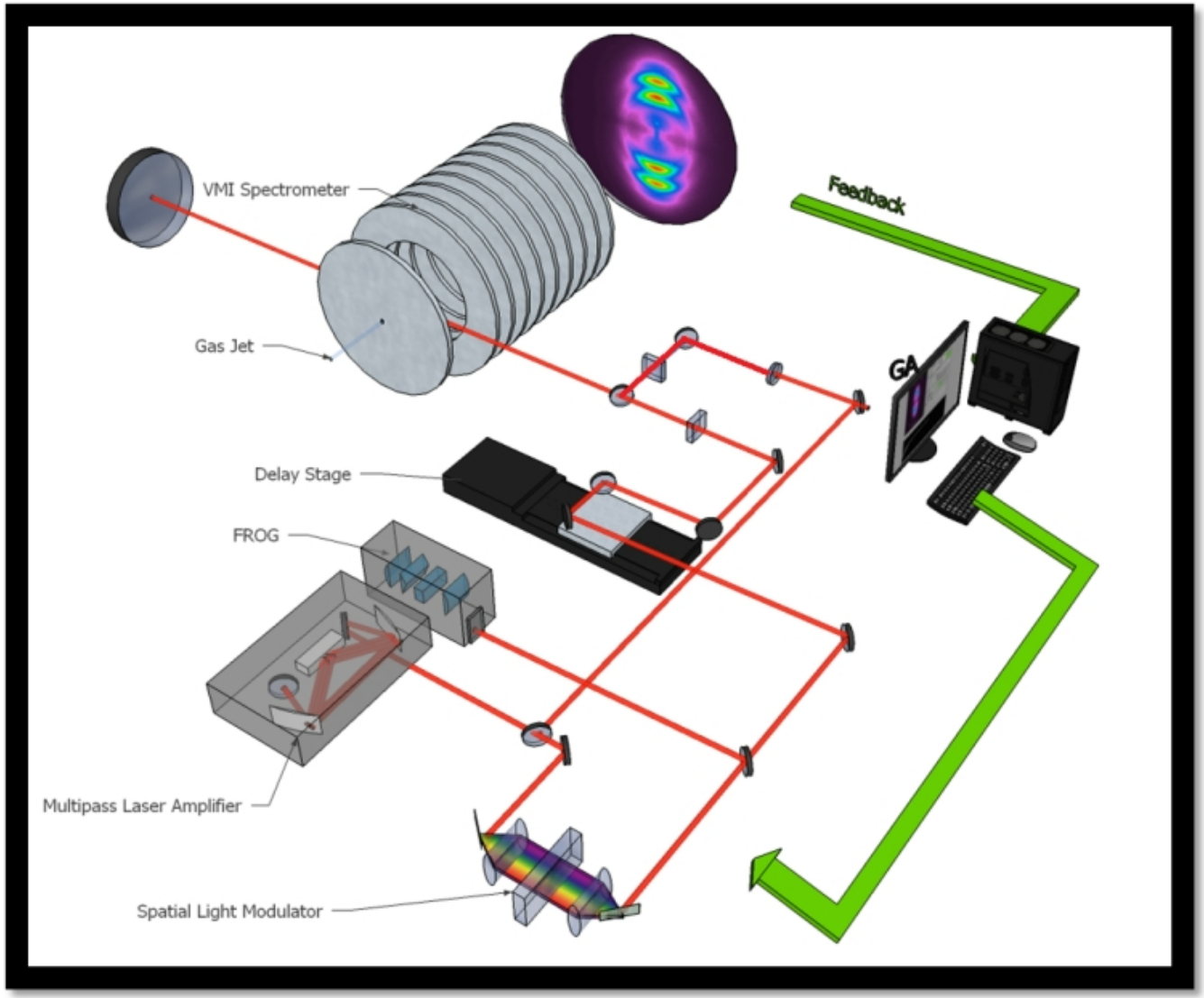
Complete femtosecond alignment (fine adjustments) of SLM setup to achieve 4f

Build another arm of beam path to form pump-probe configuration (figure next slide)

Run coherent control experiment with SLM as the pulse shaper on the reaction:

$C_2D_6^{2+} + 2e^- \rightarrow CD_4\text{-}CD_2^{2+} \rightarrow C_2D_4^{2+} + D_2$ transition state $\rightarrow$ $C_2D_3^+ + D_3^+$

SLM Pump-Probe Schematic Diagram



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http://www.frog.gatech.edu/talks.html.

E. Wells *et al.*, *Nat. Comm.* **4** (2895), 2013.

Ayhan Tajalli, *Génération et caractérisation d'impulsions façonnées - Application au contrôle spatio-temporel de la lumière diffusée* (2012).

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Antoine Monmayrant, Sébastien Weber, Béatrice Chatel, J. Phys. B: At. Mol. Opt. Phys. **43** (2010).

T. Baumert *et al.*, Rev. Sci. Instrum., **74**, 4950 (2003).

Dann Sprunken, *a 2D Spatial Light Modulator for spatial-temporal shaping* (2008).

Acknowledgements

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