Aspects and Applications of THz FELs

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Outline

Aspects of THz - FELs:

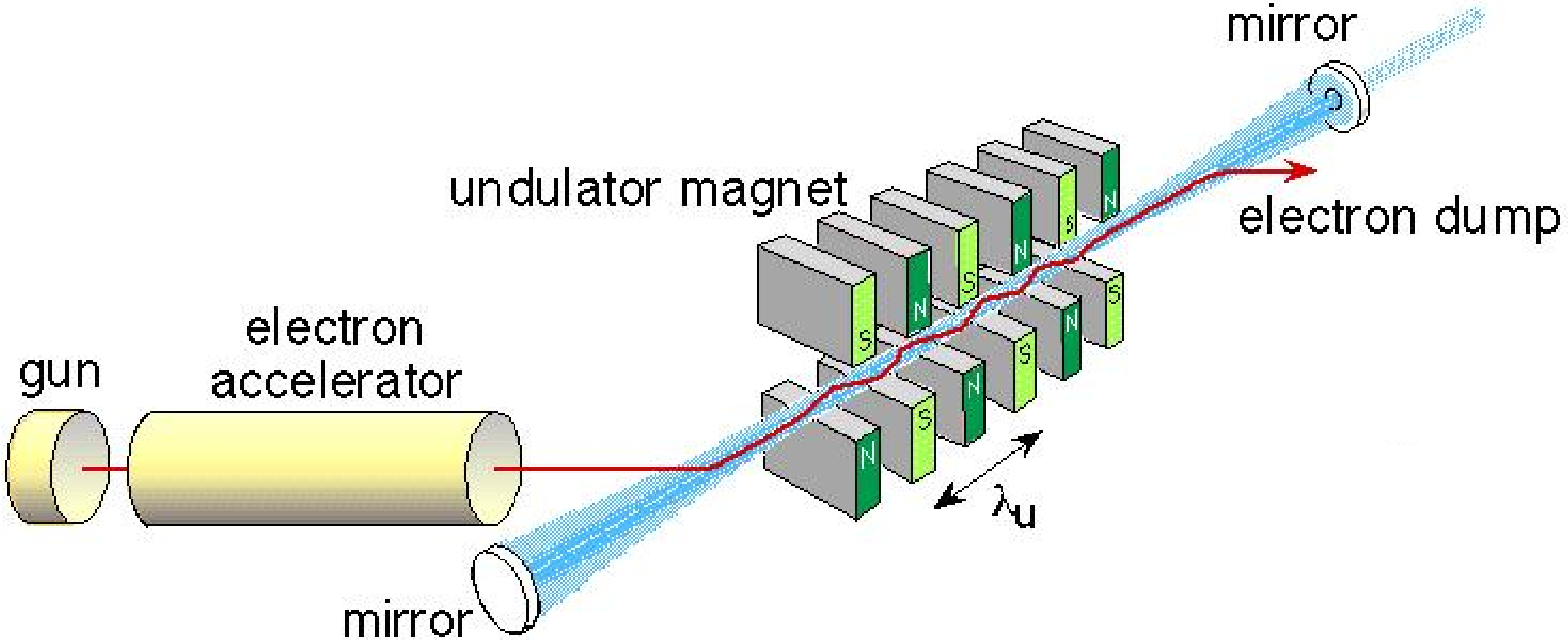
- Radiation bandwidth
- Resonator issues
- Slippage effects

Applications:

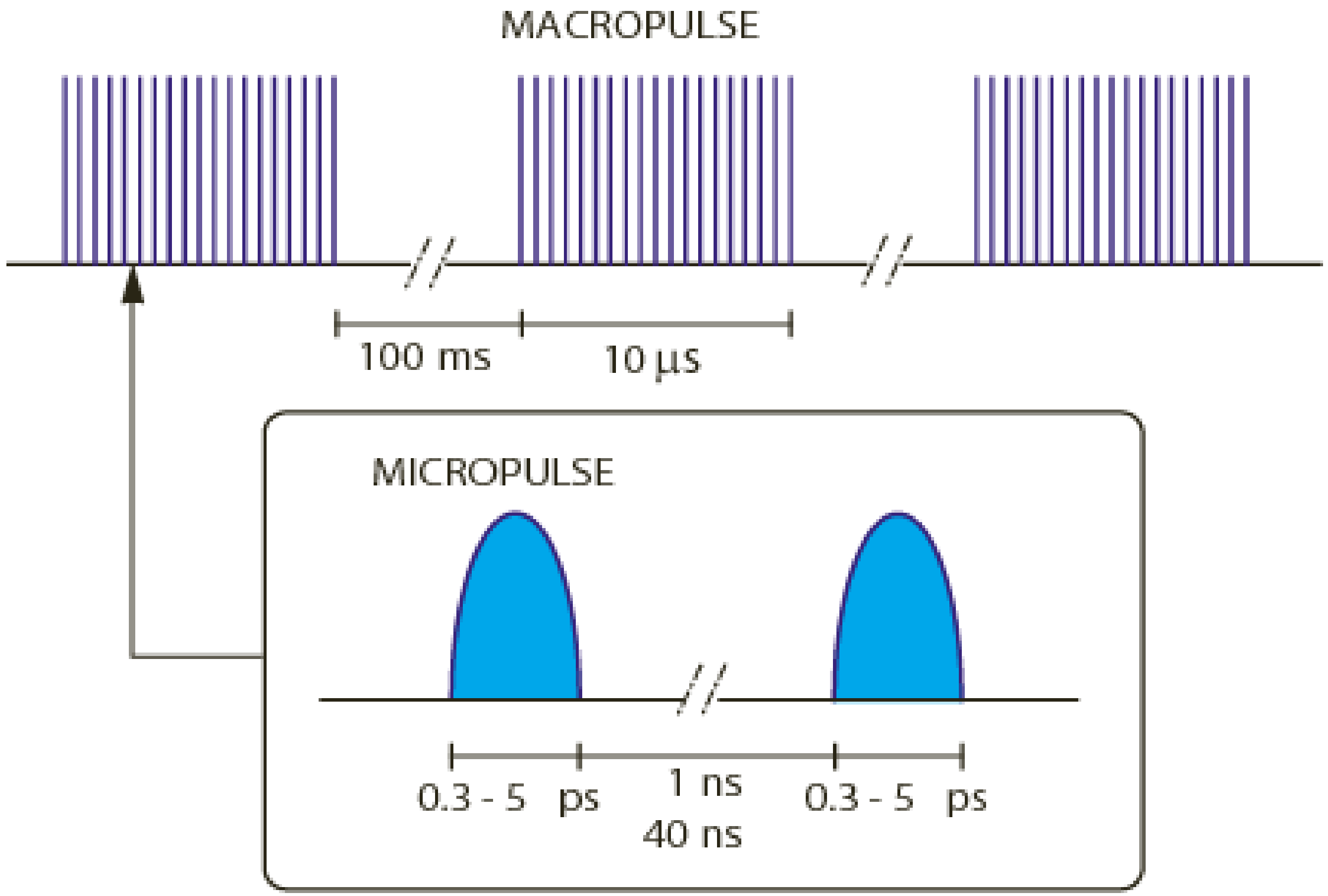
- nonlinear optics
- nano-spectroscopy
- 'action spectroscopy'

The FELIX laboratory

Generic layout

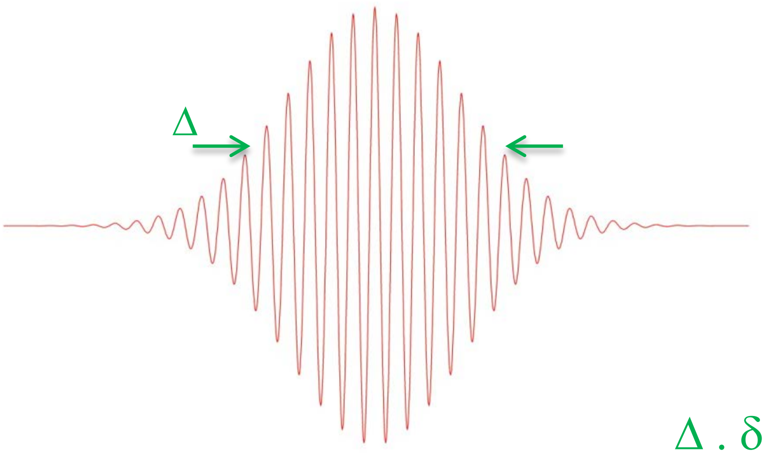


Radiation bandwidth: linac pulse structure

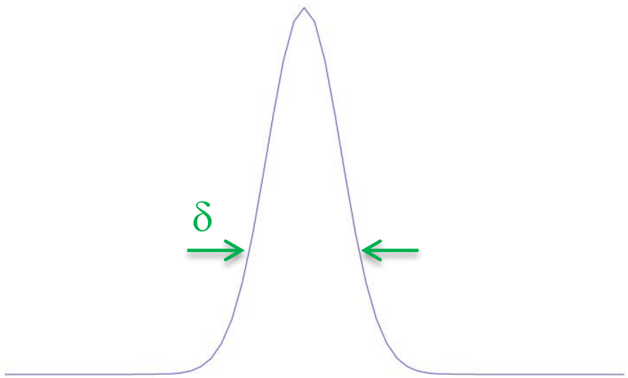


Radiation bandwidth: Fourier transform

pulse shape



spectrum

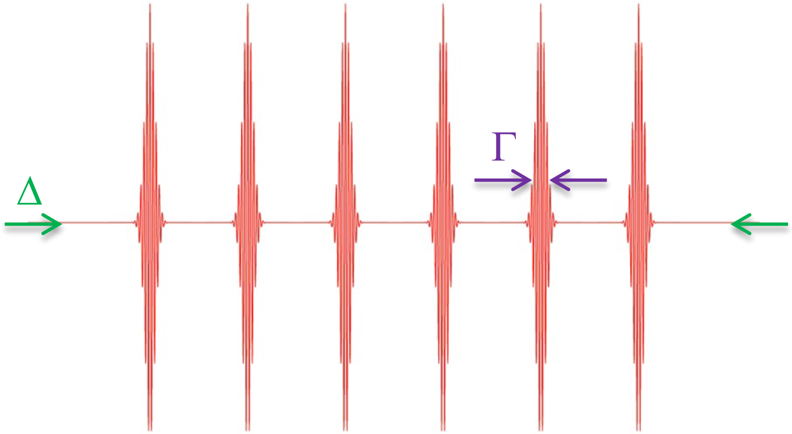


$\Delta \,.\, \delta \; \varepsilon$ constant

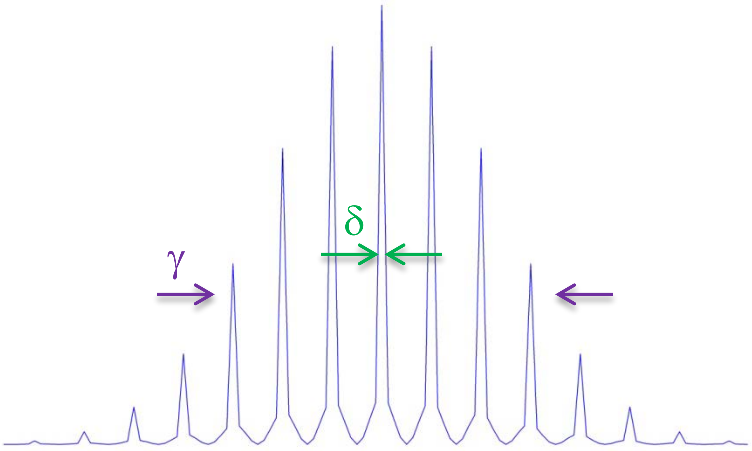
'transform limited' : no frequency / phase fluctuations of the carrier

Radiation bandwidth: Fourier transform

multiple pulses



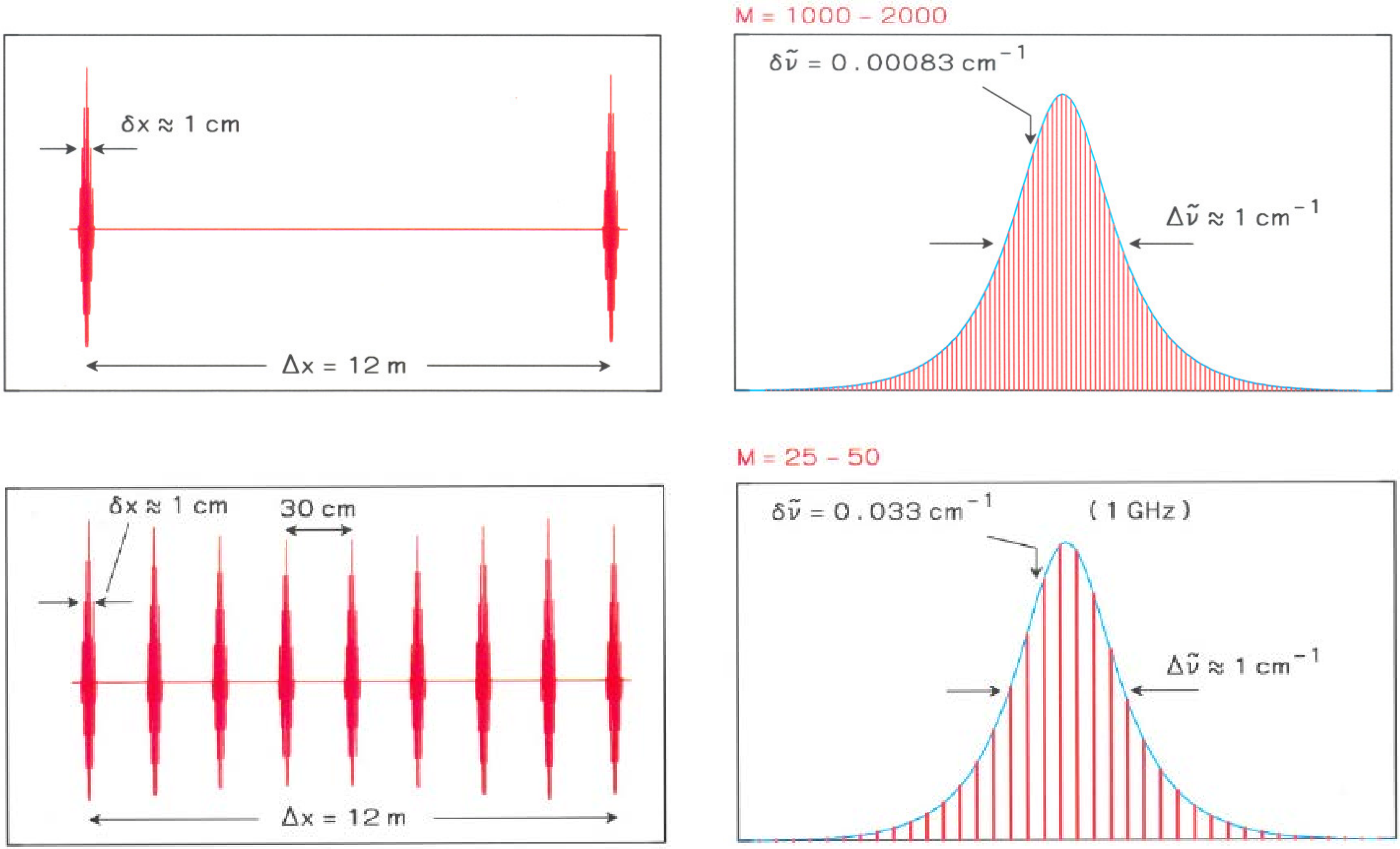
multiple spikes



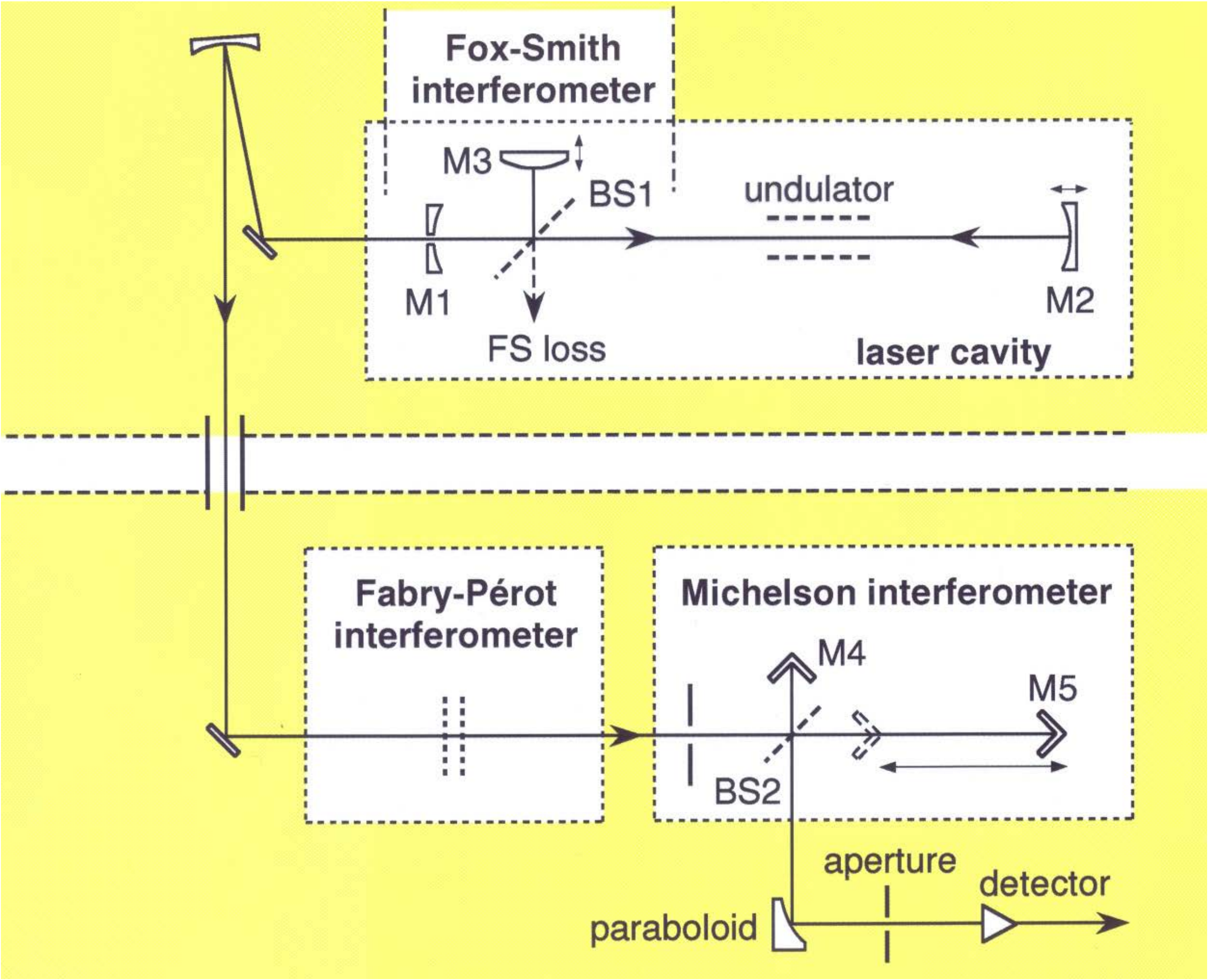
$$\Gamma \,.\, \gamma = O(1)$$

$$\Delta \,.\, \delta = O(1)$$

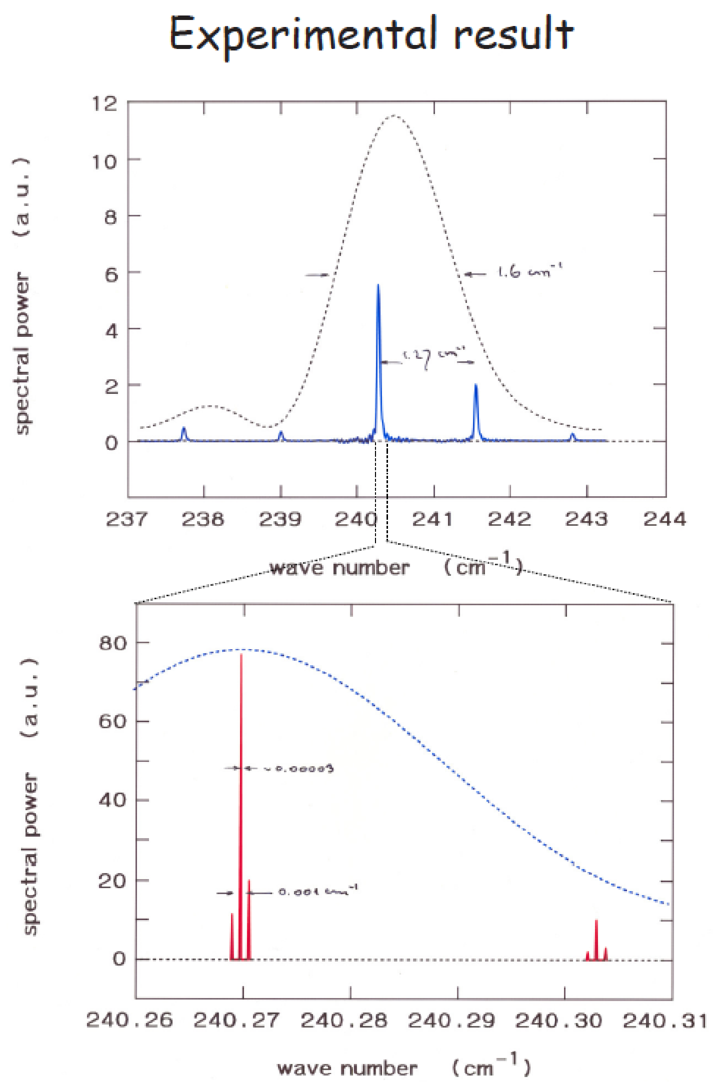
Radiation bandwidth: phase locking



Phase locking & single mode selection



Phase locking & single mode selection

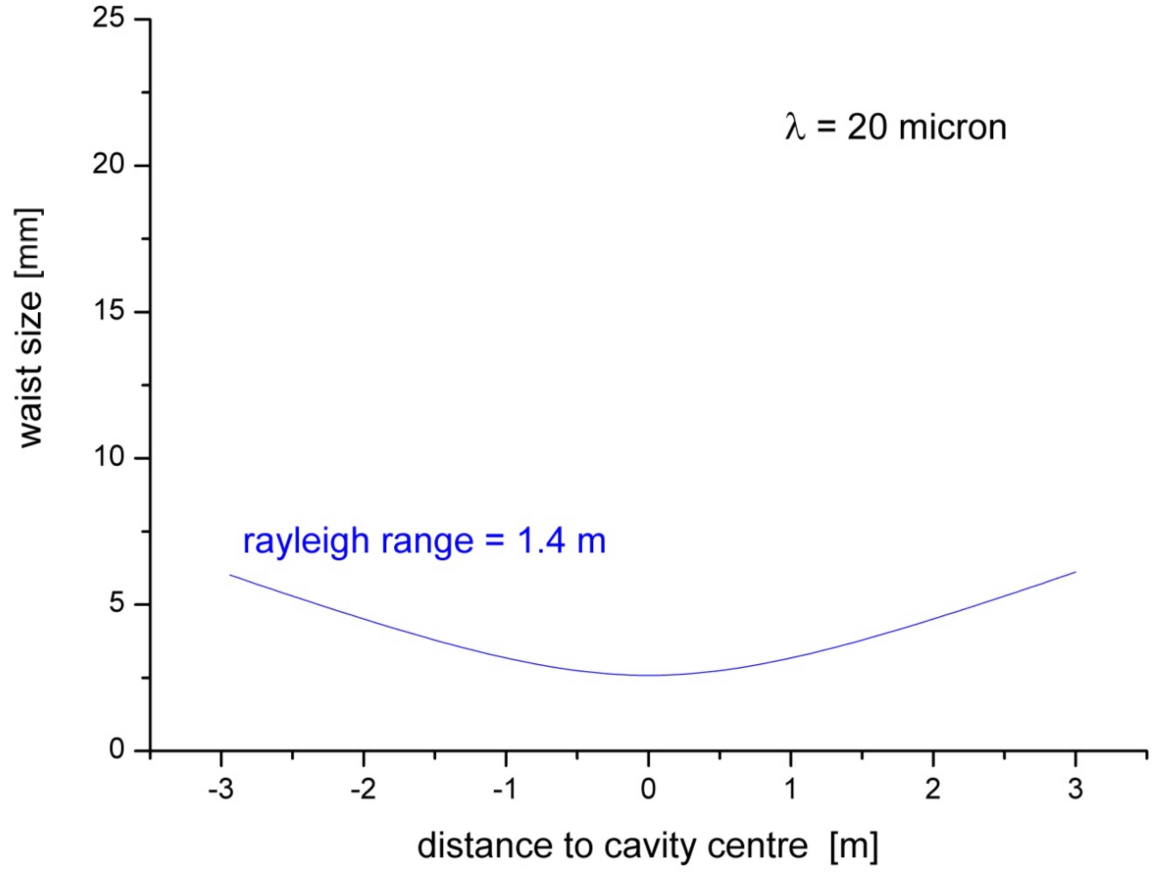


Resonator issues

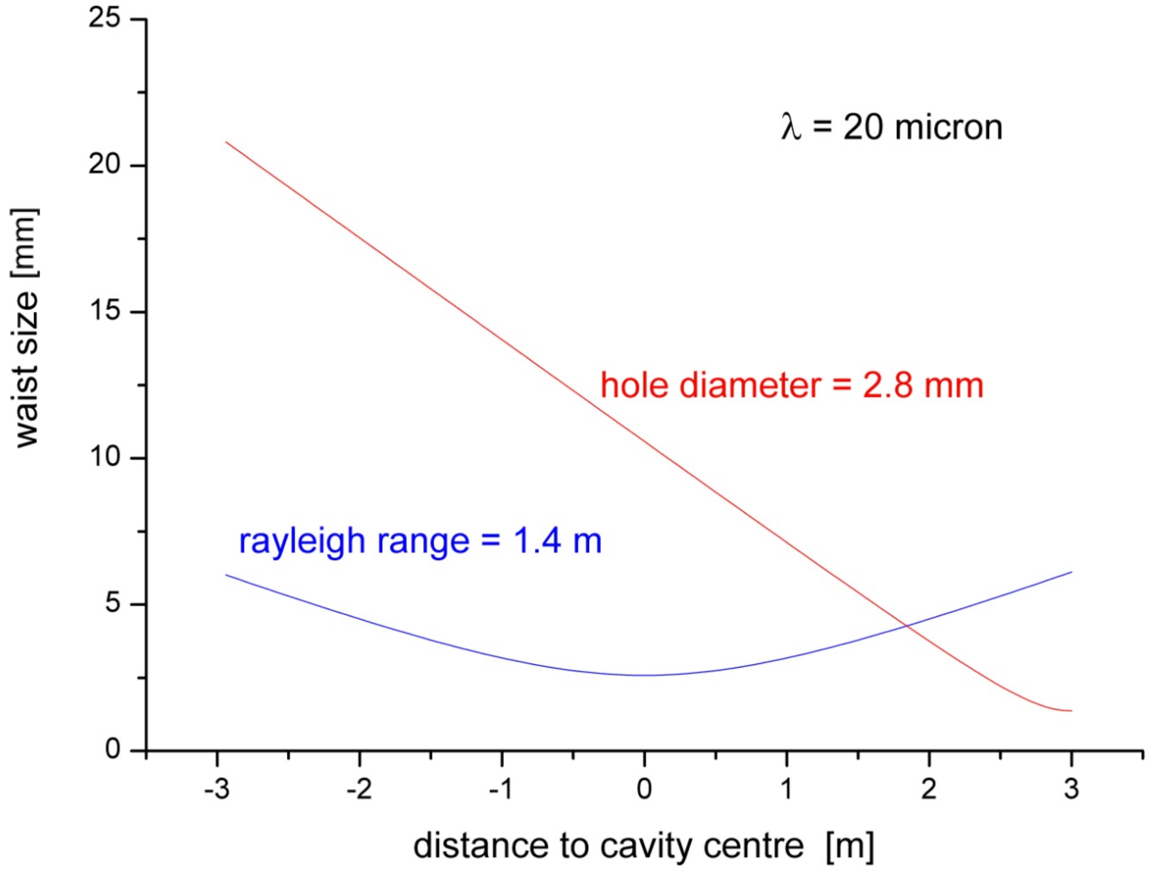
out-coupling schemes:

- 'semi-transparent' mirrors
- beam splitter
- hole coupling
- edge coupling

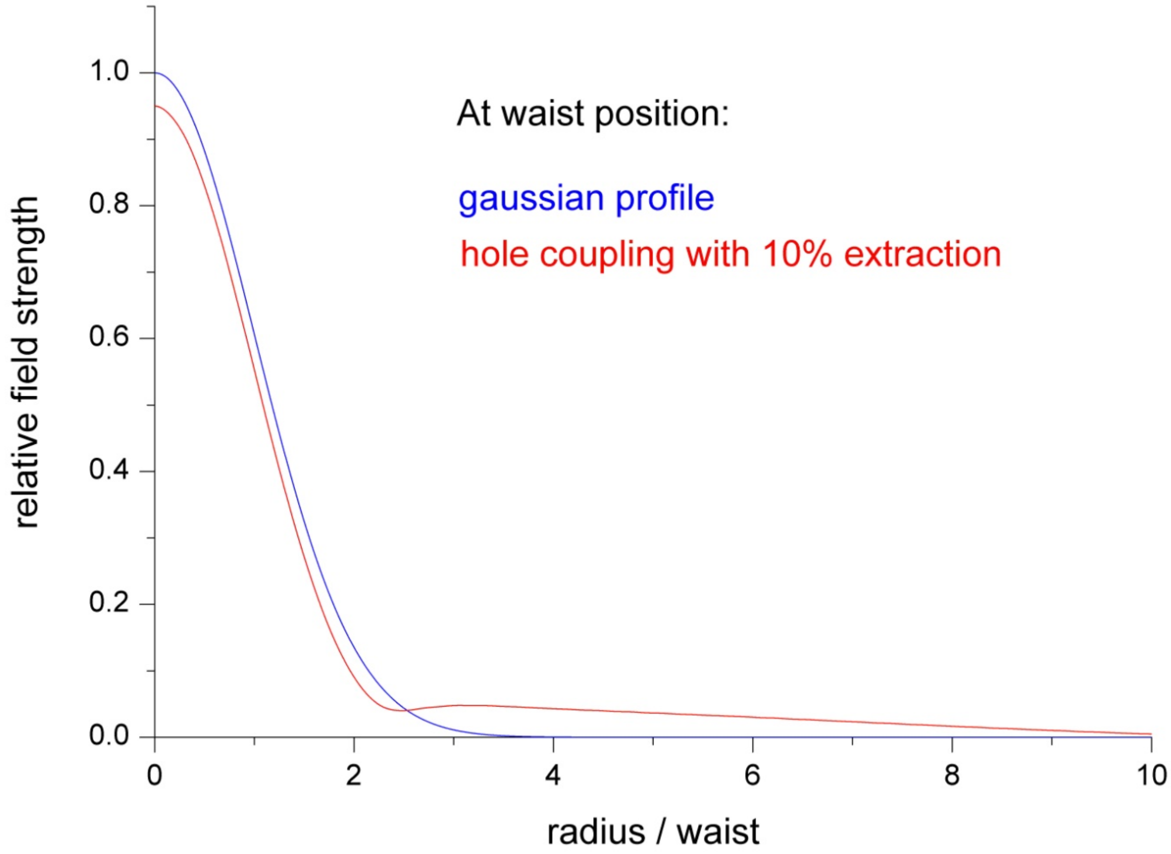
Resonator issues: on-axis hole coupling



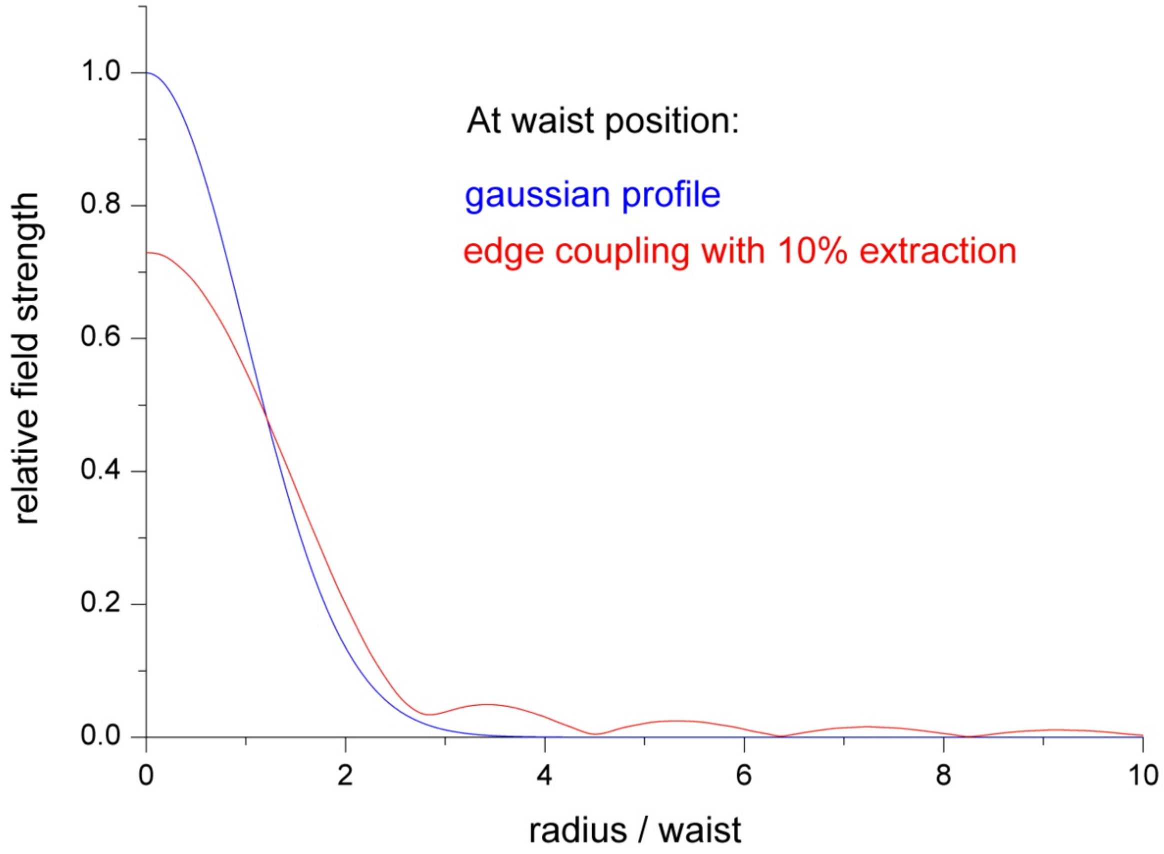
Resonator issues: on-axis hole coupling



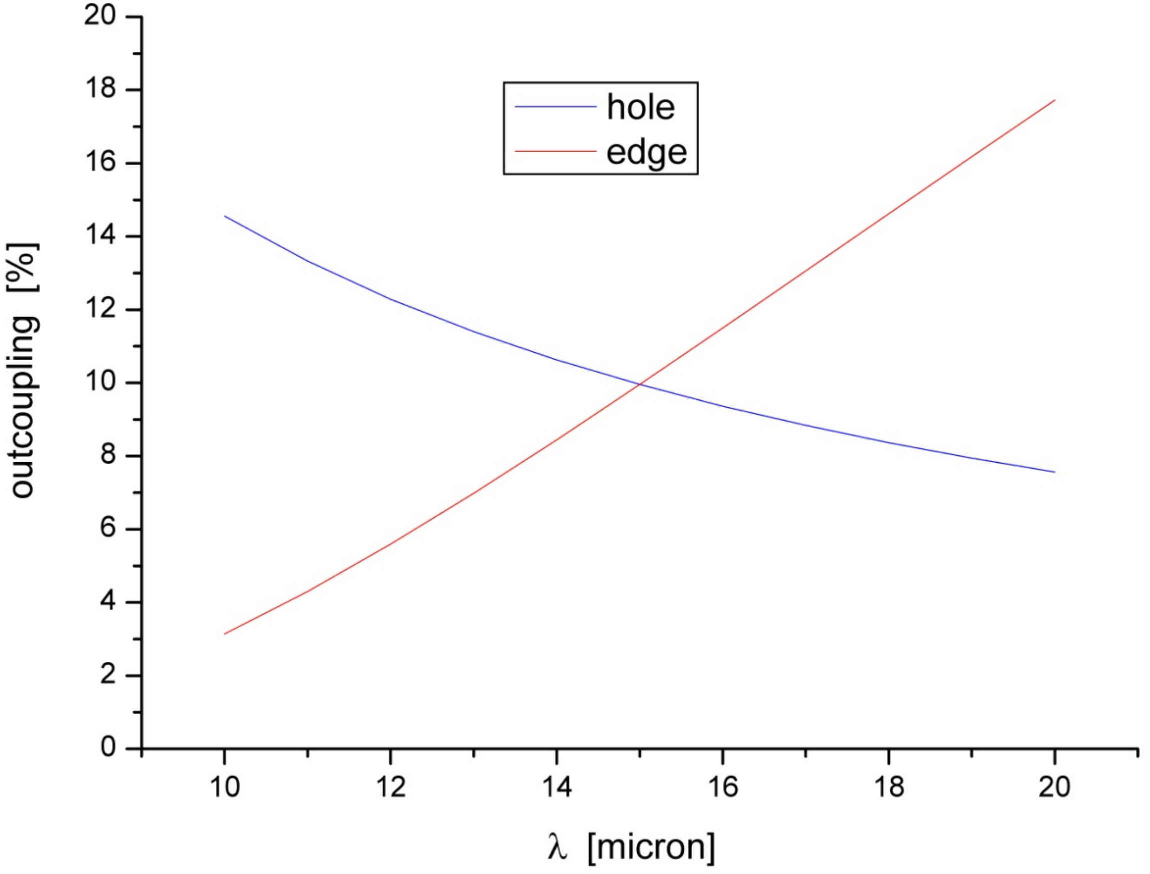
Resonator issues: on-axis hole coupling



Resonator issues: edge-coupling

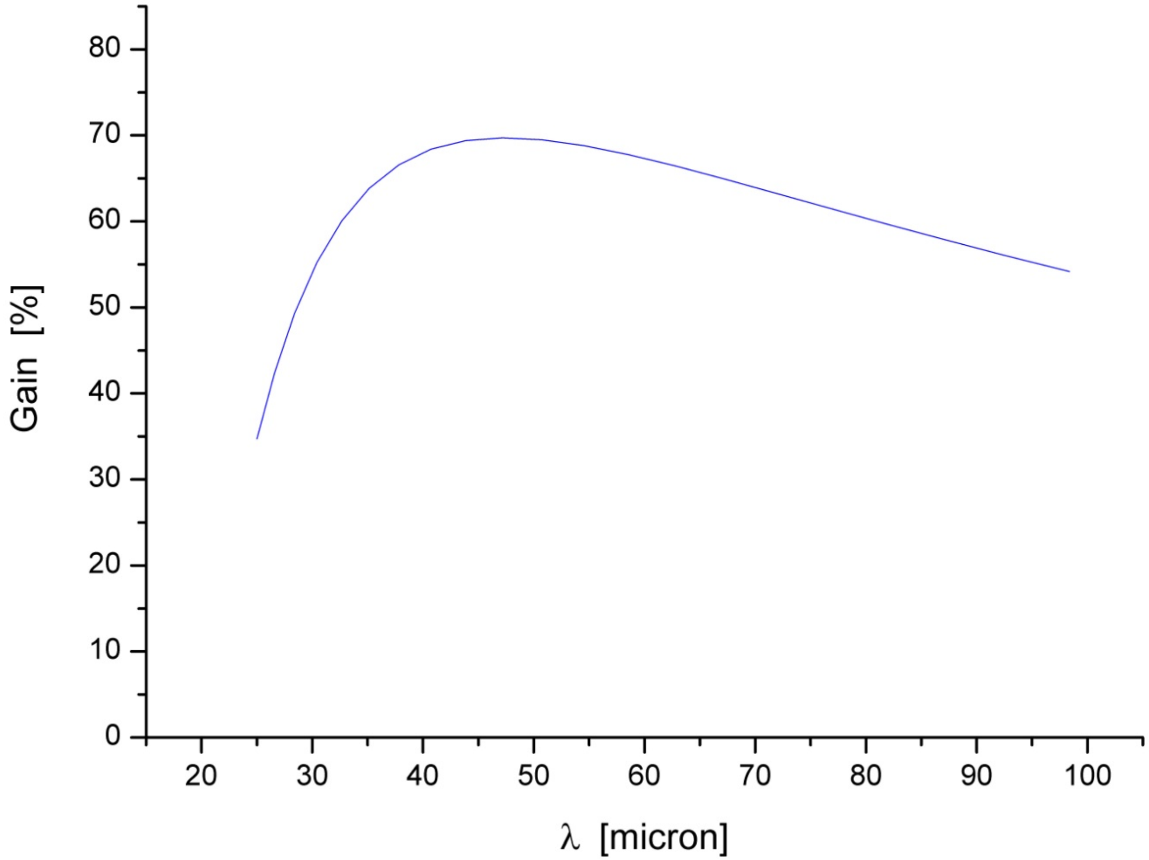


Hole vs edge coupling: λ - dependence

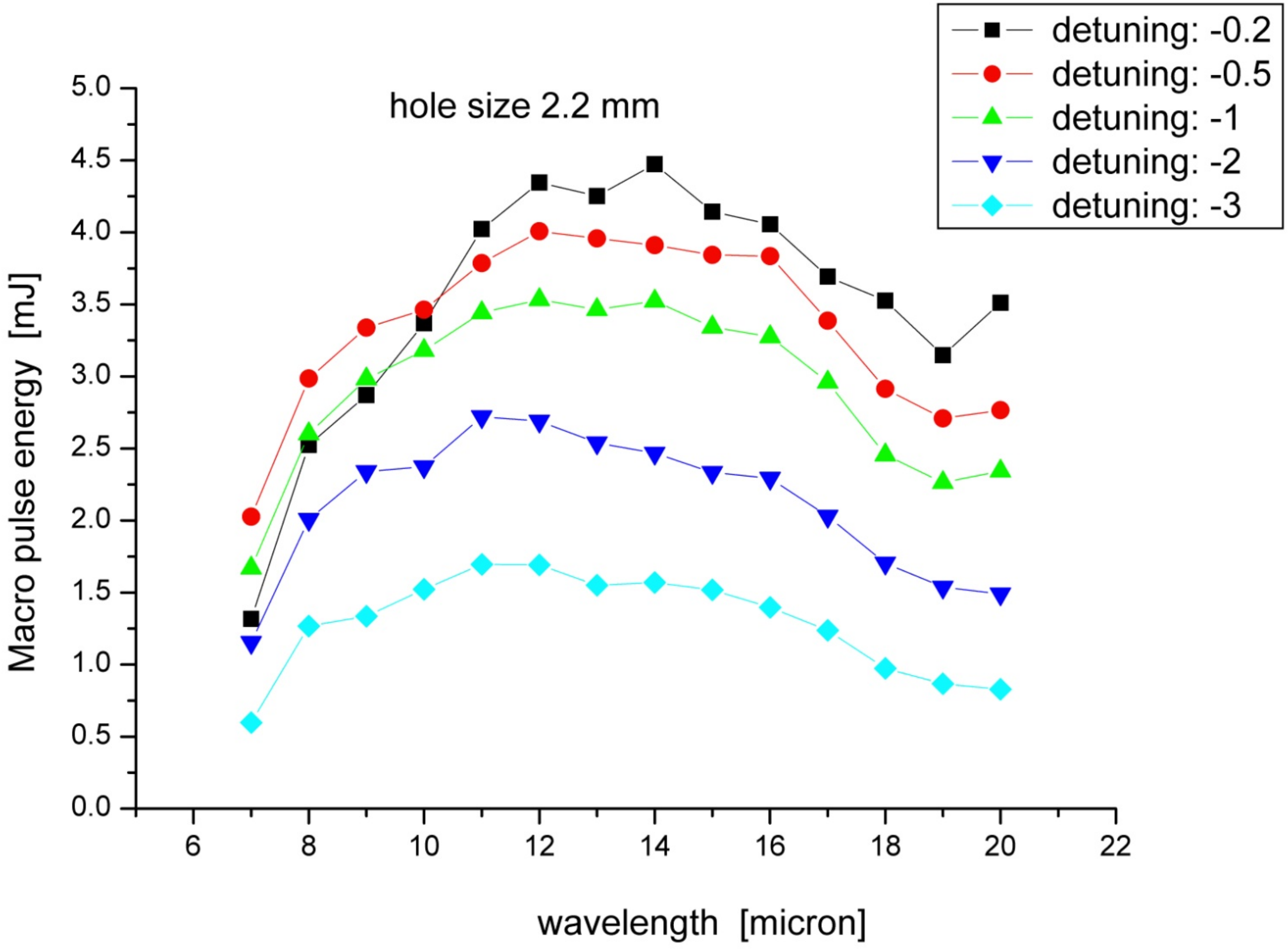


Hole vs edge coupling: λ - dependence

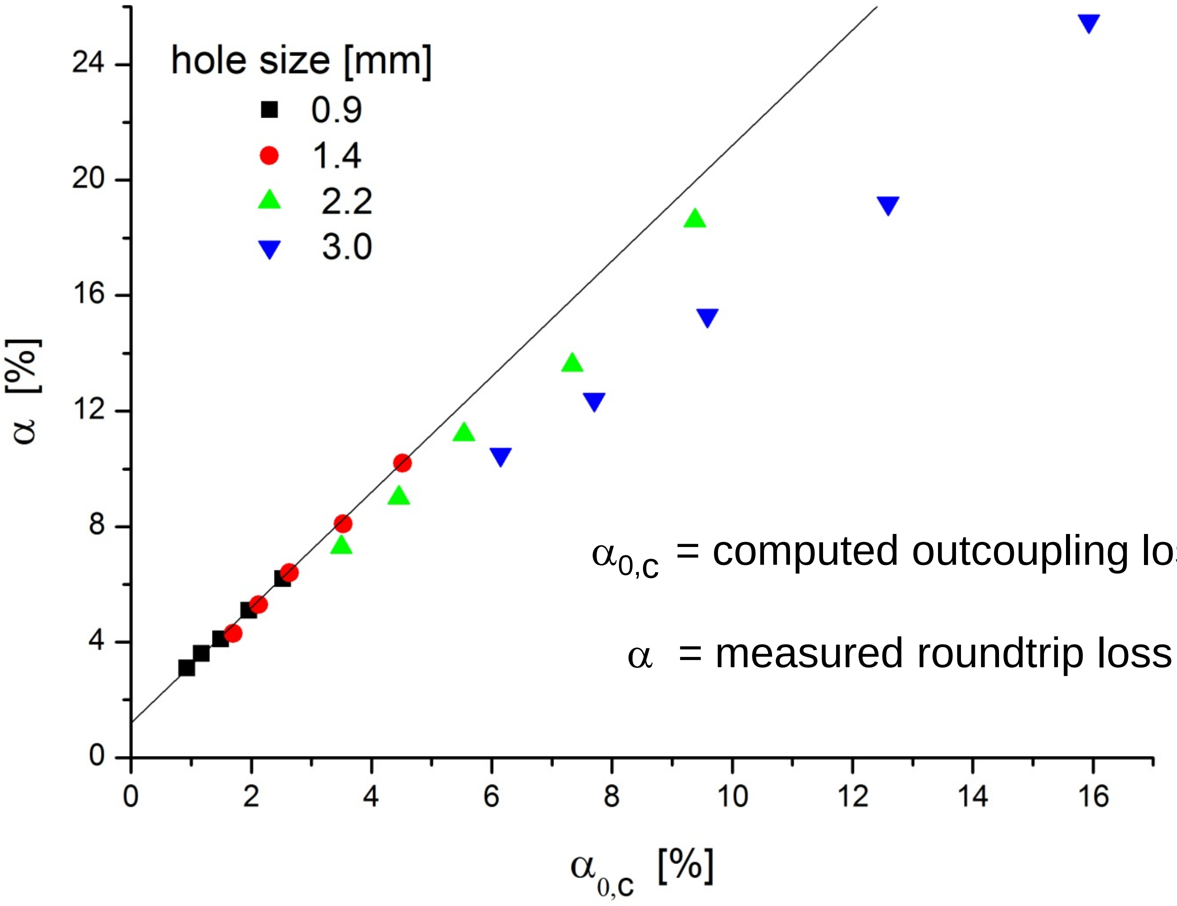
Typical gain curve



Hole coupling: tuning curves



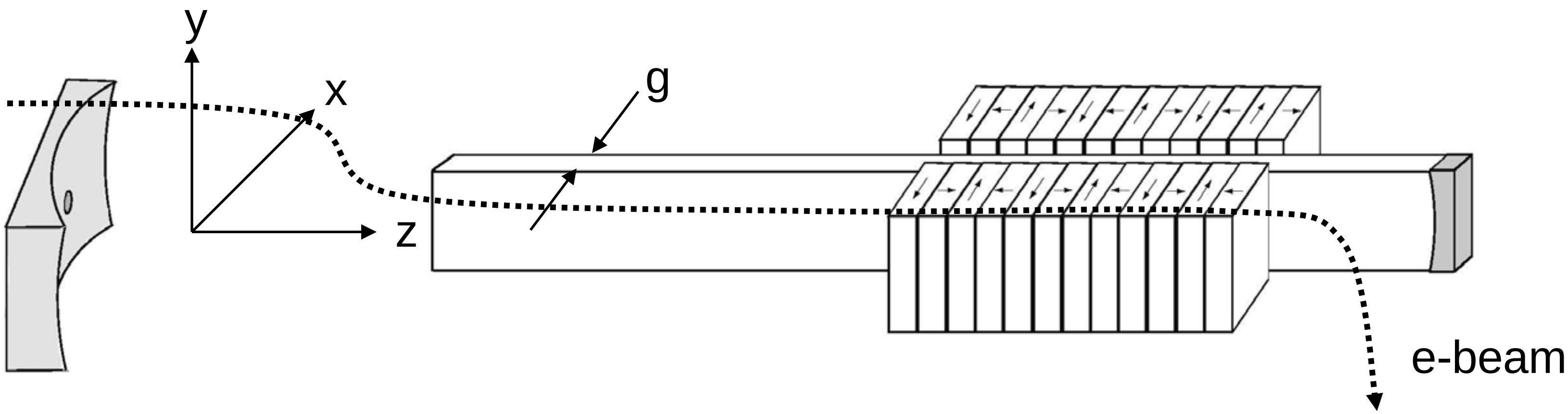
Hole coupling: measured losses



$\alpha_{0,c}$ = computed outcoupling loss

α = measured roundtrip loss

Partial-waveguide resonator



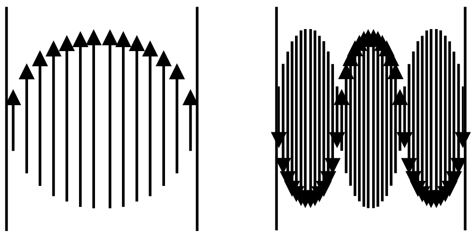
Eigenmodes are a combination of TE and Hermit-Gaussian modes:

$$\Psi_{m,n}(x,y,z) \sim H_m\left(\frac{\sqrt{2}y}{w(z)}\right)\sin\left(\frac{nx\pi}{g}\right)\exp\left(\frac{-y^2}{w^2(z)} + i\left(\frac{k_n^z y^2}{2R(z)} - \left(m+\frac{1}{2}\right)\tan^{-1}\frac{z}{z_r}\right)\right)$$

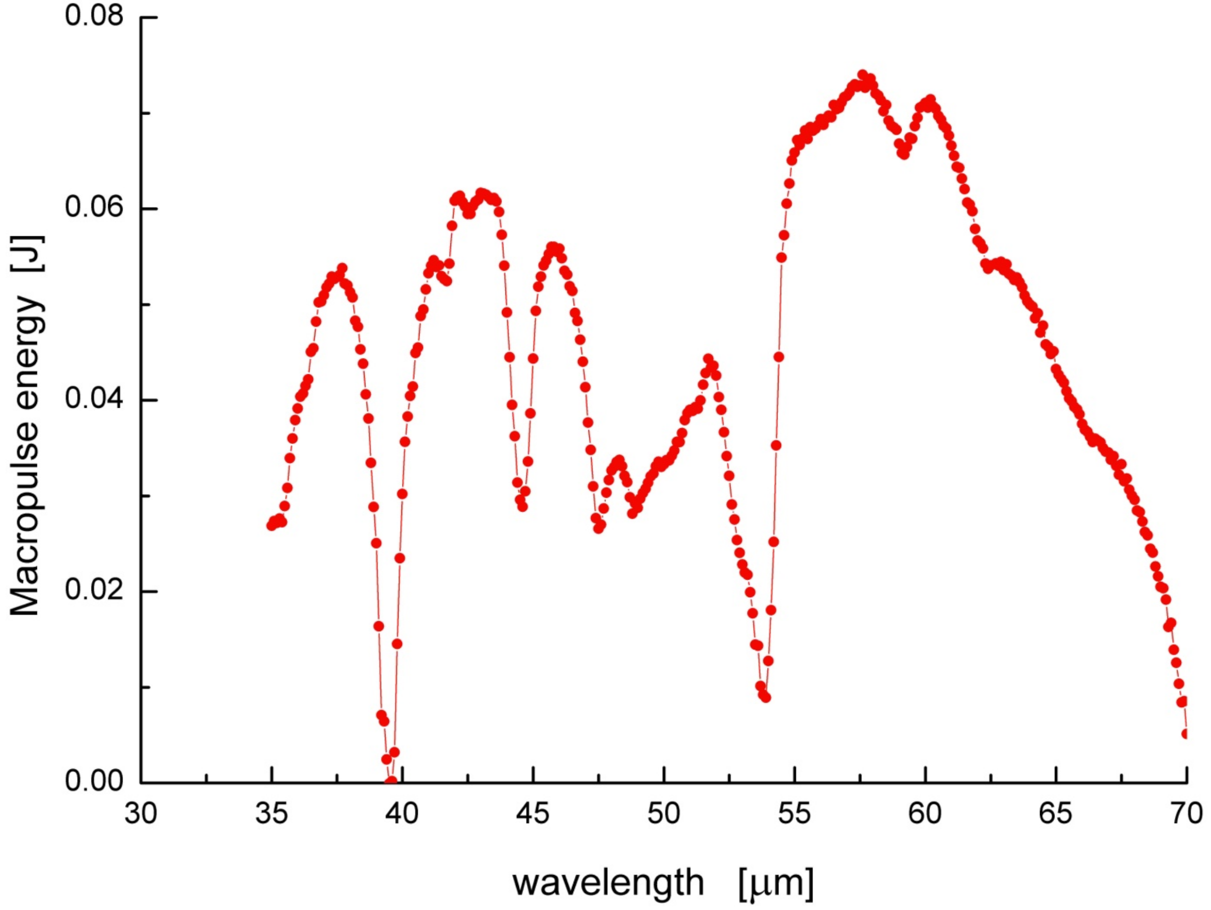
with $w(z) = w_0 \cdot \sqrt{1+\frac{z^2}{z_r^2}}$, $k_n^z = \sqrt{k^2 - n^2 k_\perp^2}$, $R(z) = z + \frac{z_r^2}{z}$

and $k_\perp = \frac{\pi}{g}$, where k is the wave vector in vacuum, z_r the Rayleigh range and w_0 the waist

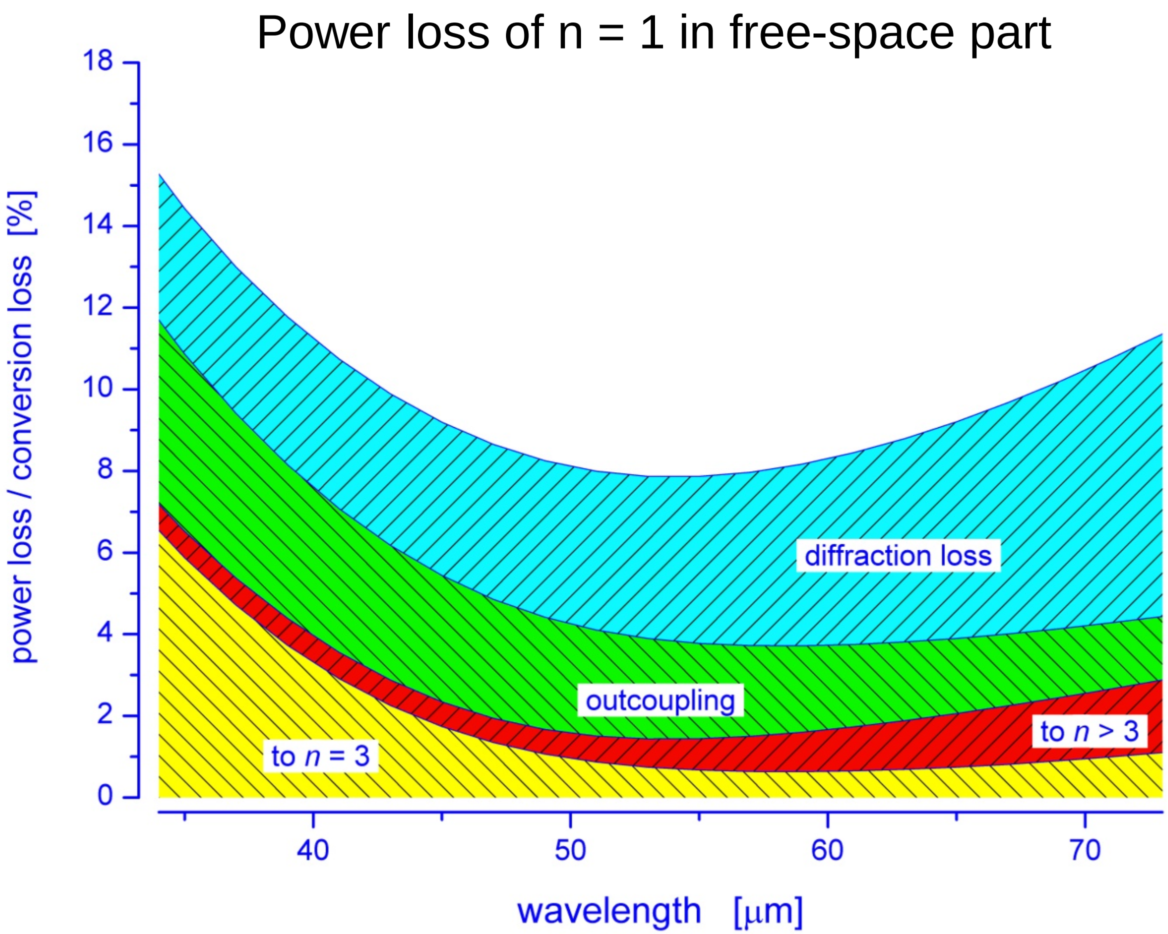
n=1 n=3



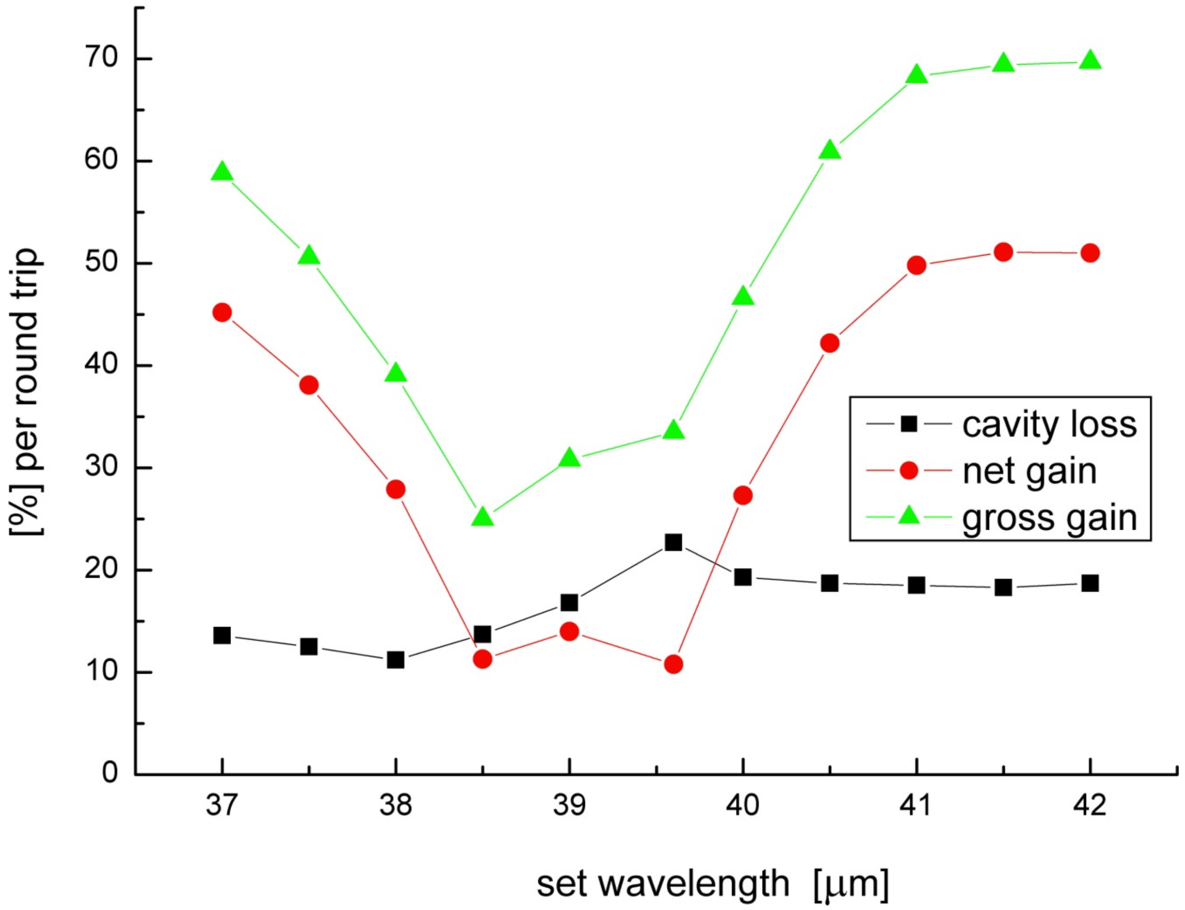
Typical tuning curve FELIX waveguide FEL



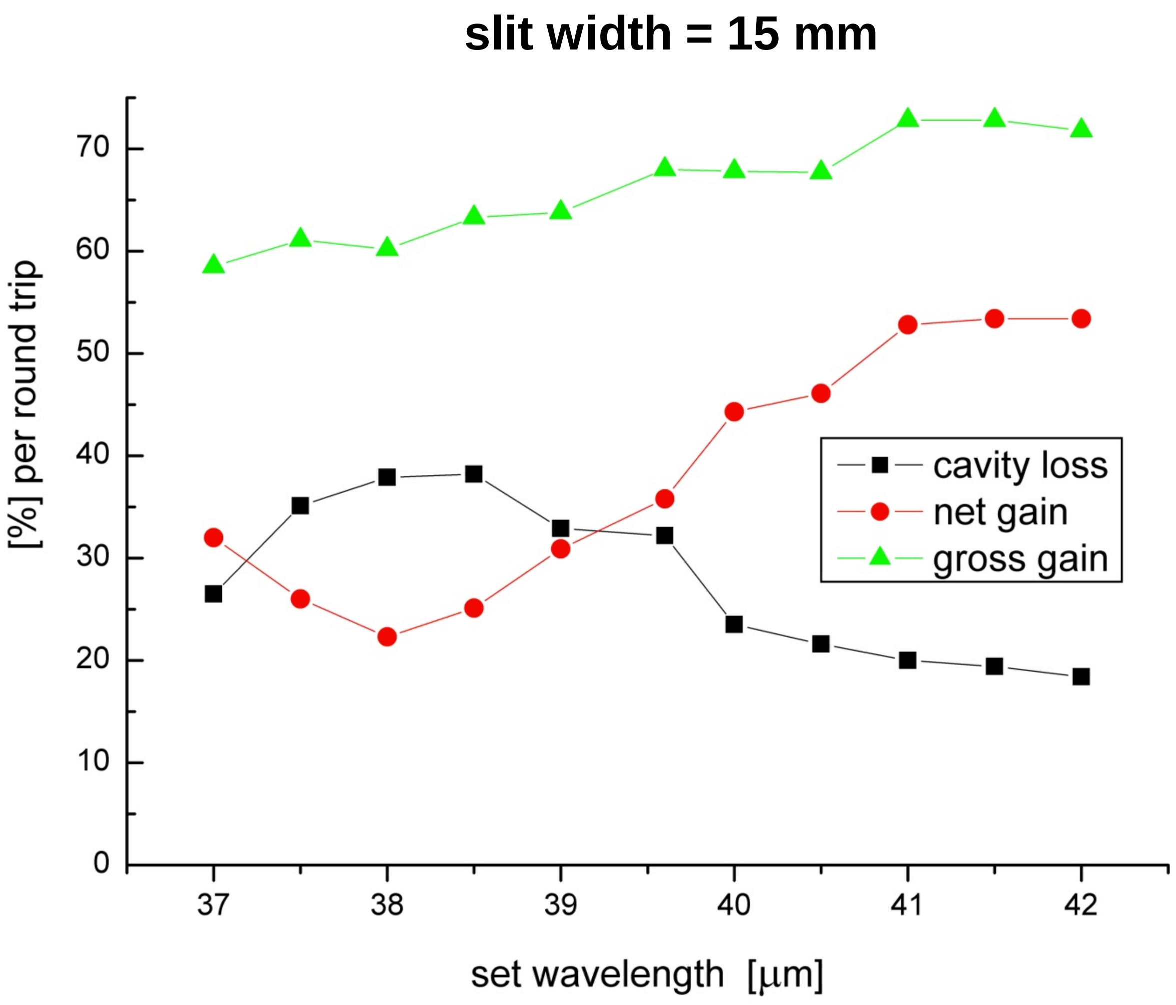
Mode conversion in free-space part



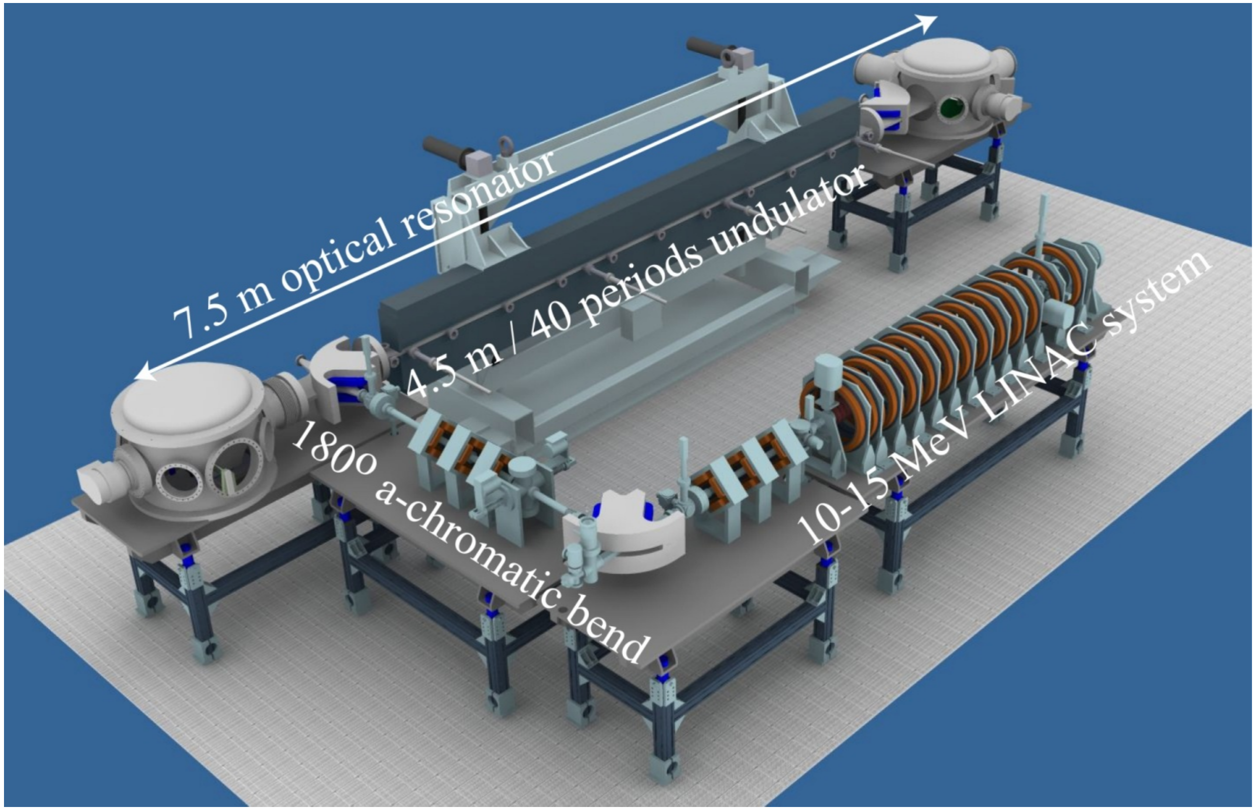
Measured roundtrip gain and loss



Measured roundtrip gain and loss cont.

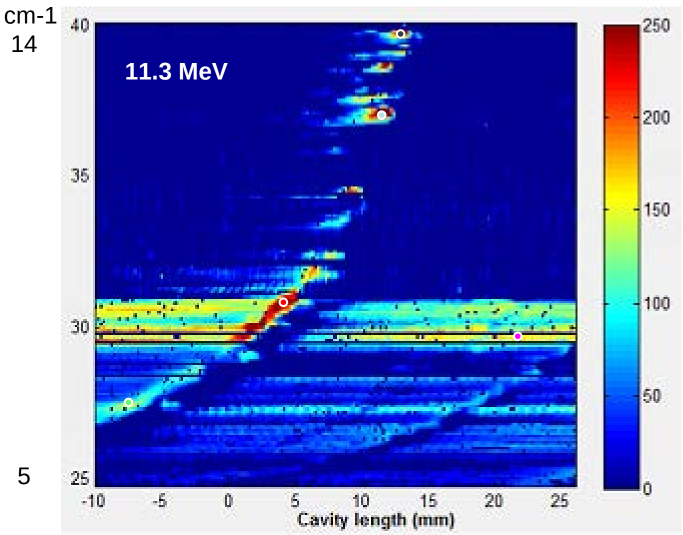


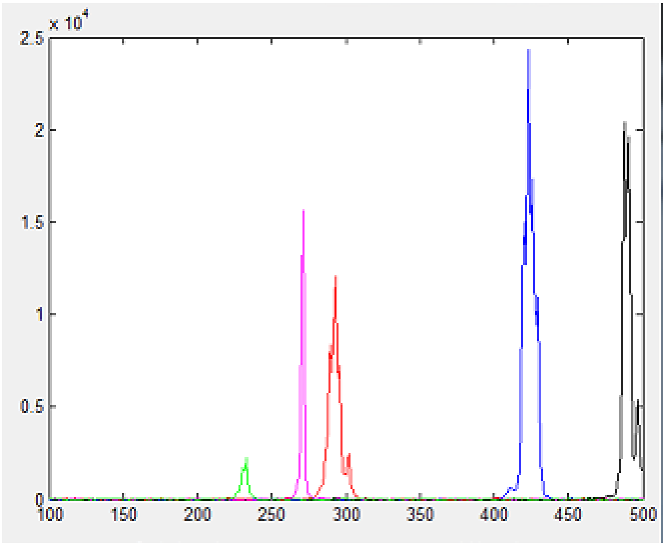
'FLARE' in Nijmegen

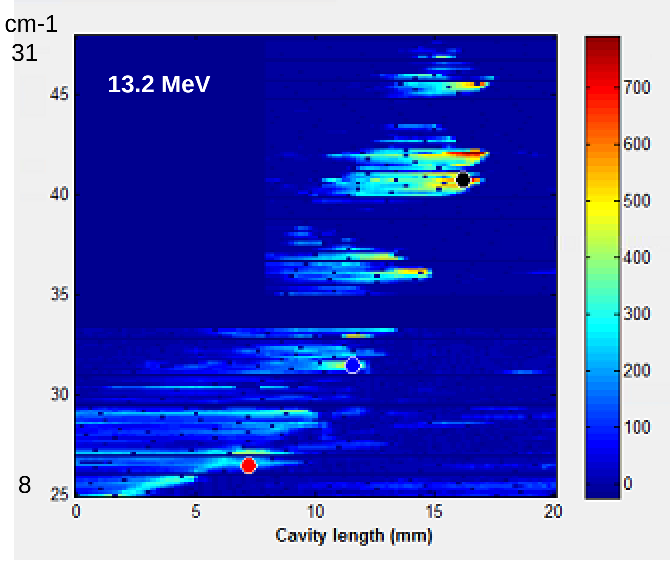


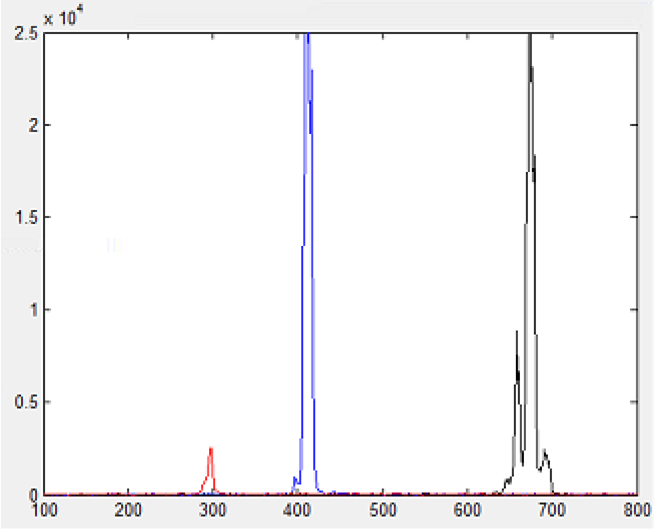
e-beam:	10-15 MeV, 3 GHz, 10 μs, 10 Hz
wavelength range:	100 – 1500 μm
special feature:	narrow-band mode

FLARE tuning gaps







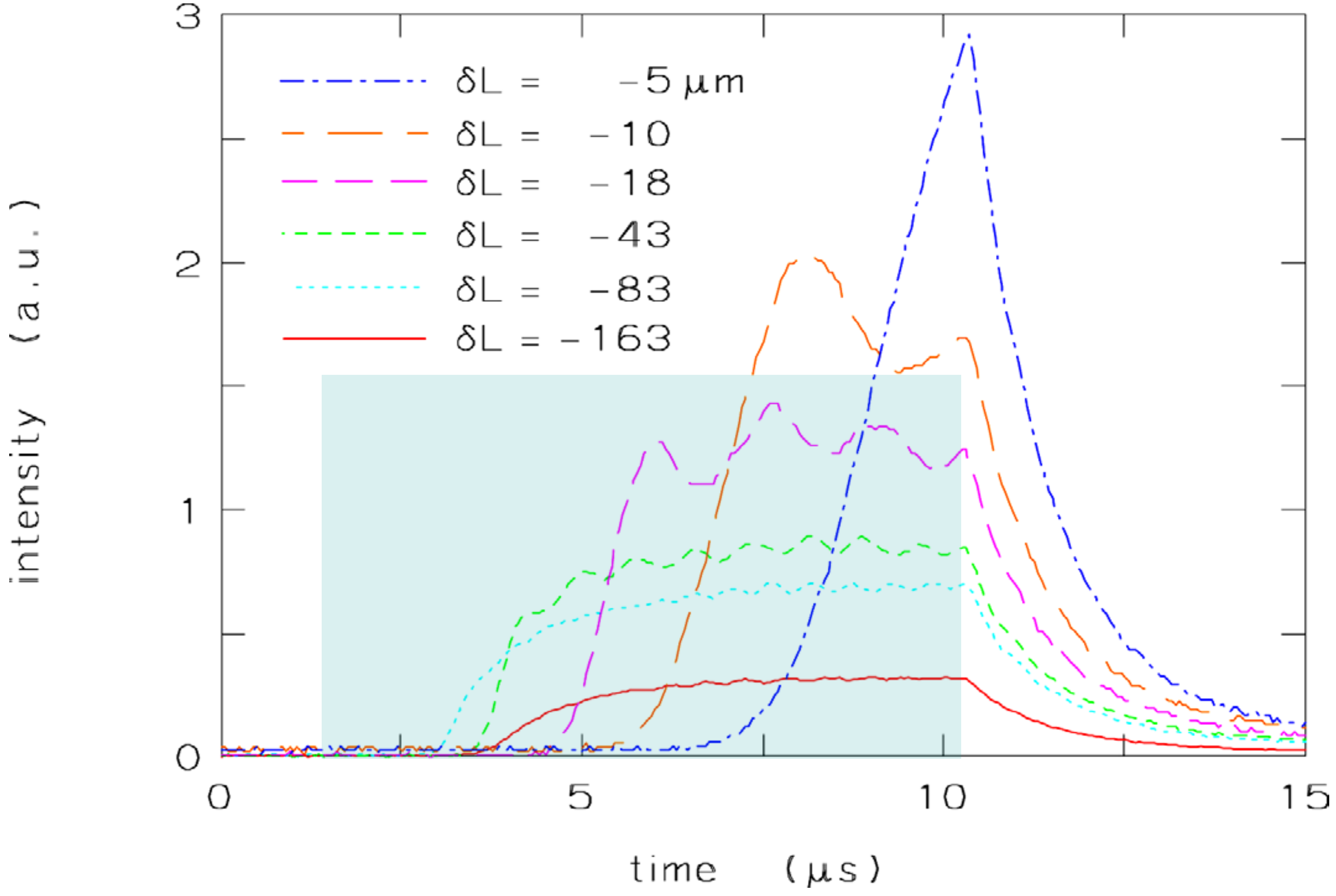


Slippage effects

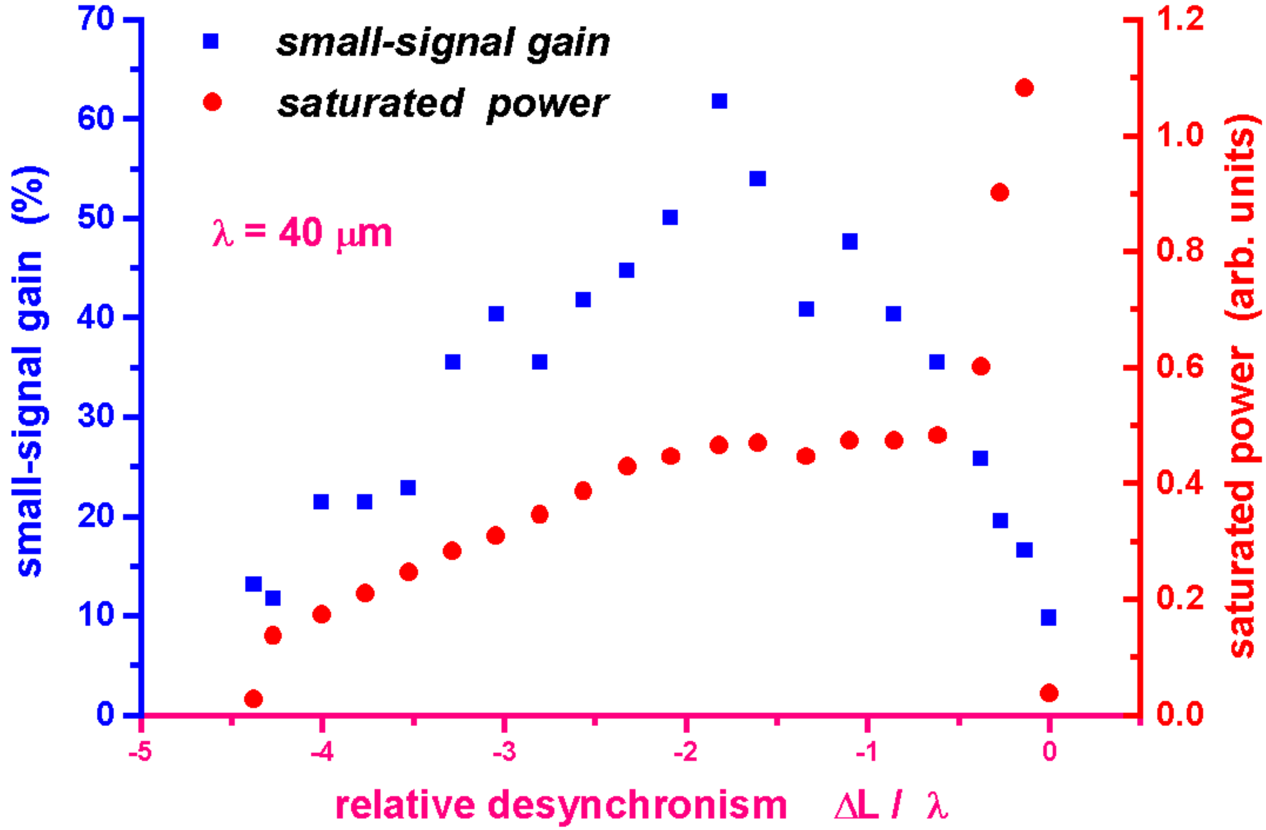
- Lethargy
- Efficiency enhancement
- Limit-cycles
- Bandwidth tuning

Slippage effects

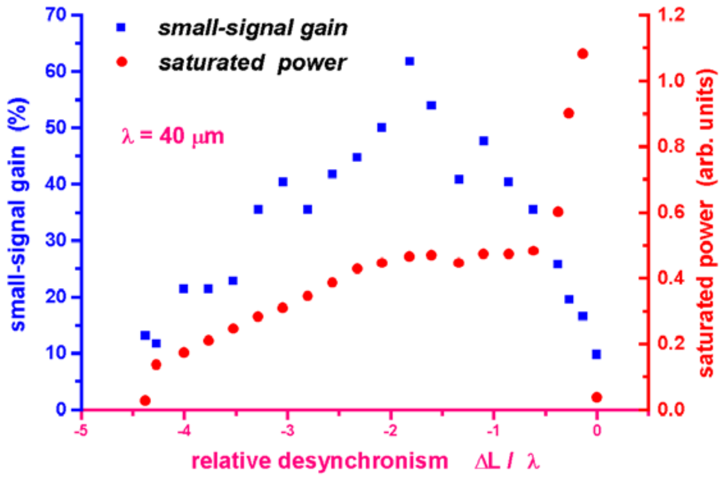
FELIX macropulse shape at λ = 40 μm



Slippage effects: gain and saturation



Slippage effects: gain and saturation

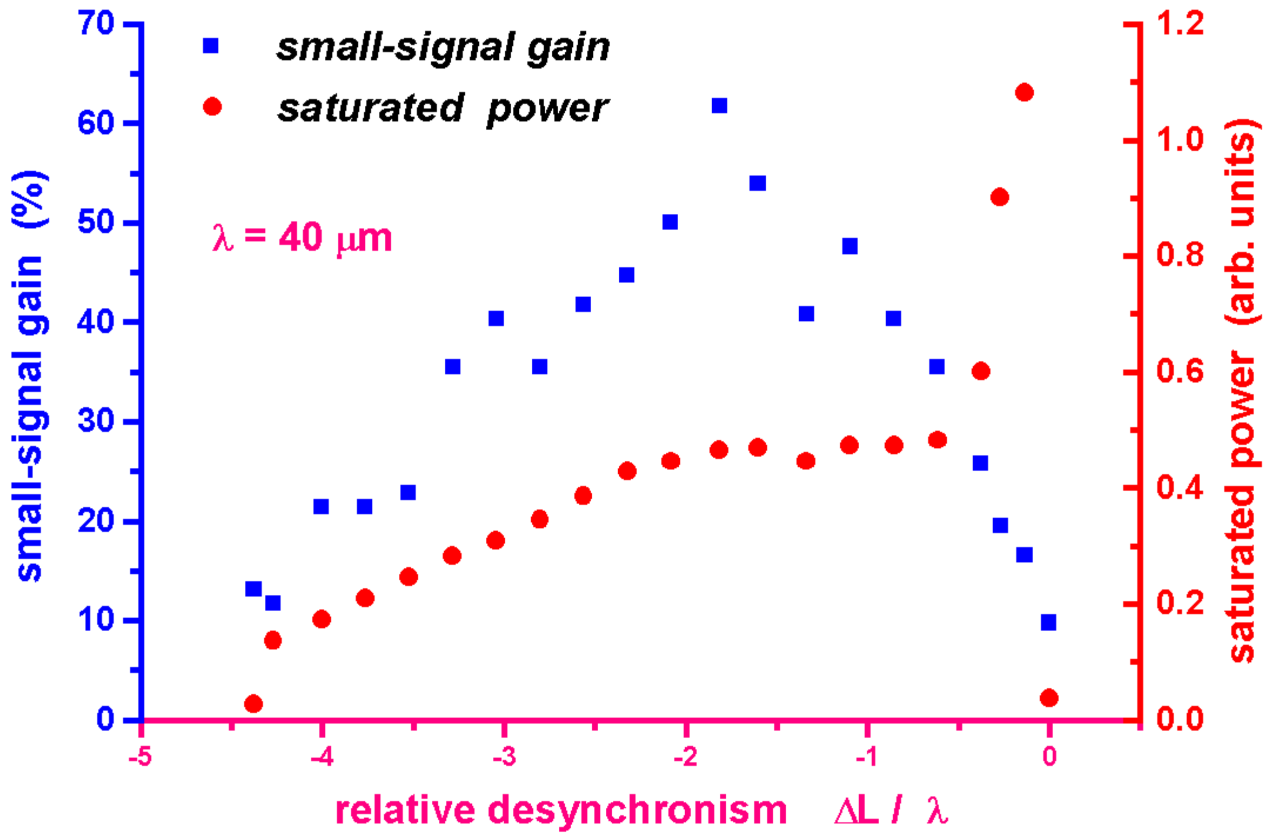


Slippage correction:

$$g_o \longrightarrow \text{Gain} \sim \frac{g_o}{\left(1 + \frac{N_u \lambda}{3\sigma_z}\right)}$$

$$g_o \sim I \sim \frac{Q_b}{\sigma_z} \longrightarrow \text{Gain} \sim \frac{Q_b}{\left(\sigma_z + \frac{N_u \lambda}{3}\right)}$$

Slippage effects: gain and saturation

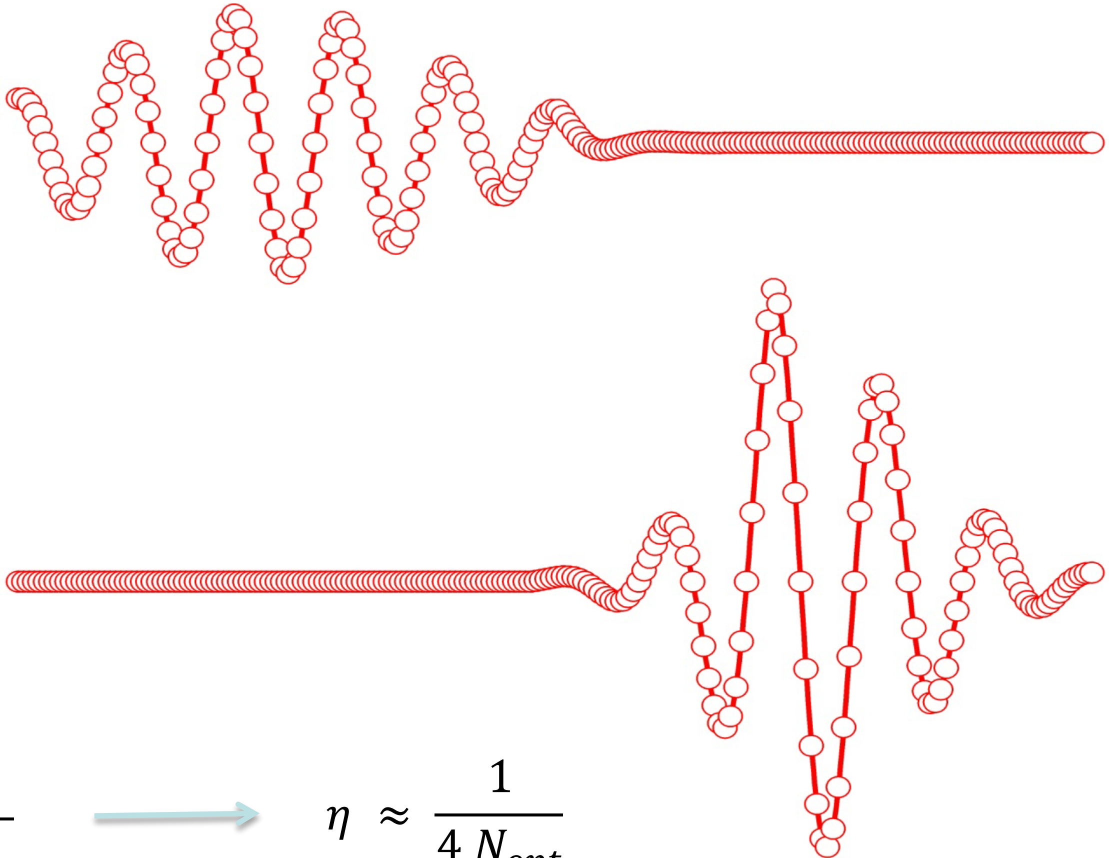


Slippage effects: efficiency enhancement

short pulse propagation

entrance

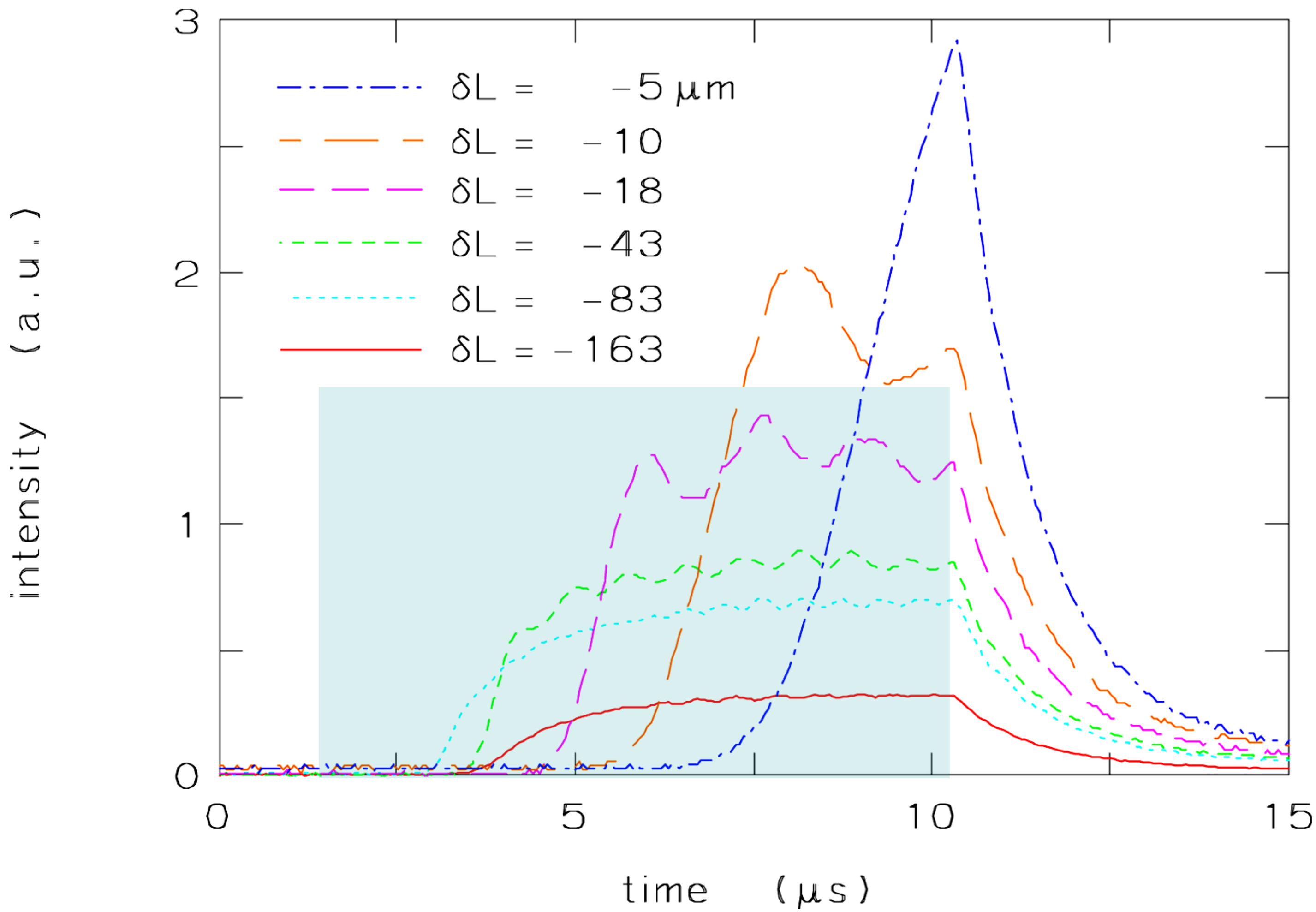
exit



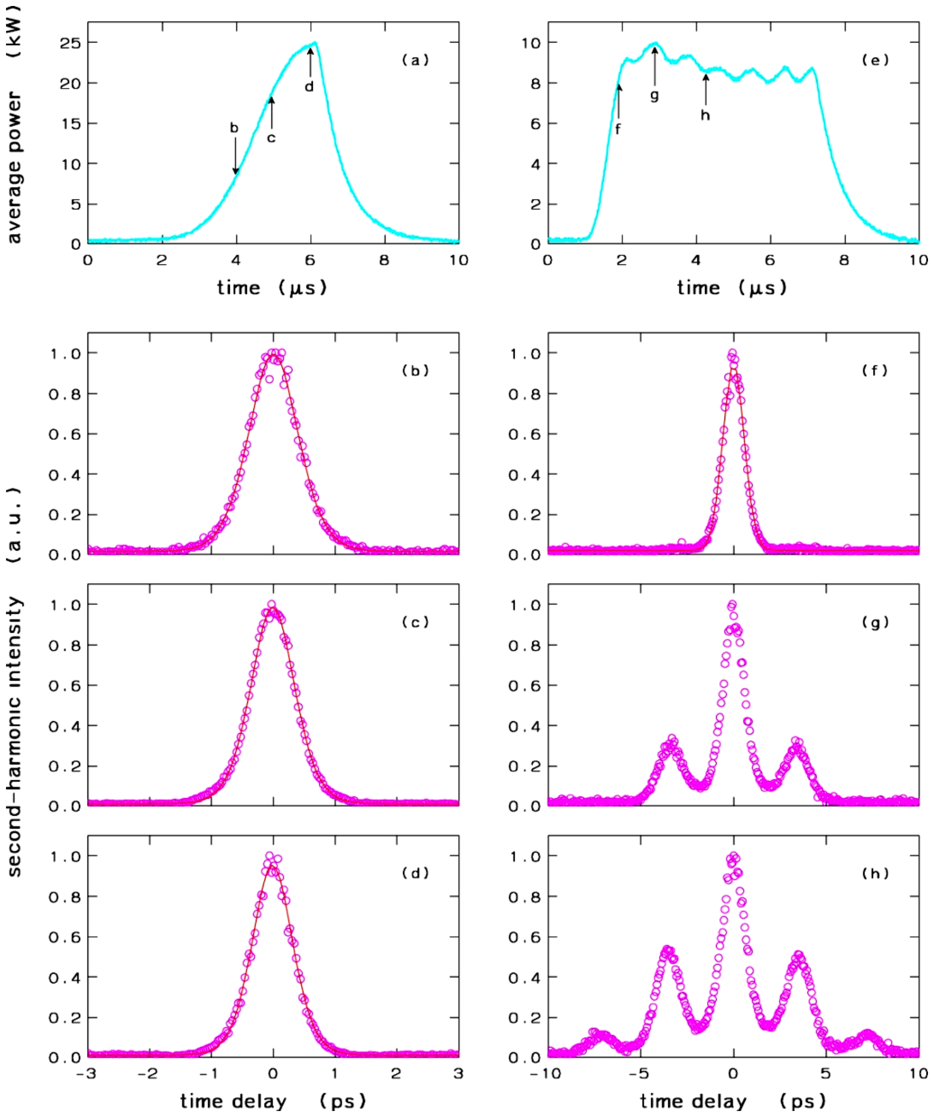
$$\eta \approx \frac{1}{4\,N_u} \longrightarrow \eta \approx \frac{1}{4\,N_{opt}}$$

Slippage effects

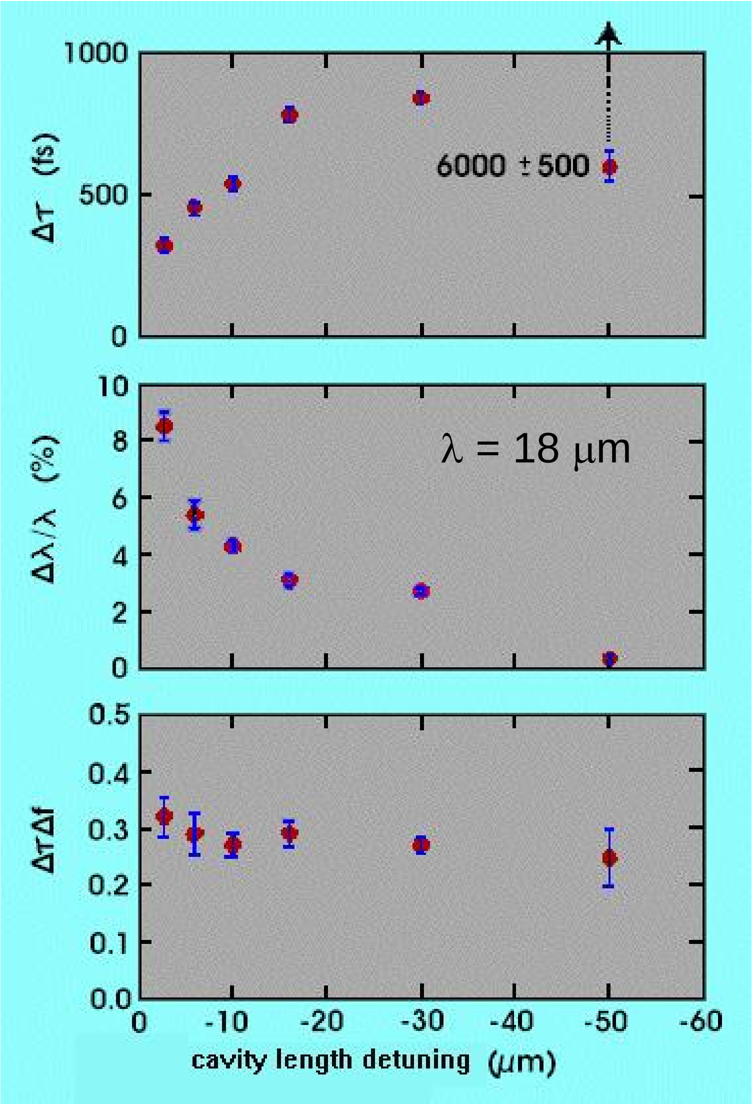
FELIX macropulse shape at λ = 40 μm



Slippage effects: limit cycles



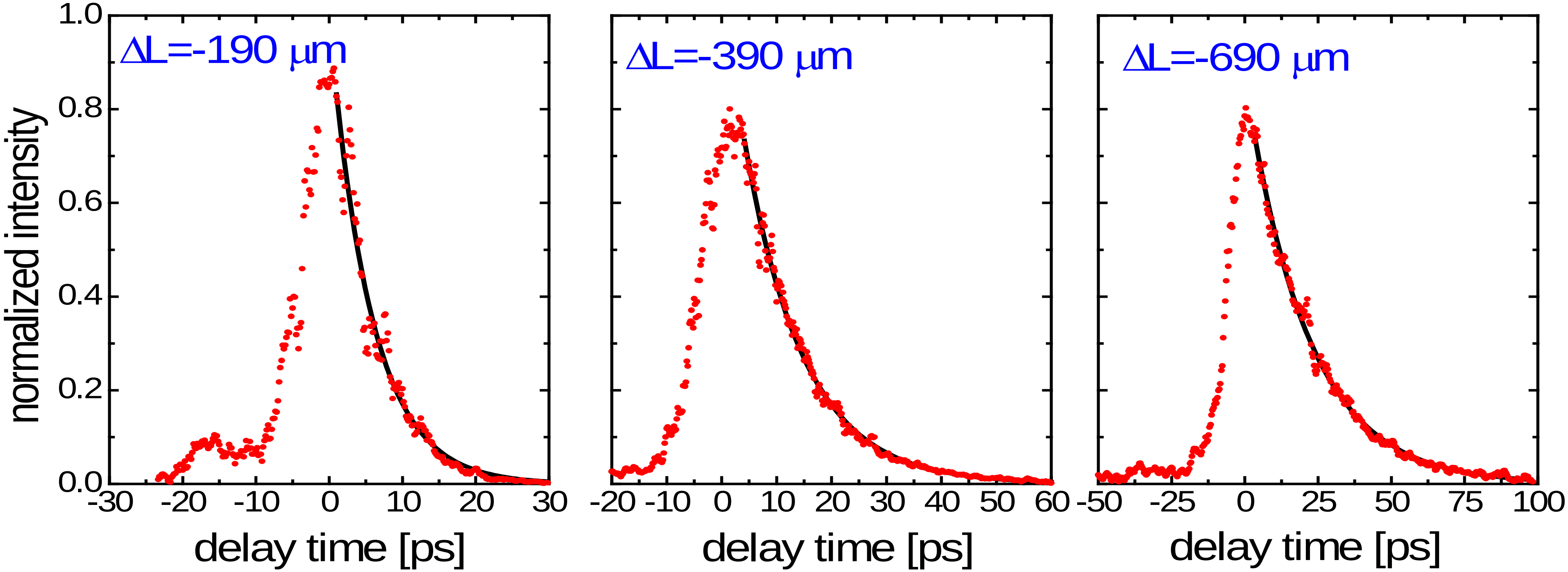
Slippage effects: bandwidth tunability



- bandwidth 0.4 - 6% [FWHM]
- near transform limited

Slippage effects: pulse length tunability

FELIX micro-pulse shape at λ = 150 mm
for different cavity detunings

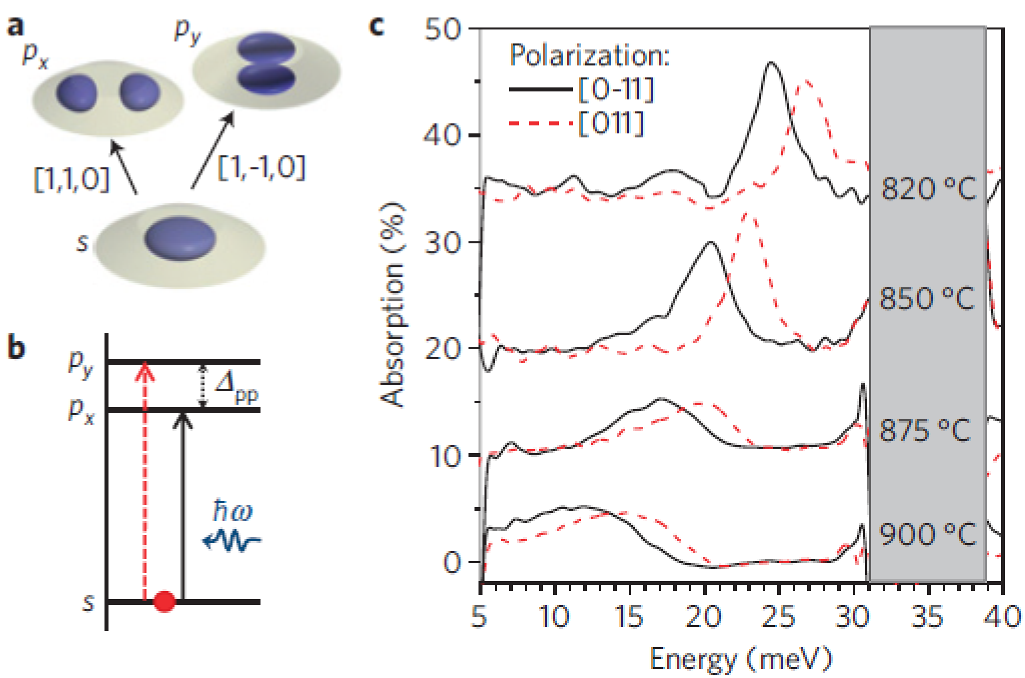


Exponential leading edge has a time constant:

$$\tau = \frac{2\Delta L}{\alpha c}$$

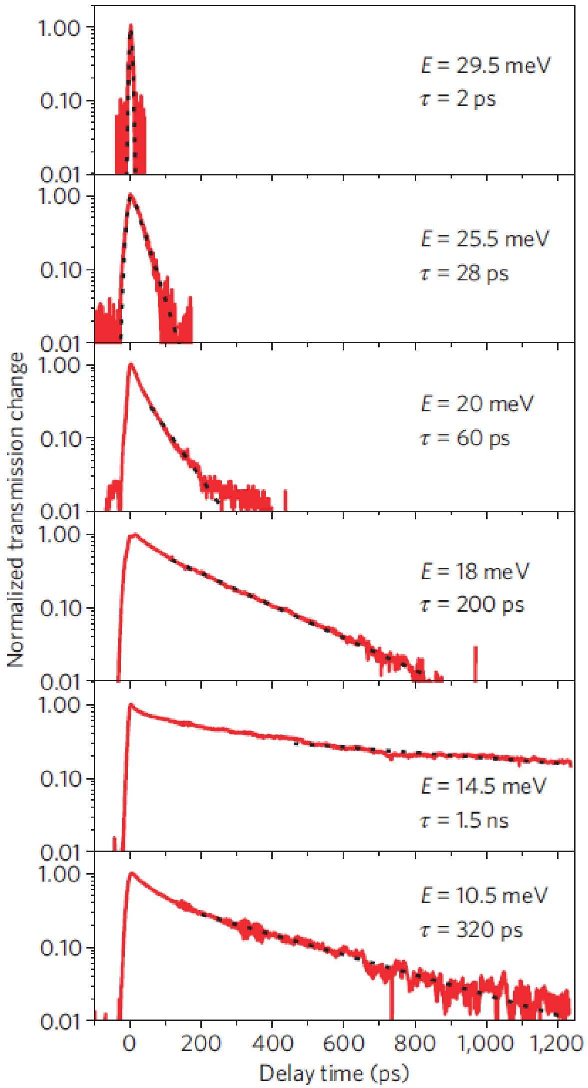
where α are the cavity losses and ΔL is the cavity detuning from synchronism

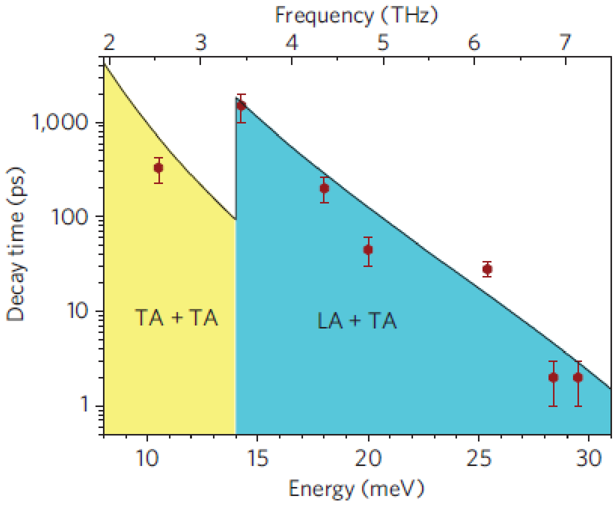
Nonlinear optics: lifetime of quantum-dot intersubband levels



InGaAs self-assembled quantum dots

Nonlinear optics: lifetime of quantum-dot intersubband levels

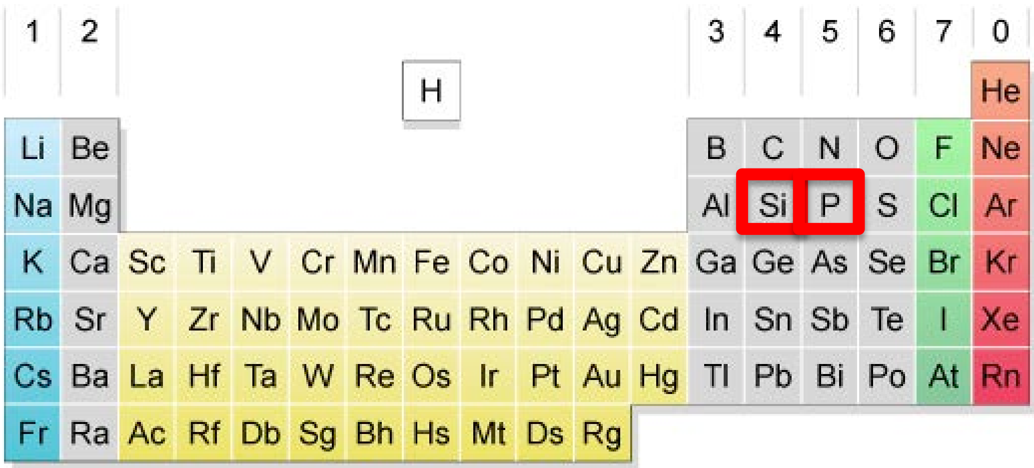




Lifetime strongly depends on energetically available decay channel

***FELBE:** E.Zibik et al., Nature Materials **8** (2009) 803*

A hydrogen-like atom in a silicon chip



- Binding energy:

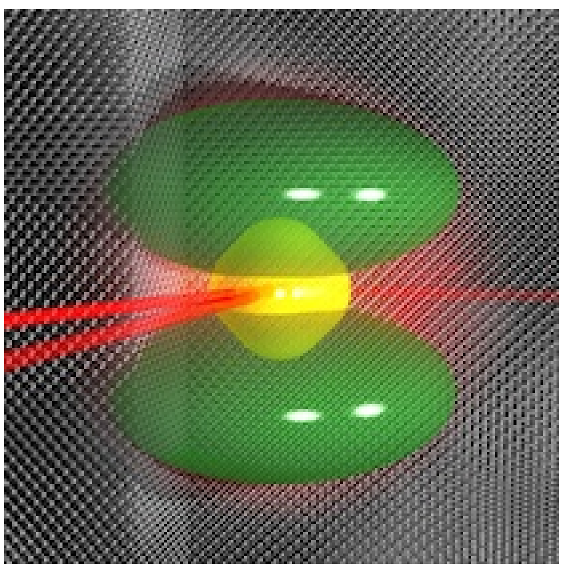
$$E_R = \frac{1}{2}\left(\frac{e^2}{2h}\right)^2 \frac{m_e}{\varepsilon^2}$$

- Bohr radius

$$a_0 = \frac{h^2}{\pi \cdot e^2}\frac{\varepsilon}{m_e}$$

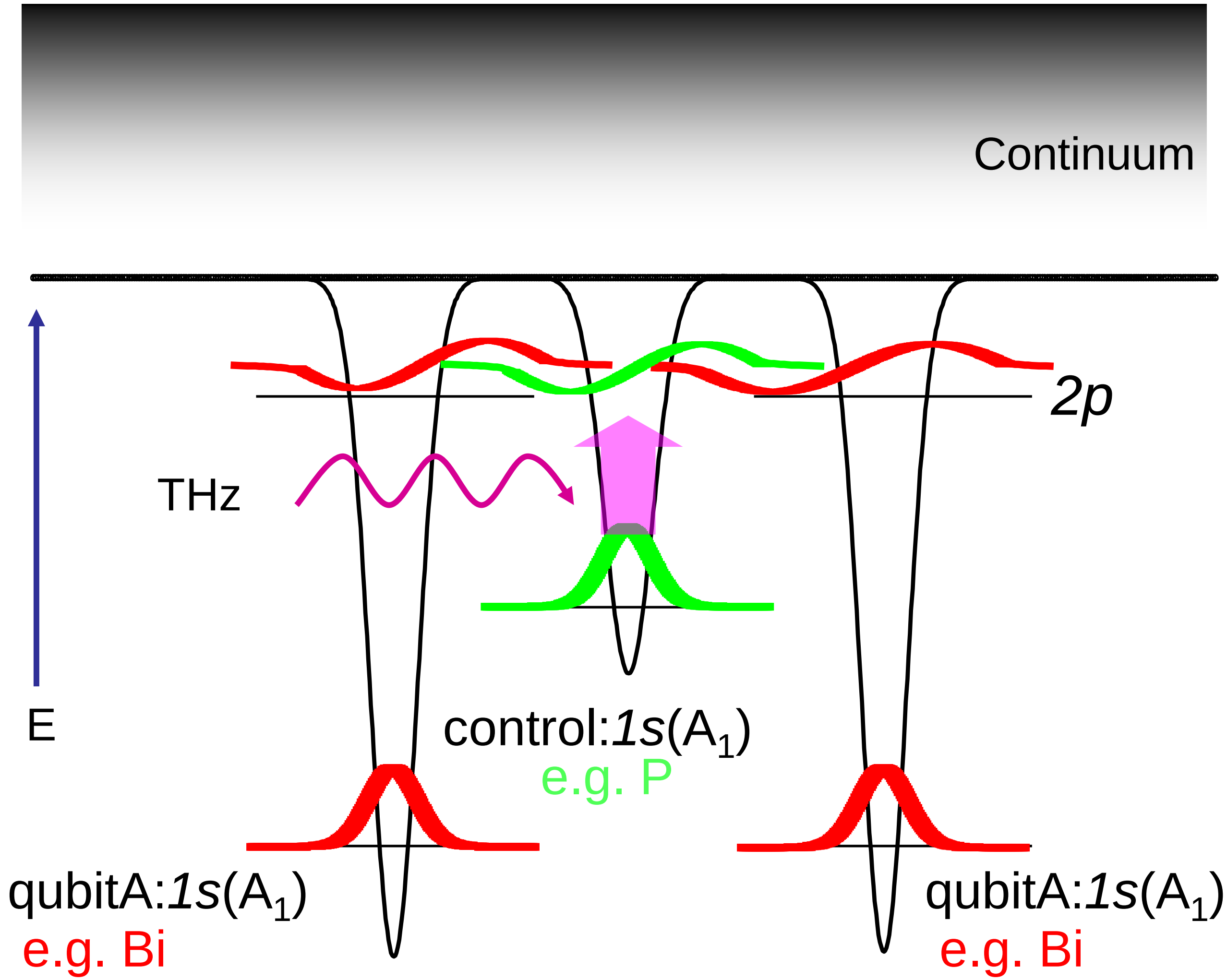
- Characteristic field

$$B_0 = \frac{\pi}{4}\left(\frac{e}{h}\right)^3 \frac{m_e^2}{\varepsilon^2}$$



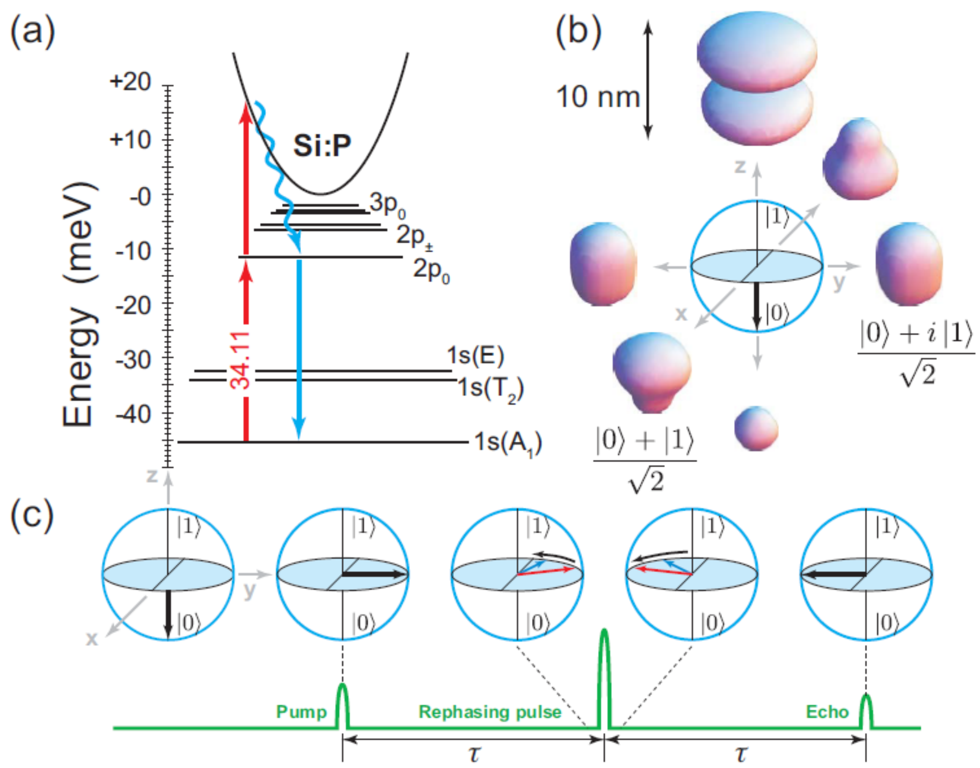
	H	Si:P
ε_r	1	11.4
m_e	1	0.19
E_R	13.6 eV	0.020 eV
a_0	0.056 nm	3.2 nm
B_0	117,000 T	32 T

QIP proposal: THz control of entanglement

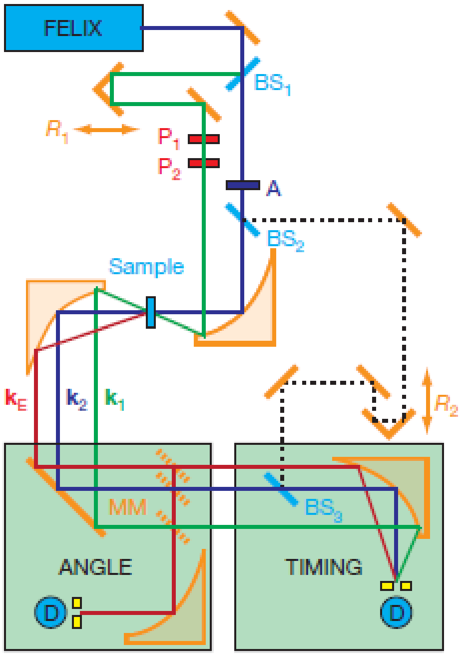


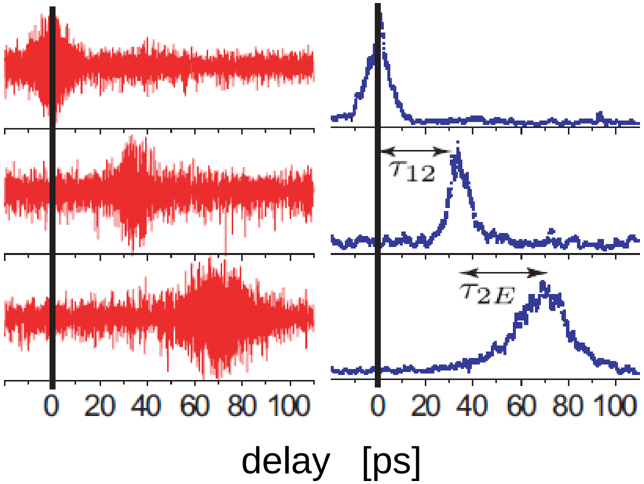
A. M. Stoneham *et al*, *J. Phys. C,* 15, L447, 2003.

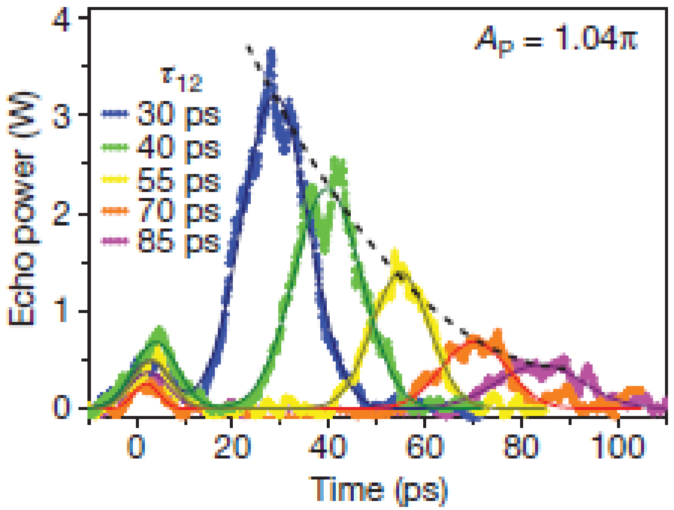
Coherent control of orbital states: Hahn echo in Si:P



Coherent control of orbital states: Hahn echo in Si:P



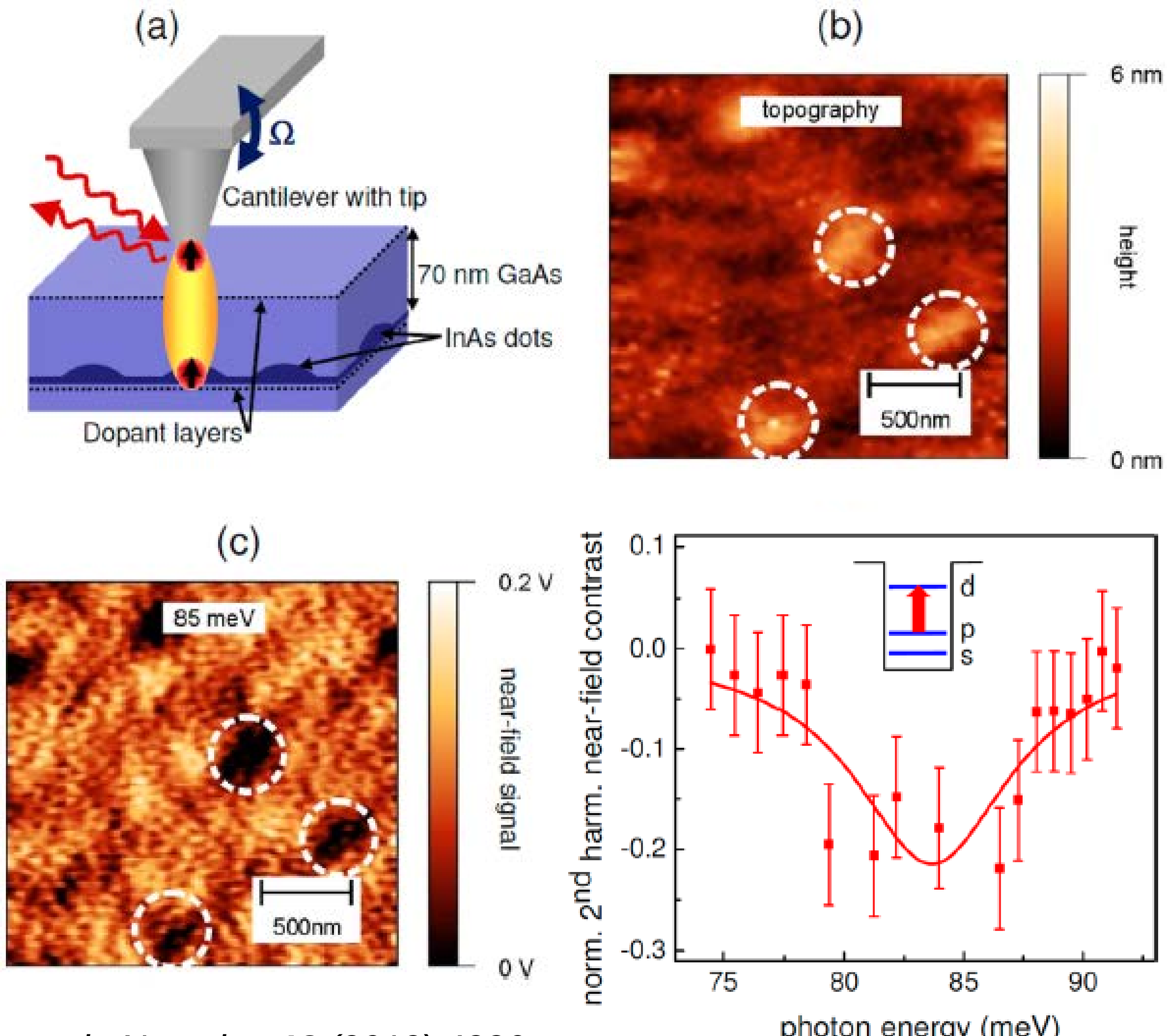




T2 ~ 160 ps lifetime

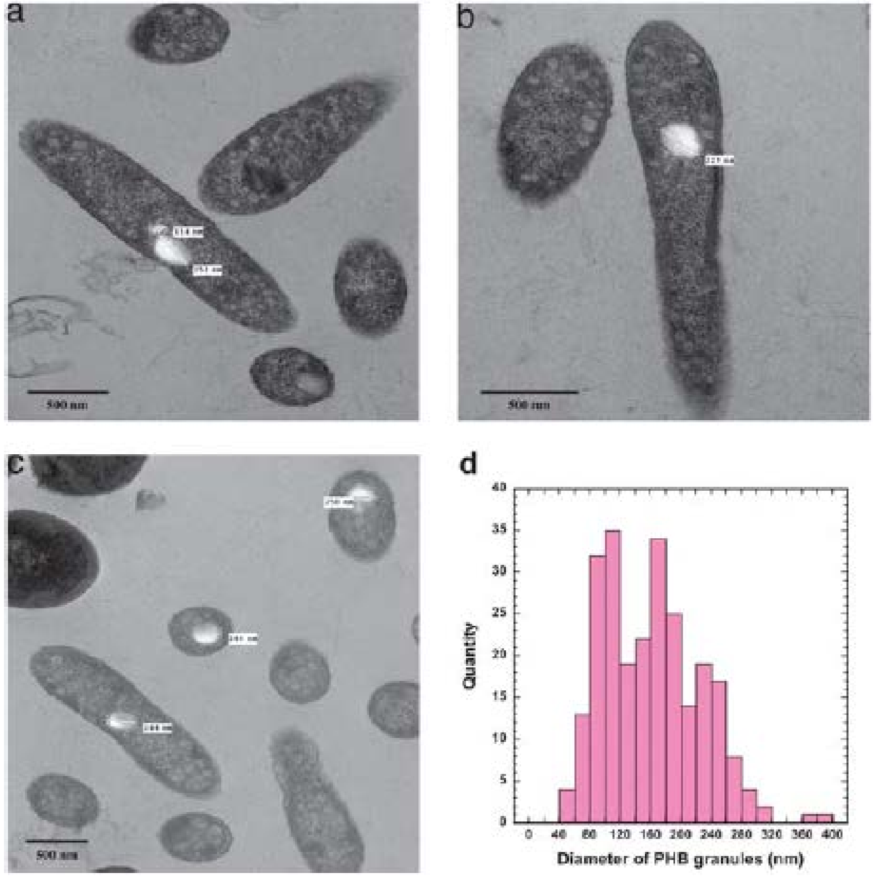
*Greenland et al, Nature **465** (2010) 1057*

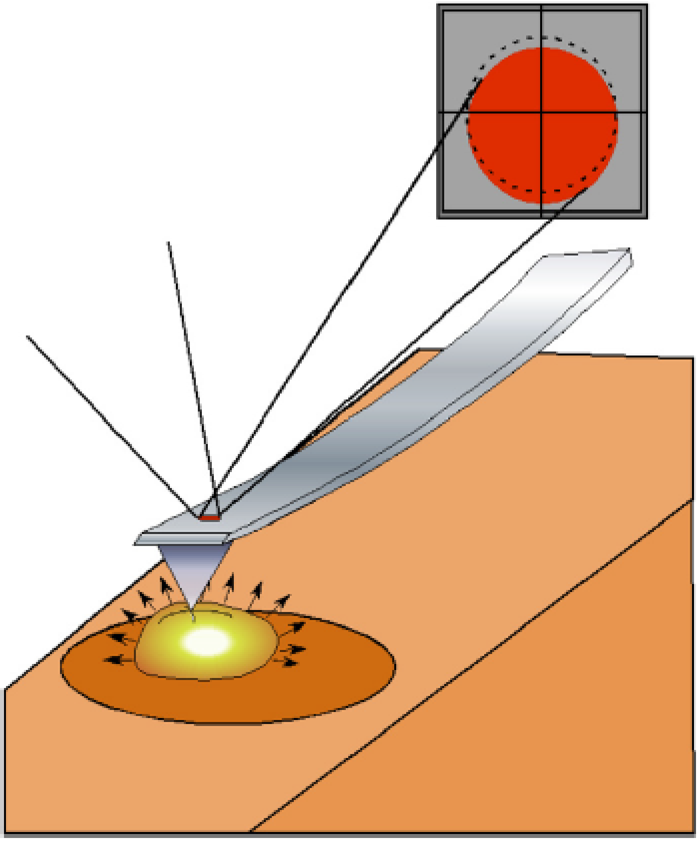
Nano spectroscopy: single quantum dot



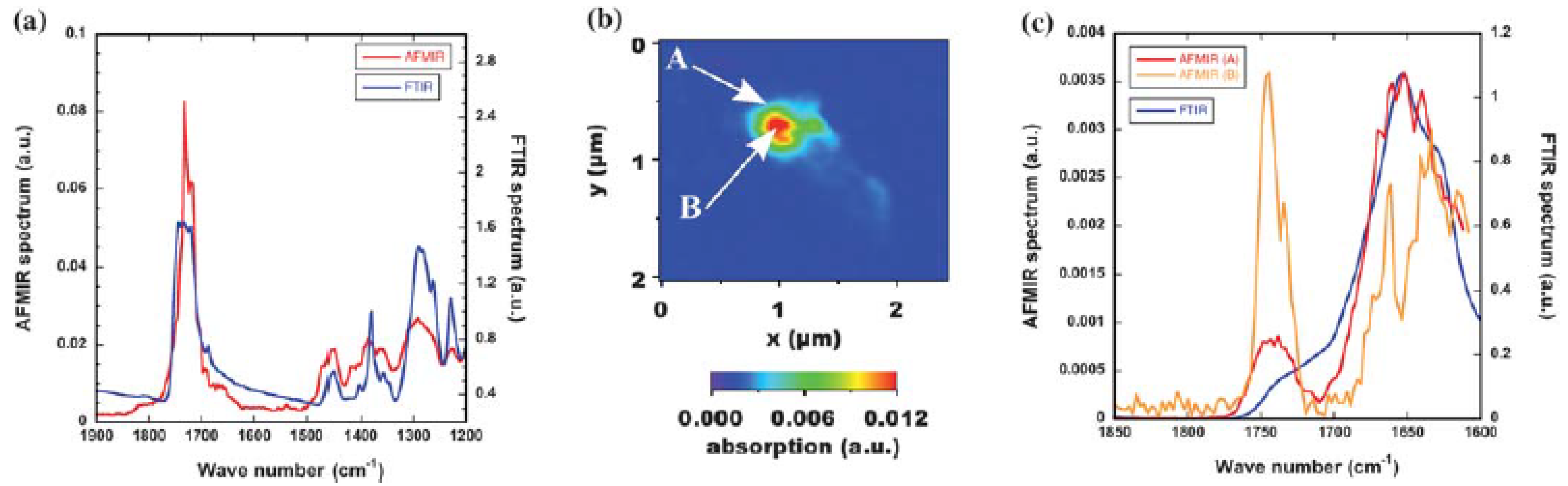
FELBE: *R. Jacob et al., Nano lett.**12** (2012) 4336*

Nano spectroscopy: PHB inside bacteria



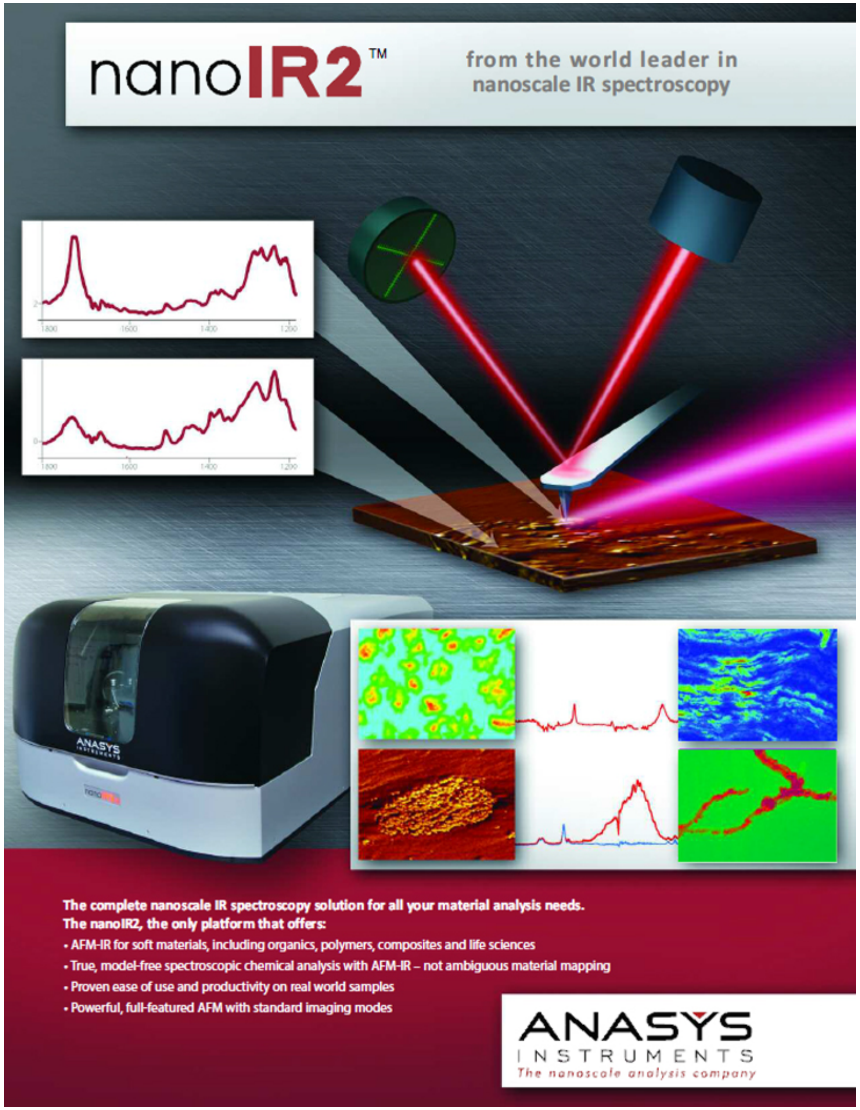


Nano spectroscopy: PHB inside bacteria

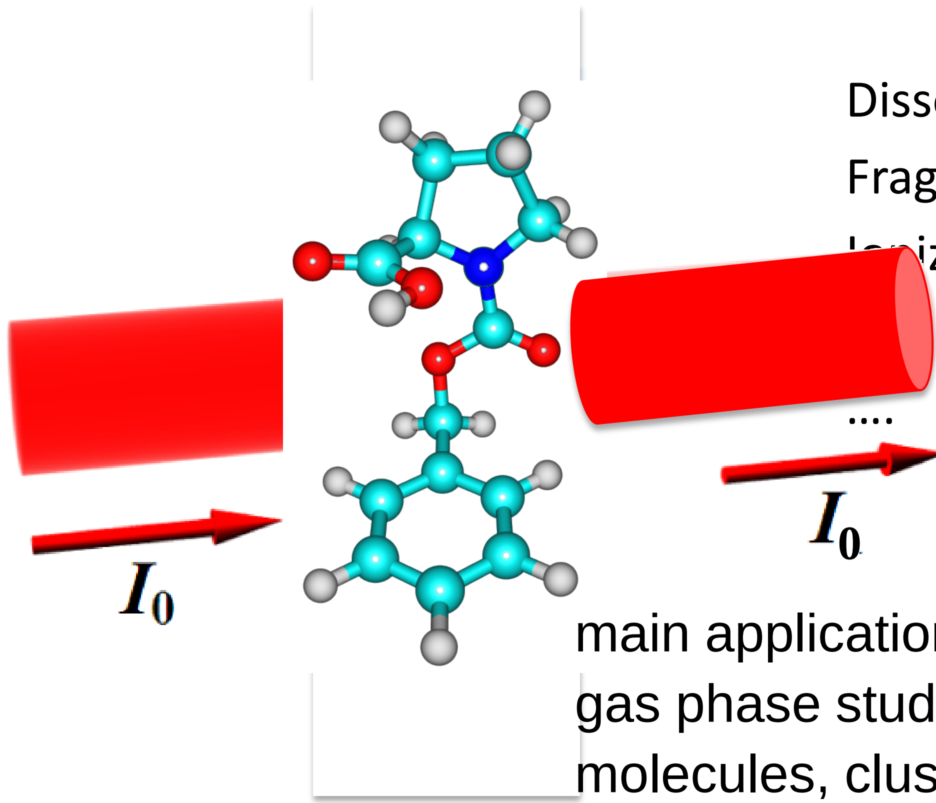


CLIO: *C. Mayet et al., Analyst* ***135*** *(2010) 2540*

Nano spectroscopy: spin-off



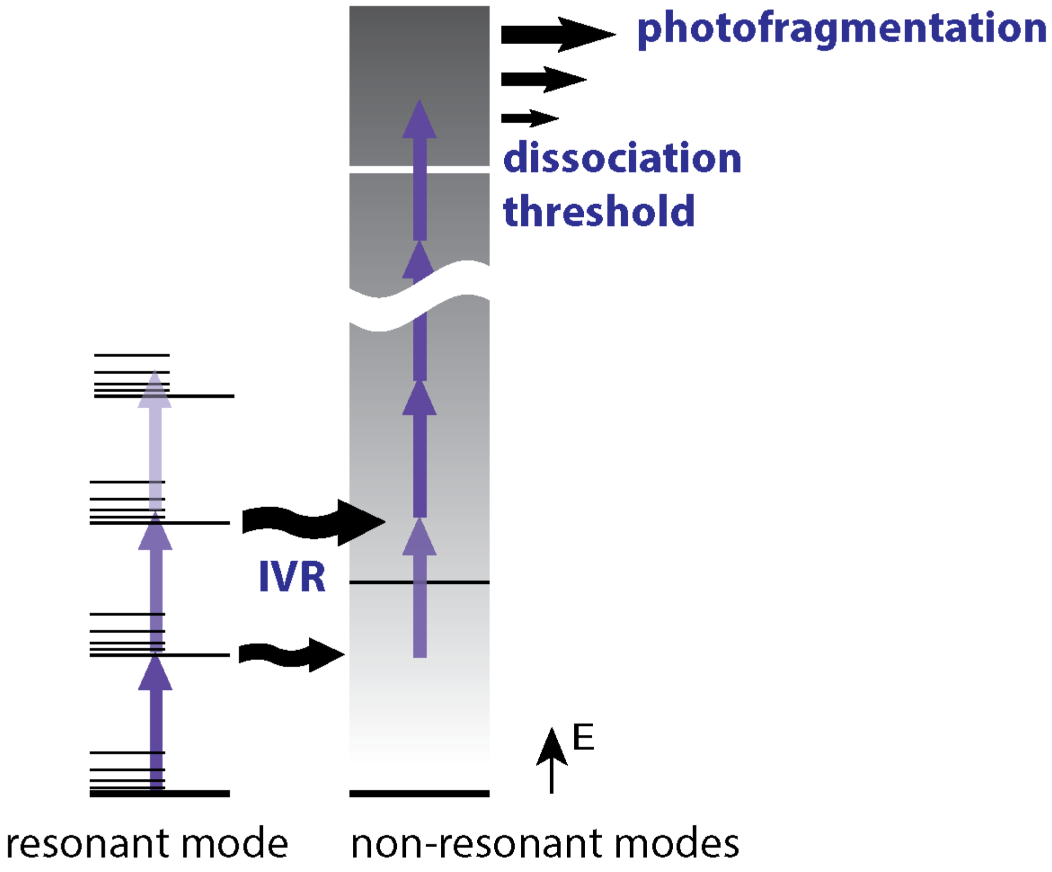
Infrared Action Spectroscopy



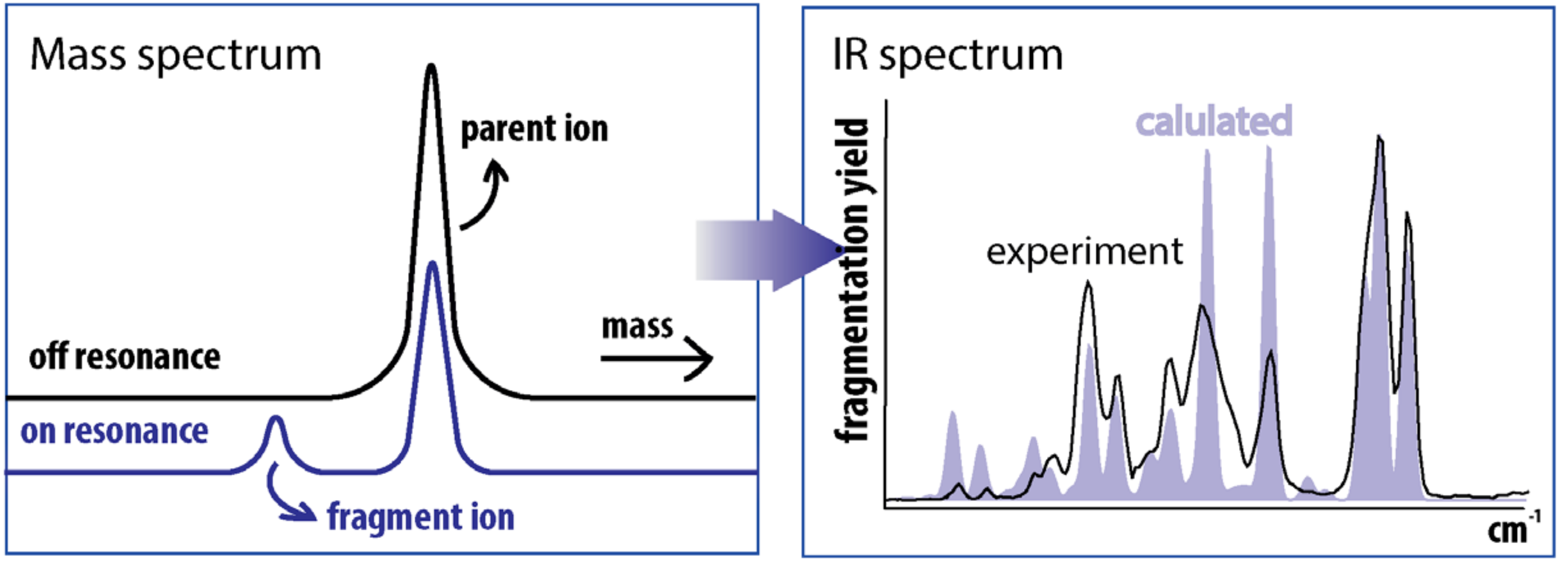
Dissociation
Fragmentation
Ionization
Electron Loss
....

main application:
gas phase studies
molecules, clusters, complexes
neutral, ionic species
molecular beams, ion traps

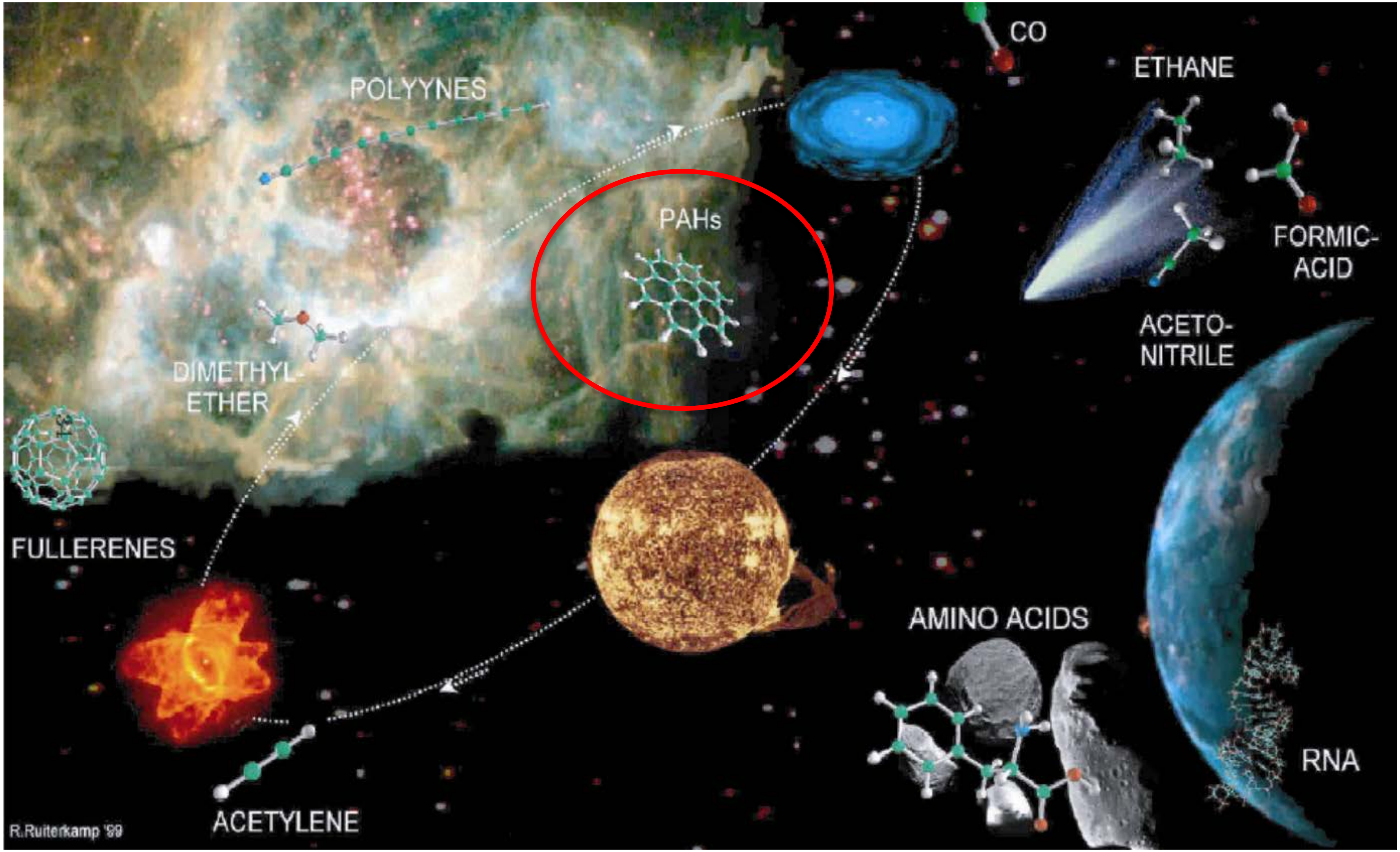
Mechanism, Infrared Spectrum, Molecular Structure



Mechanism, Infrared Spectrum, Molecular Structure

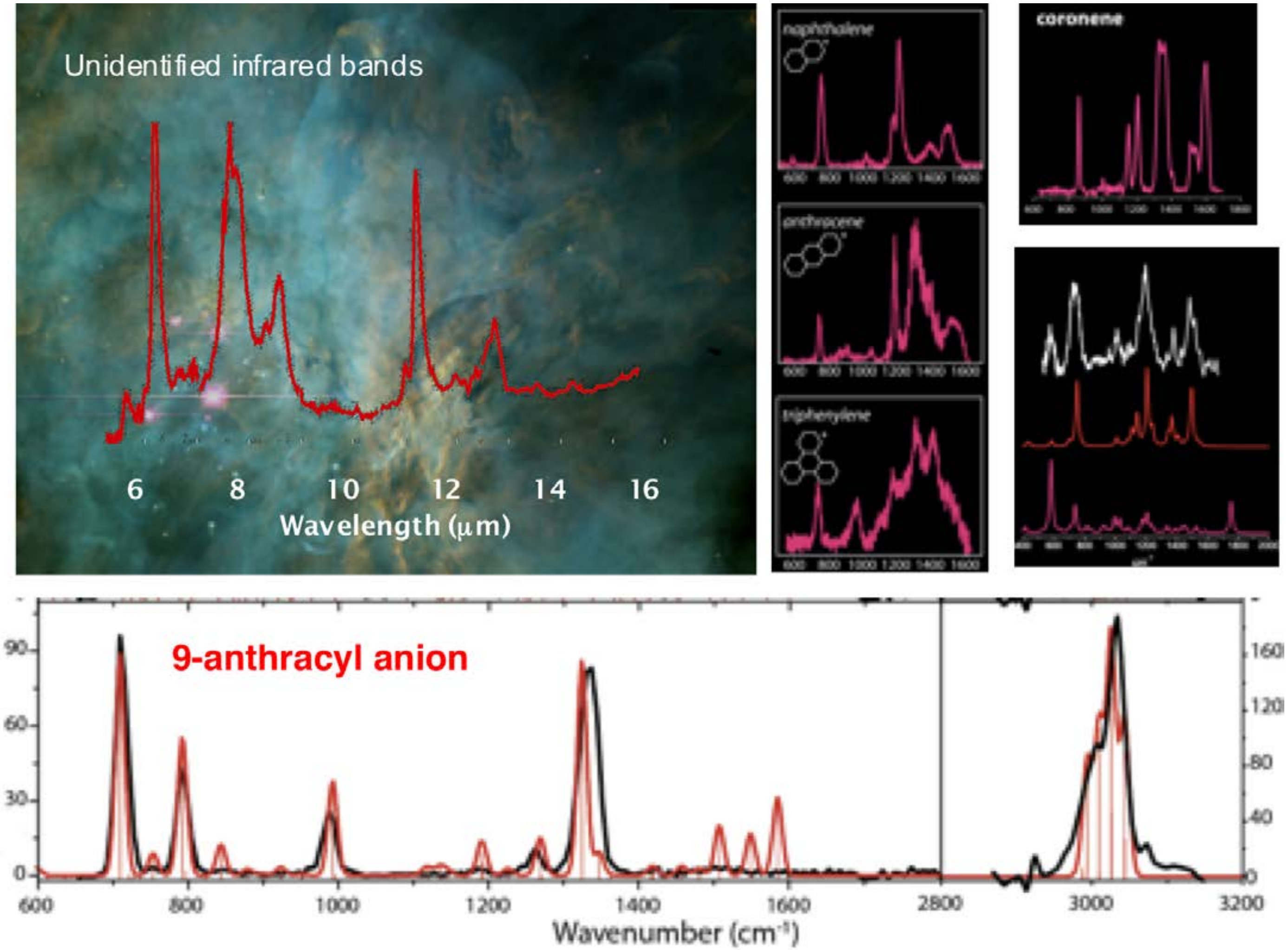


Large organic compounds in space



P. Ehrenfreund

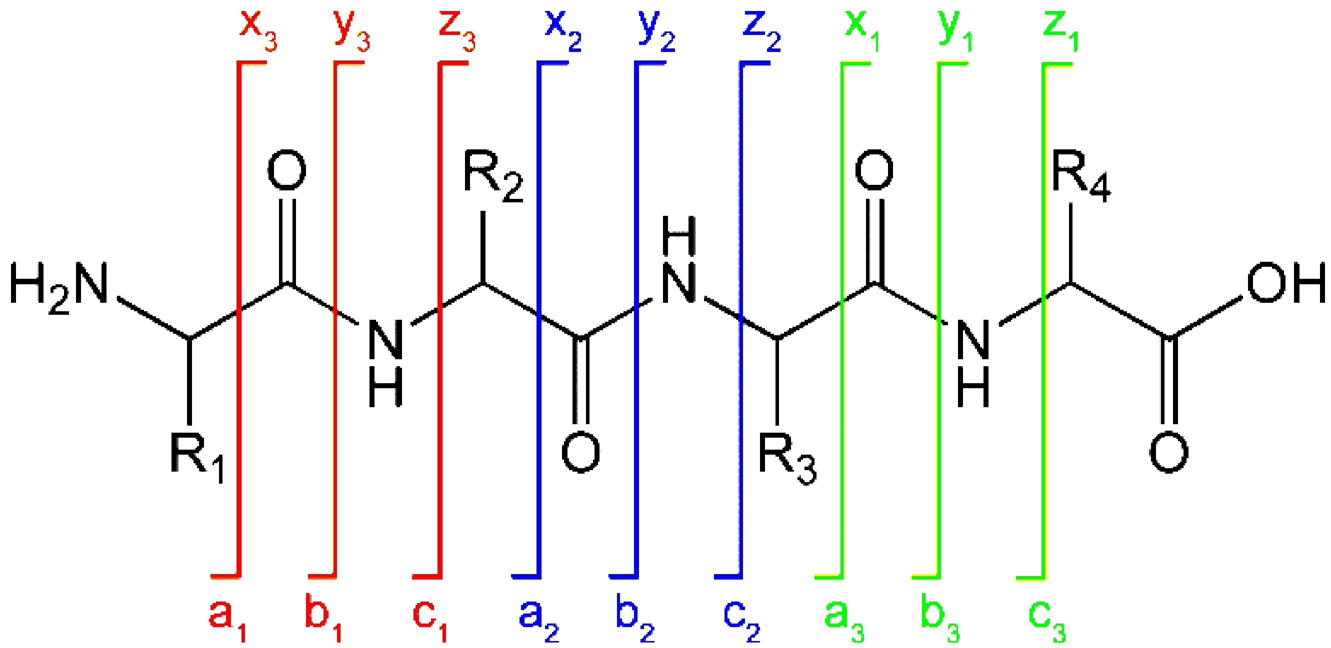
Gas-phase infrared spectra of ionic PAHs



ApJ **542** 404 (2000)
ApJL **560** L90 (2001)
JPC-A **105** 8302 (2001)
JPC-A **107** 782 (2003)
ApJ **591** 968 (2003)
ApJ L66 706 (2009)
JCP 131 184307 (2009)
ANIE 50 7004 (2011)
ApJ 83 746 (2012)
ApJ 170 787 (2014)
…..

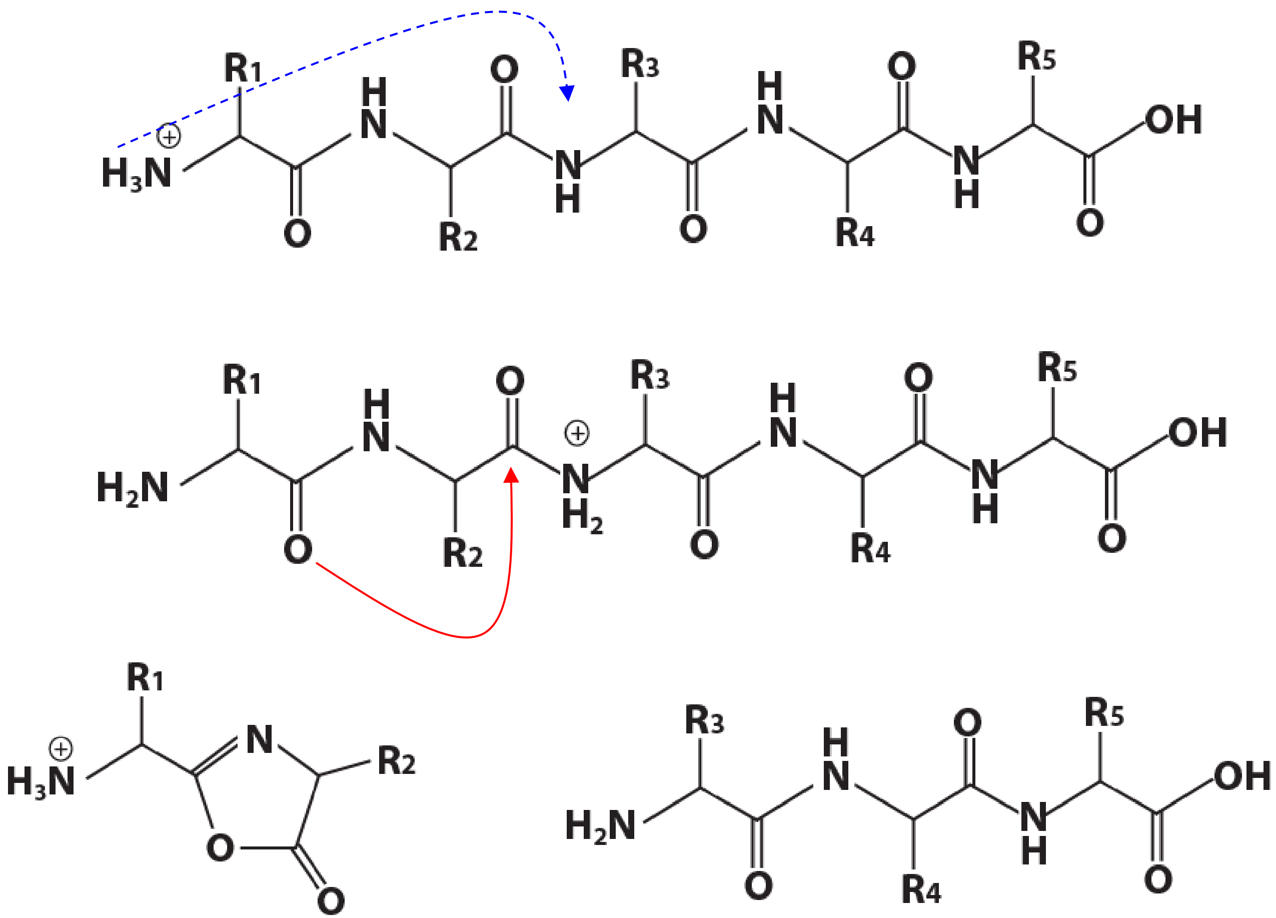
- Investigate various and larger systems in various states – cation, anion, protonated, deprotonated etc.

Understanding Mechanisms of Peptide sequencing



collisional activation → **mobile proton** → **nucleophilic attack** → **amide bond cleavage** → ***b/y* fragments**

b/*y* fragmentation pathway



mobile proton

nucleophilic attack

amide bond cleavage

b-fragment: oxazolone

y-fragment: truncated peptide

Do all b_2 ions have oxazolone structures?

ELSEVIER

International Journal of Mass Spectrometry 210/211 (2001) 71–87

International Journal of Mass Spectrometry

www.elsevier.com/locate/ijms

Do all b_2 ions have oxazolone structures? Multistage mass spectrometry and ab initio studies on protonated *N*-acyl amino acid methyl ester model systems†

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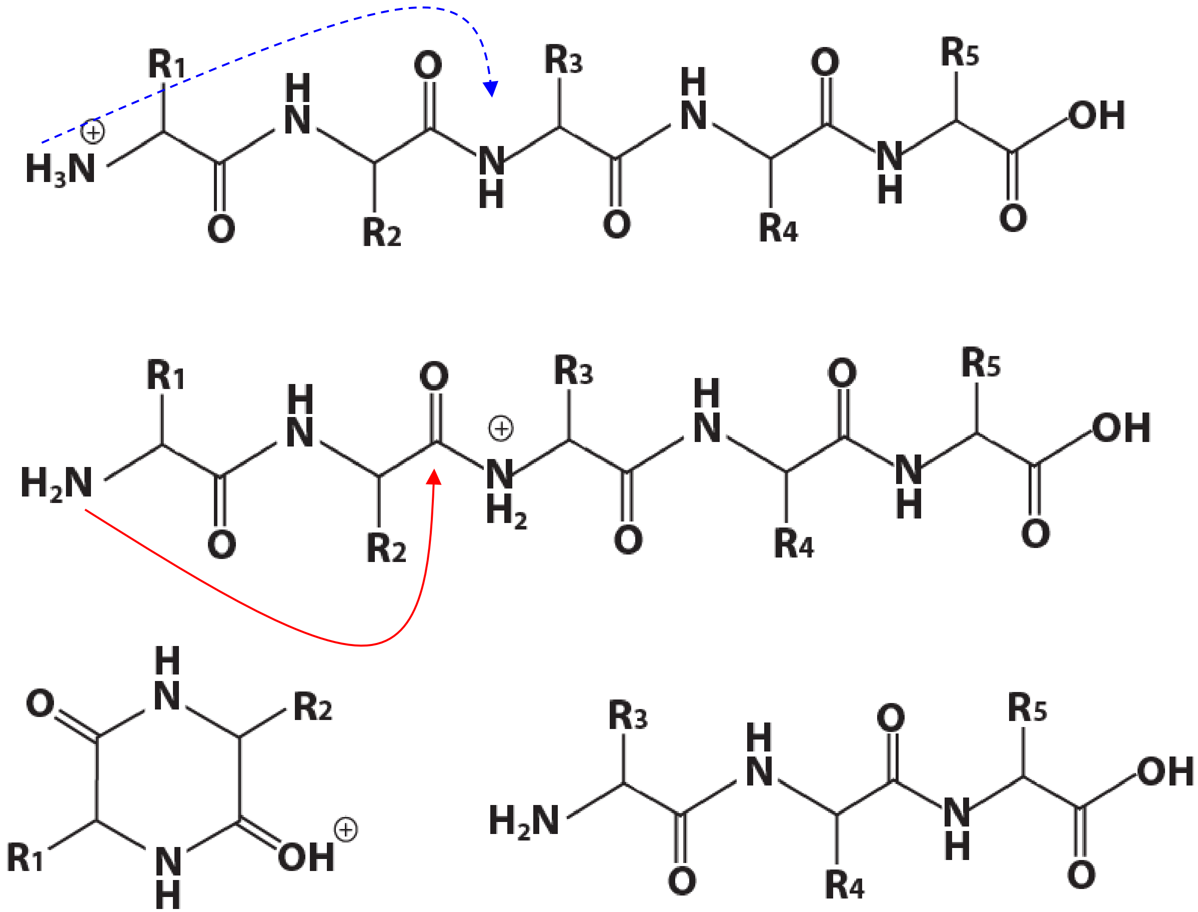
Received 5 December 2000; accepted 19 February 2001

Alternative *b/y* pathway: N-terminus as nucleophile



mobile proton

nucleophilic attack

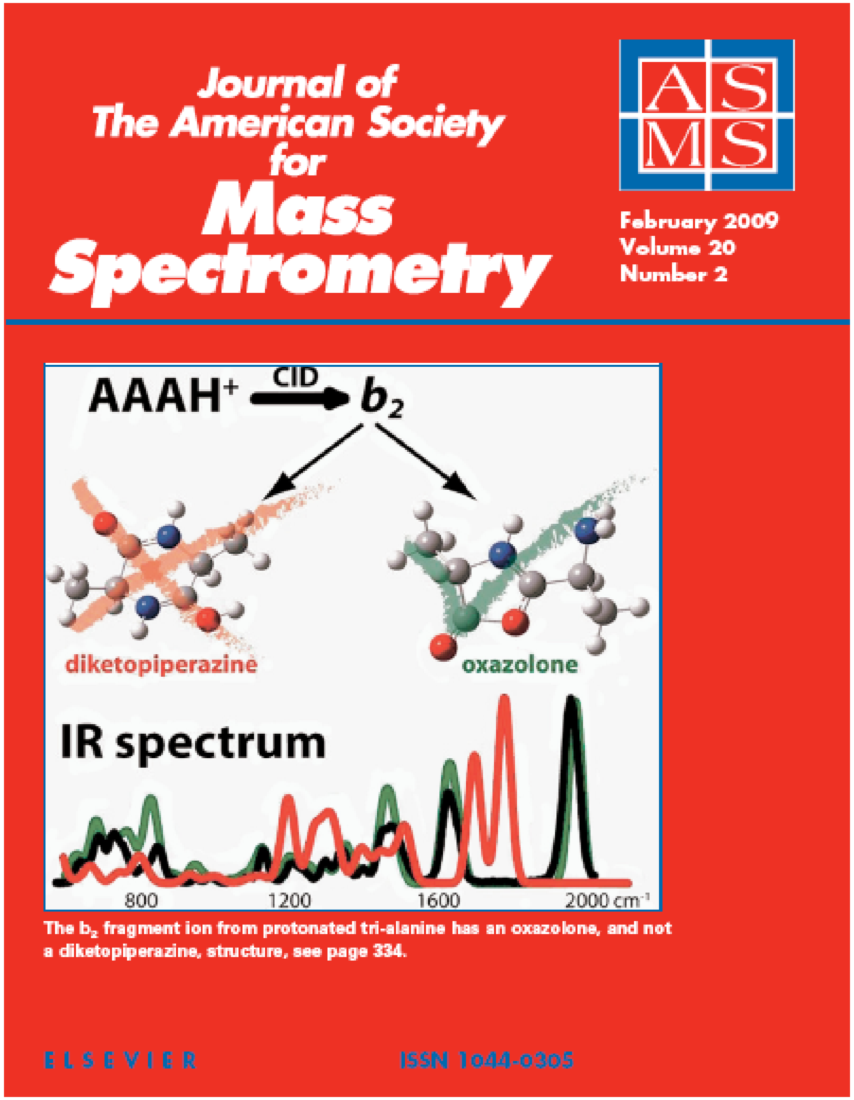
amide bond cleavage

b-fragment: diketopiperazine

y-fragment: truncated peptide

Lower in energy than oxazolone !

Oxazolone fragment structure identified by spectroscopy



b_2 from AAA

Oomens, Young, Molesworth, van Stipdonk, *JASMS* **2009**, *20*, 334

b_2 from AGG

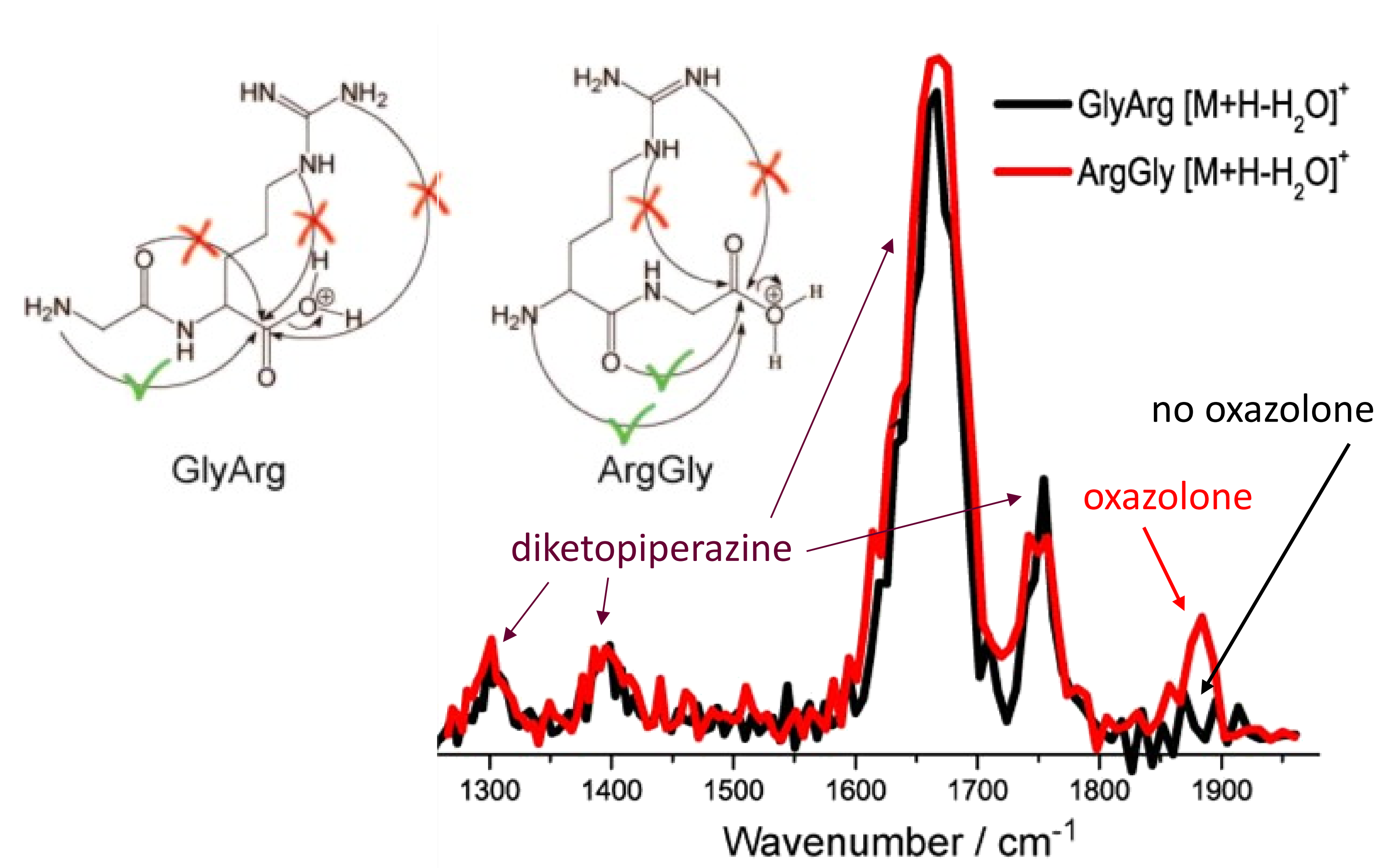
Yoon, Chamot-Rooke, Perkins, Hilderbrand, Poutsma, Wysocki *JACS, 2***009**, *20*, 334

b_2 from GGG

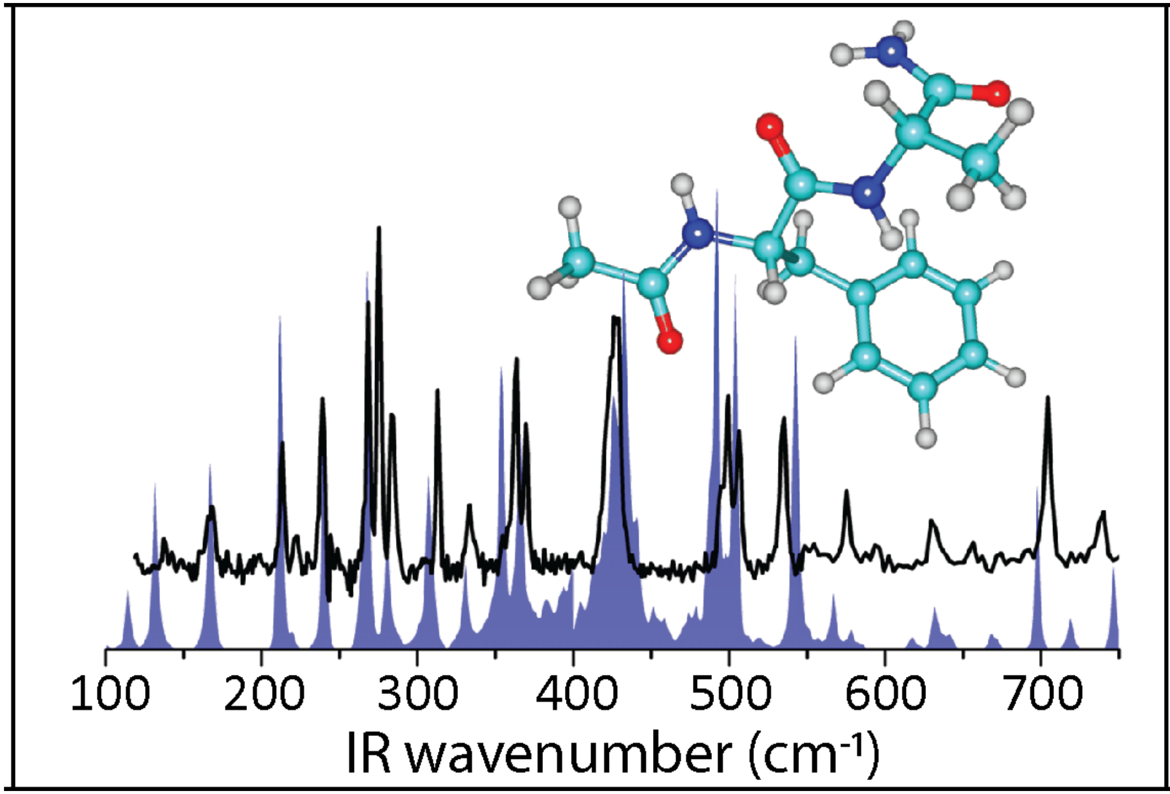
Chen, Steill, Oomens, Polfer, *JACS, 2***009**, *191*, 18272

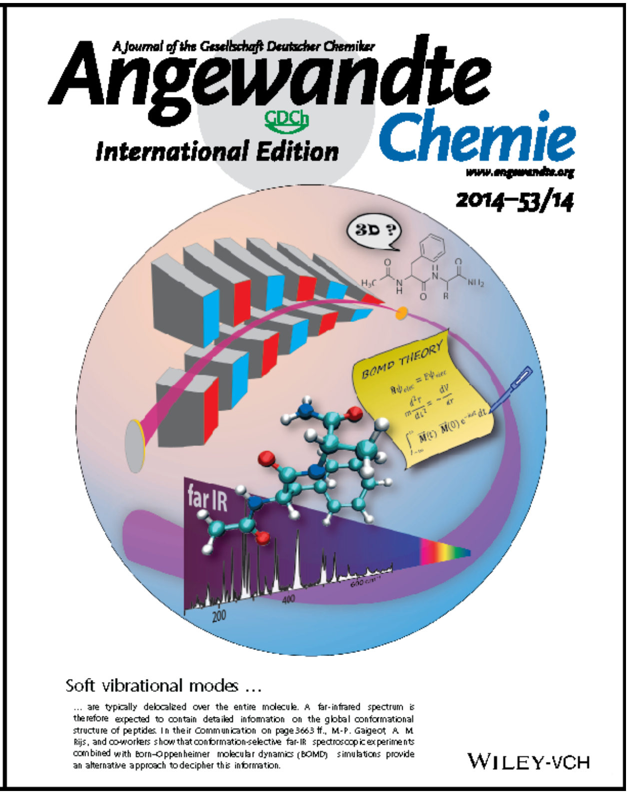
N-terminus as nucleophile: Arg

H_2O-loss from protonated GlyArg and ArgGly



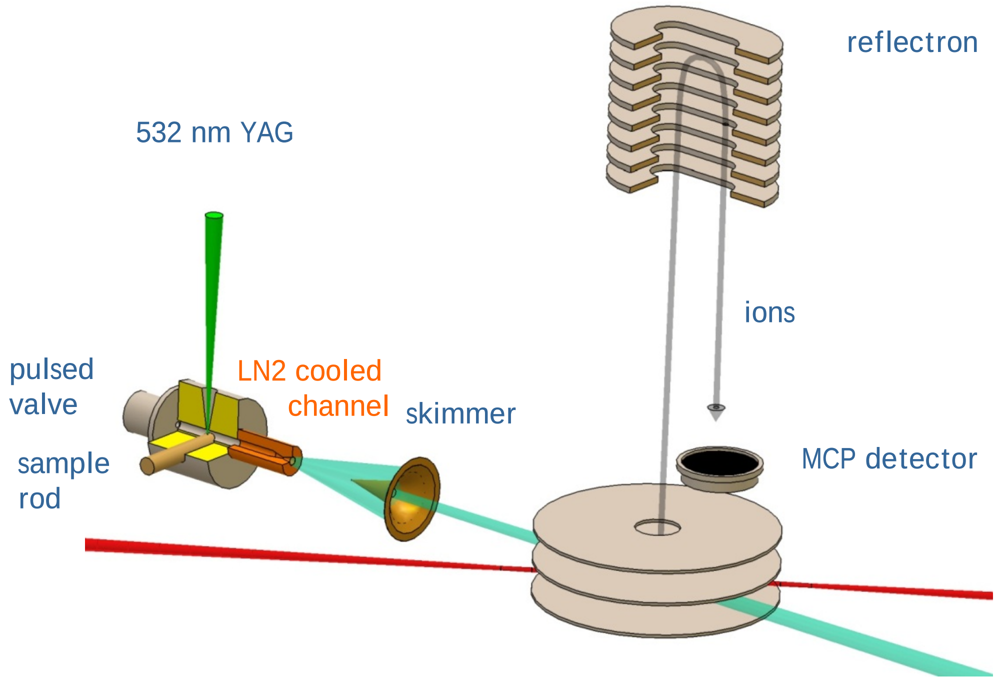
Spectroscopy in the THz range



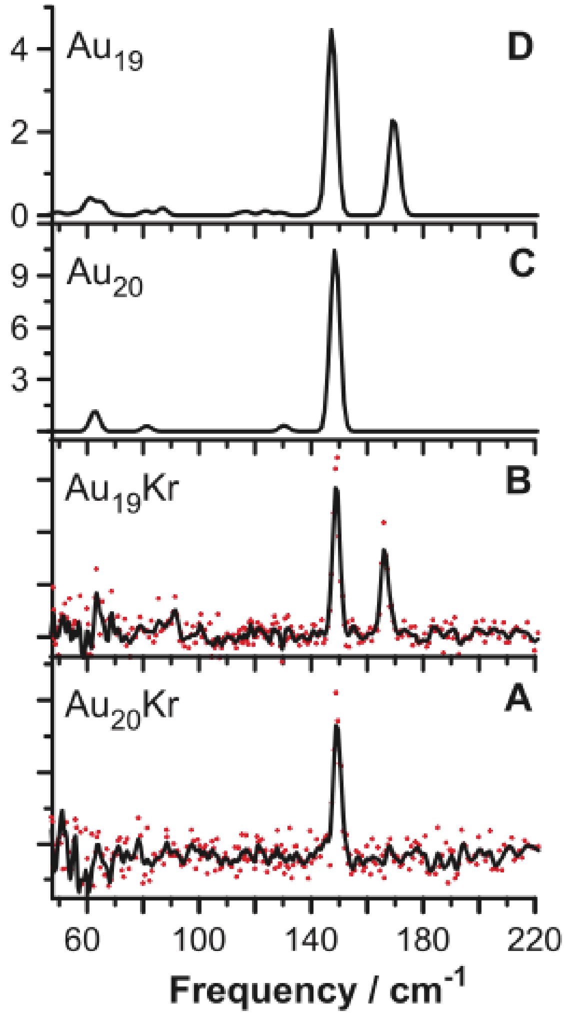


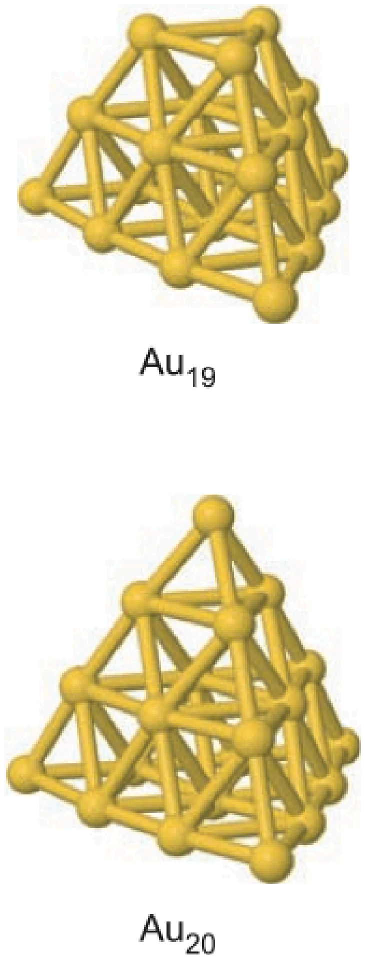
Production of clusters

- Smalley-type cluster source
- Laser vaporization of metal rod in presence of He
- Clusters cooled to ~ 100K
- *Optional: reaction gas added in reaction channel (prior to expansion)*
- Interaction with laser(s)
- Mass spectrometric detection



Catalysis: Structure determination of metal clusters

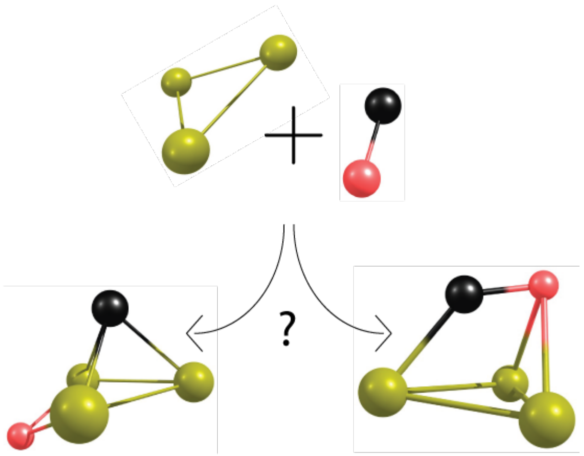


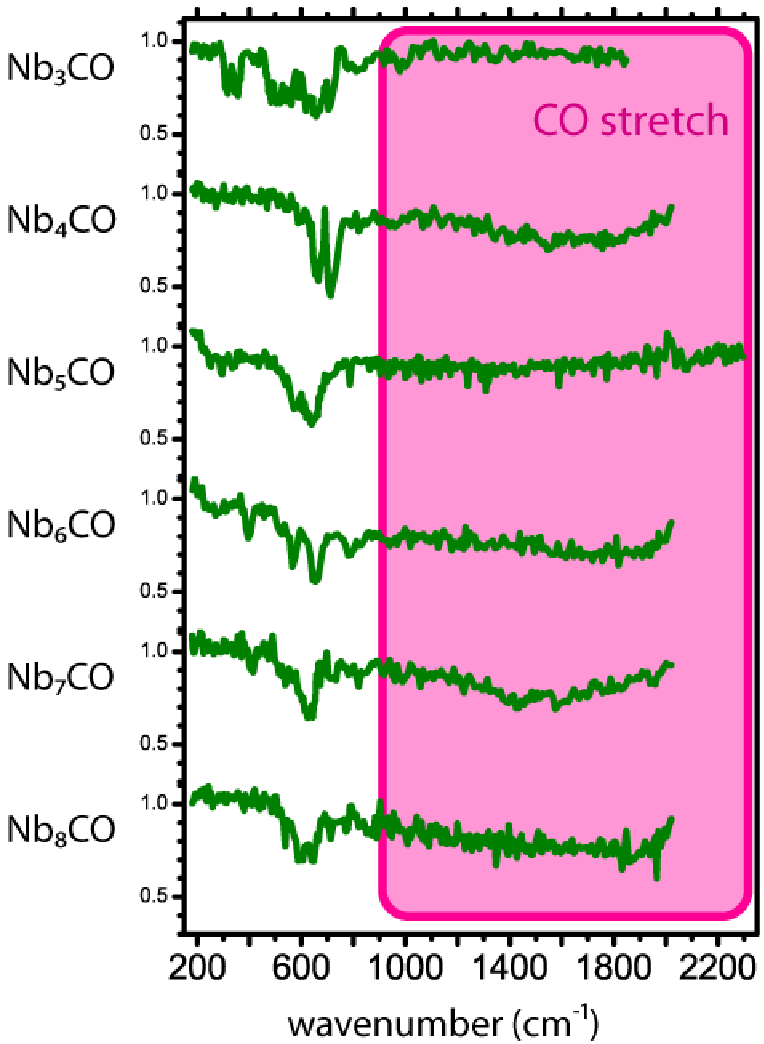


Gruene, *Science* **321**, 674 (2008).

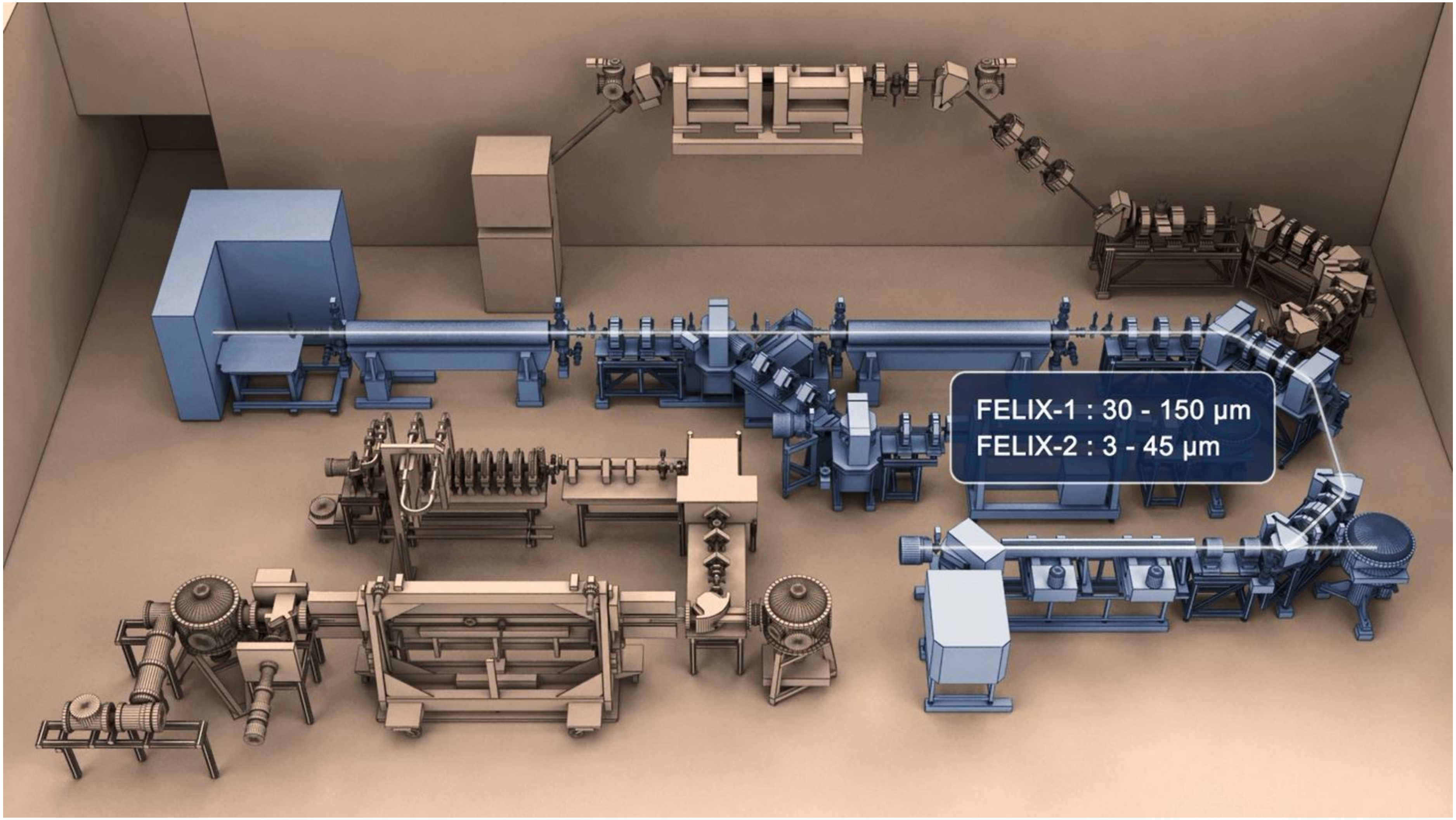
Catalysis: Molecular vs dissociative adsorption

CO adsorption on metal clusters

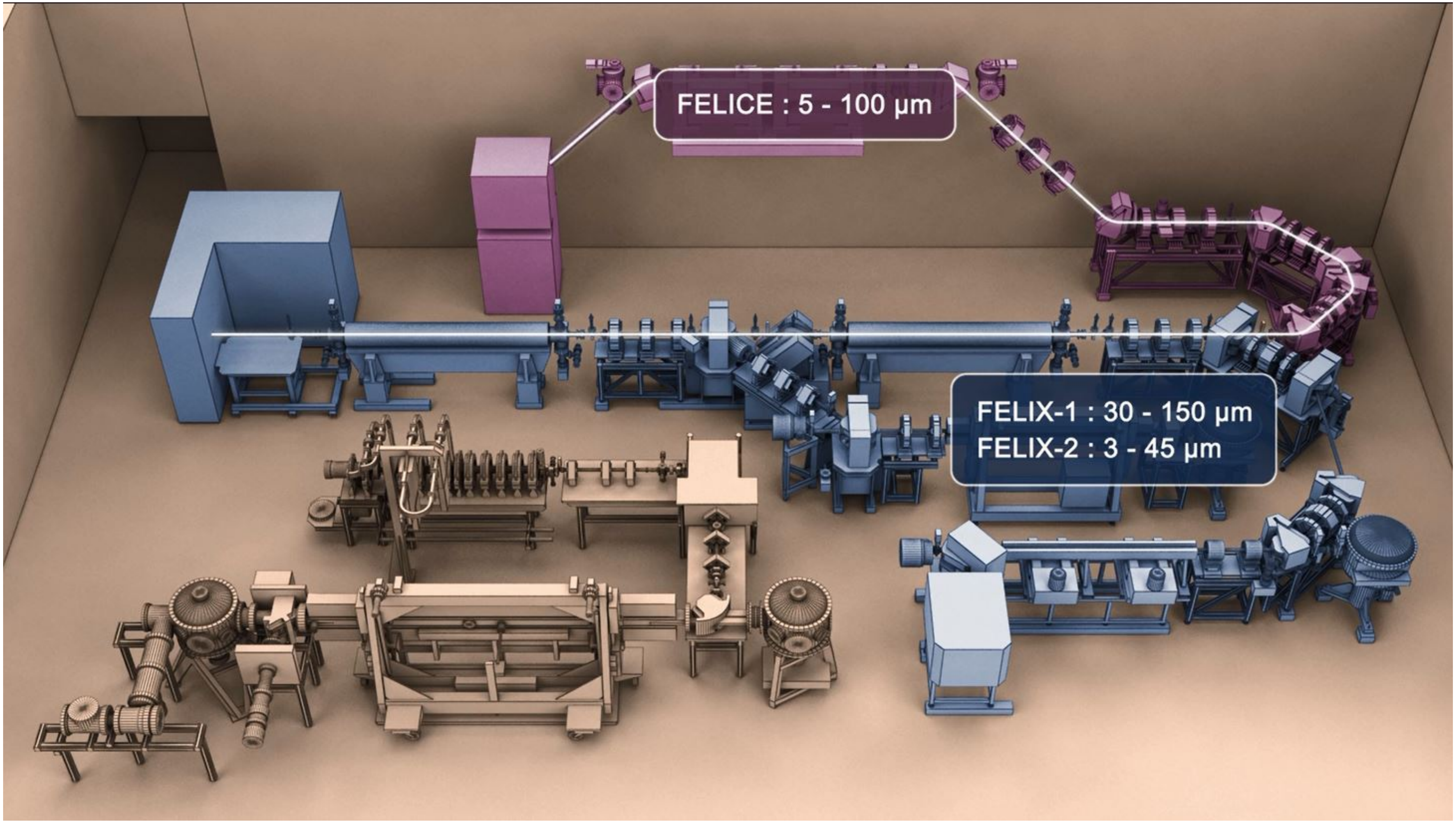




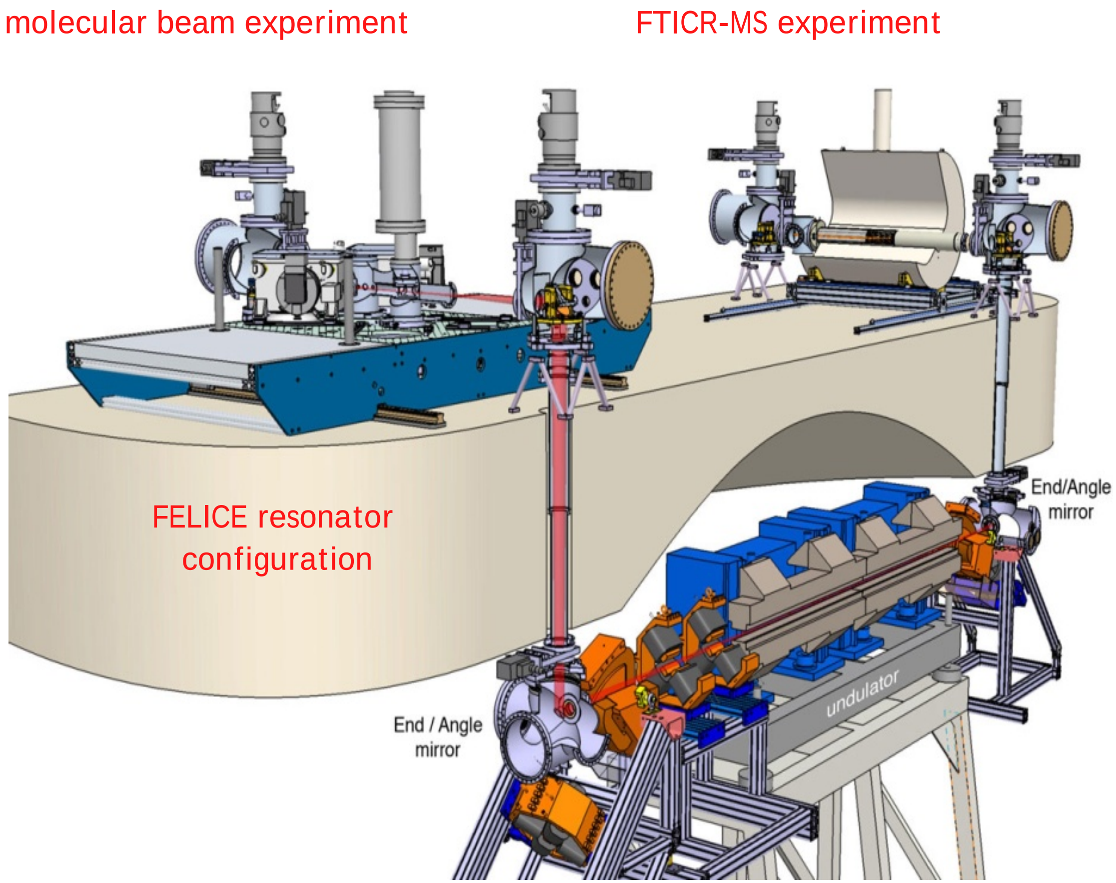
The FELIX Laboratory



The FELIX Laboratory



The FELICE beam line

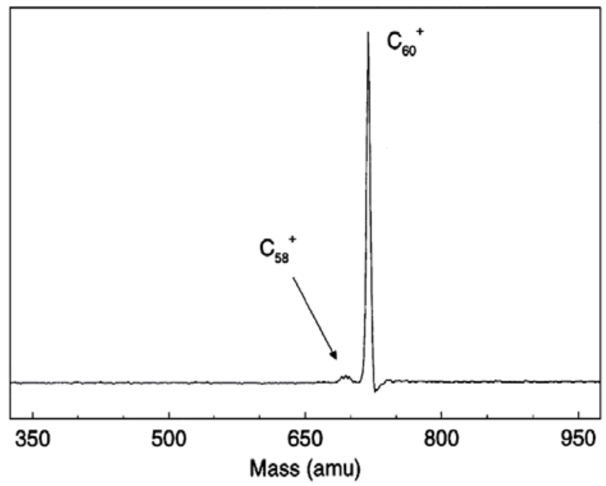


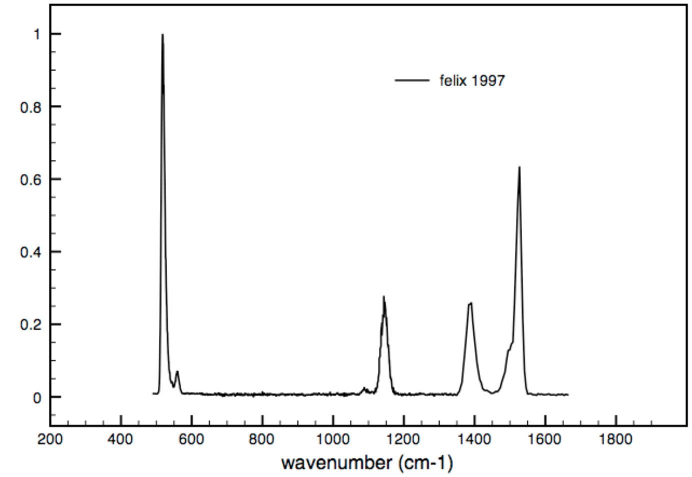
FELIX vs FELICE

mass spectra

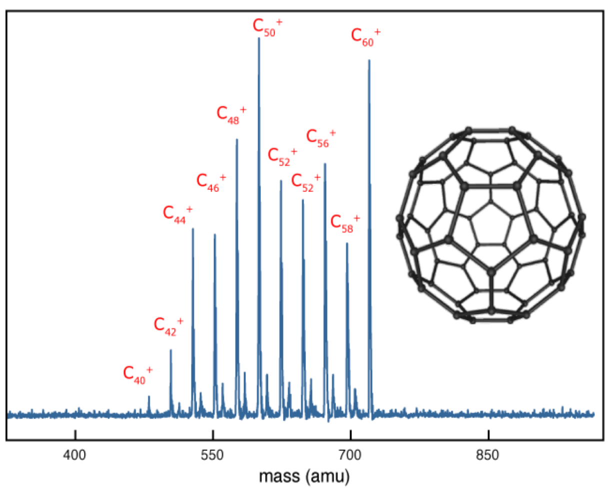
IR spectra

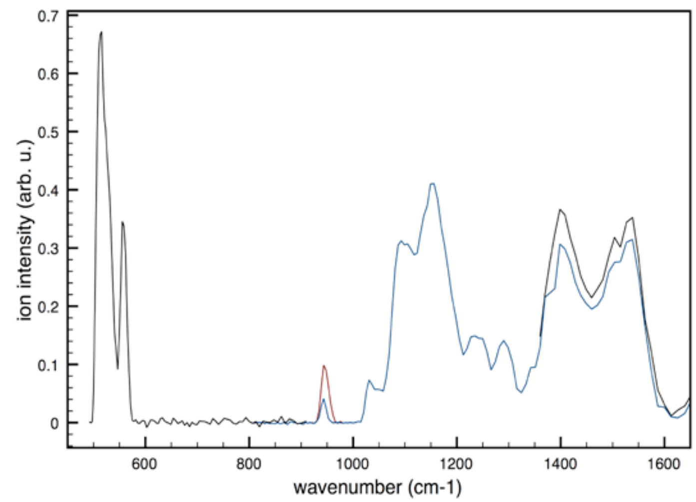
FELIX





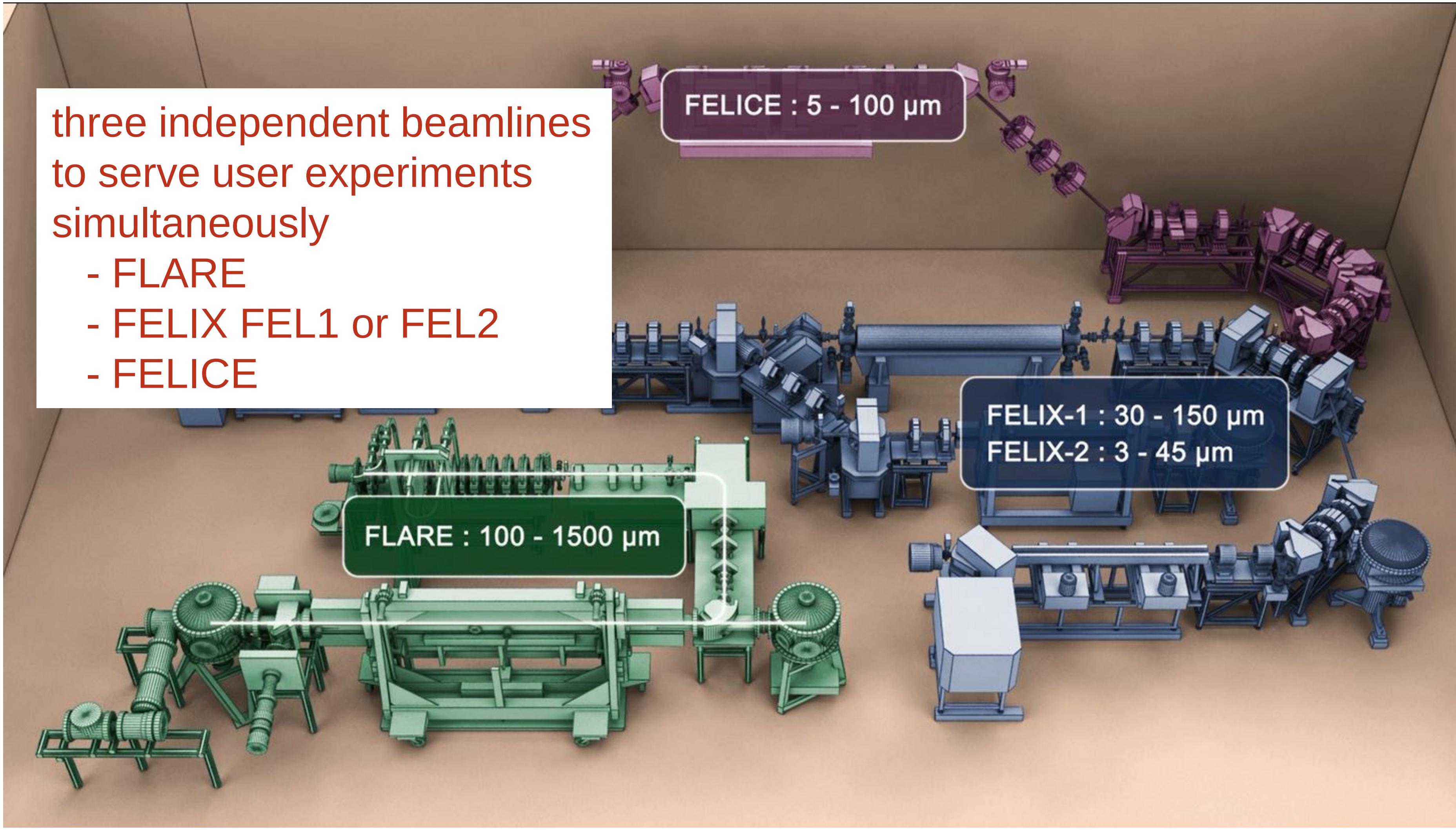
FELICE





The FELIX Laboratory

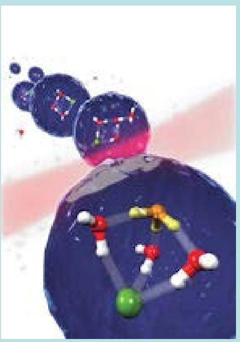
three independent beamlines to serve user experiments simultaneously
- FLARE
- FELIX FEL1 or FEL2
- FELICE



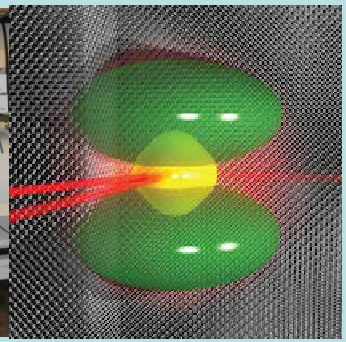
FELIX facility @ Nijmegen: User Laboratories

User laboratory 1 – FLARE & FELIX

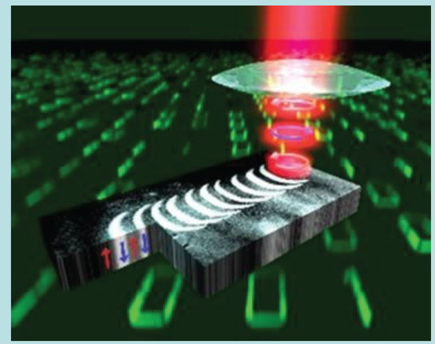
He-droplet machine
Havenith (Bochum)



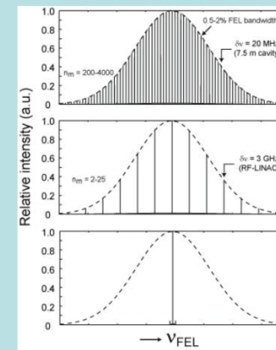
Dilution Refrigerator
EPSRC, Aeppli, Murdin



Ultrafast laser system
Kimel & Rasing (RU)



FLARE Diagnostic
Station

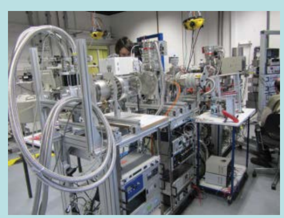


muli-purpose
station with
optical table

To be defined

(currently construction of
FLARE beam line)

Cold 22-pole ion trap
Schlemmer (Cologne)



Molecular beam
apparatus

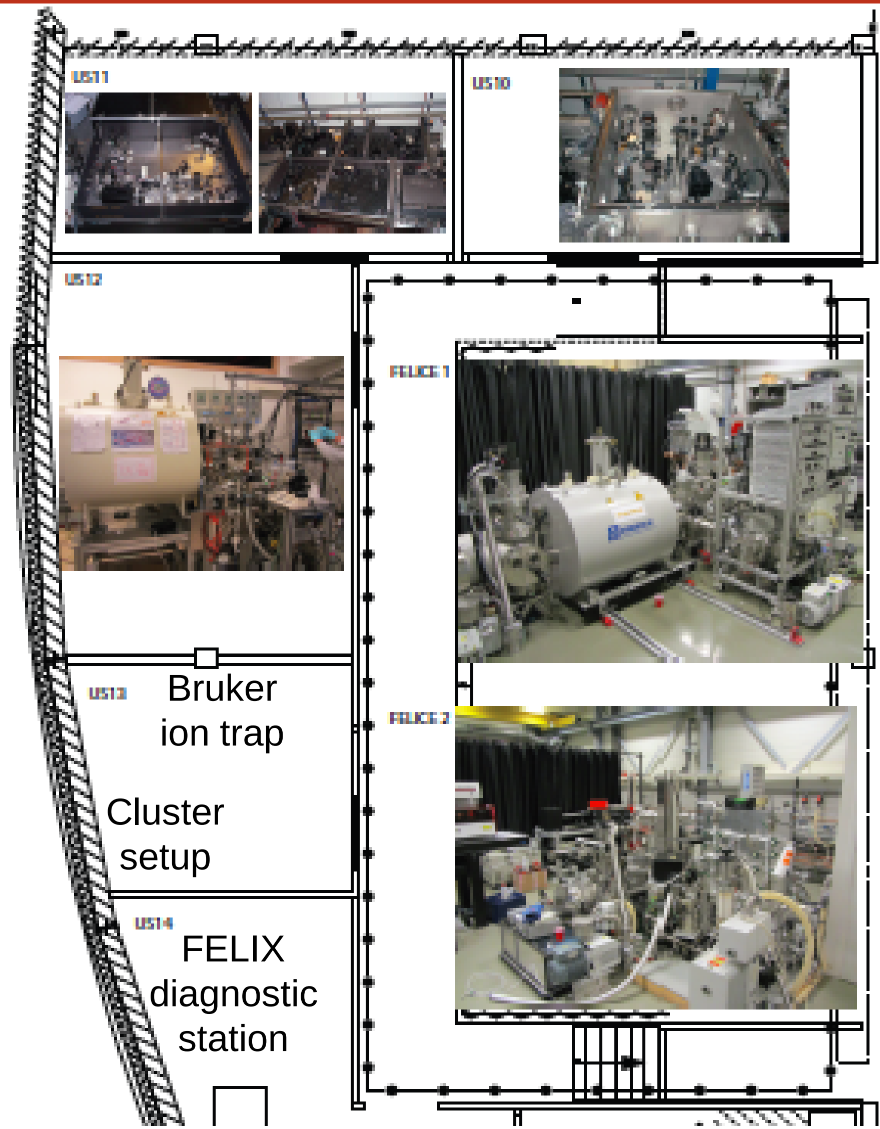
Paul type Ion trap

FELIX facility @ Nijmegen: User Laboratories

Ultrafast laser systems

Non-linear optics laboratory

Versatile FTICR mass spectrometer



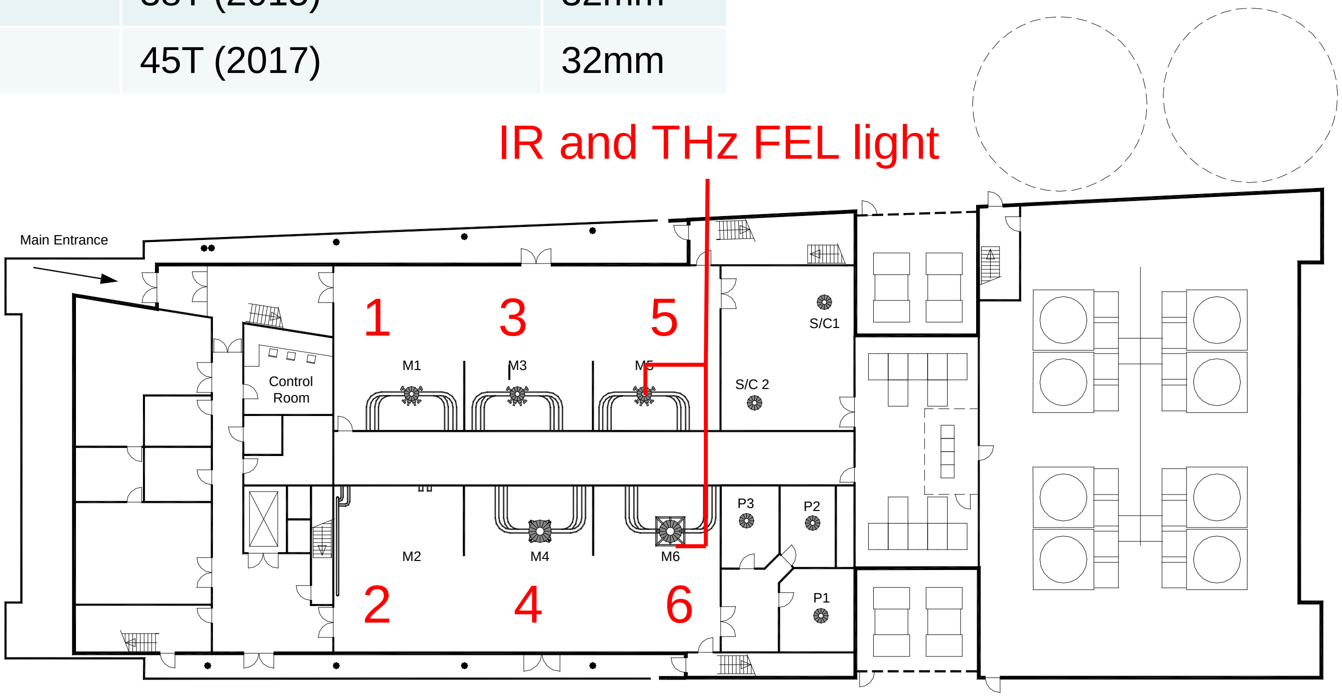
FELICE
FTICR mass spectrometer

FELICE
cluster apparatus

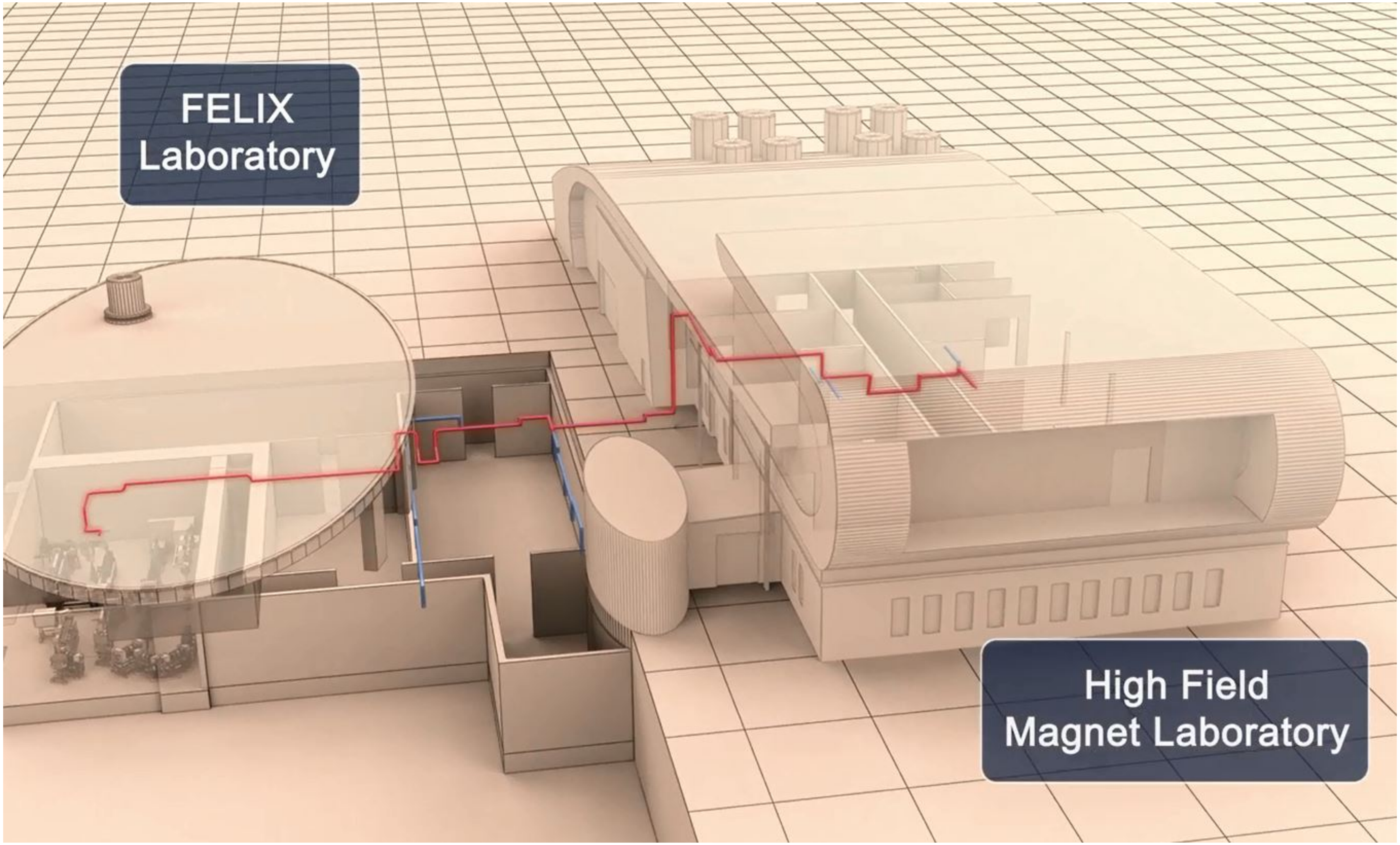
User laboratory 2 - FELIX & FELICE

Our neighbour: HFML

Site	Magnet	Planned	Bore
1	32T (Optics above)		50mm
2	38T		32mm
3	33T (FIR above)	38T (2015)	32mm
4	30T (hybrid, 10MW)		50mm
5	33T	38T (2015)	32mm
6		45T (2017)	32mm



Combination of THz radiation and Magnetic Field

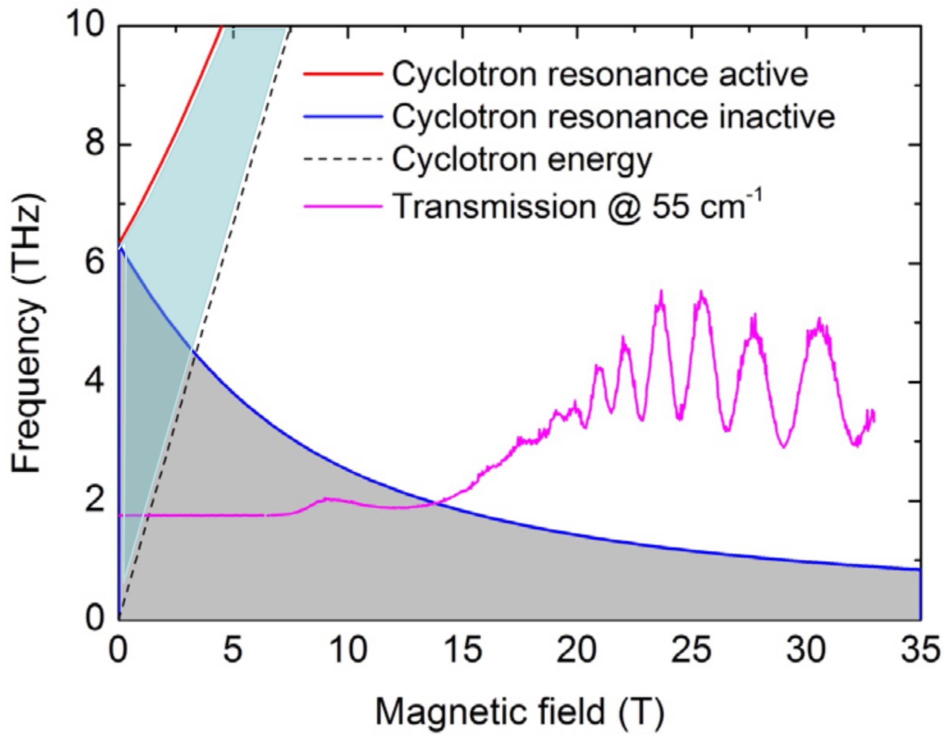


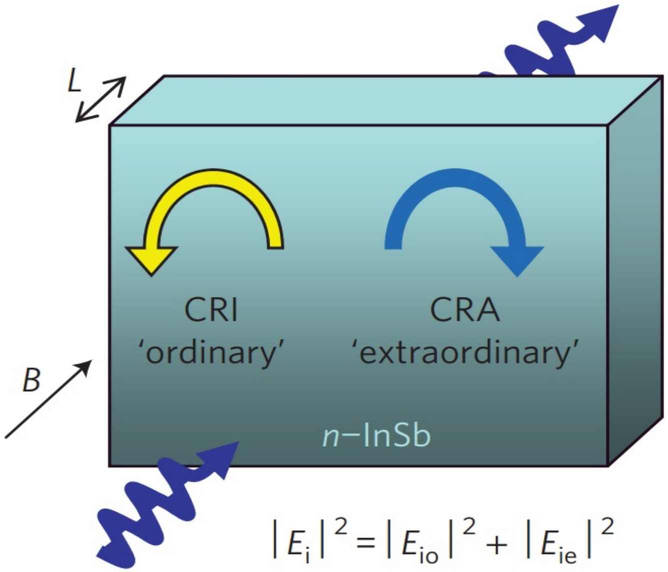
Magneto-plasma oscillations: n-type InSb

Te-doped n-InSb

$\omega_p = 6.33$ THz
$m_{eff} = 0.021\ m_0$
$n = 1.66 \times 10^{17}\ cm^{-3}$

$$\frac{1}{4\pi}\left(\sqrt{\omega_c^2 + 4\omega_p^2} \pm \omega_c\right)$$

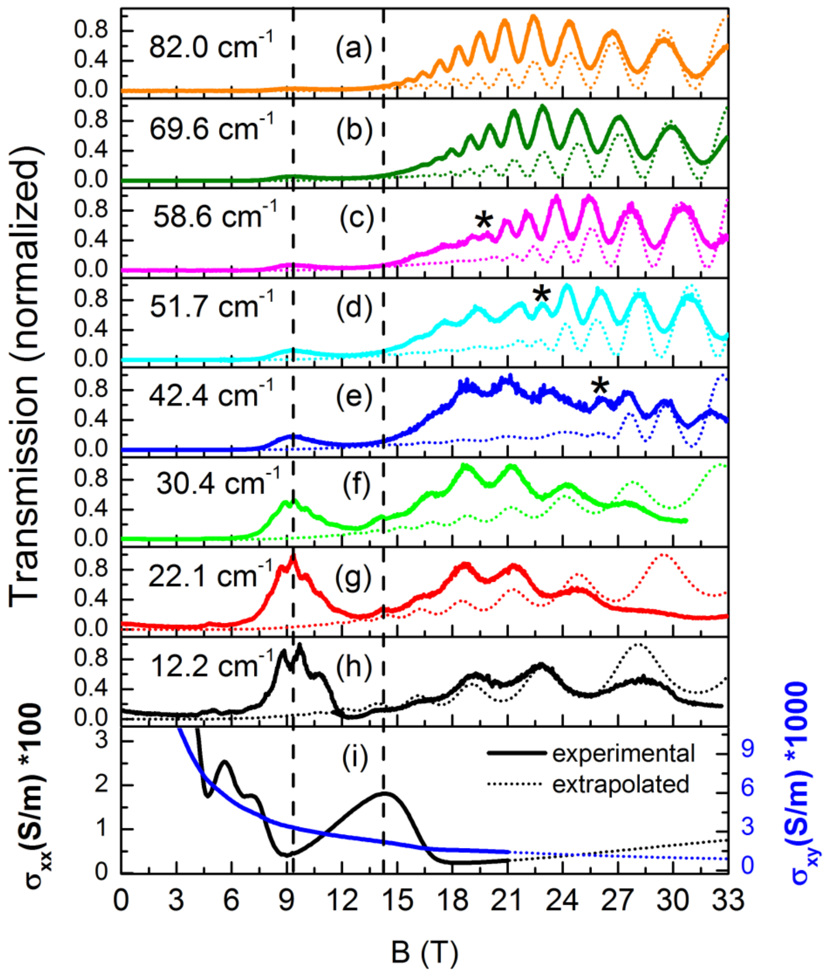




$$|E_i|^2 = |E_{io}|^2 + |E_{ie}|^2$$

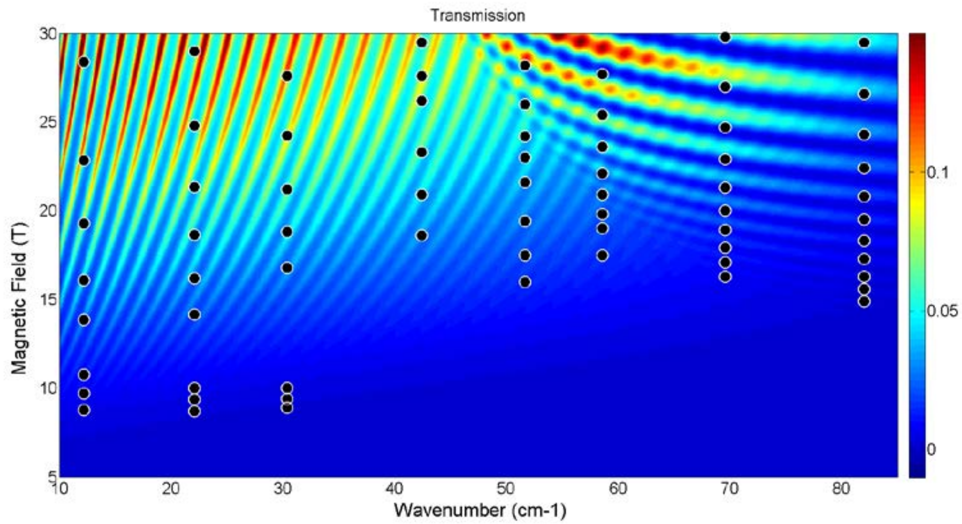
Magneto-plasma oscillations: n-type InSb

Experiment



Calculations

$$\tilde{n}_{\pm}^{2} = \tilde{\varepsilon}_{xx} \pm i\tilde{\varepsilon}_{xy} = \kappa - \frac{\omega_p^2}{\omega(\omega \pm \omega_c + i\nu)} + \tilde{\varepsilon}_{ph}$$



Thank you for your attention