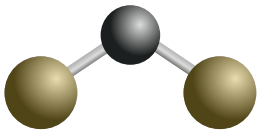
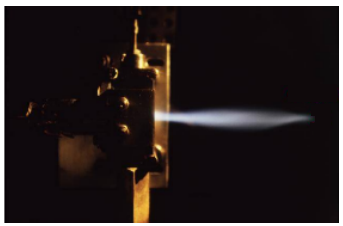
Vibrational Spectroscopy of Small Silicon-Carbides

DISILICONCARBIDE

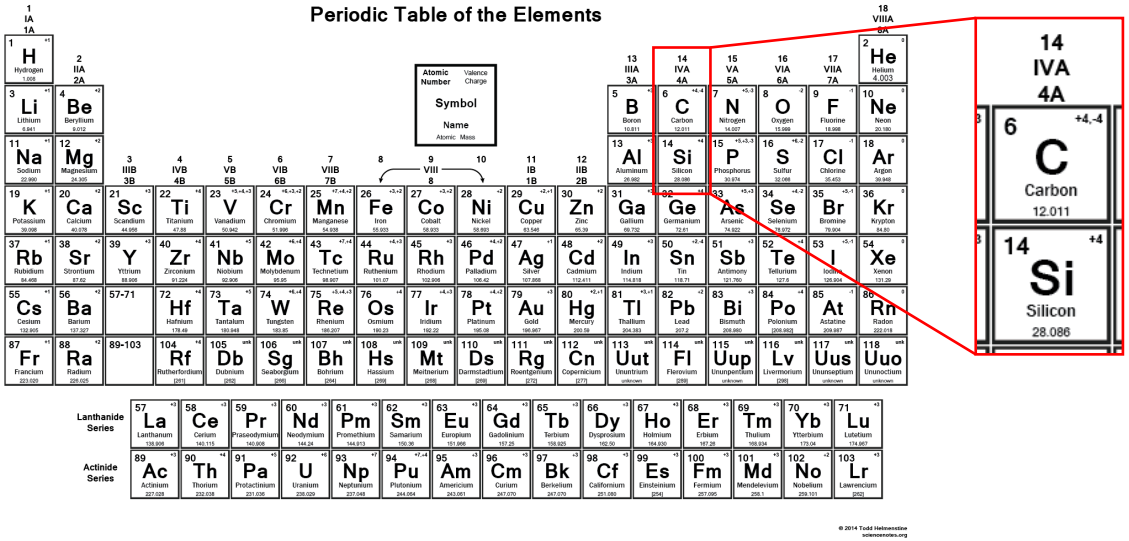
Daniel Witsch
University of Kassel
Laboratory Astrophysics

Outline





Small Silicon-Carbon Molecules



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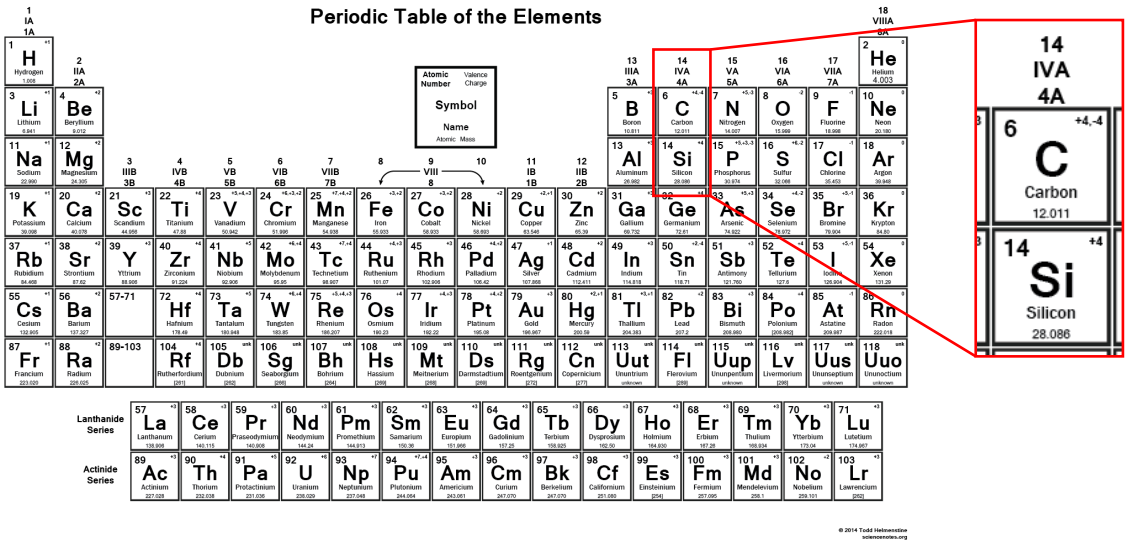

Bonding properties:

- C: single or multiple bonds
- Si: multidirectional bonds

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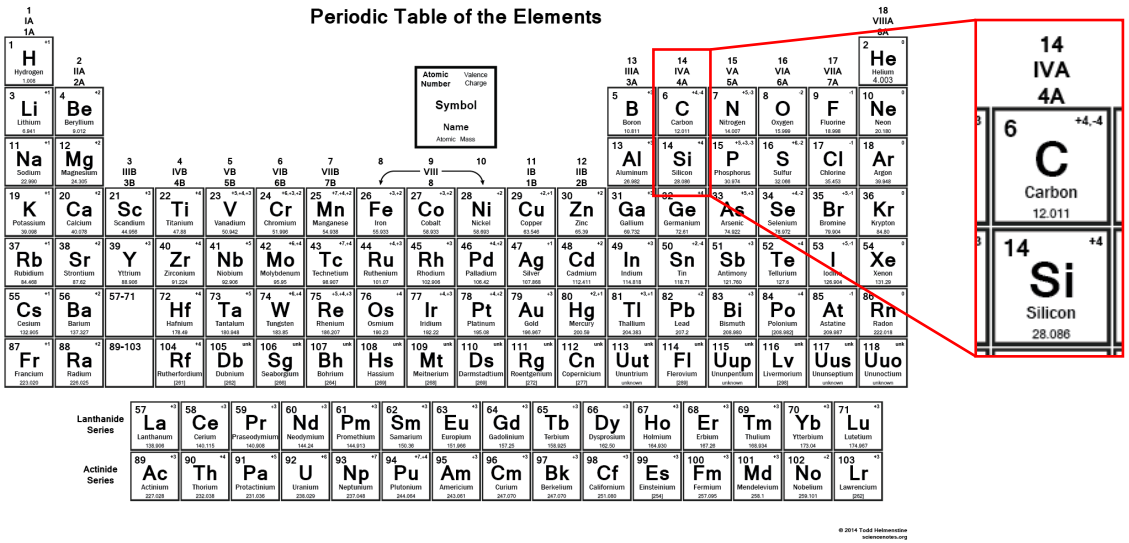

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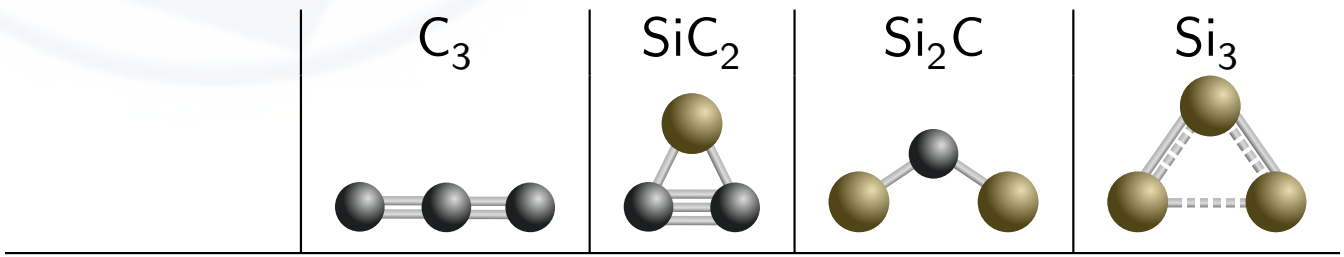

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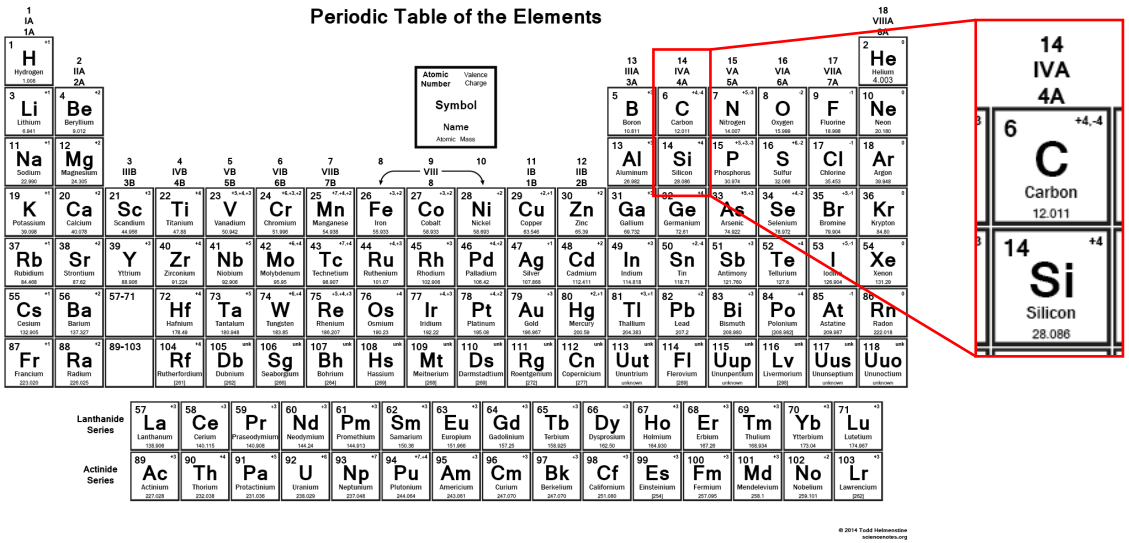

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	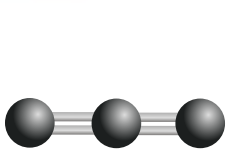	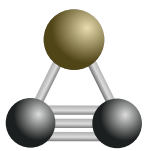	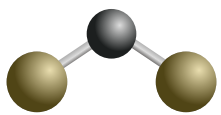	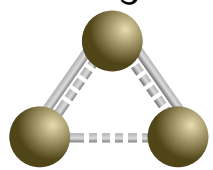
Rotation	-	✓	✓	✓

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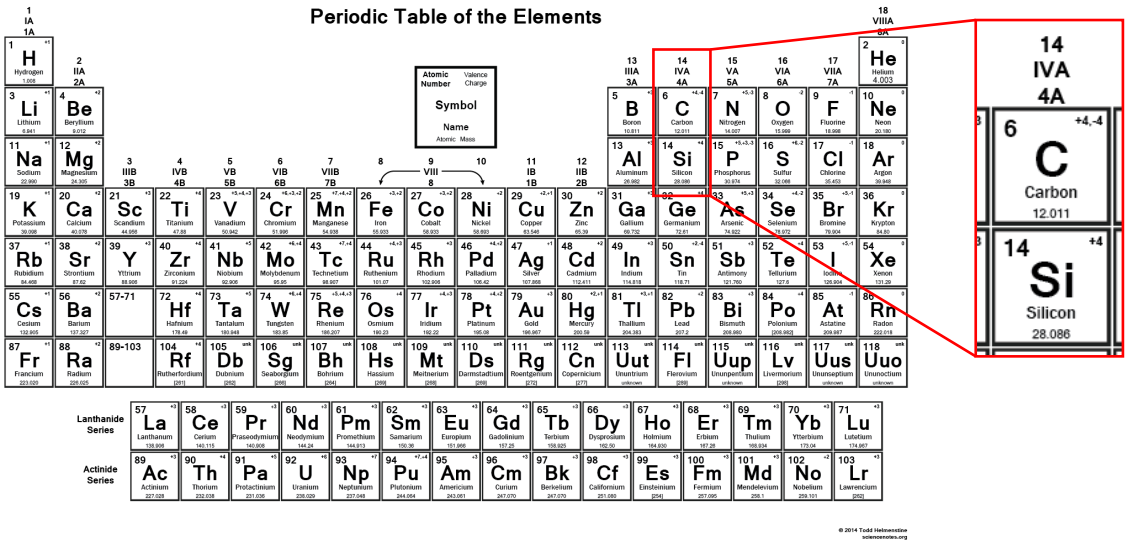

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	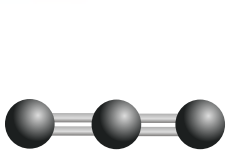	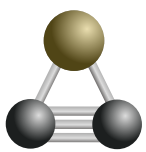	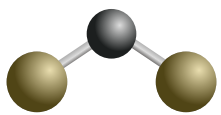	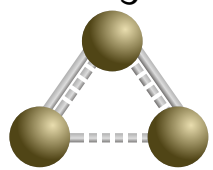
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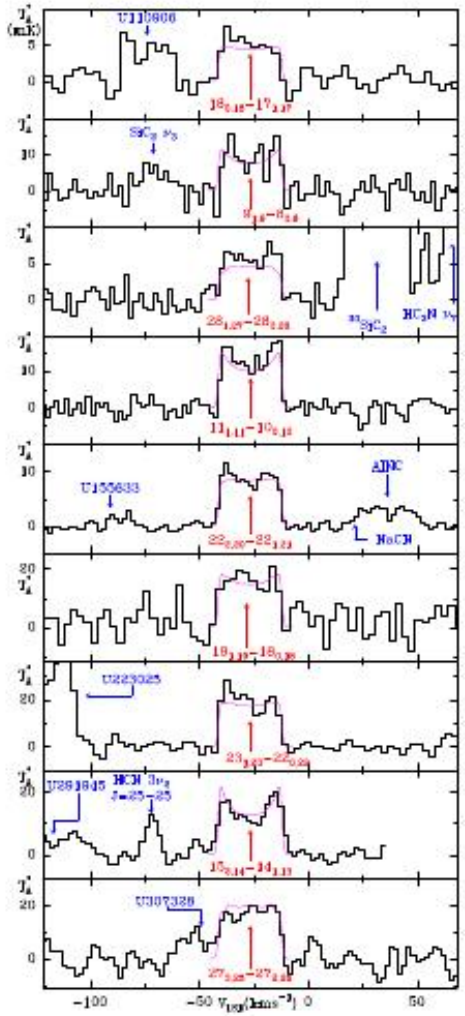
Vibration	✓	✓	✗	✗

Disiliconcarbide (Si_2C)

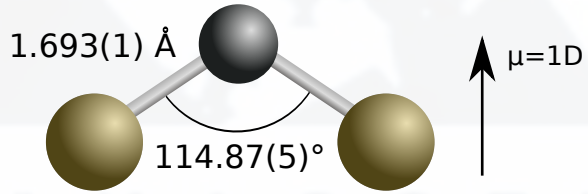


Detection of Si_2C towards IRC+10216

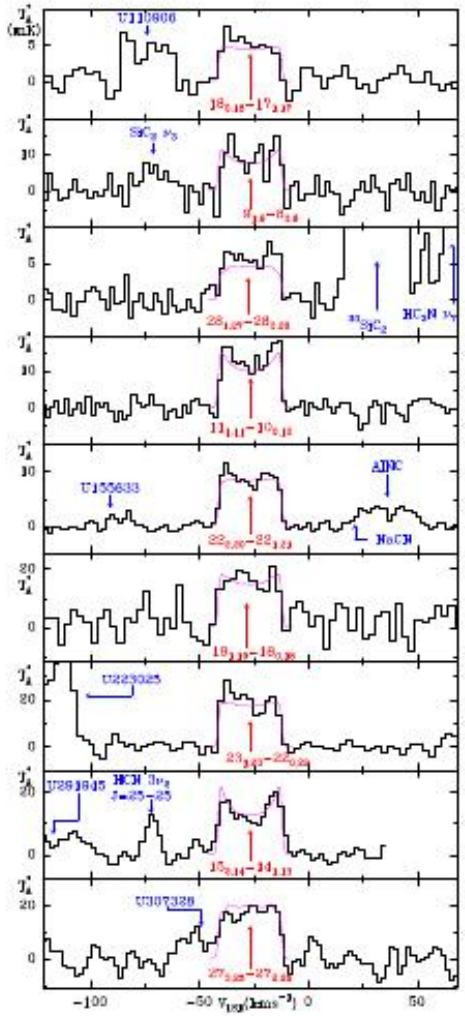
J. Cernicharo *et. al.*, Ap.J.Lett. L3 2015

Disiliconcarbide (Si_2C)

Vibrational Groundstate Structure



M.C. McCarthy *et. al.*, J.Phys.Chem.Lett 6 (2015), 2107



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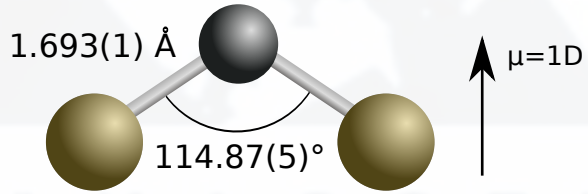

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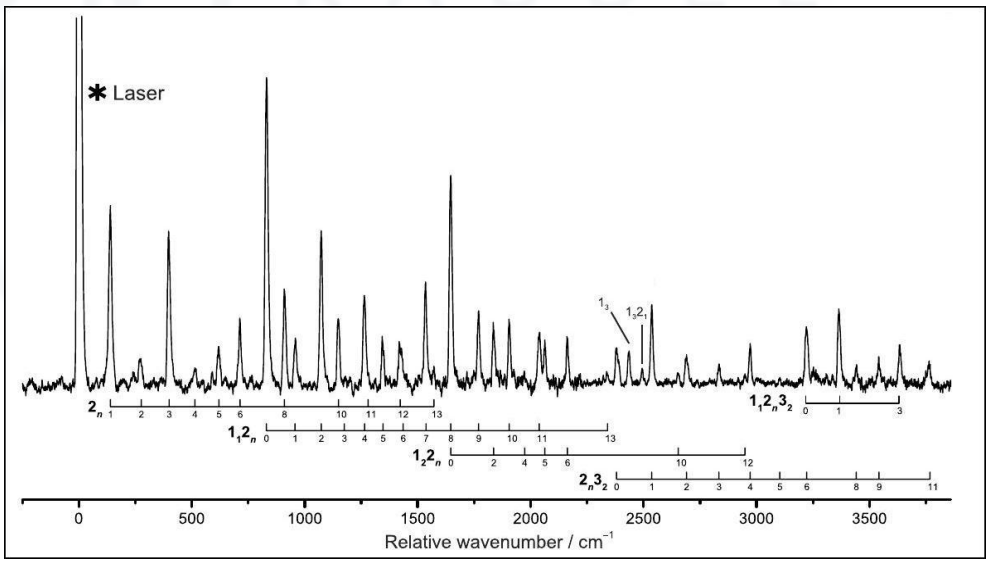
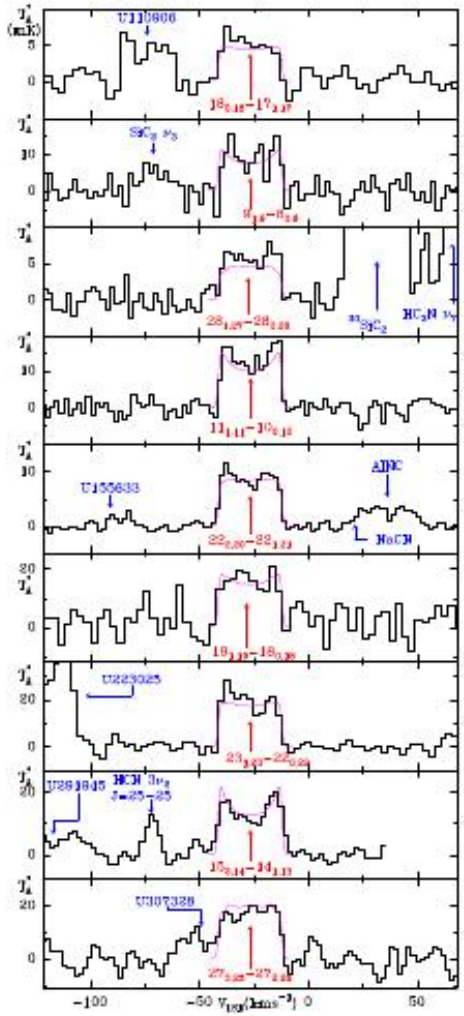


M.C. McCarthy *et. al.*, J.Phys.Chem. 142 (2015), 231101



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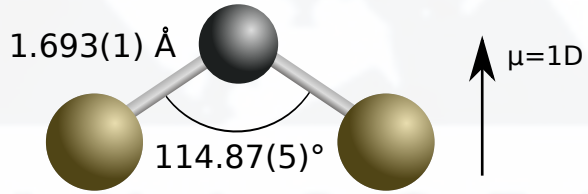

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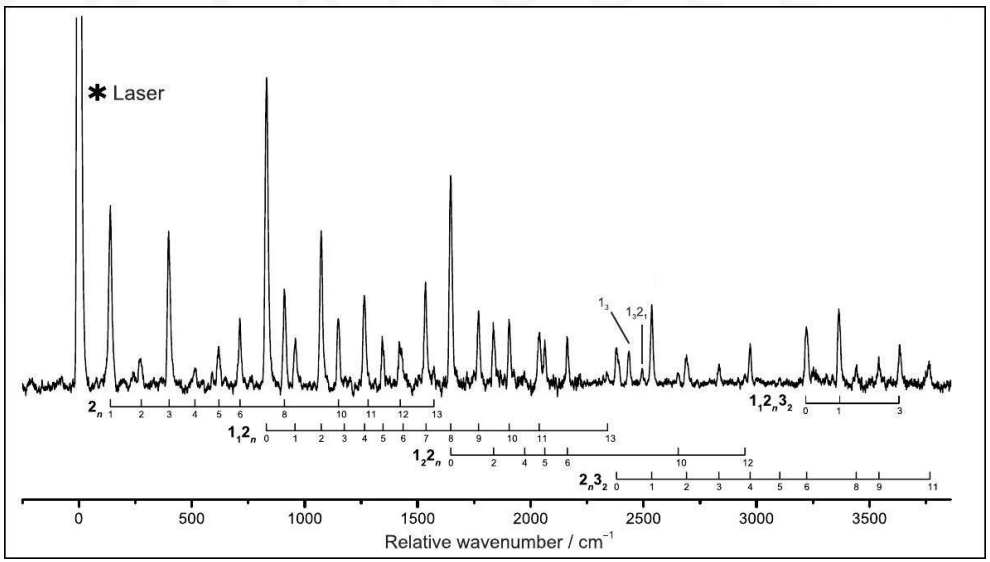

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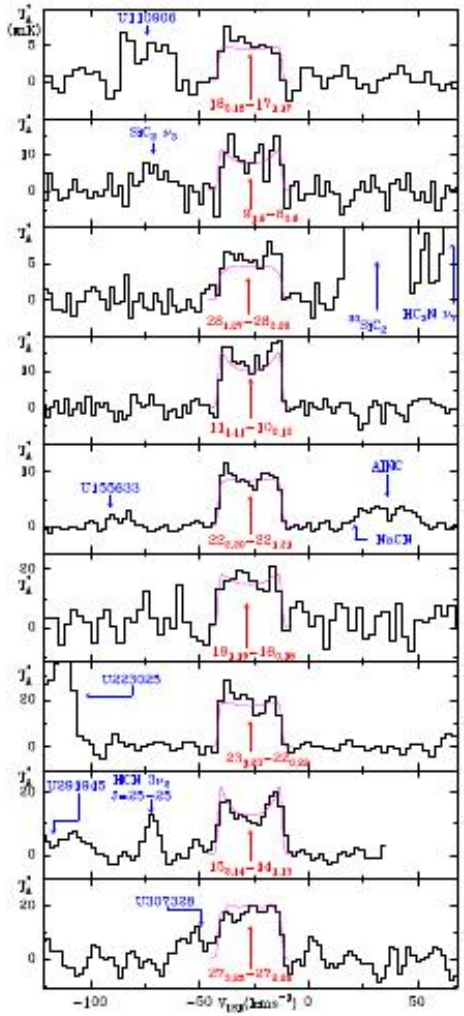
	ν_1	ν_2	ν_3
Frequency $[\mathrm{cm}^{-1}]$	143	831	1198
Calculated[b] $[\mathrm{cm}^{-1}]$	142	838	1207
Intensity[b] $[\mathrm{km/mol}]$	1.0	16.5	198.2

[b] from CCSD(T)/cc-pwCVQZ calculations (S. Thorwirth)



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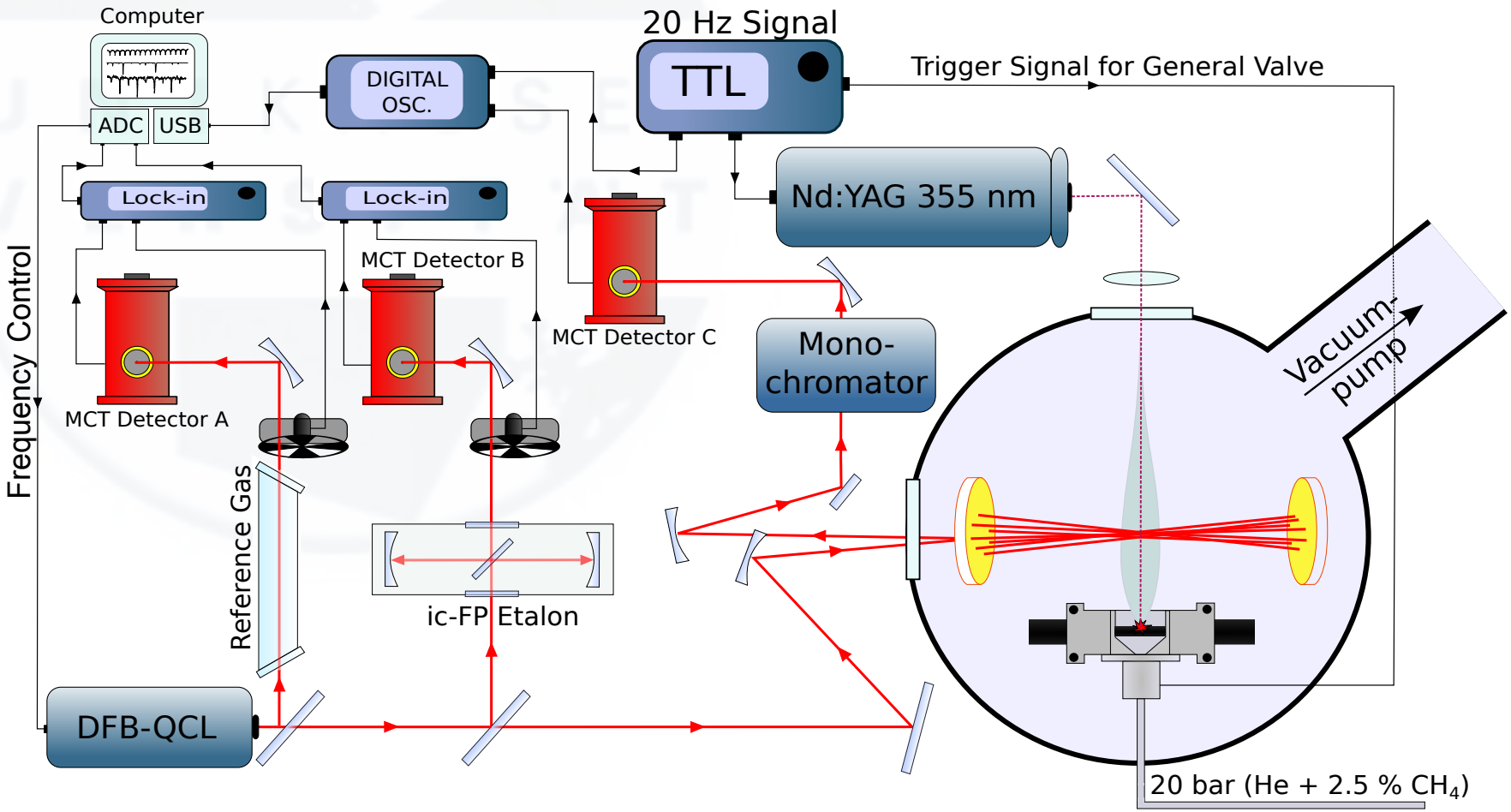
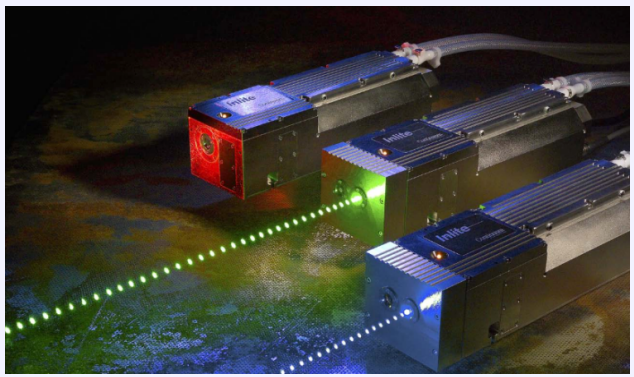
The Apparatus



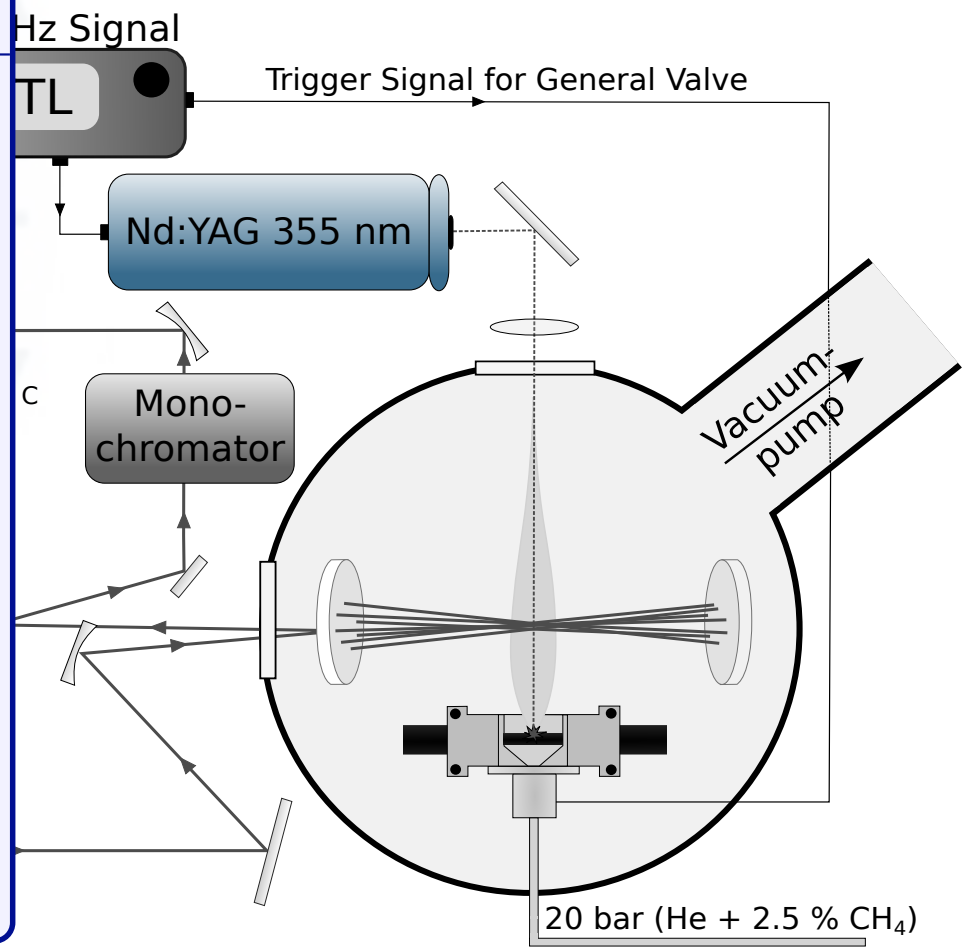
The Apparatus

Pulsed Nd:YAG



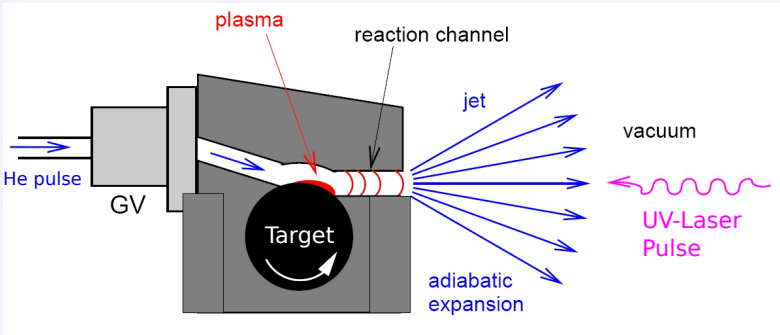
www.continuumlasers.com

Details	
Wavelength	355 nm
Pulse energy	40 mJ
Repetition rate	20 Hz
Pulse length	7 - 9 ns



The Apparatus

Cluster Source



Details	
Rot. Temperature	$\sim 30\,\mathrm{K}$
Target Material	Si
Buffergas	He+2.5 %CH_4
Backing Pressure	$20\,\mathrm{bar}$

The Apparatus

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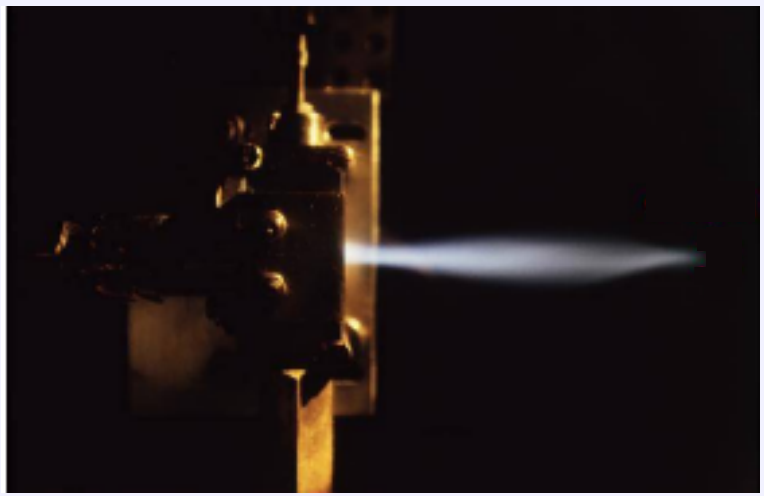

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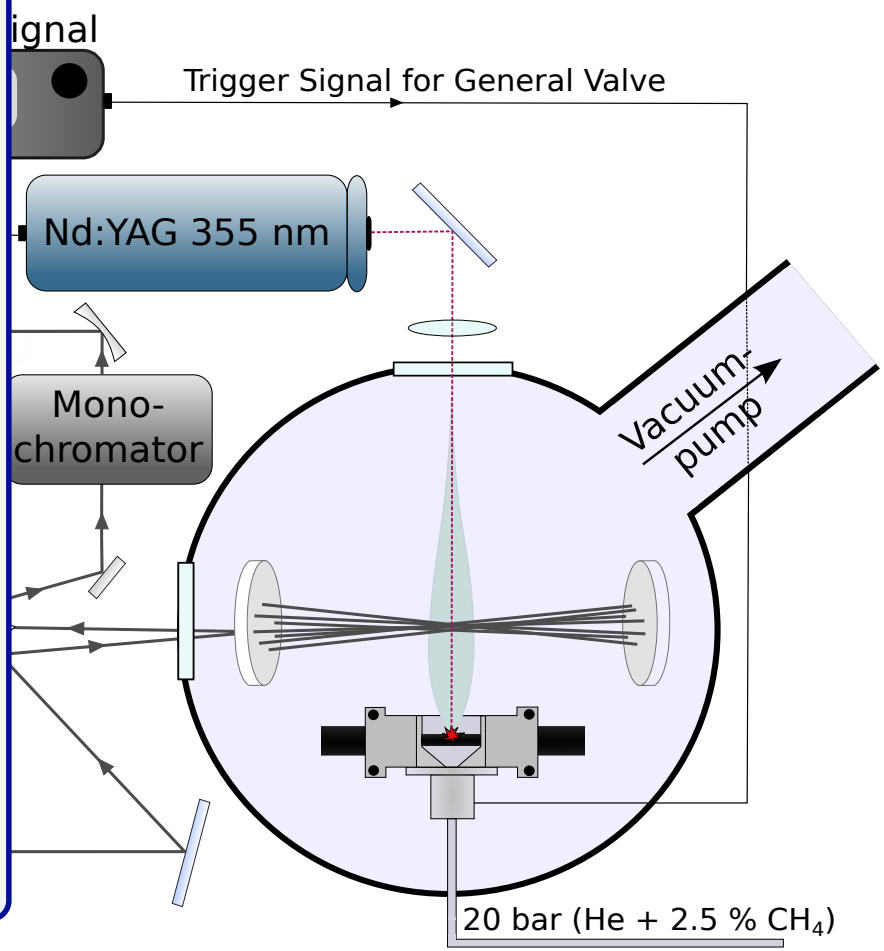
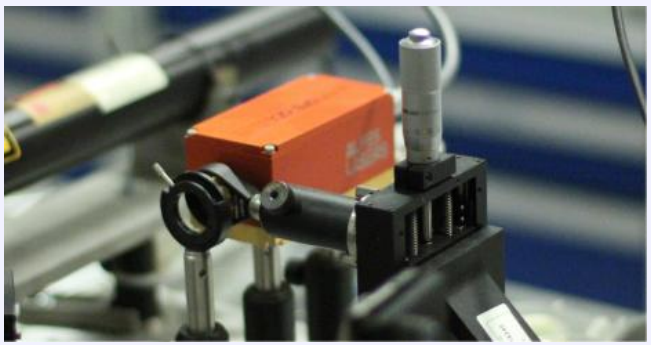


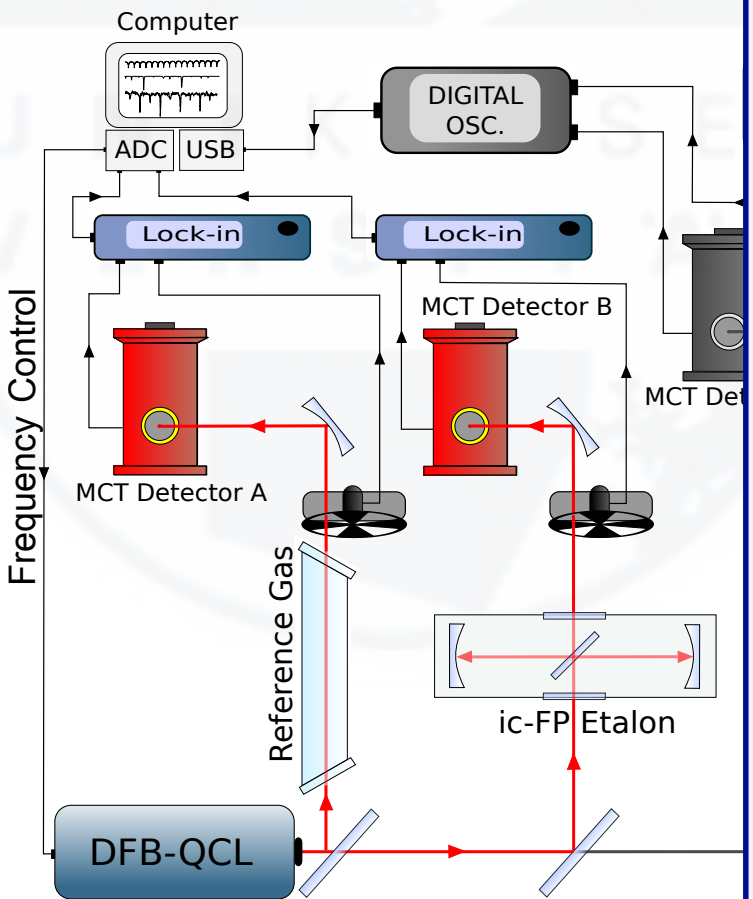
The Apparatus

Quantum Cascade Laser



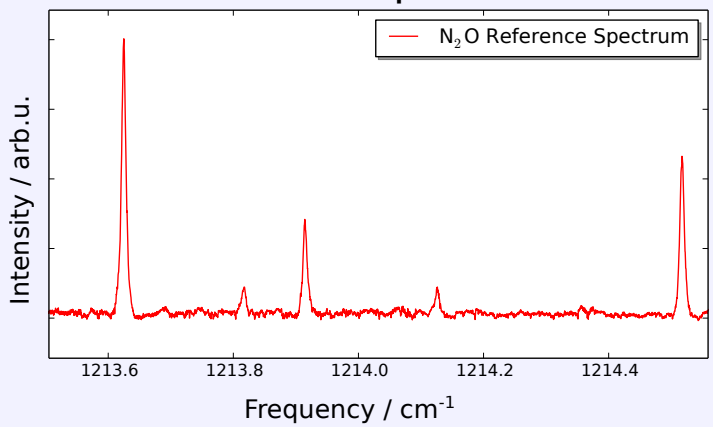
Details	
Frequency	1208 - 1217 cm^{-1}
Power (cw)	up to 20 mW
Herriott-type multi-pass optics with up to 50 passes	

The Apparatus

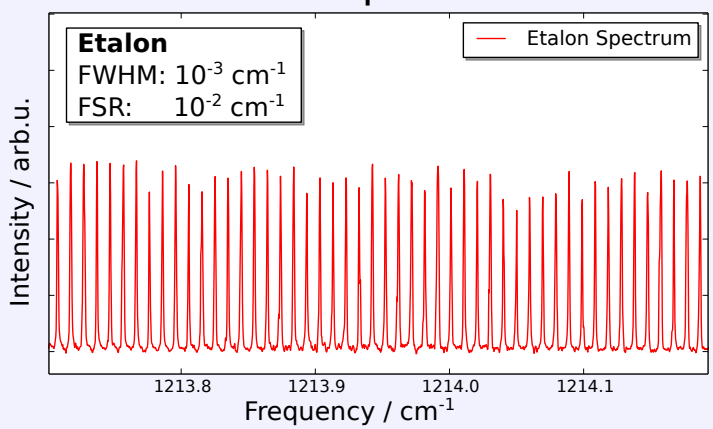


Frequency Calibration

Reference Spectrum

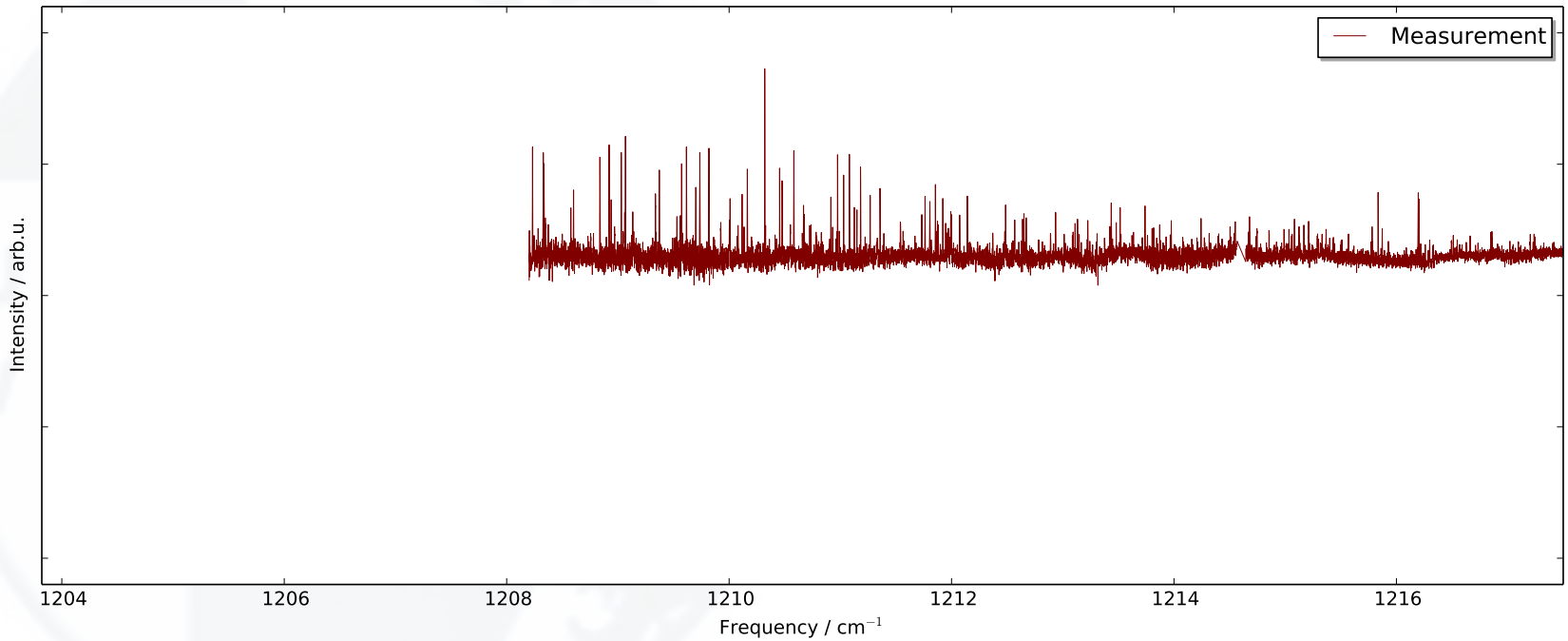


Etalon Spectrum



Calibration Accuracy: $\Delta\nu \approx 10^{-4}\,\mathrm{cm}^{-1}$

Rovibrational Spectrum of a Silicon-Carbide



- 120 absoprtion features
- Calculated band origin[b]
  $\nu_3 = 1207.34\,\mathrm{cm}^{-1}$
- D's, H's taken from J. Chernicharo *et. al.*
- Assignment needed

Rovibrational Spectrum of a Silicon-Carbide

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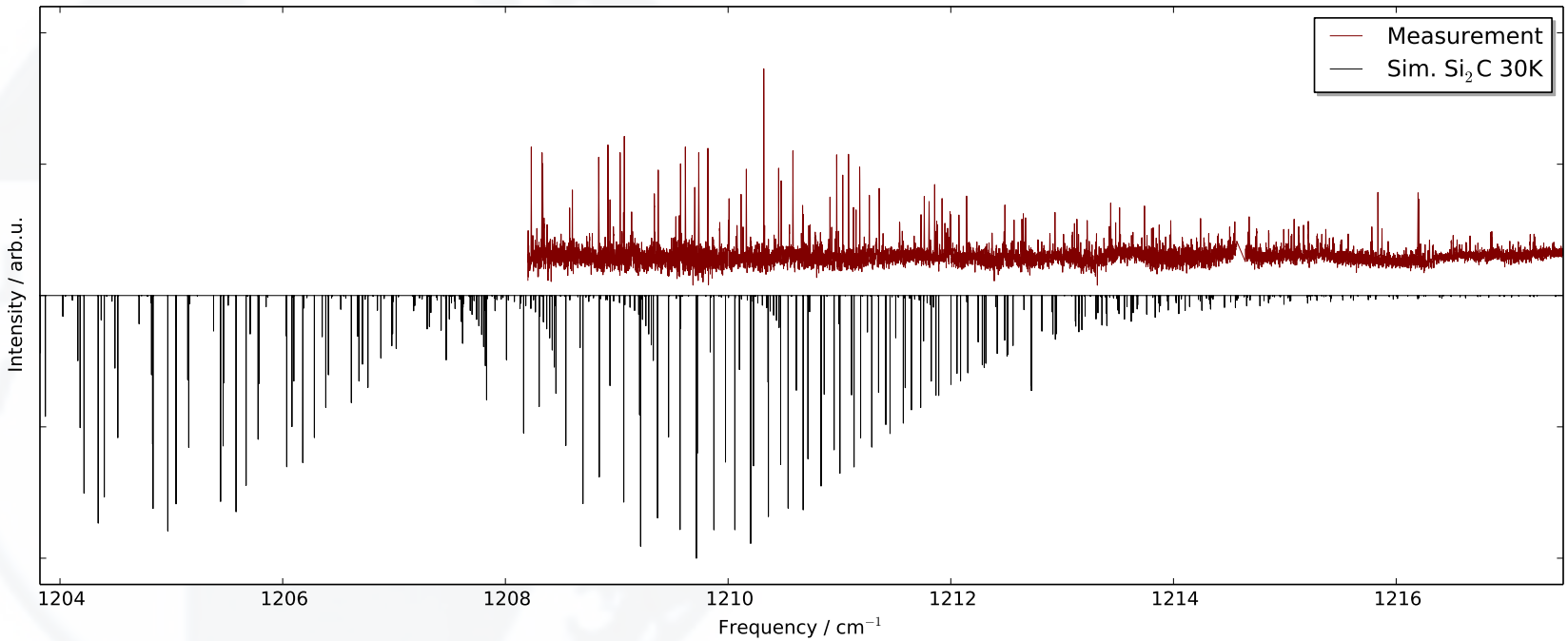

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Molecular Constants of Si_2C		
	Ground[a]	$\nu_3 = 1$[b]
A	64074.3366(44)	60347.75
B	4395.51772(41)	4463.64
C	4102.13098(62)	4156.22

values in MHz

[a] J. Cernicharo *et. al.*, Ap.J.Lett. L3 2015

[b] α_i from CCSD(T)/cc-pwCVQZ calculations (S. Thorwirth)

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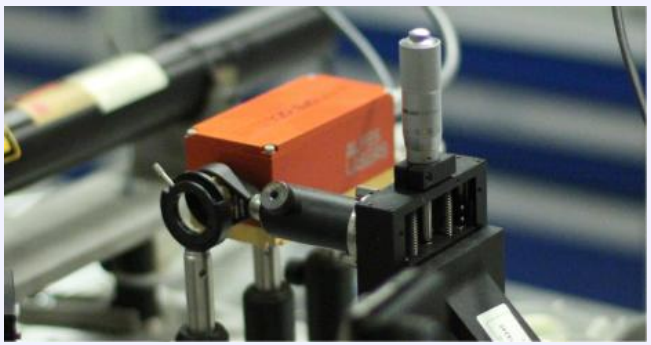
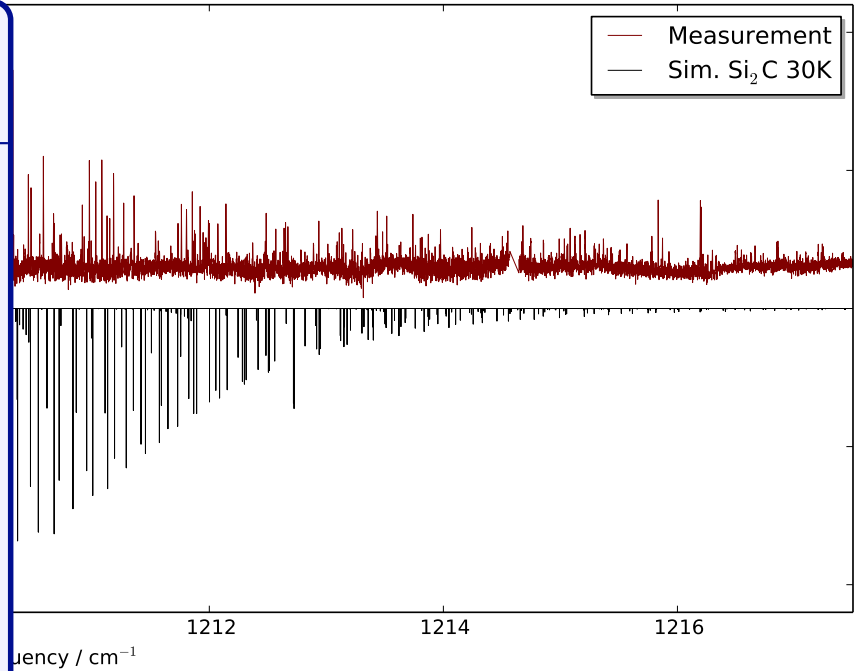


Details	
Frequency	1200 - 1208 cm^{-1}
Power (cw)	up to 100 mW



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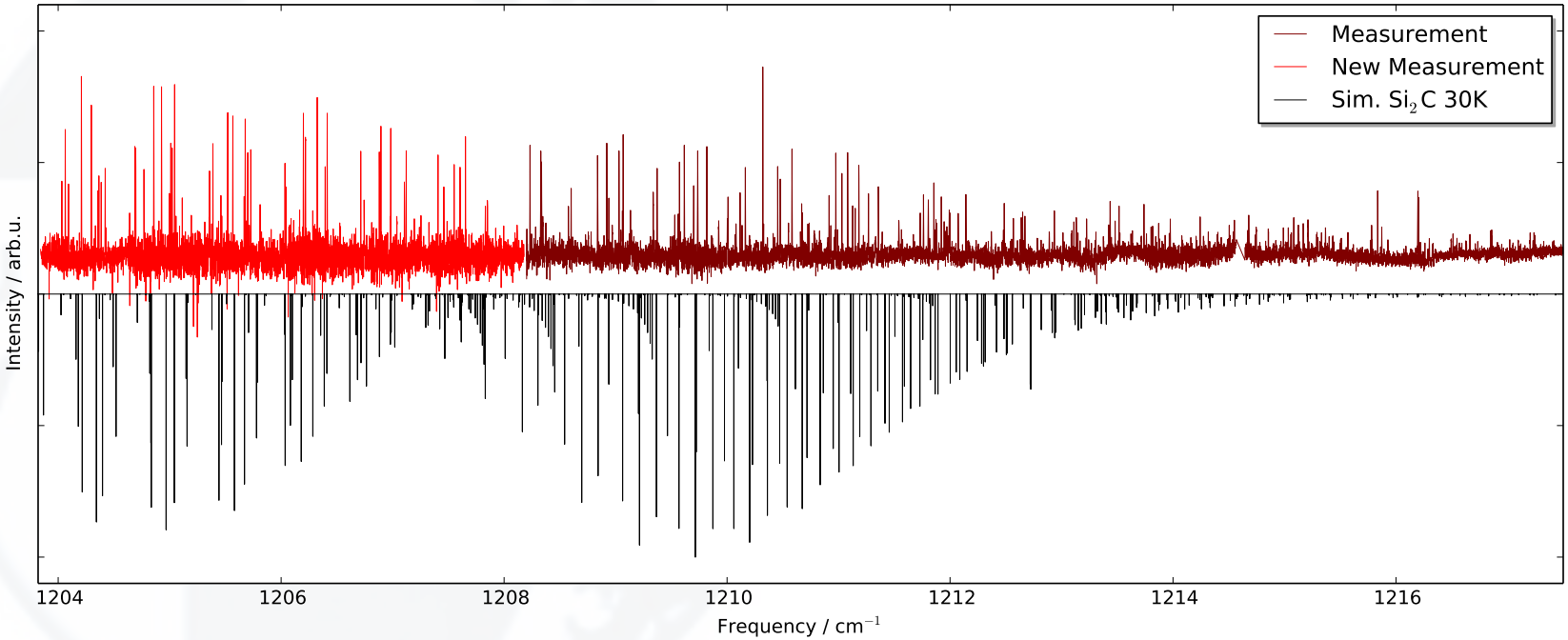

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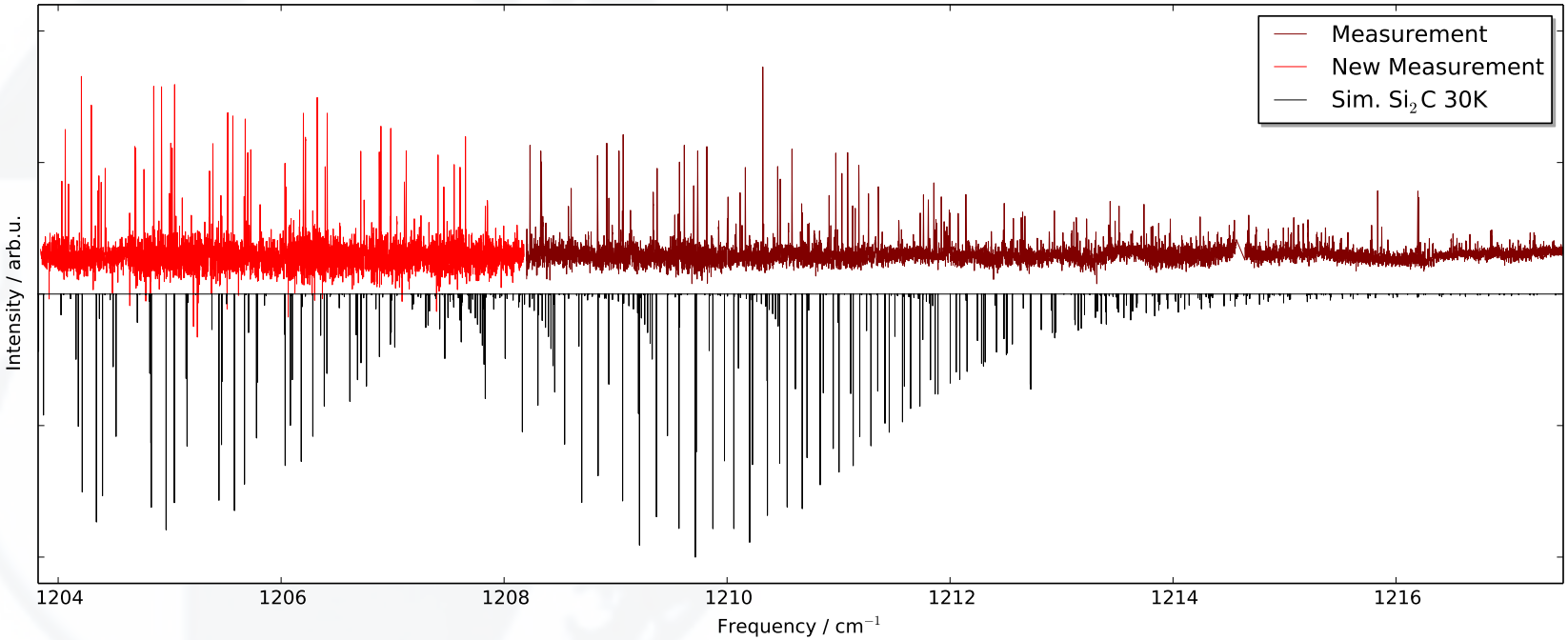

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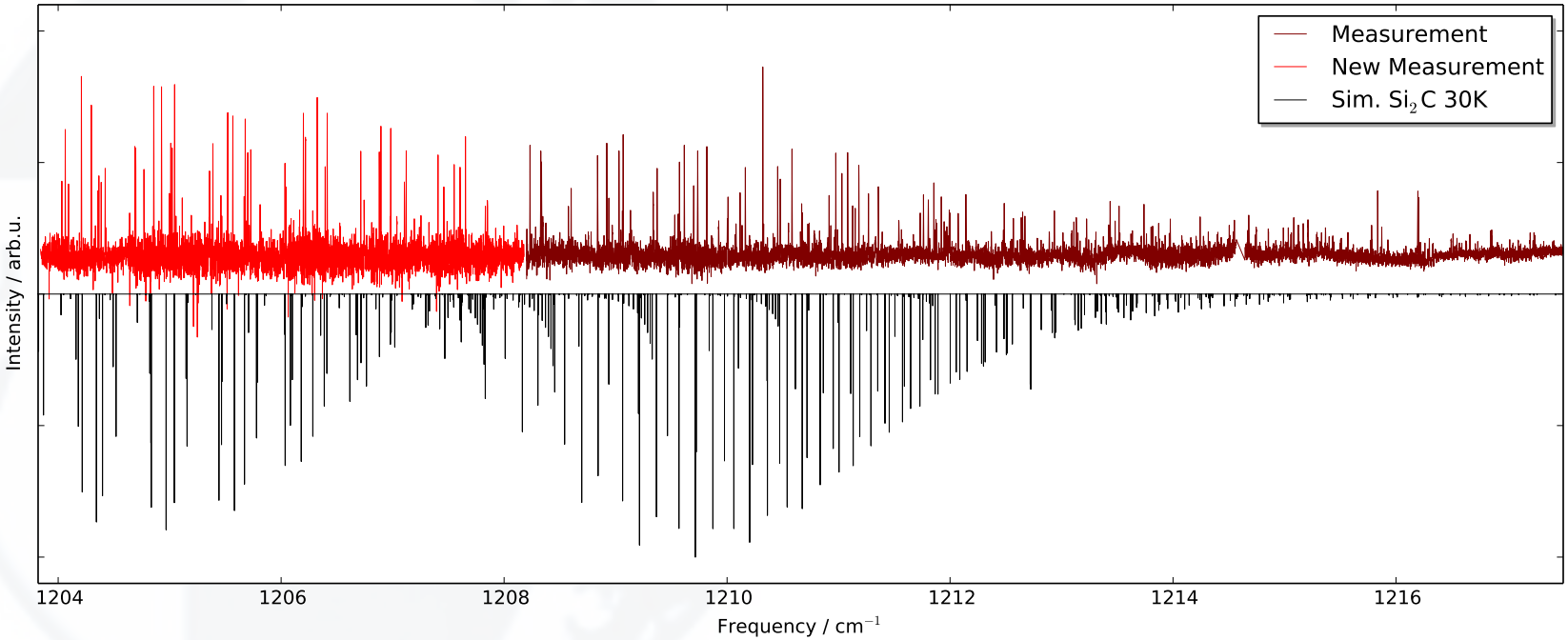

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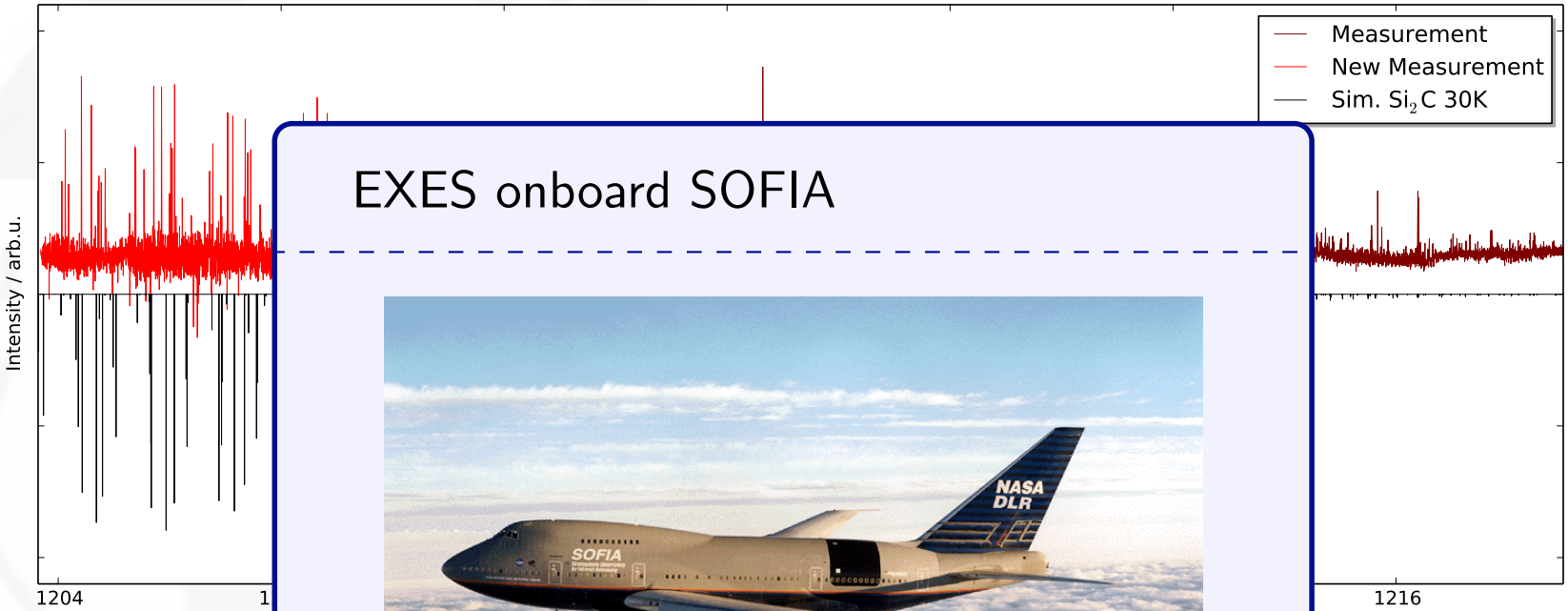

EXES onboard SOFIA

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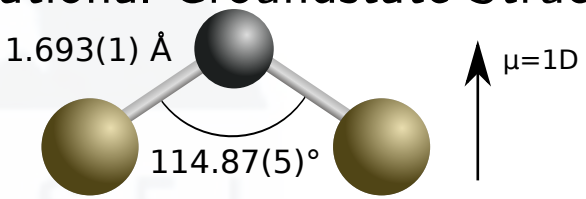
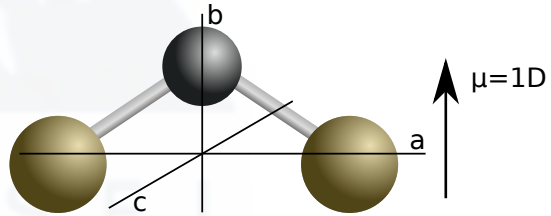


M.C. McCarthy *et. al.*, J.Phys.Chem.Lett 6 (2015), 2107

The Molecule

Vibrational Groundstate Structure



M.C. McCarthy *et. al.*, J.Phys.Chem.Lett 6 (2015), 2107

What do we know?

- electronic ground state: $^1\mathrm{A}_1$
- $I(\mathrm{Si}) = 0$ symmetric spin wavefuntion
- asymmetric stretching vibration (vib. excited state: B_2 symmetry)
- a-type transition ($\Delta K_a = 0$ and $\Delta K_c = \pm 1$)

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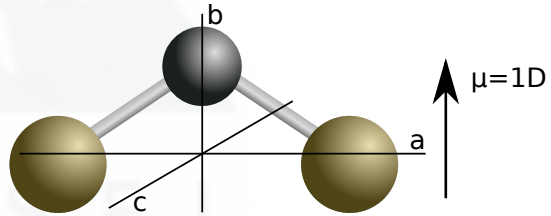

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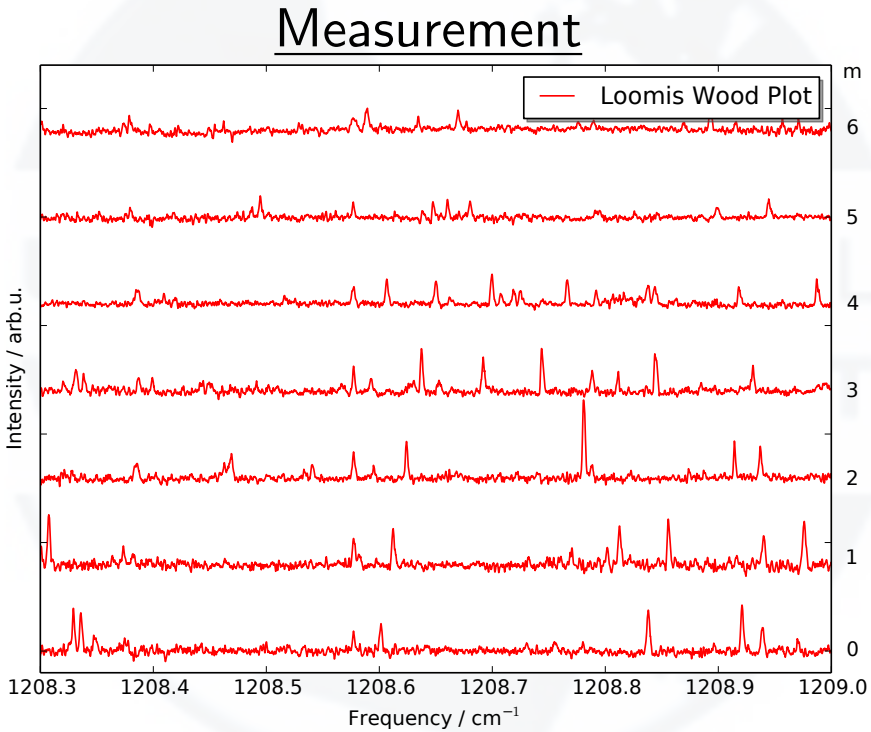
Similar Systems?

- H_2O?
  - similar structure
  - BUT mass in O-atom
- Cl_2O
  - Distortion due to coupling with symmetric stretching vibration

Data Analysis



several series have been found

$$\begin{aligned} \text{Shift} &= b \cdot m + d \cdot m^2 \\ b &\approx 0.74\,\text{cm}^{-1} \\ d &\approx 0.01\,\text{cm}^{-1} \end{aligned}$$

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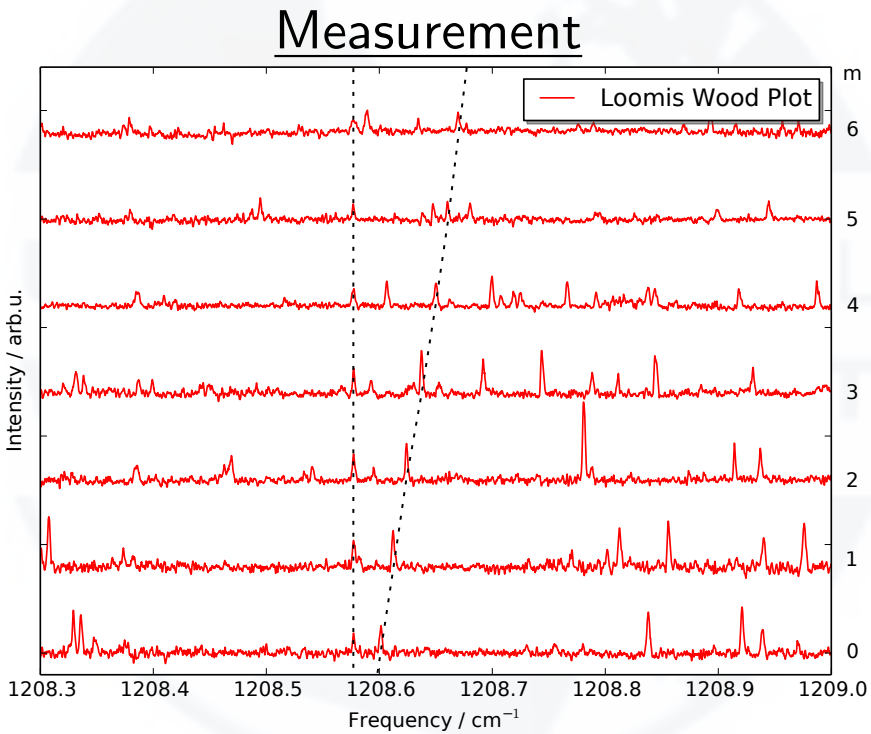

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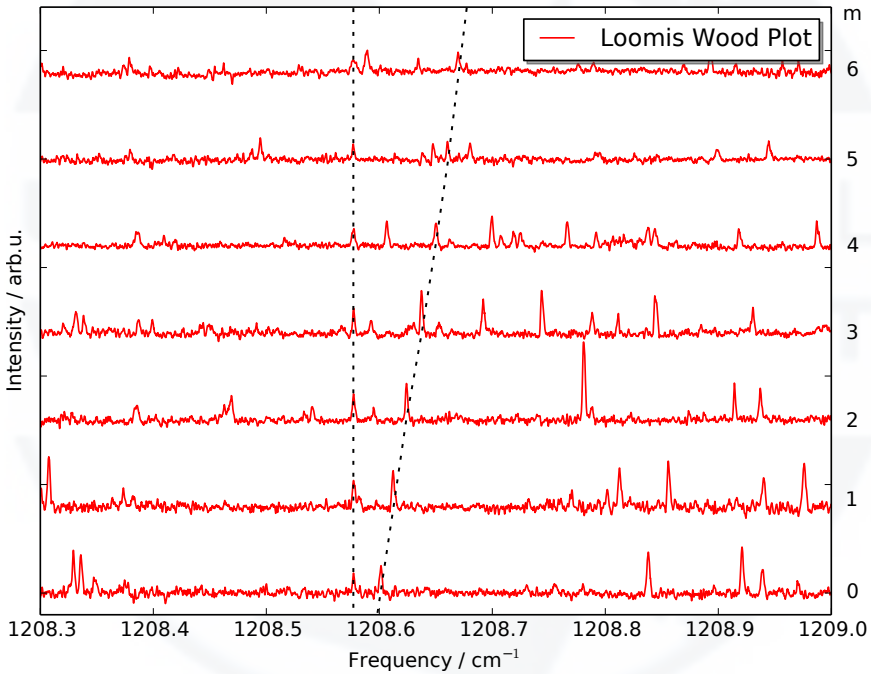
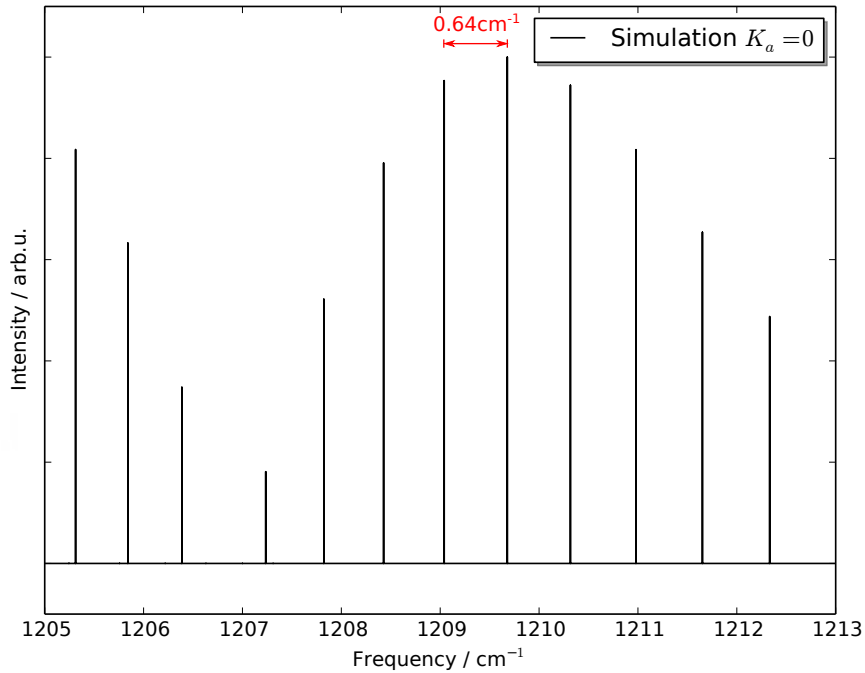
Measurement



Simulation



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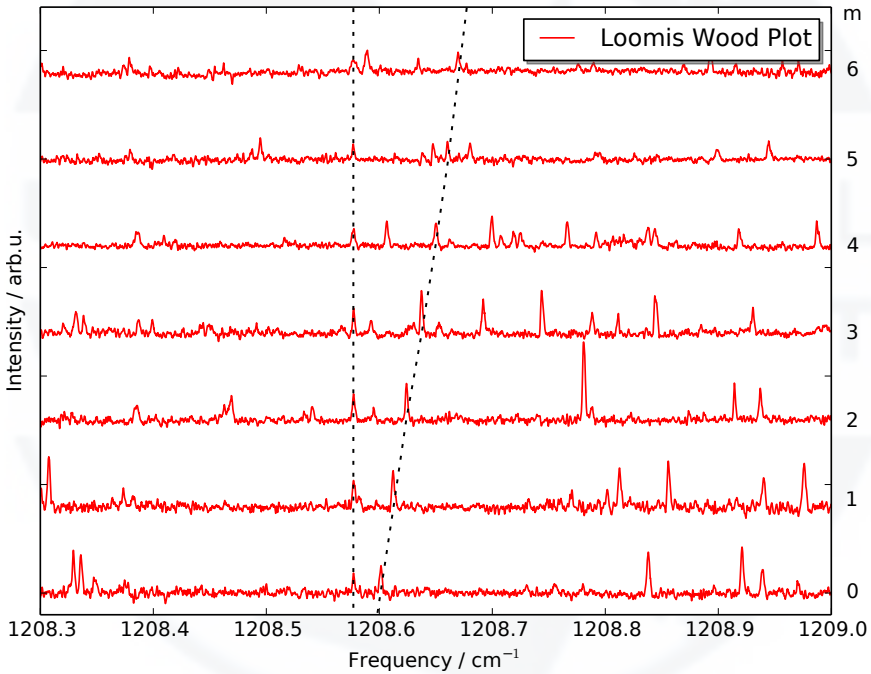

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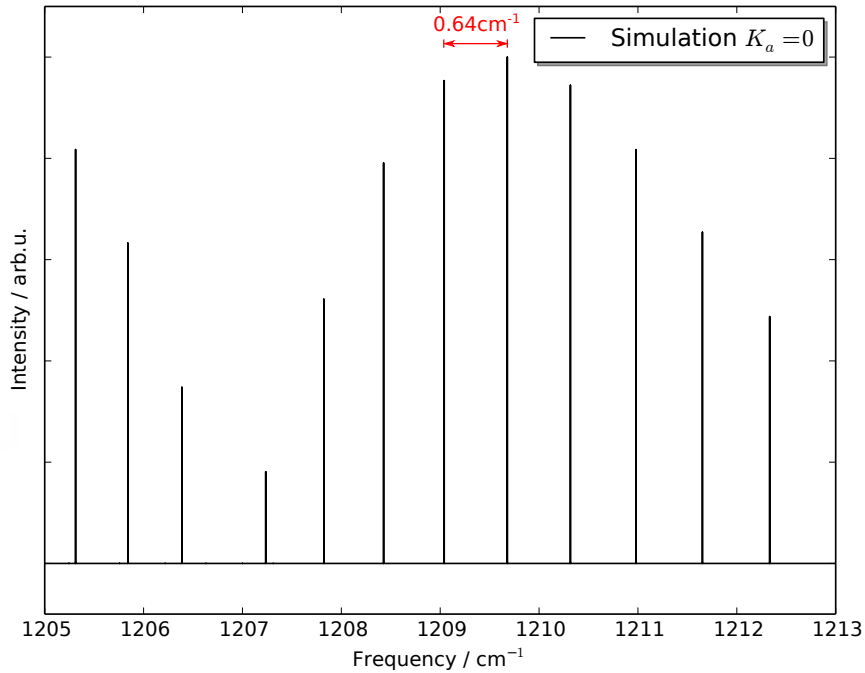
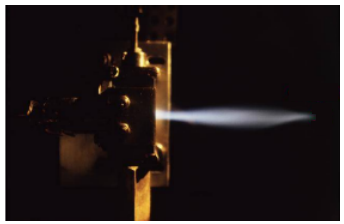
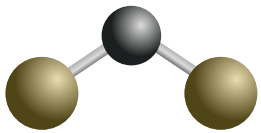


- B'' and C'' seem to be larger than calculated values
- Different Shifts between series ($0.68 < \frac{b}{\text{cm}^{-1}} < 0.8 \Rightarrow$ Distortion?)
- Assignment needed:
  - New PGopher feature

Summary

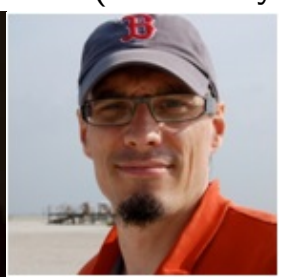


- Experimental setup to produce cold and small silicon-carbides
- Rovibrational spectrum of the ν_3 band of Si_2C
- B'' and C'' are 1 to 3 % larger than calculated values for Si_2C
- Prepared for interstellar search with EXES onboard SOFIA

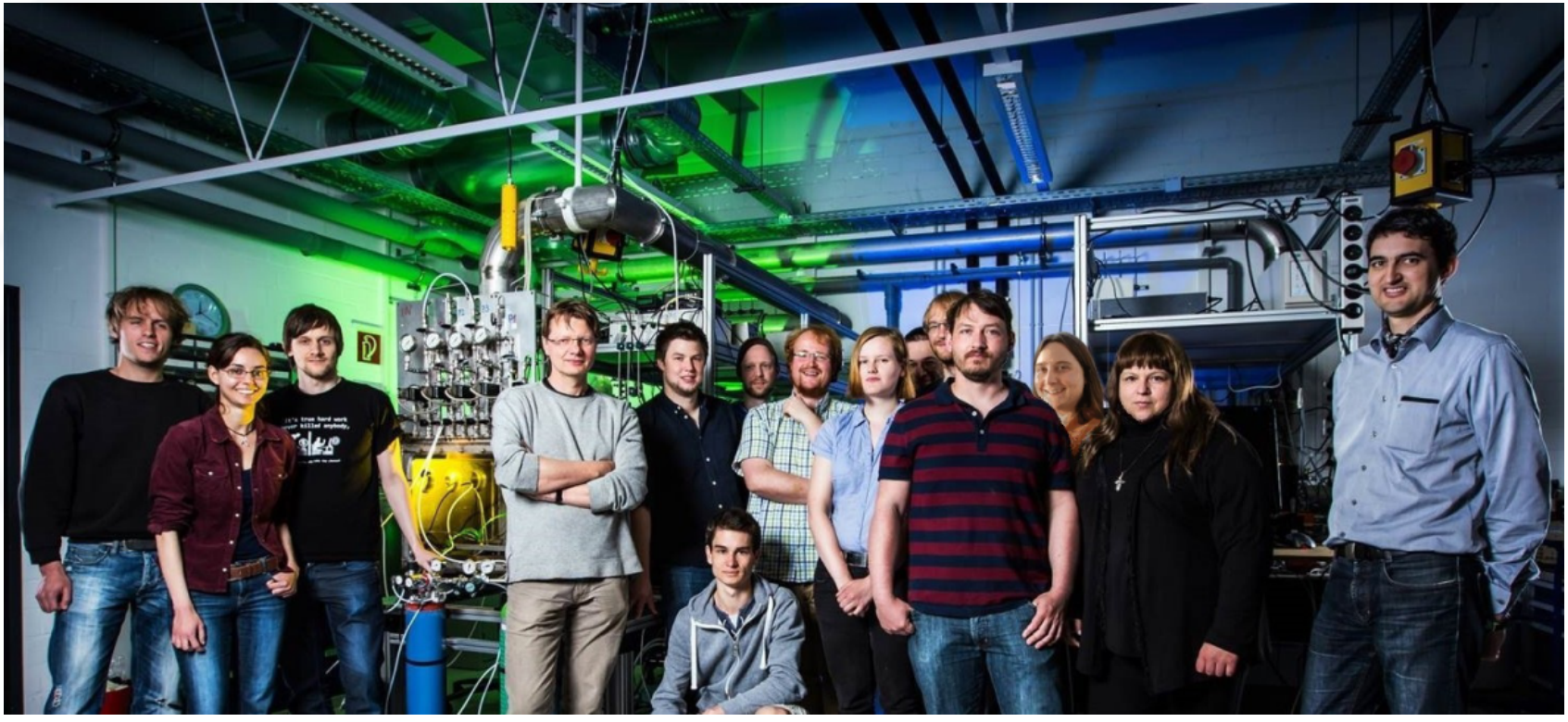


Acknowledgement

- **Jürgen Gauss** (University of Mainz)
- **Sven Thorwirth** (University of Cologne)



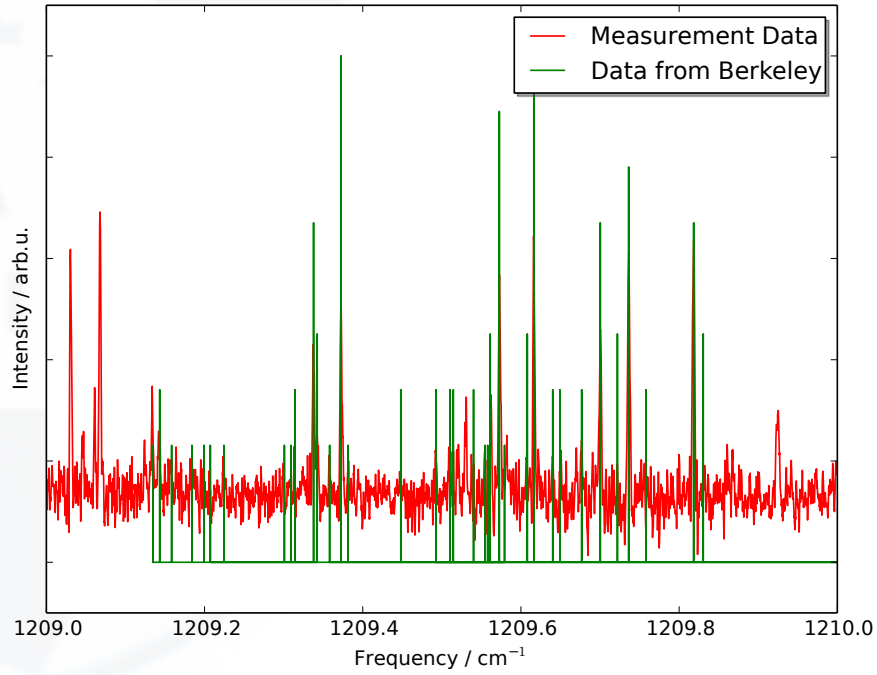
Funding by
DFG Deutsche Forschungsgemeinschaft



Thank you for your attention!

Appendix

Data from Berkeley



Berkeley:

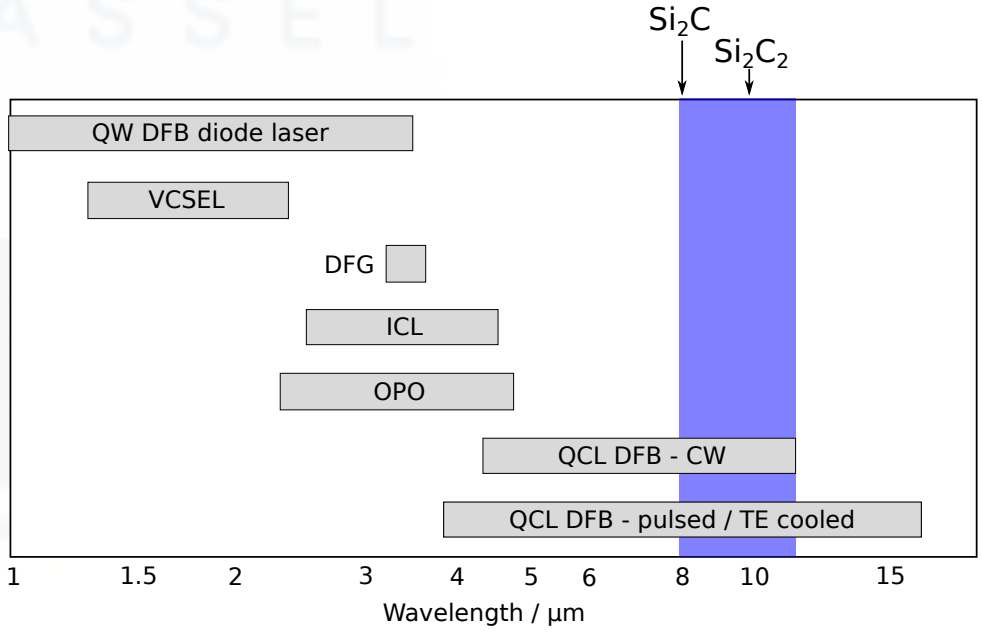
- Ablation Laser: Excimer Laser
- Silicon-Carbide (SiC) Rod
- pure Helium Carrier-Gas

Kassel:

- Ablation Laser: Nd:YAG
- pure Silicon (Si) Rod
- Carrier-Gas: 2.5% CH_4 in Helium

Radiation Sources

- Asymmetric stretching vibration of Si_2C expected to be at 8 μm (1250 cm^{-1})
- Common radiation sources for IR-spectroscopy

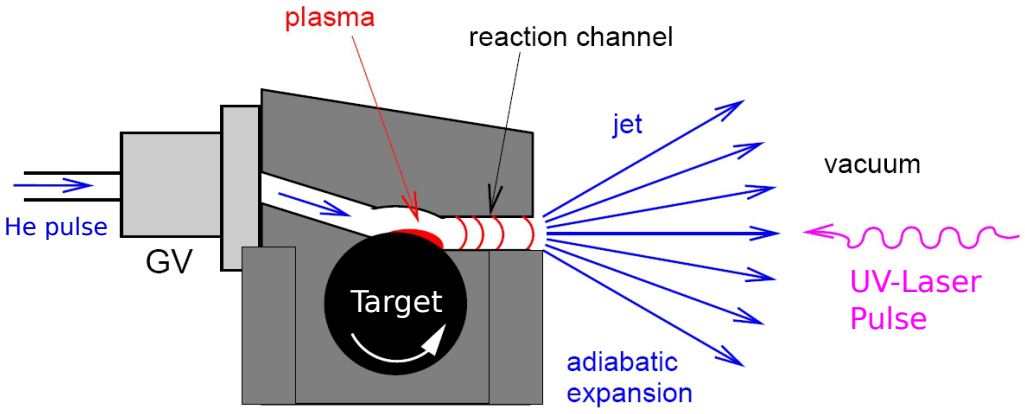


Hodgkinson *et al.*, Meas. Sci. Technol. 24 (2013)

- Lack of radiation sources above 5 μm
- Two main tasks: Production and Investigation

Cluster Source

Laborastrophysik
Universität Kassel



Laser Ablation Source	
Laserfrequency	355 nm
Pulse energy	40 mJ
Repetition rate	20 Hz
Helium gas	20 bar
Background pressure	0.1 mbar
Target material	SiC