Time-resolved photoelectron imaging using hollow-core photonic crystal fibres and capillaries

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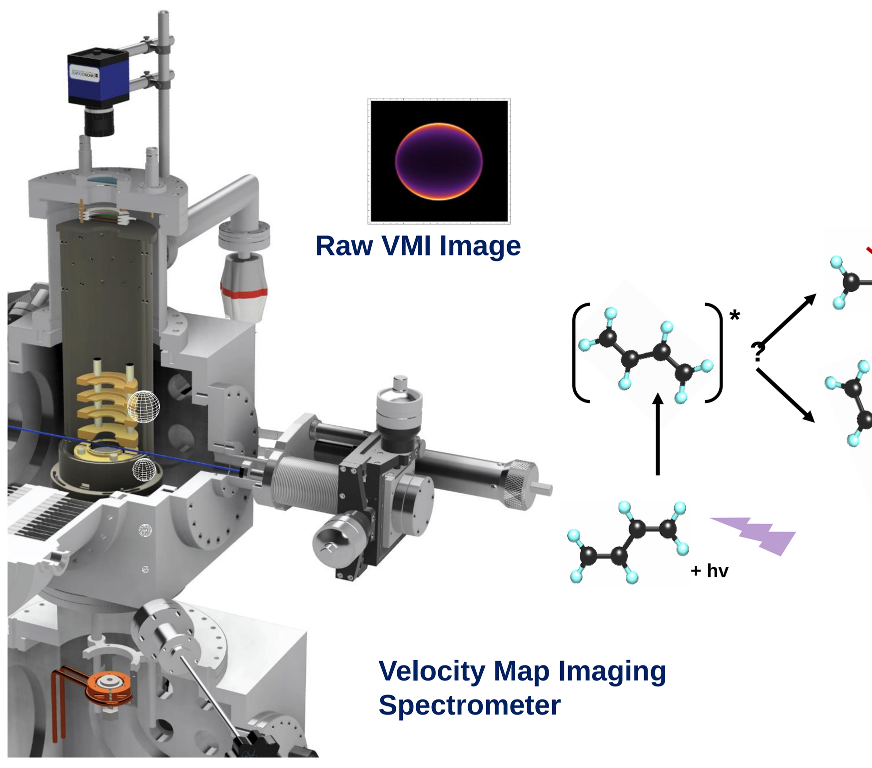
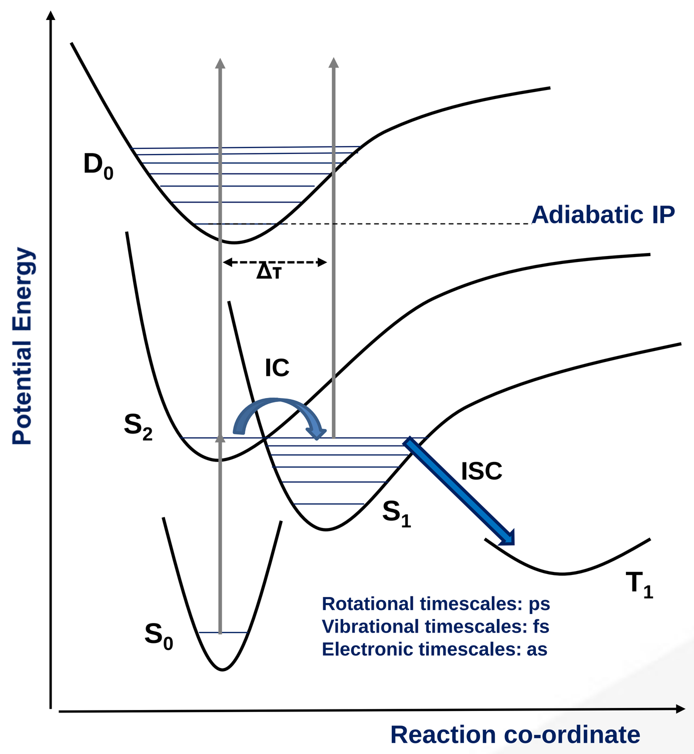
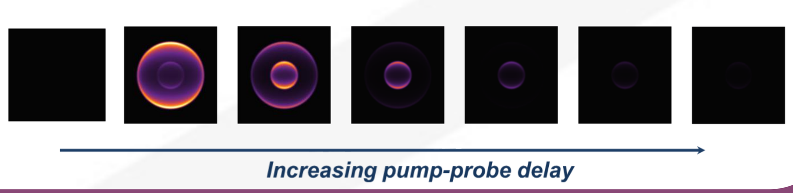
Time-resolved Photoelectron Imaging (TRPEI) Setup

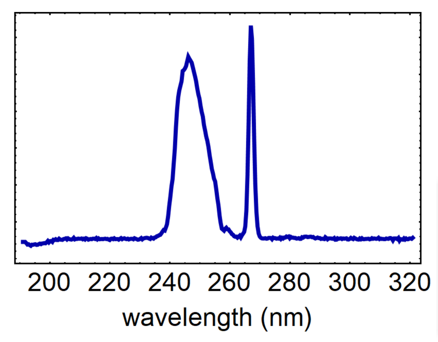




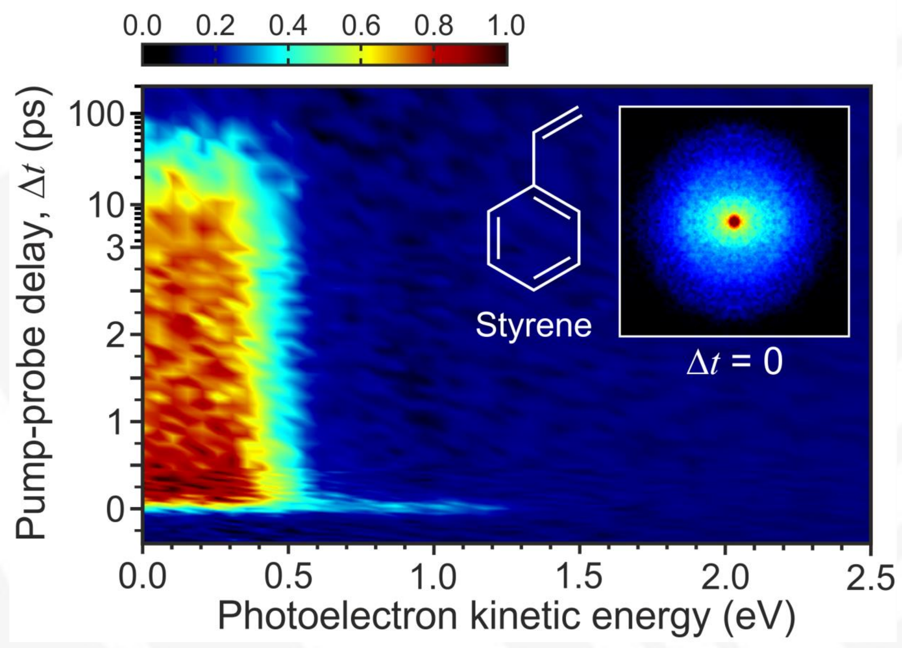
- Gas phase pump-probe measurements under ultrahigh vacuum conditions with Velocity Map Imaging (VMI)
- Time-, energy- and angle-resolved data

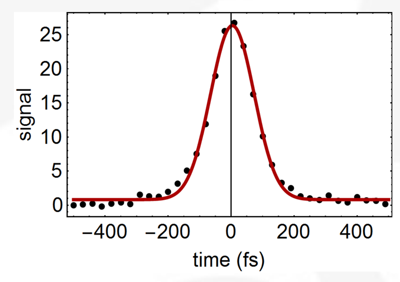


First Demonstration-Styrene



Ionisation scheme (1+1')
Pump Photon (HC-PCF): 248 nm
Probe Photon (BBO): 267 nm



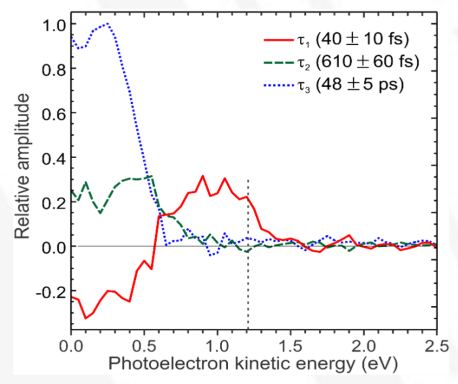


Cross Correlation: ~160 fs

$S_0 \xrightarrow{h\nu} S_2 \xrightarrow{IC} S_1 \xrightarrow{IC} S_0$

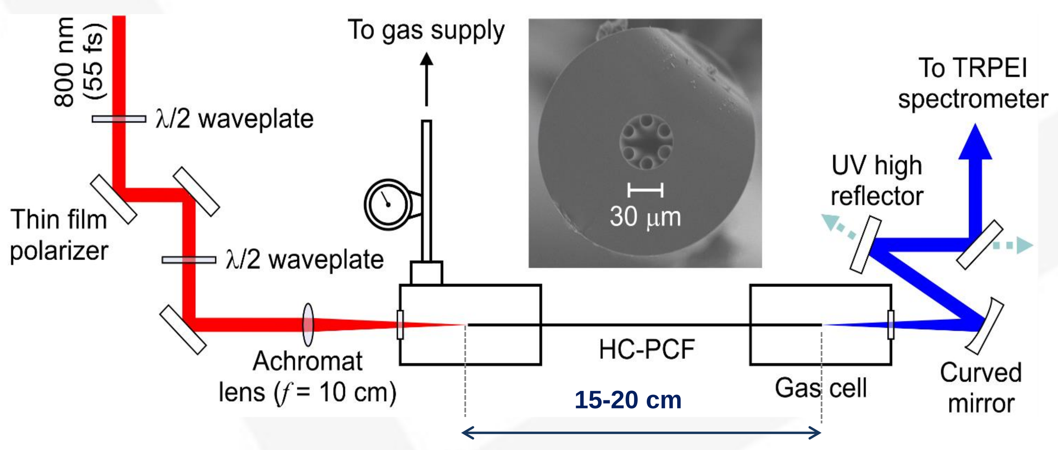
Time-resolved photoelectron spectrum (top-down view). The time scale is linear from -450 fs to 3 ps and logarithmic beyond that. Pump and probe photon at 247 and 267 nm, respectively [1].

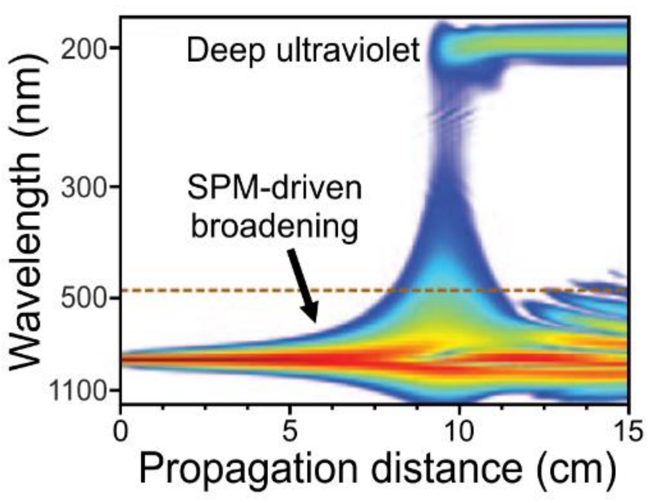
Decay Associated Spectra (DAS) plot and the three time constants associated with the fitted data [1]

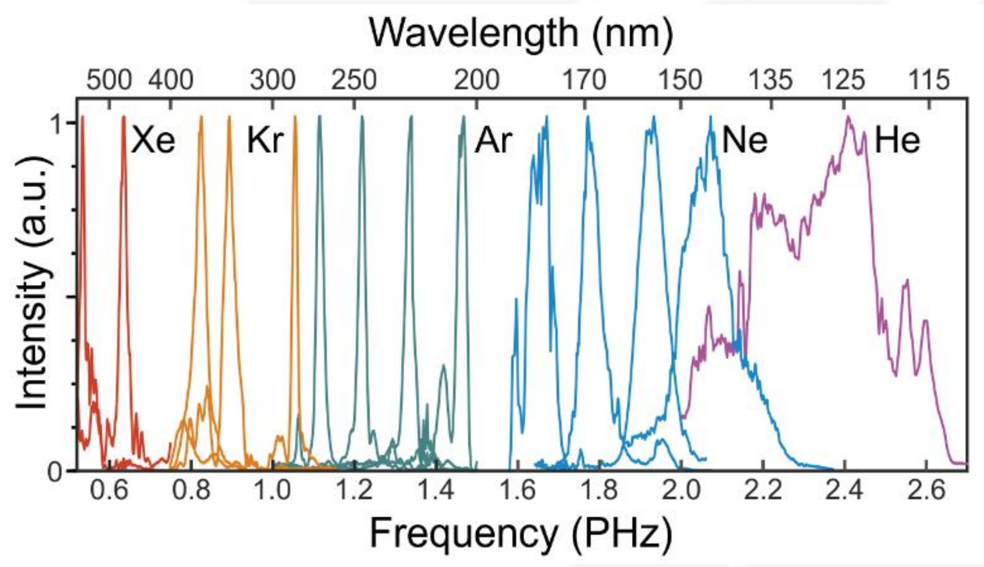


Tuneable UV Generation Light Source: Hollow Core-Photonic Crystal Fibre (HC-PCF)

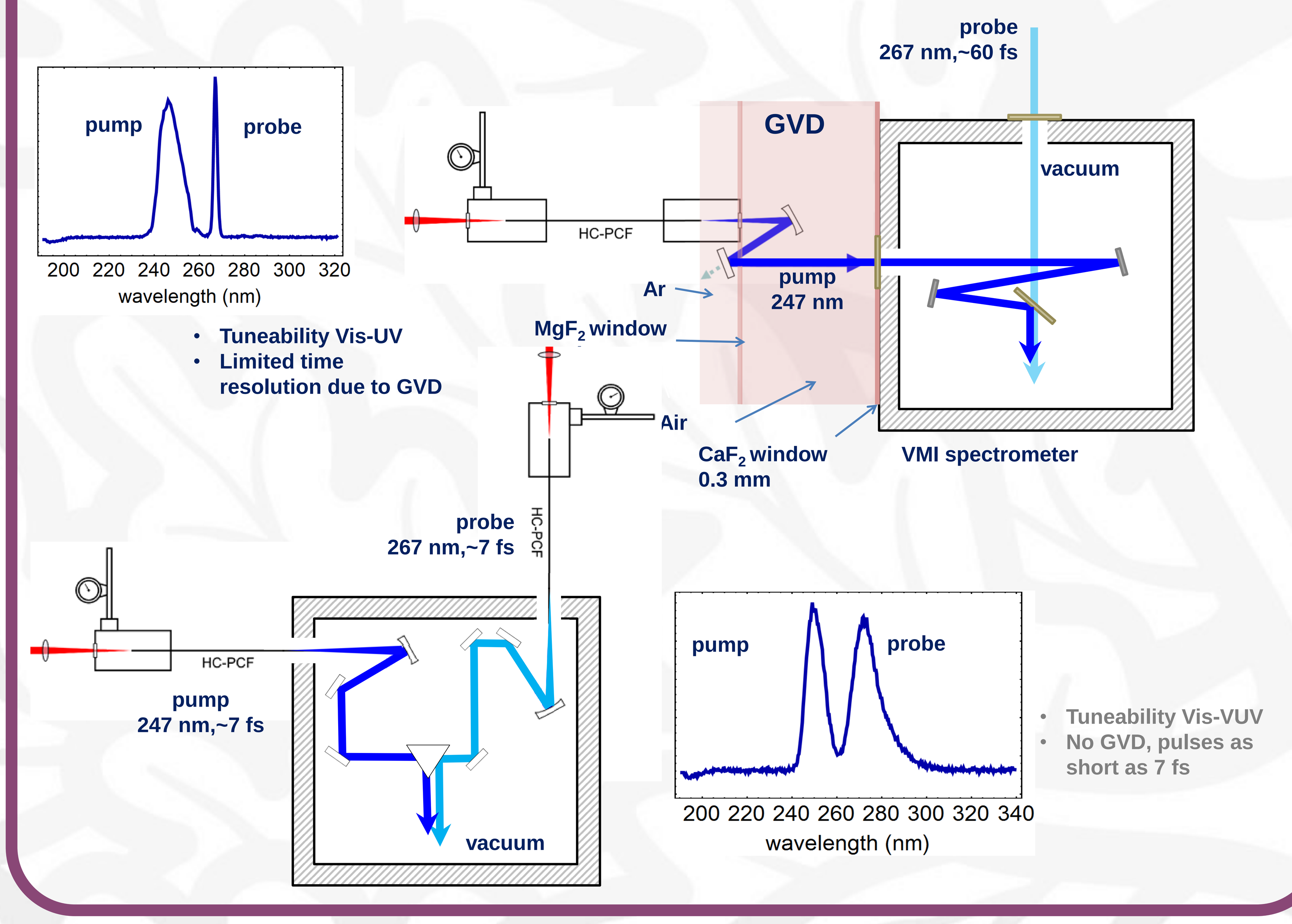
- Broadly tuneable over UV/VUV region 350-120 nm
- Vastly improved efficiency (2-3 orders of magnitude)
- Significantly improved time resolution (<10 fs, with two fibres)







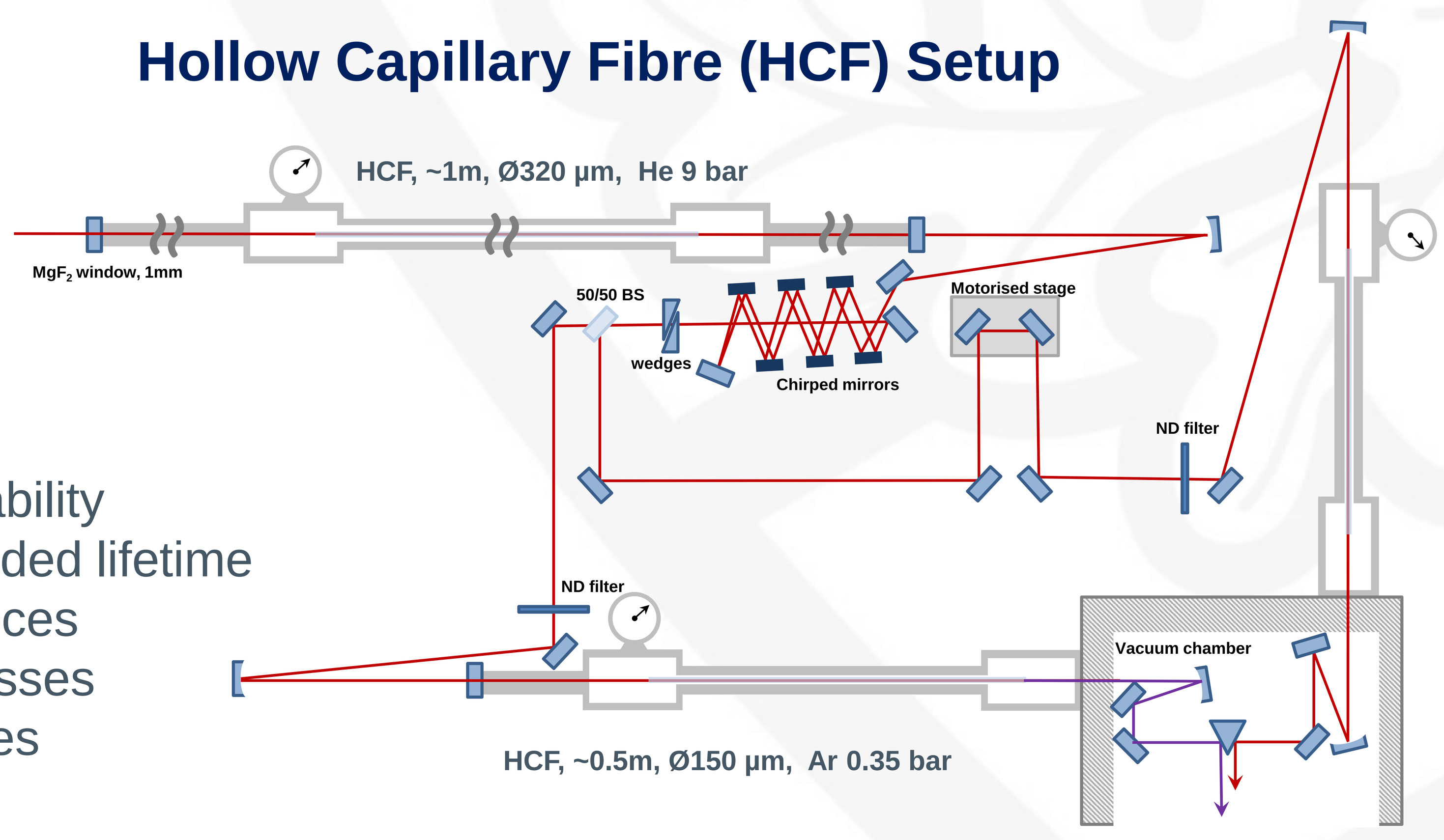
Exploiting the time resolution
Incorporate fibre in both pump and probe beams

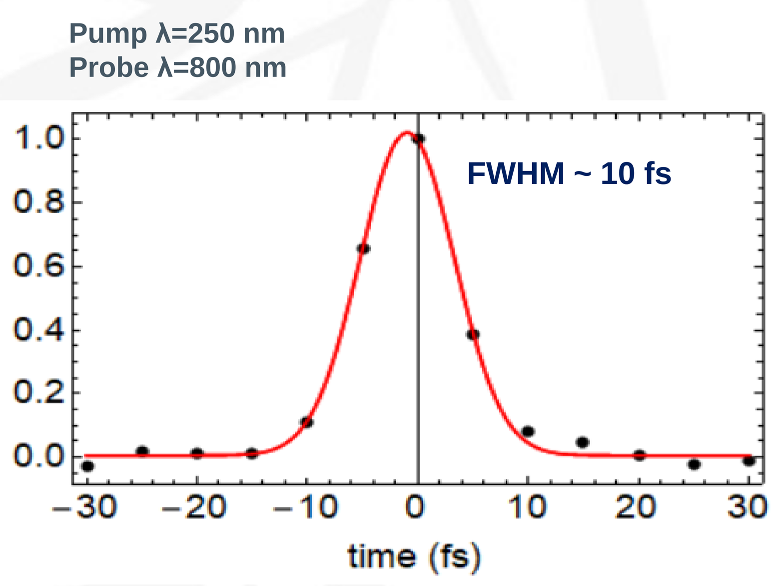


Hollow Capillary Fibre (HCF) Setup

Comparison

HC-PCF Ø 25 µm	HCF Ø 150 µm
Low input/output power	High energy sustainability
Limited lifetime	Long durability-extended lifetime
Cladding resonances	No cladding resonances
	High transmission losses
	Ultrashort input pulses





Butadiene cross corelation

Conclusion & Future Work

- Ar filled HC-PCF→ New source in ultrafast spectroscopy
- Tuneability from visible to VUV
- Care should be taken to manage the Group Velocity Dispersion of generated pulse
- Generate light in the VUV range

Acknowledgments

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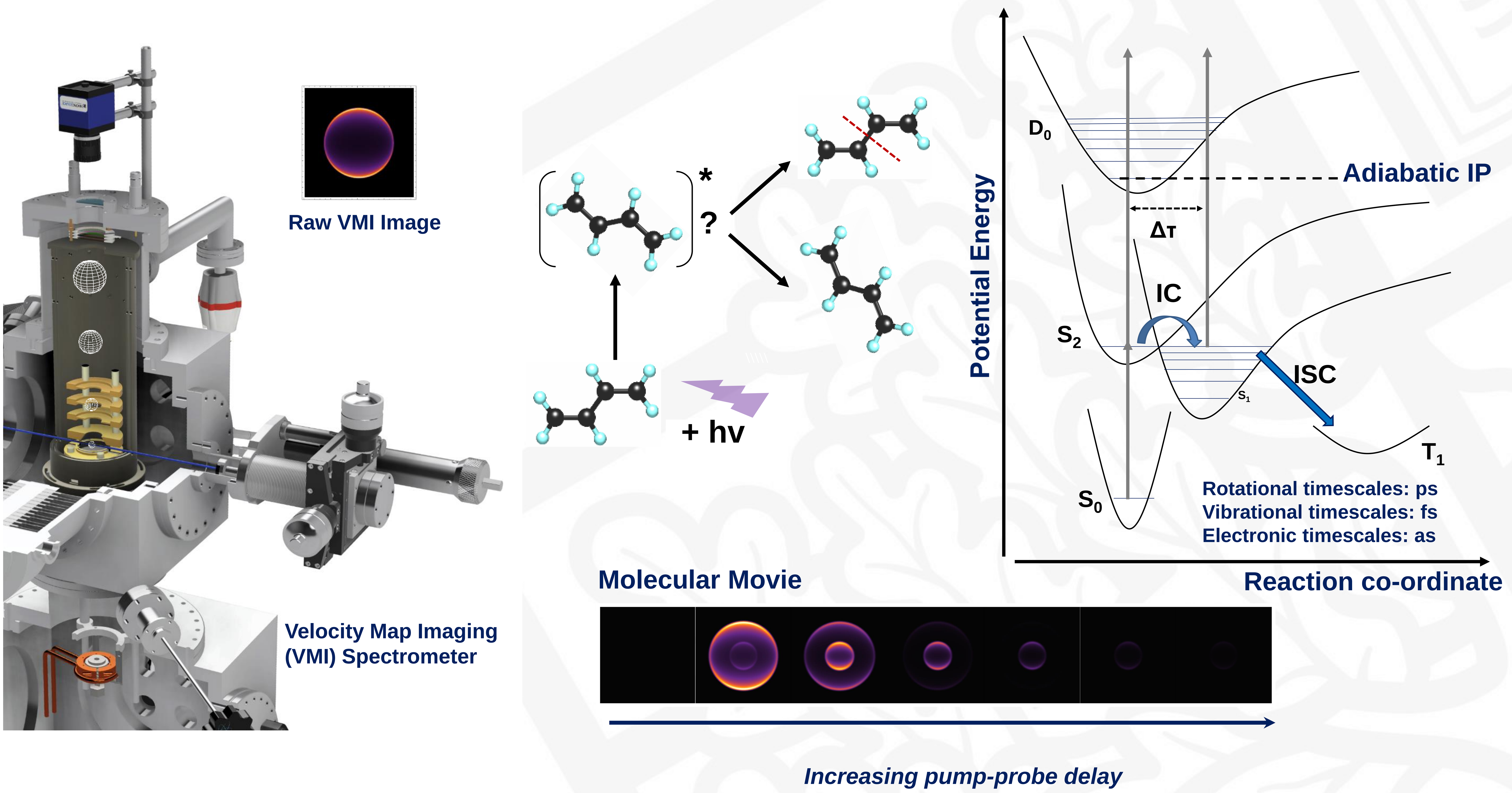
References

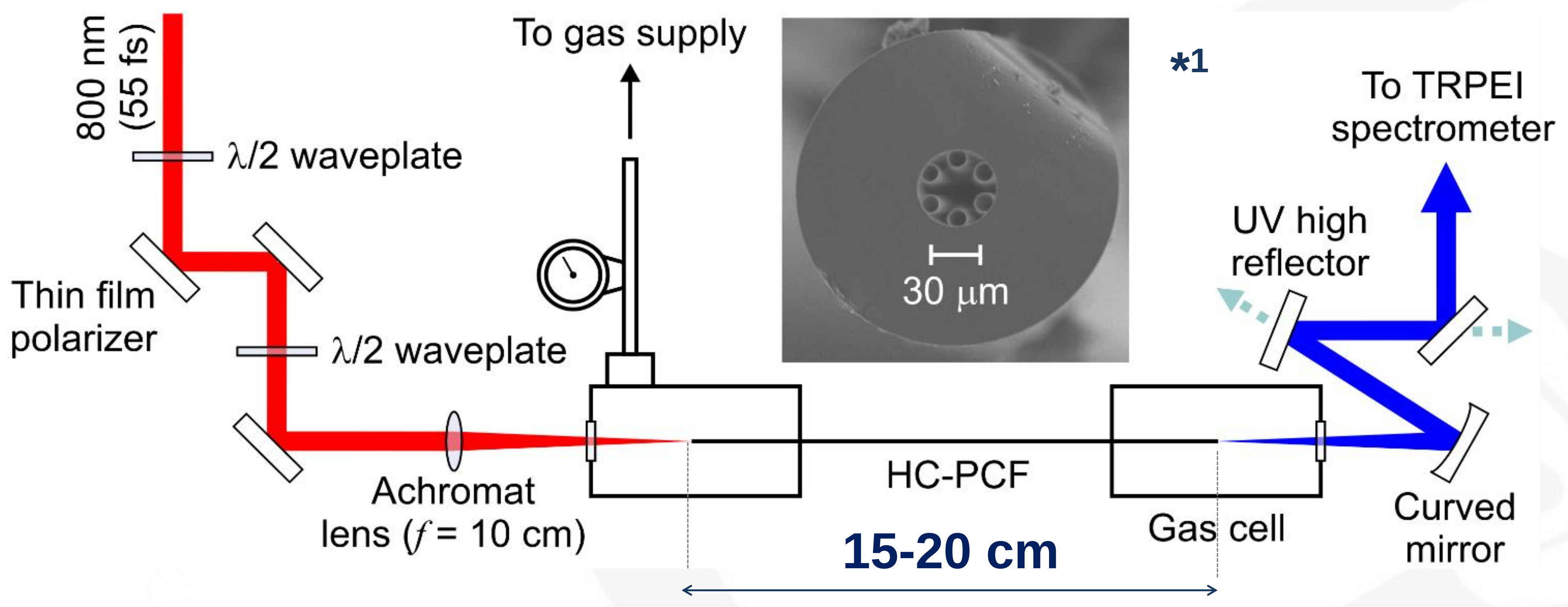
1) Kotsina *et al.*, J. Phys. Chem. Lett., **10**(4), 715 (2019)
2) Markos *et al.*, Rev. Mod. Phys., **89**, 045003 (2017)

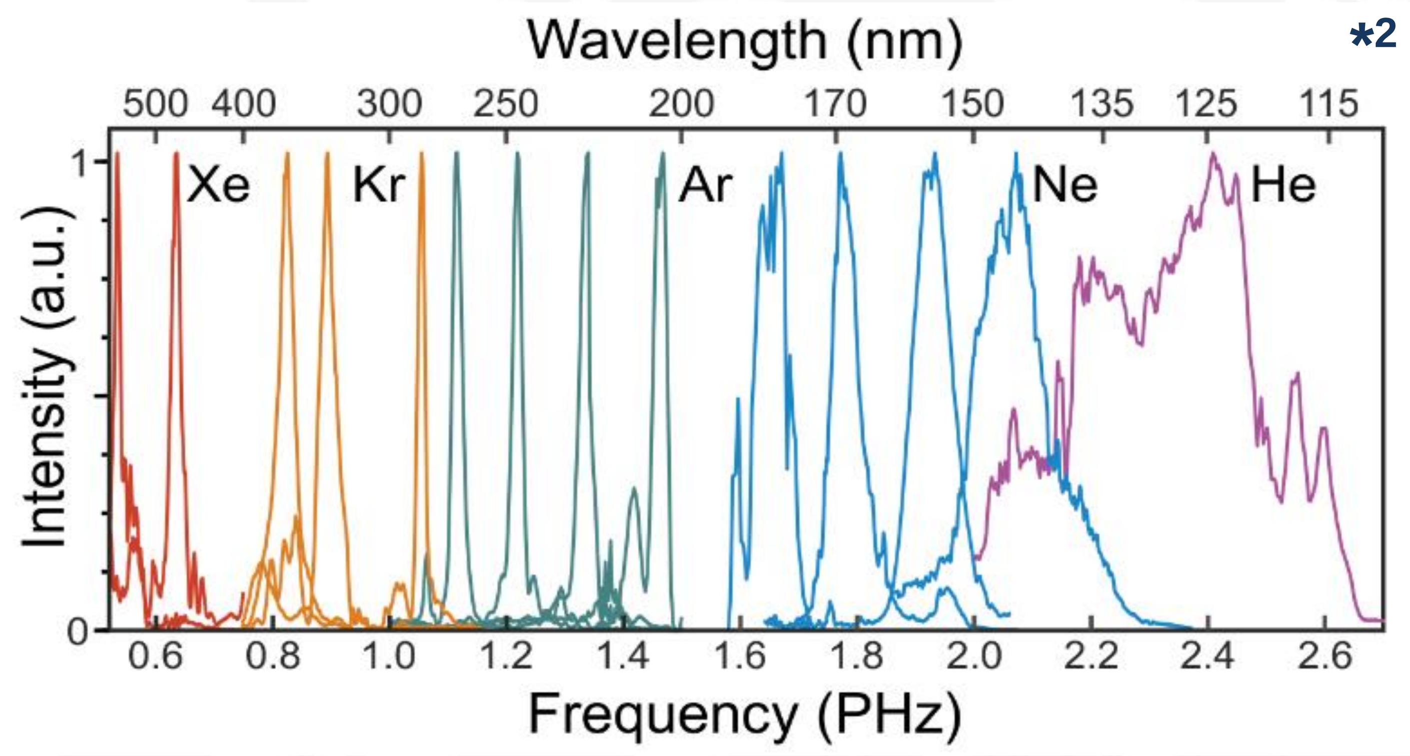
EPSRC Engineering and Physical Sciences Research Council

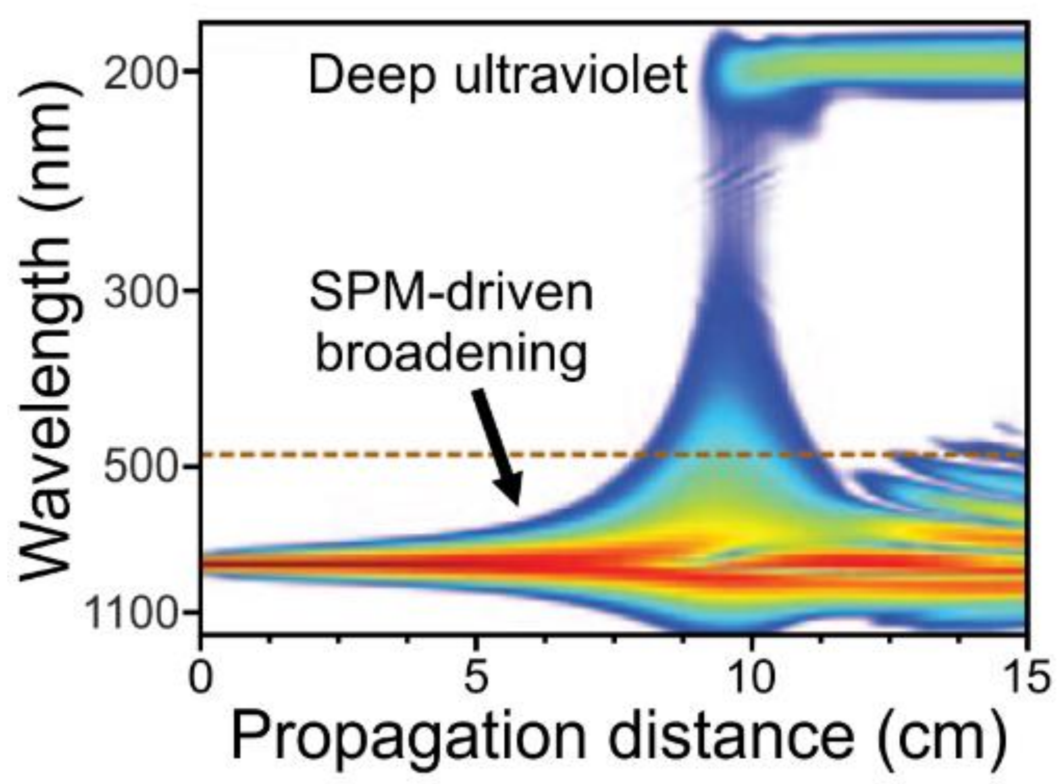
Time-Resolved Photoelectron Imaging



Tuneable UV Generation Light Source
Hollow Core-Photonic Crystal Fibre (HC-PCF)







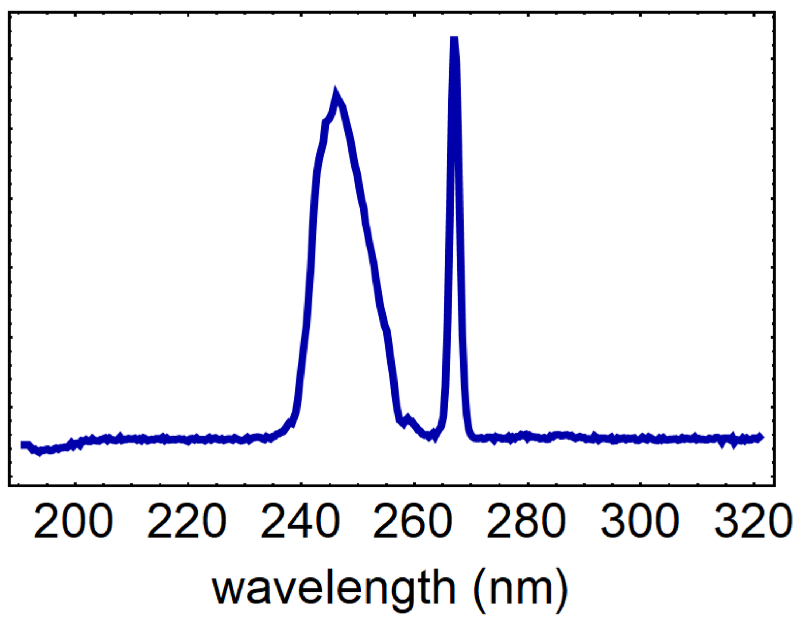
Broadly tuneable over UV/VUV region 350-120 nm

Vastly improved efficiency (2-3 orders of magnitude)

Ultrashort pulses (<10 fs)

*[1] Kotsina *et al.,* J. Phys. Chem. Lett., **10**(4), 715 (2019)
*[2] Markos *et al.*, Rev. Mod. Phys., **89**, 045003 (2017)

TRPEI and HC-PCF Experiment



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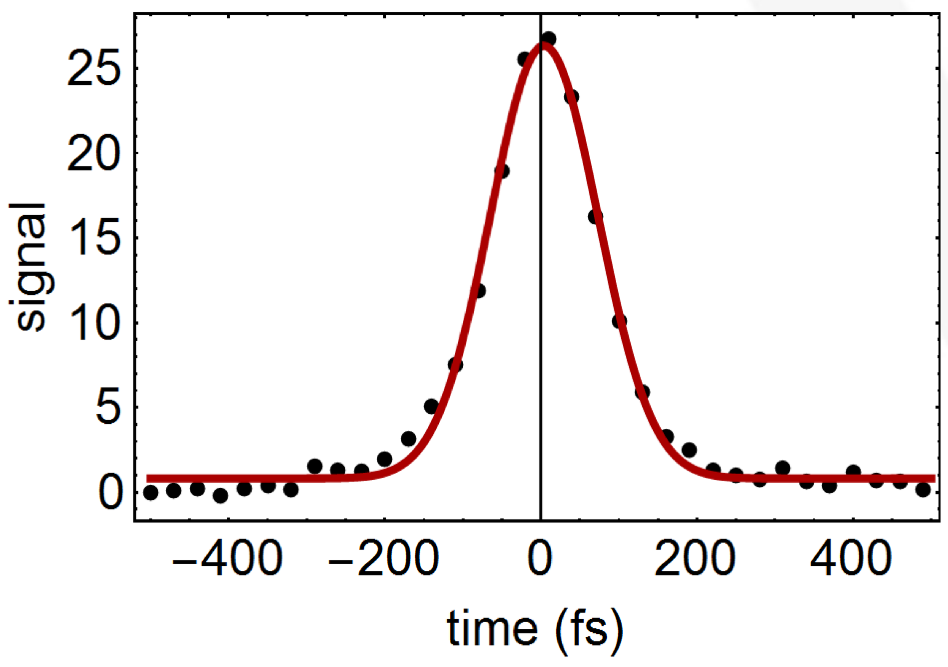

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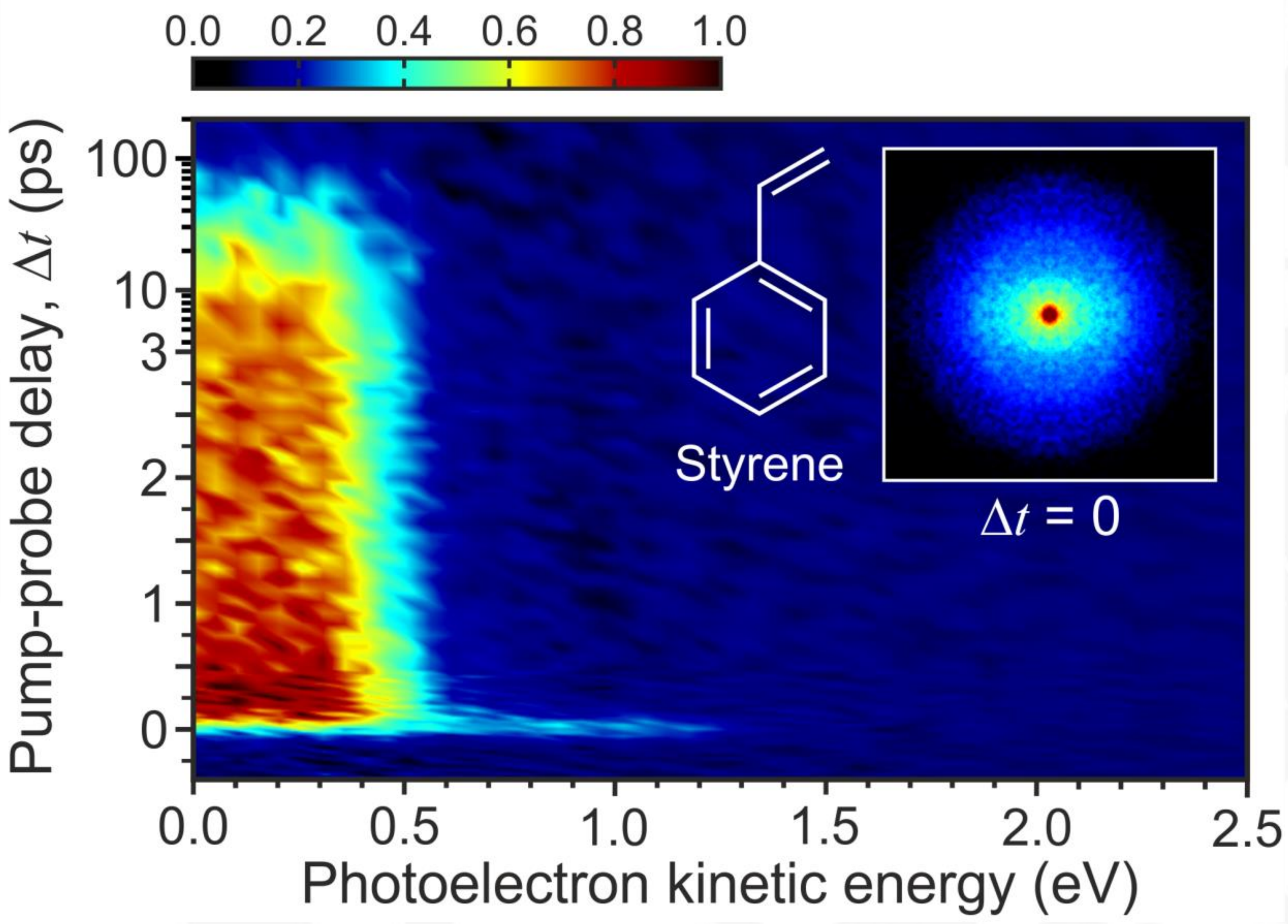
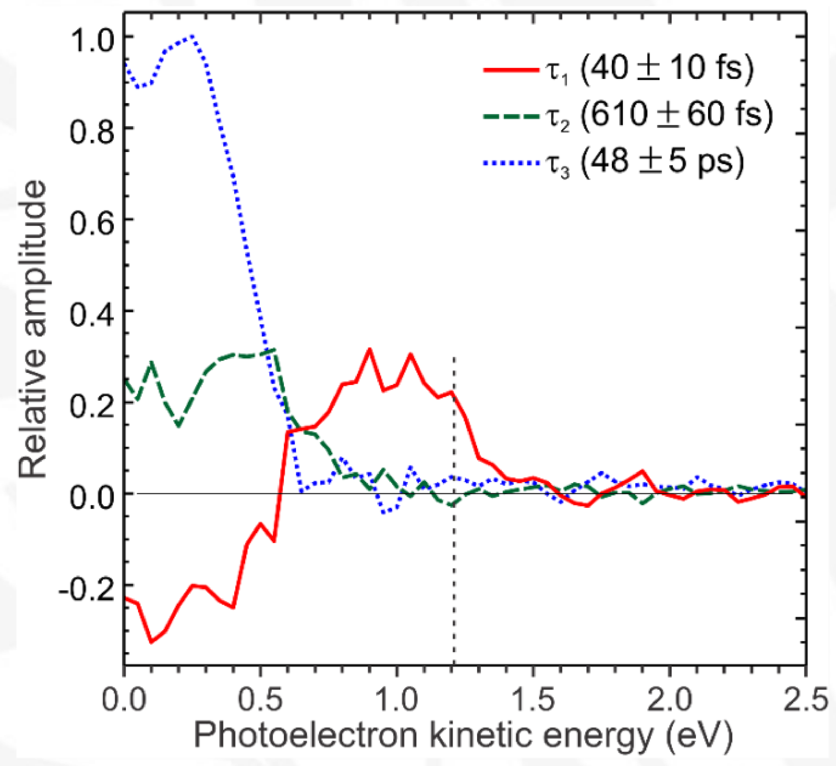
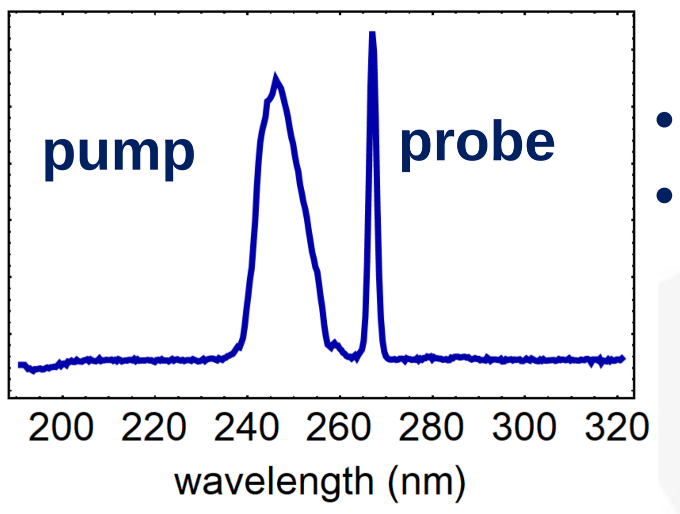


Styrene Photoelectron Spectrum

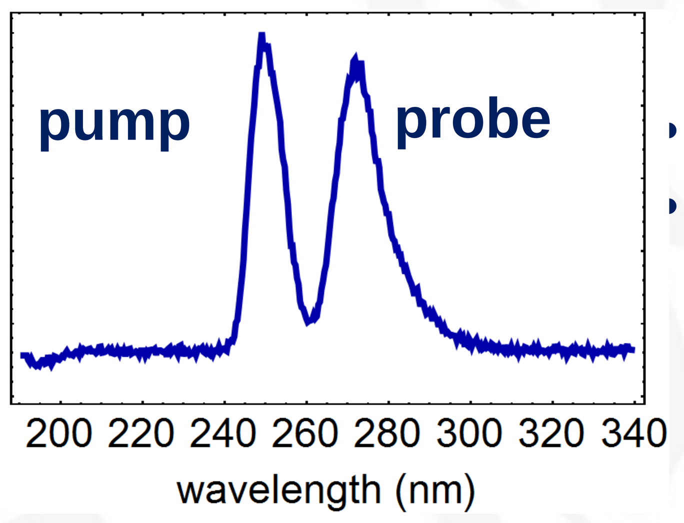


Decay Associated Spectra (DAS) Plots

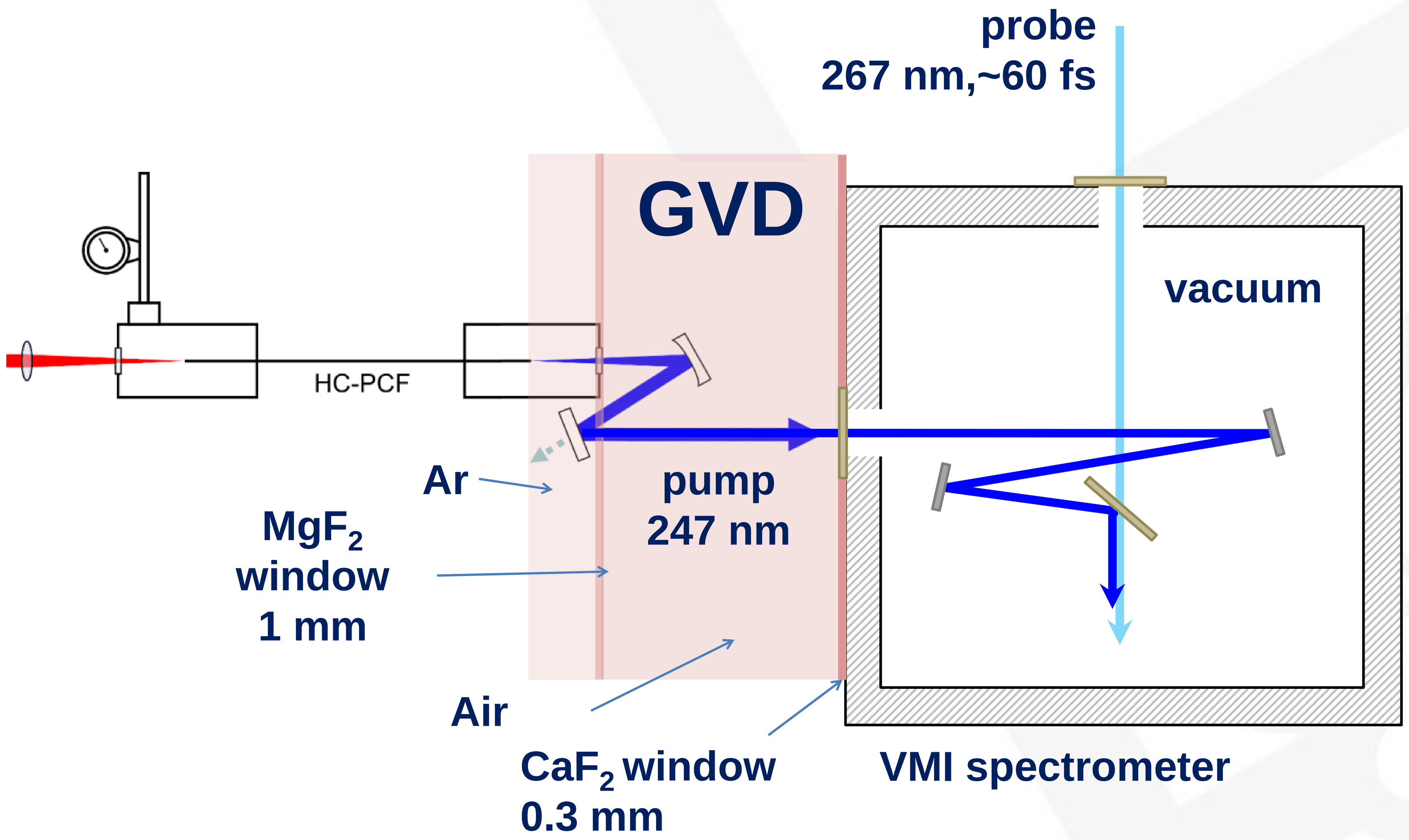
Exploiting the time-resolution
Incorporate fibre in both pump and probe beams

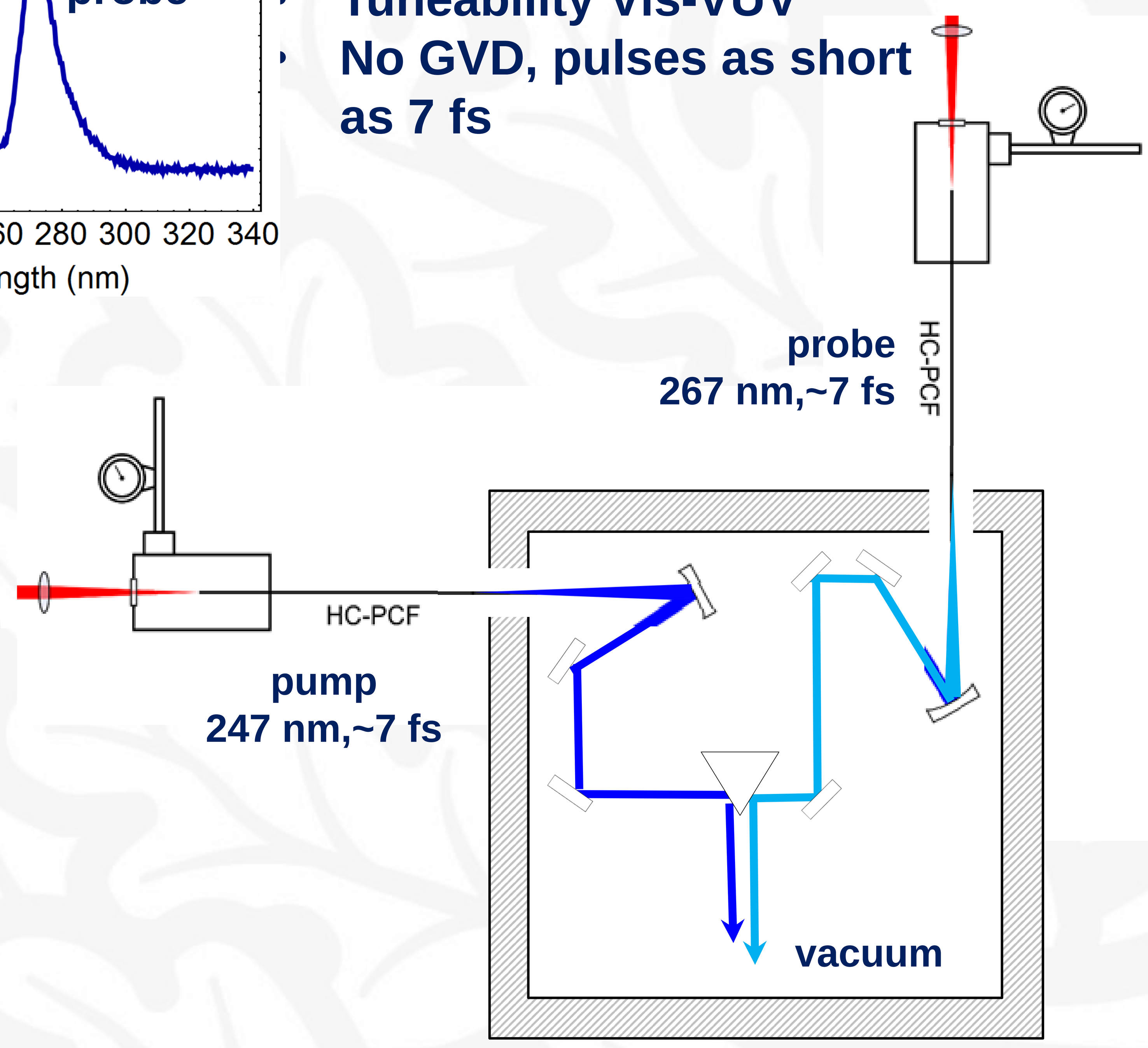


- Tuneability Vis-UV
- Limited time resolution due to GVD



- Tuneability Vis-VUV
- No GVD, pulses as short as 7 fs





TRPEI and HCF Experiment