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# *Optically-Pumped Terahertz Sources and Applications*

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Terahertz Photonics Group
IEMN CNRS Lille University

cnrs dépasser les frontières

Université de Lille

# Introduction



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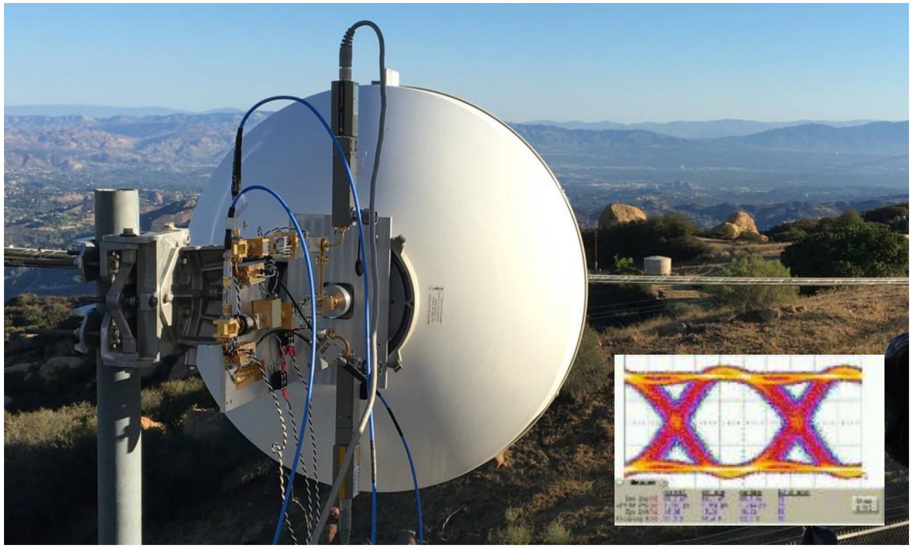
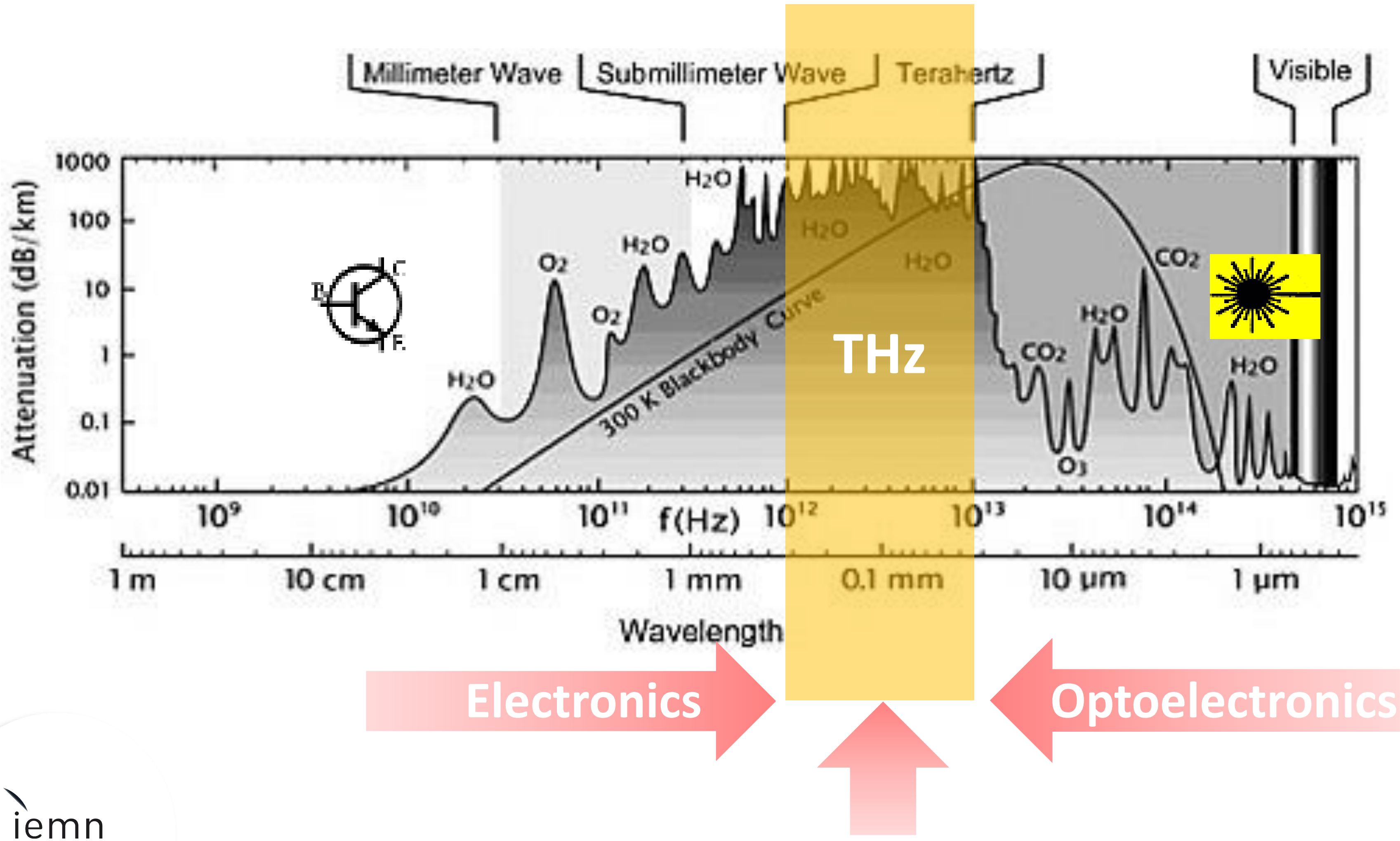
# THz ? Driven by applications



- Strong interactions with matter
- Huge bandwidths
- Imaging

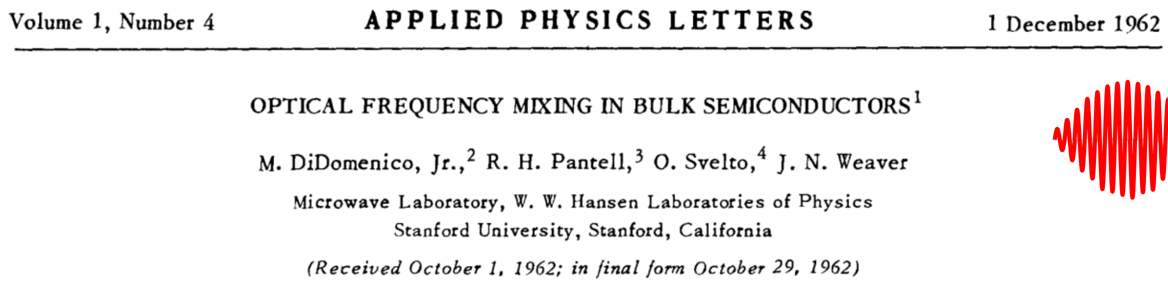
# How to generate THz waves ?

# The THz band



Millimeter Wave
Submillimeter Wave
Terahertz
Visible
Attenuation (dB/km)
1000
100
10
1
0.1
0.01
H2O
O2
300 K Blackbody Curve
CO2
O3
THz
10⁹
10¹⁰
10¹¹
f(Hz)
10¹²
10¹³
10¹⁴
10¹⁵
1 m
10 cm
1 cm
1 mm
0.1 mm
10 µm
1 µm
Wavelength
Electronics
Optoelectronics

# THz Photomixing

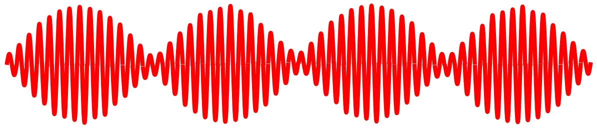

Volume 1, Number 4 APPLIED PHYSICS LETTERS 1 December 1962

OPTICAL FREQUENCY MIXING IN BULK SEMICONDUCTORS[1]

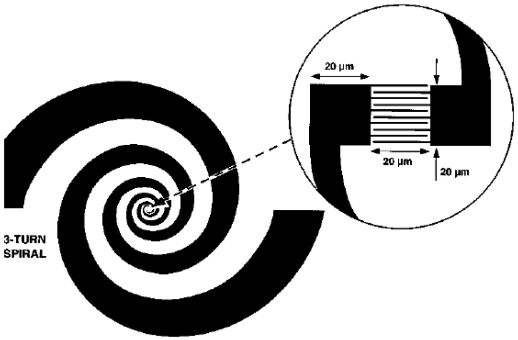
M. DiDomenico, Jr.,[2] R. H. Pantell,[3] O. Svelto,[4] J. N. Weaver

Microwave Laboratory, W. W. Hansen Laboratories of Physics
Stanford University, Stanford, California

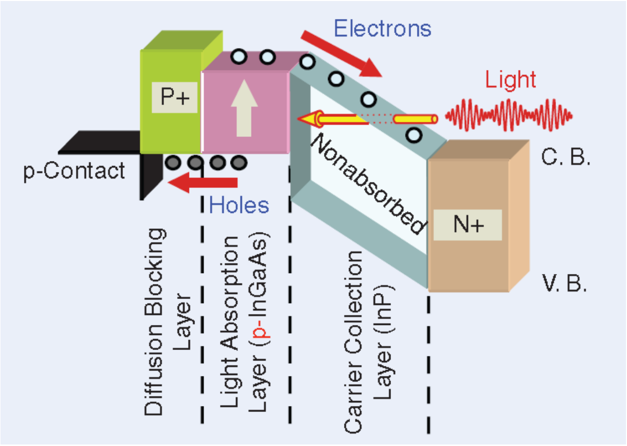
(Received October 1, 1962; in final form October 29, 1962)



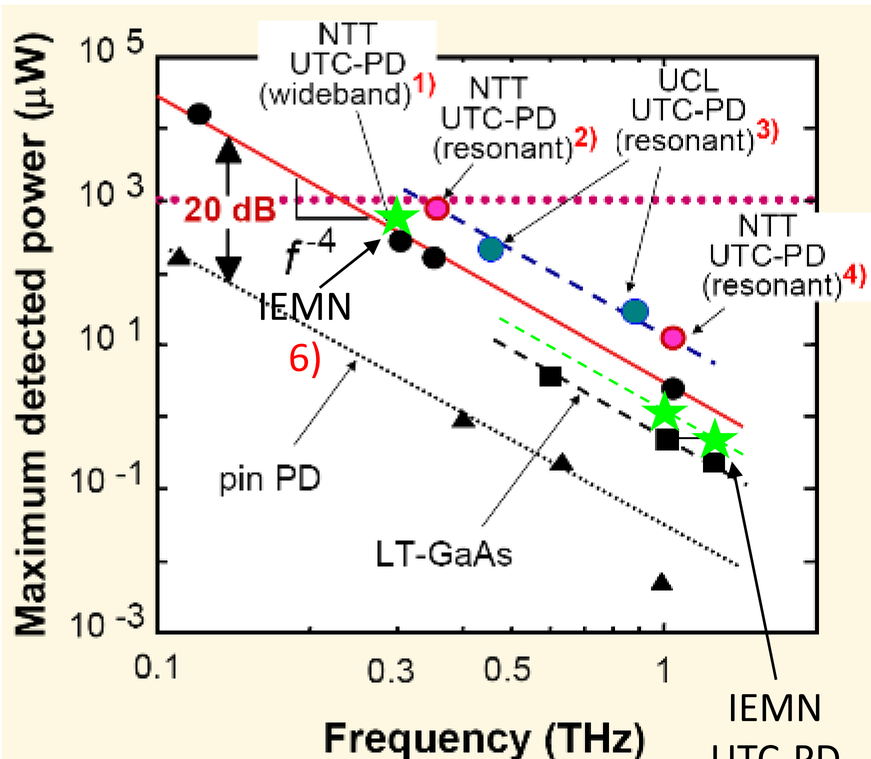
- LT-GaAs Photoconductor: Elliott Brown *et al.* (1995)

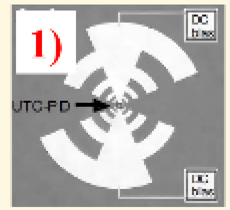


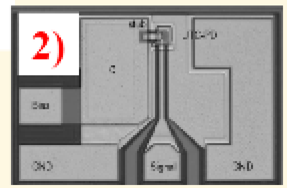

- UTC photodiodes: T. Ishibashi, H. Ito *et al.* (1997)

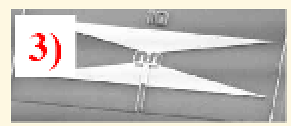


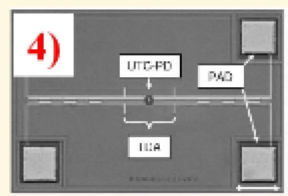
# UTC-PD: State of the art



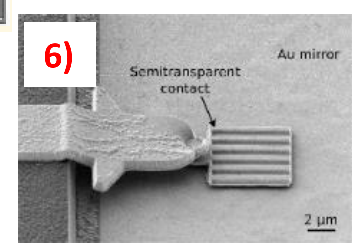








- Wideband antenna
≈1-2 μW @ 1 THz

- Resonant antenna
≈ 10-20 μW @ 1 THz
≈ 500 μW @ 350 GHz

RCE UTC-PD
0.75 mW
@300GHz

Latzel *et al.*, IEEE TTST **7**, 800 (2017)

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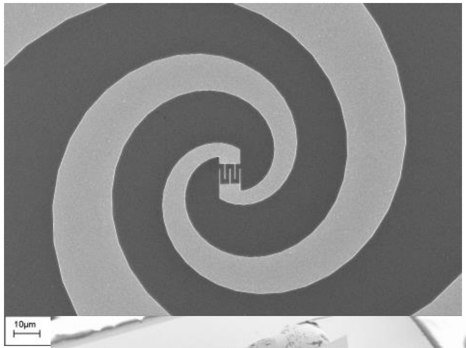
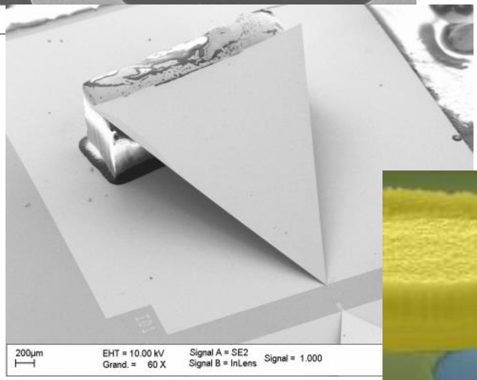
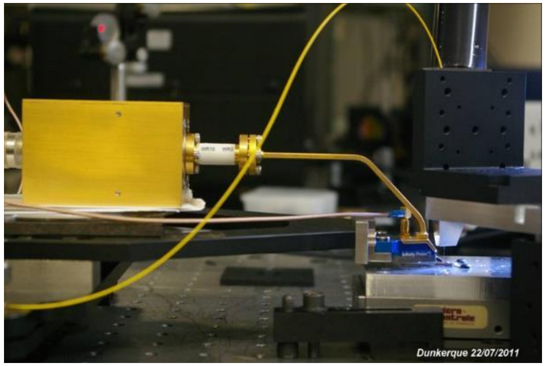
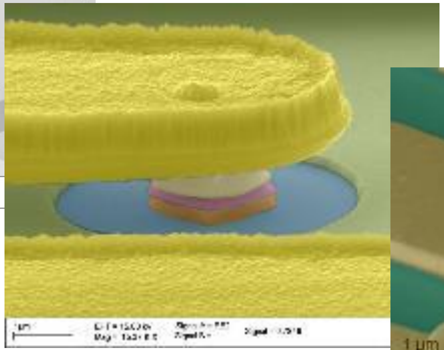
# THz Photomixing at IEMN

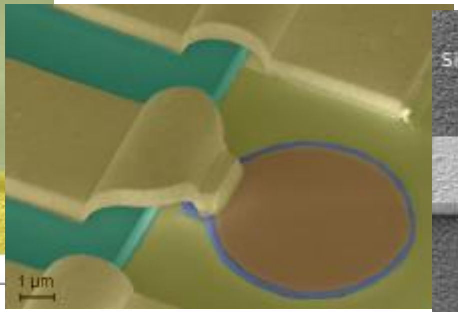
0.8 & 1.5 µm photomixers families developed by E. Peytavit, M. Zaknoune, G. Ducournau *et al.*

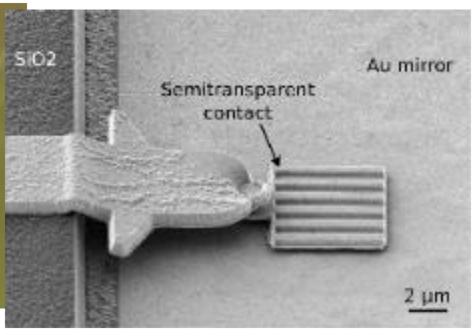





≈ 1 mW @ 300 GHz, efficiencies ≈ 0.1-1 %

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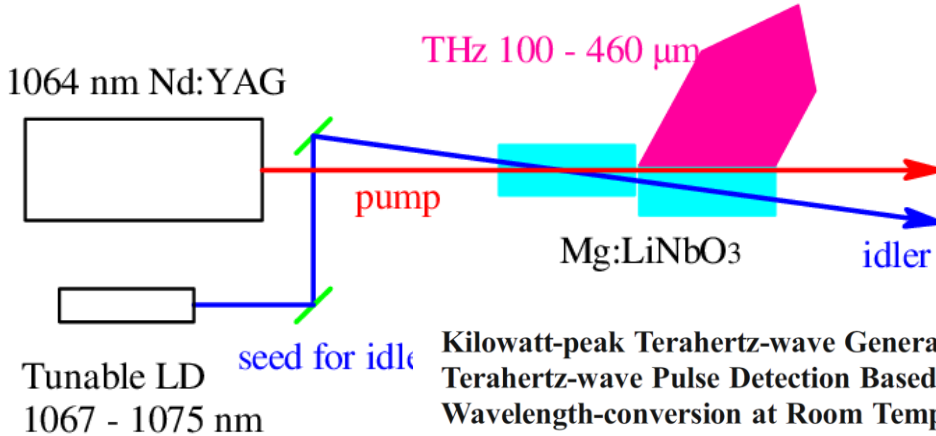
# THz generation: parametric & DFG

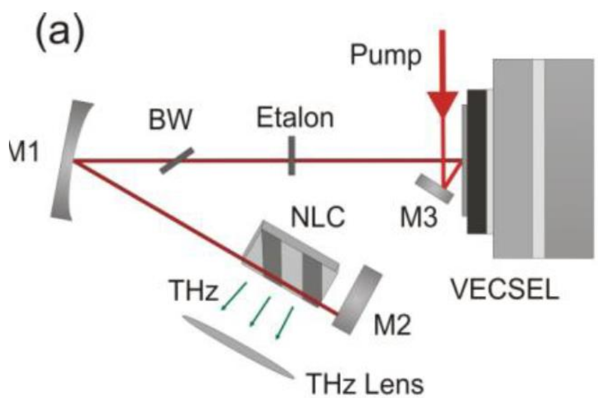
Terahertz Parametric Generation (Riken)

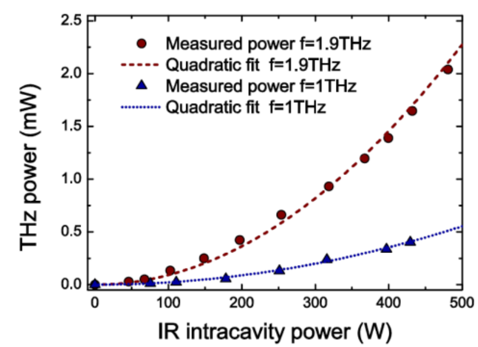



**Kilowatt-peak Terahertz-wave Generation and Sub-femtojoule Terahertz-wave Pulse Detection Based on Nonlinear Optical Wavelength-conversion at Room Temperature**

**Hiroaki Minamide · Shin'ichiro Hayashi · Koji Nawata · Takunori Taira · Jun-ichi Shikata · Kodo Kawase**

M. Scheller *et al.*, OE 18, 27112 (2010)

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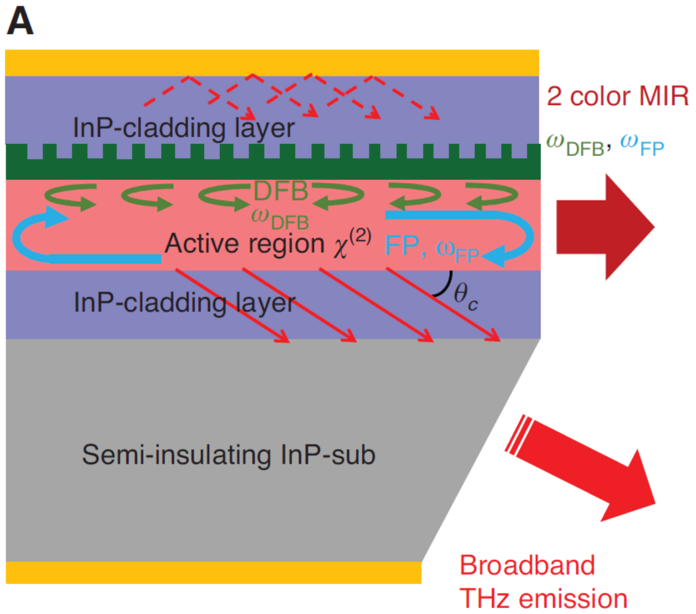
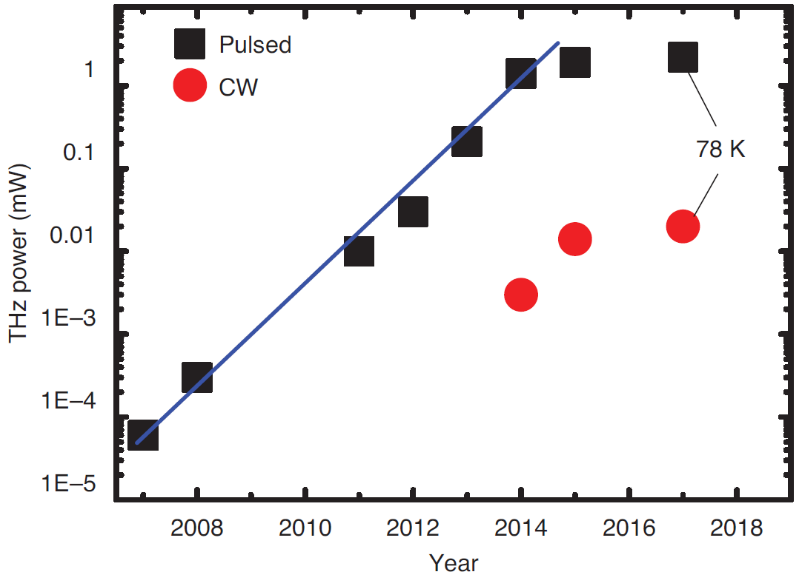
# DFG in MIR QCLs

For a review:
K. Fujita, …., M. A. Belkin, Nanophotonics 7, 1795 (2018)




Hamamatsu : 1mW/W$^2$ peak @ 2.5 THz (Mo-AM-6-1)

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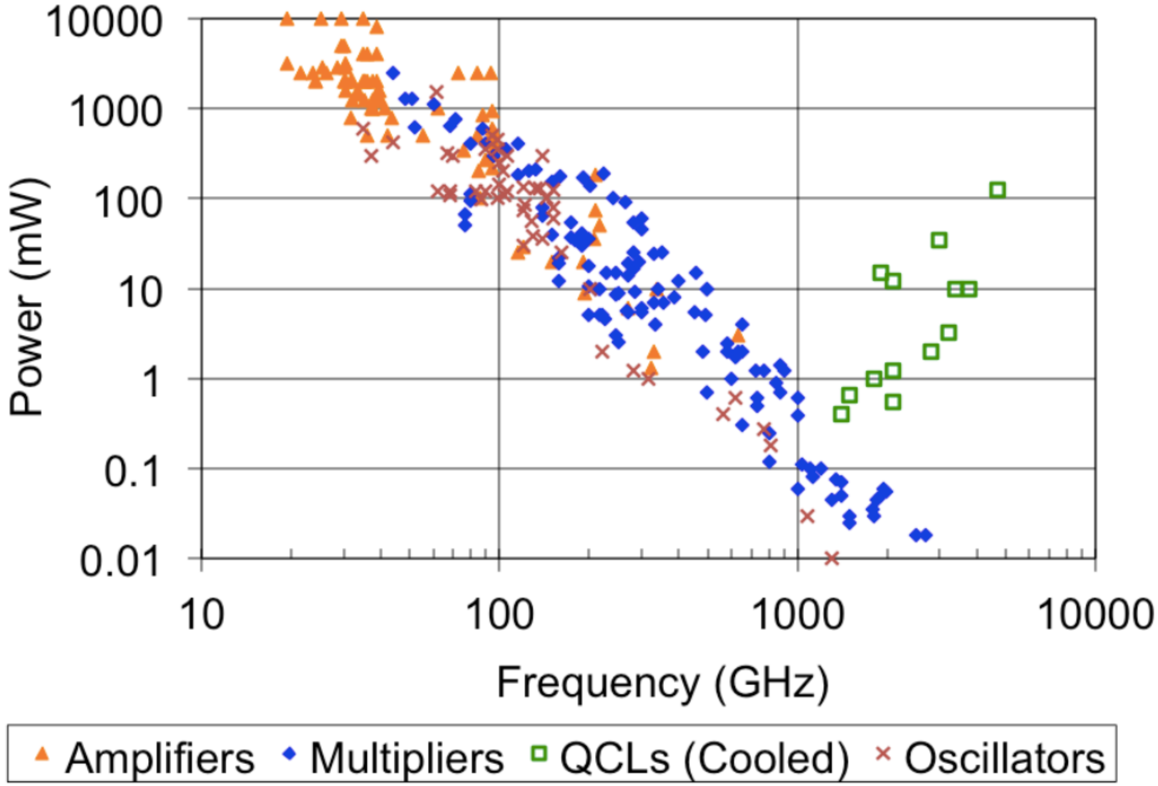
# State of the art CW solid-state THz sources



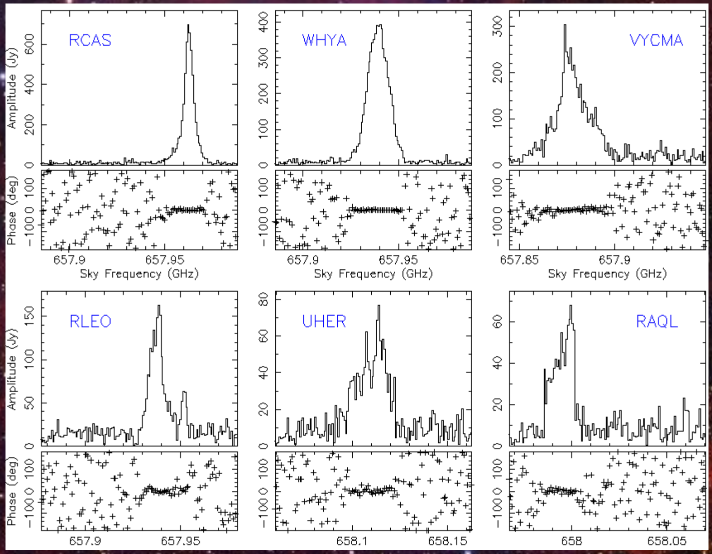

HEMT/HBT Schottky VCO/RTD

*Thanks to J. Esler (VDI)*

CW generation around and above 1 THz is **still challenging !**

# Natural THz lasers ….

Microwave and THz masers exist in the universe in molecular clouds !




658 GHz Maser, Hunter 2007

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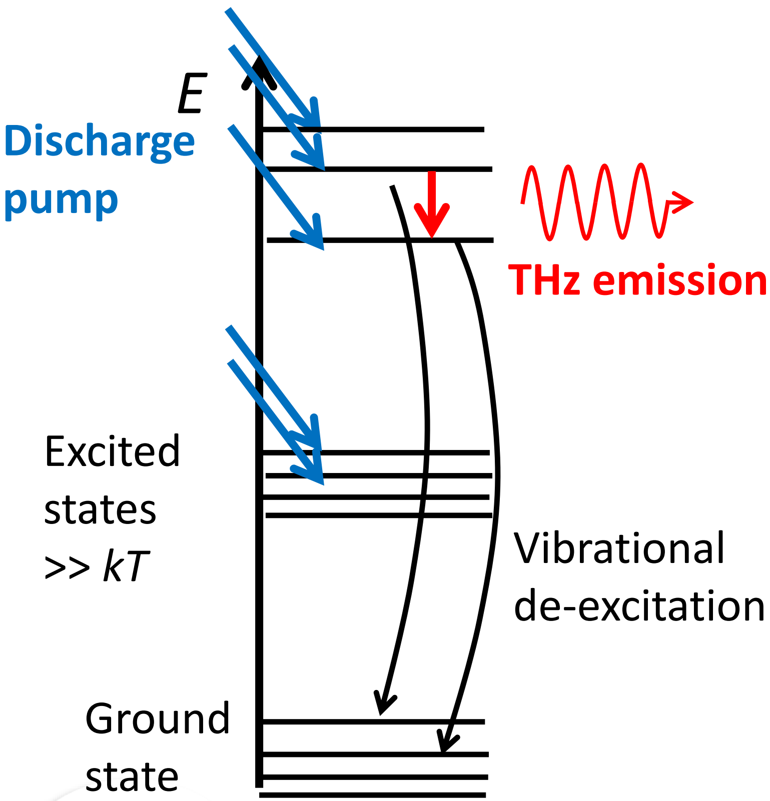
# The molecular lasers

The **molecular discharge lasers**:

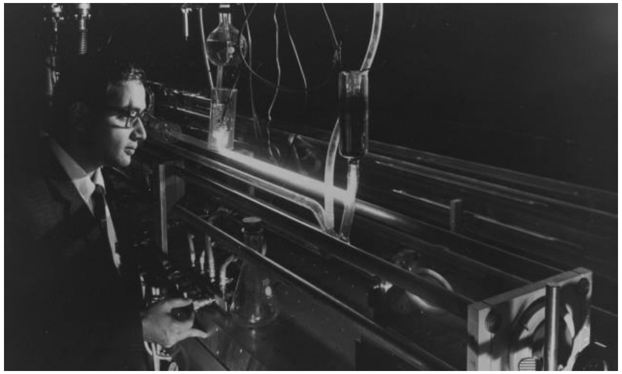
1964: The $H_2O$ laser by Crocker, Gebbie, Kimmit and Mathias

1965: Patel ($CO_2$ laser)




M. Kimmit (KJB prize 2004)

- Works with only few molecules
- Problem of dissociation
- Lethal voltage, low efficiency
- Only $CO_2$ was really successful



K. Patel

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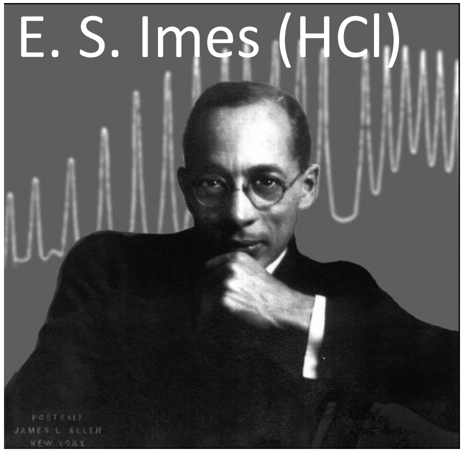
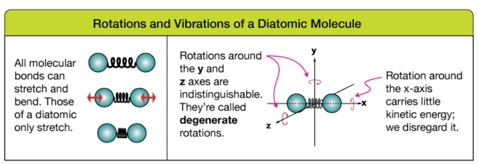
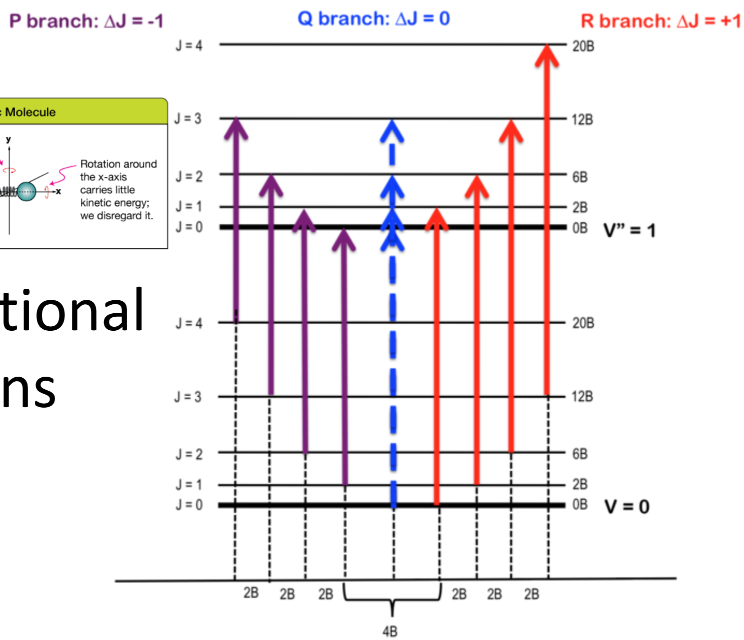
# Molecular infrared absorption

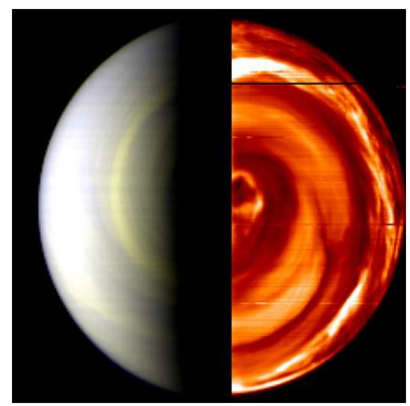







Ro-vibrational transitions



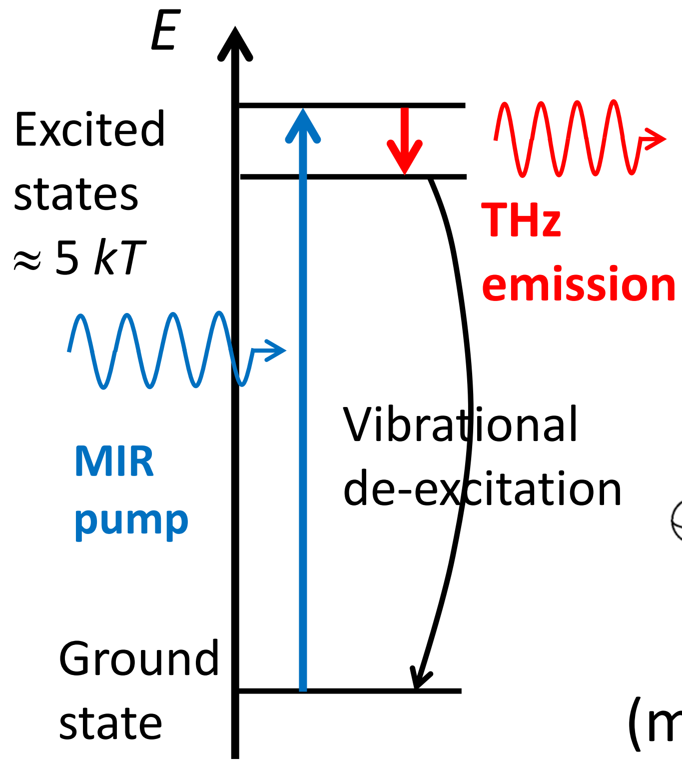
Venus greenhouse effect

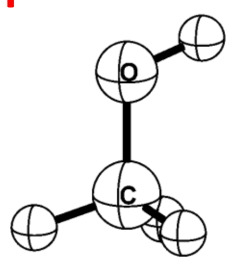
Visible IR

# Principle of OPTLs

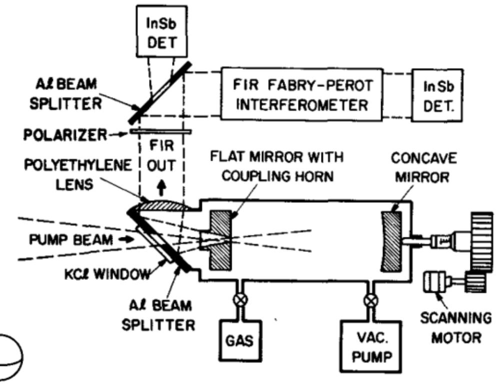
**OPTL = Optically Pumped Terahertz Laser**

(Chang & Bridges 1970, Bell Labs, Holmdel)



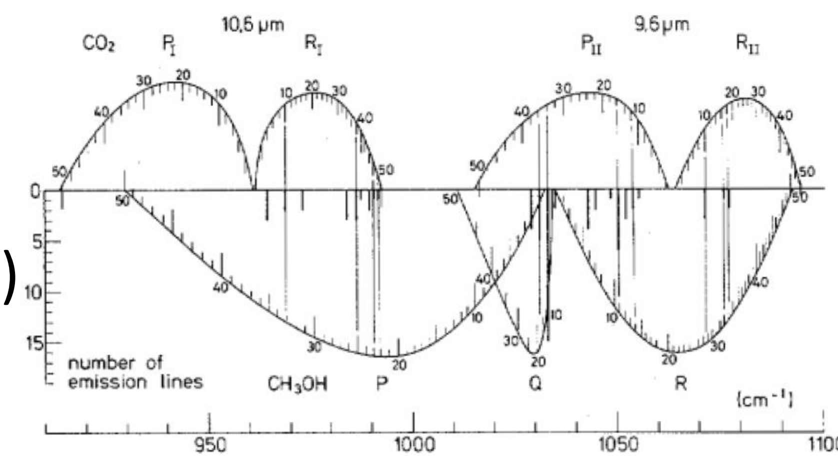




$CH_3OH$ (methanol)

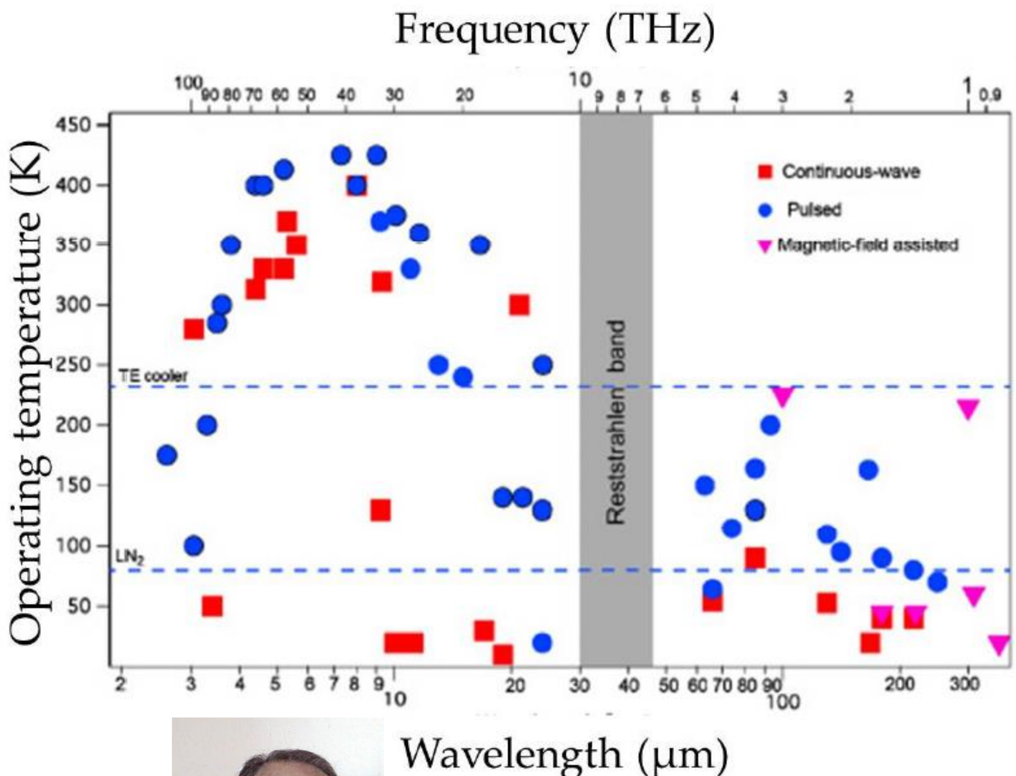


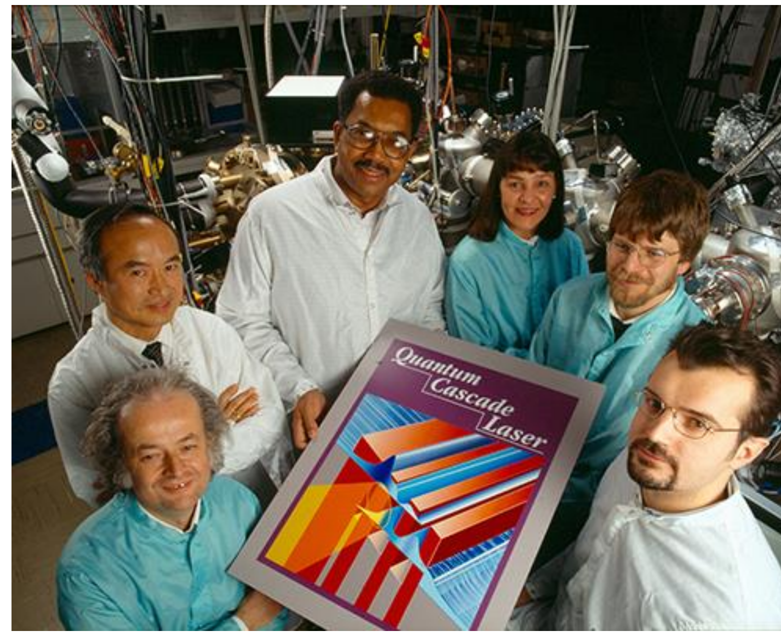

T.-Y. Chang

A MIR pump? … the $CO_2$ Laser



# A revolution in the IR world: QCLs !




Faist *et al.*, Science (1994)

- Monomodes CW QCL: external cavity, DFB
- Commercially available with 100's mW

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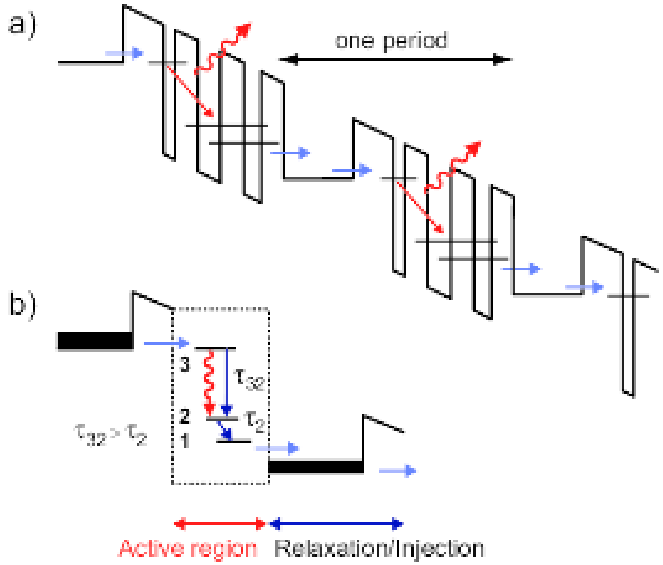
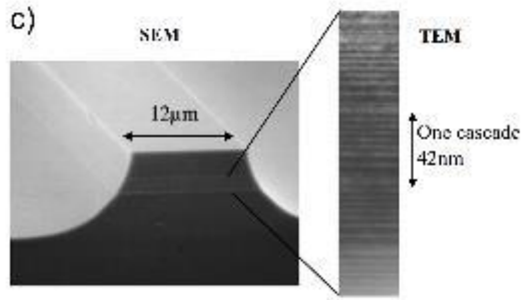
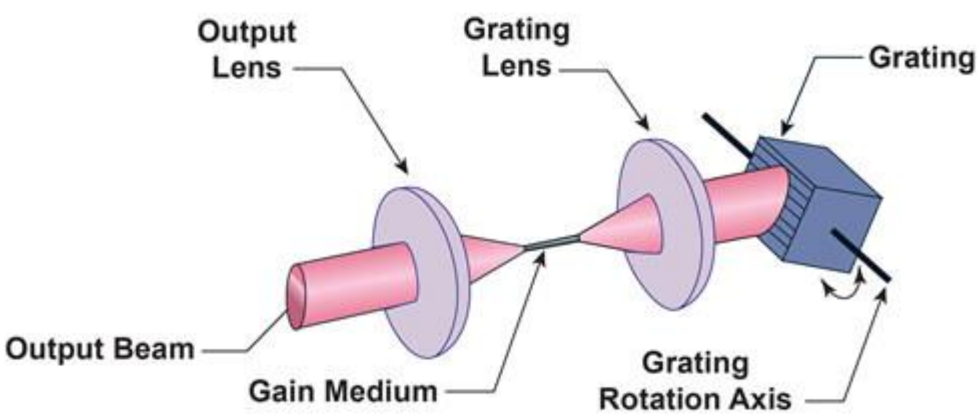
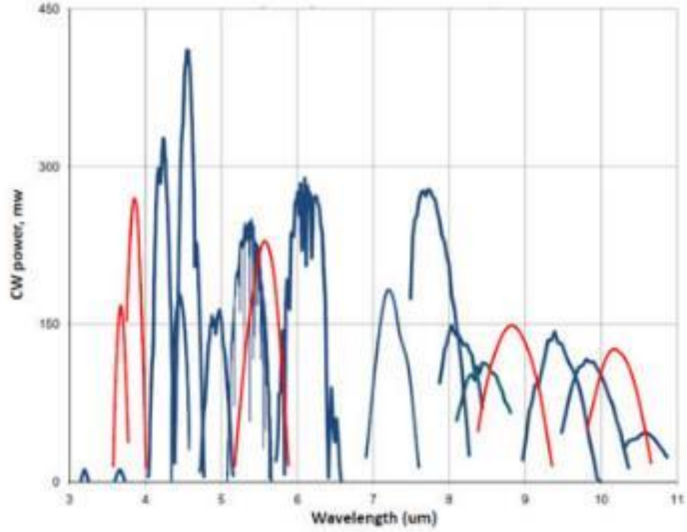
# Tunable QCLs

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# Why pumping an OPTL with a QCL ?

Comparison with $CO_2$ lasers:

**Advantages:**

- Compactness
- Low power consumption
- *Continuous tunability*

**Drawbacks:**

- Lower power
- Needs monomode QCL
- More sensitive to feedback

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**Drawbacks:**
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- More sensitive to feedback

The main advantage is: QCL allows to pump ***light molecules*** with **high dipolar moment**:
- High gain
- Fast relaxation
- Frequencies OK for applications

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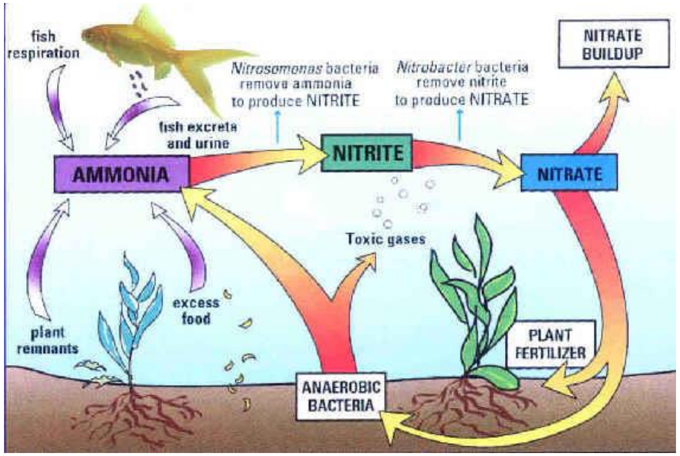
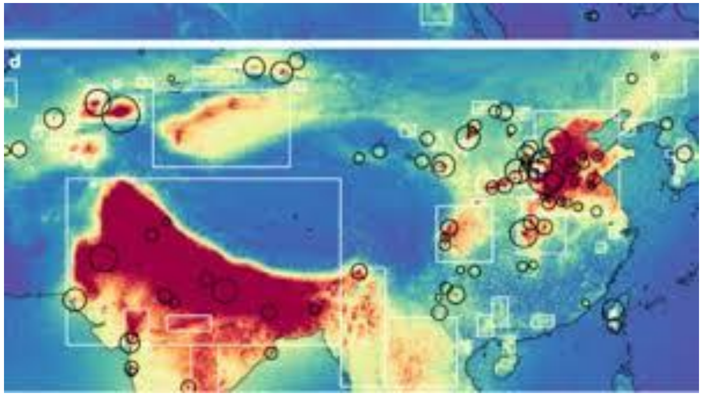
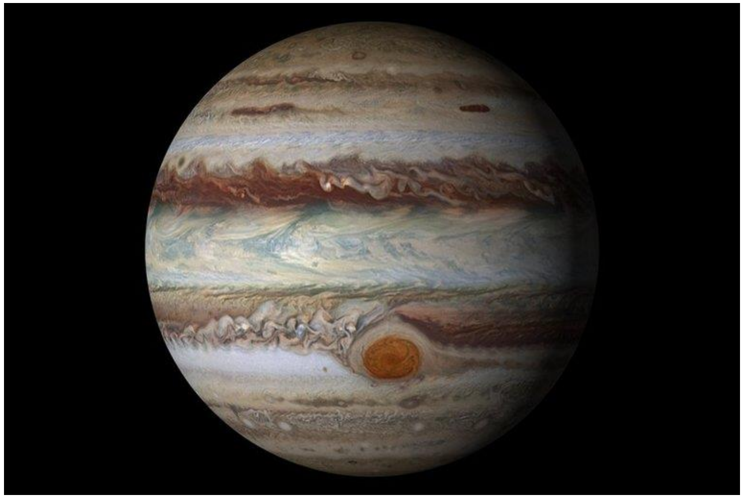
# A very common molecule…





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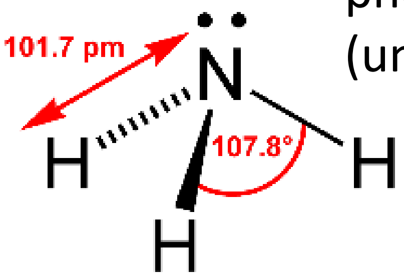
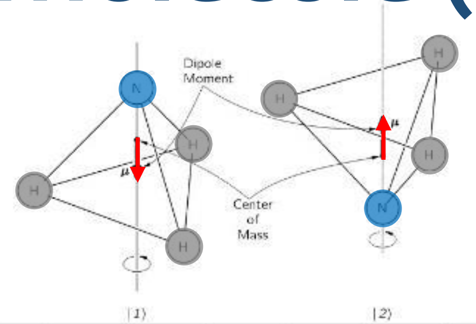
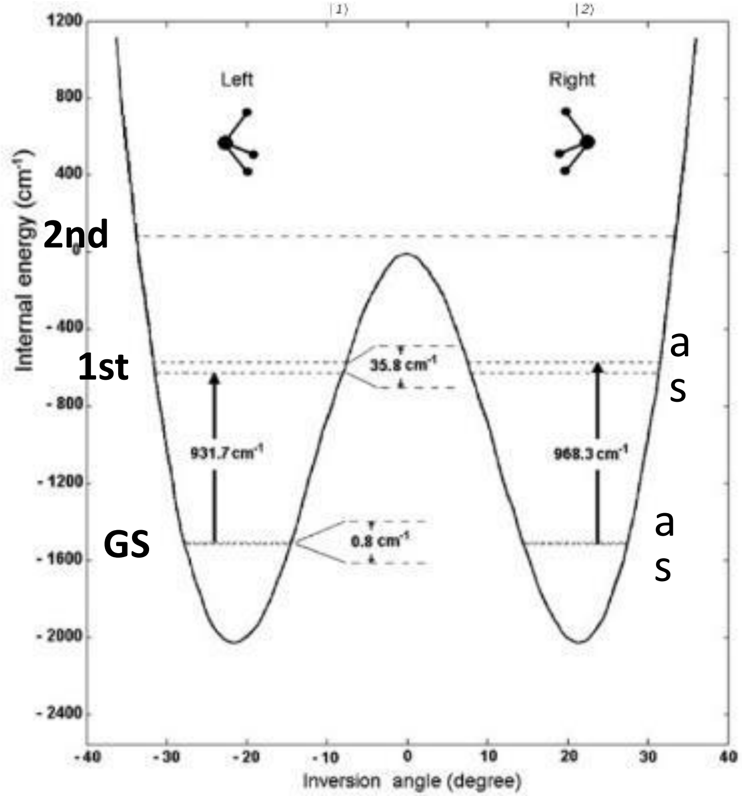
# The ammonia molecule ($NH_3$)




**« Inversion »** phenomenon (umbrella)




Permanent dipole : 1.4 Debye




Vibration-inversion double quantum well

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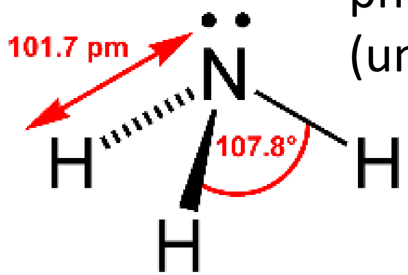
# The ammonia molecule ($NH_3$)

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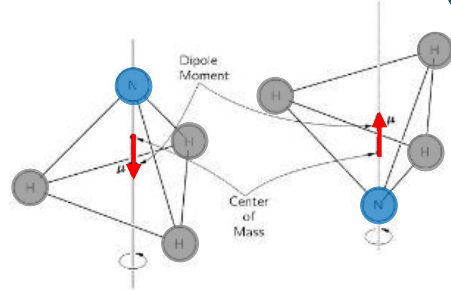


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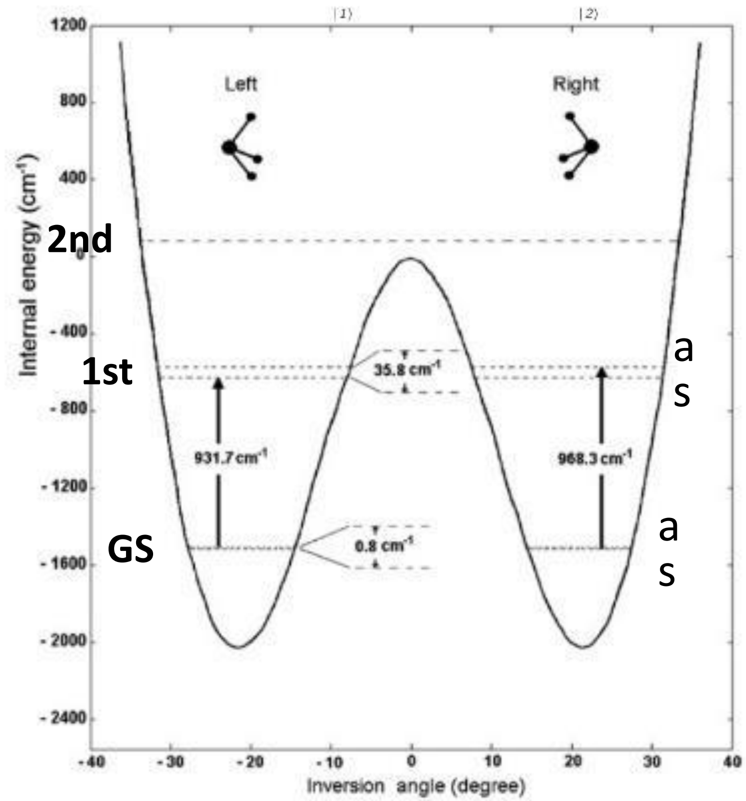


Vibration-inversion double quantum well


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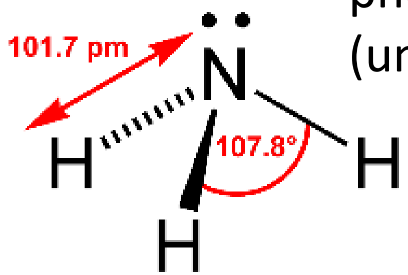


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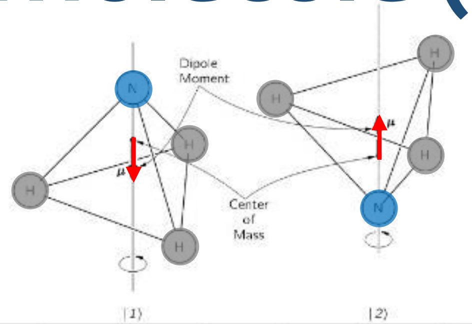
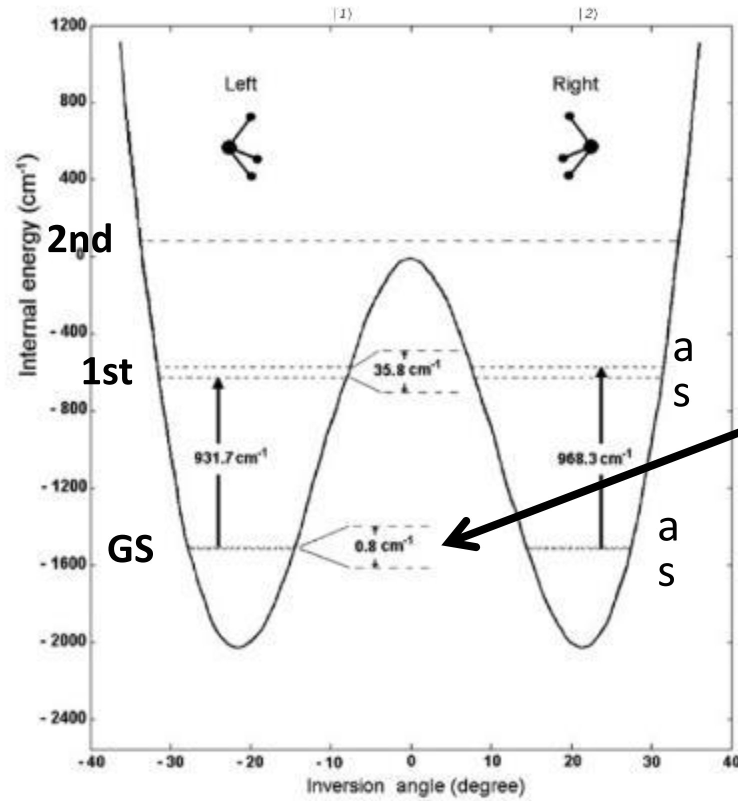
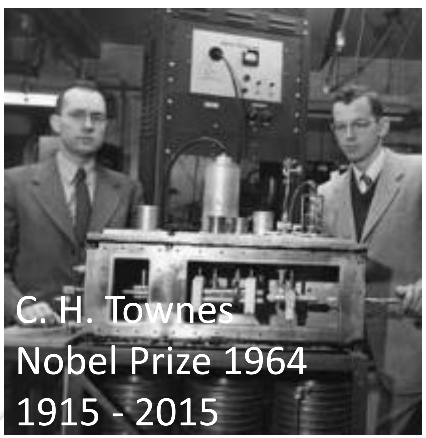



Permanent dipole : 1.4 Debye



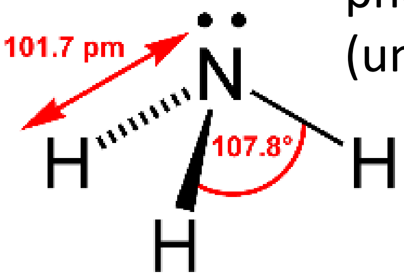

Ground state inversion splitting ≈ 24 GHz

1st Maser (Townes & Gordon, 1954)

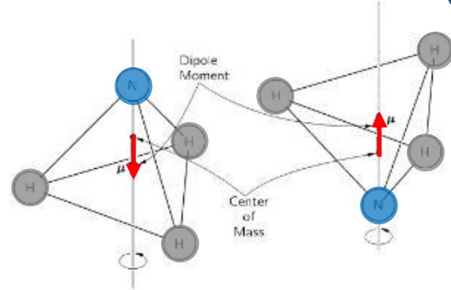



Vibration-inversion double quantum well

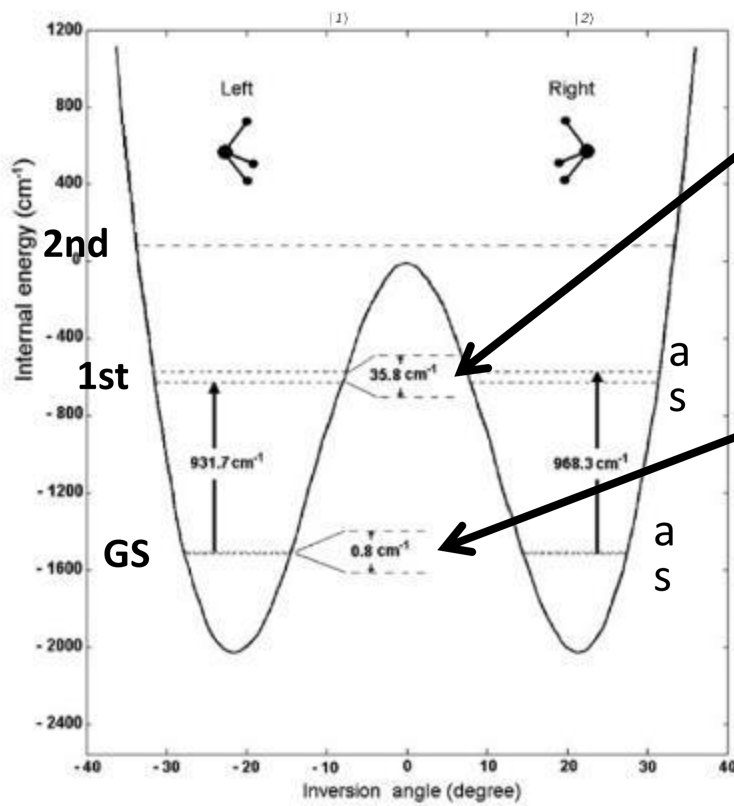
# The ammonia molecule ($NH_3$)




« **Inversion** »
phenomenon
(umbrella)




Permanent dipole :
1.4 Debye



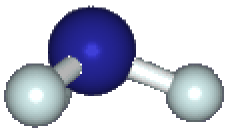

$1^{st}$ $\nu_2$ excited state
inversion splitting
**≈ 1 THz !**

Ground state
inversion splitting
≈ 24 GHz
$1^{st}$ Maser (Townes & Gordon, 1954)


Vibration-inversion double quantum well

# Pure inversion transitions in NH3

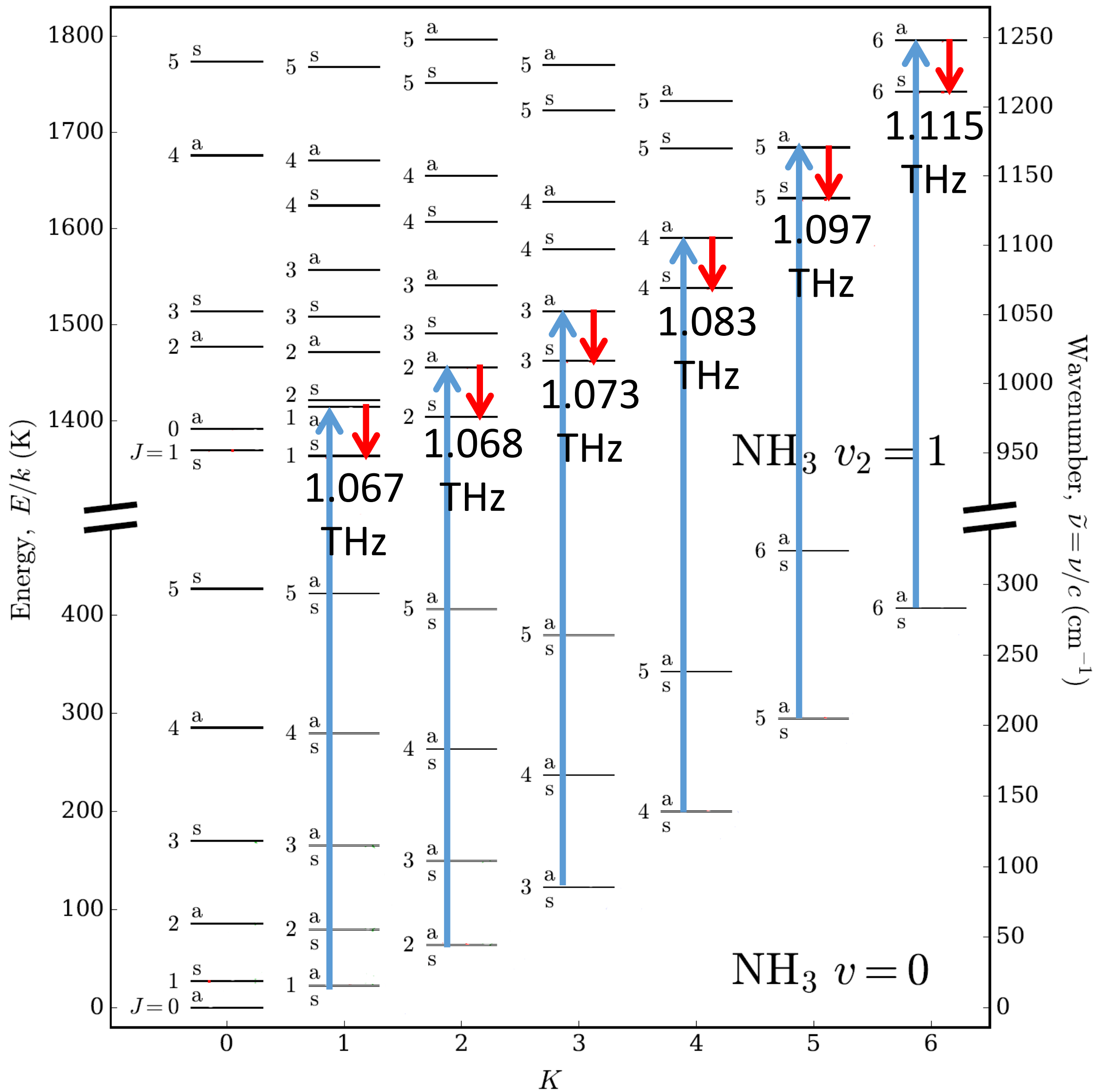
Inversions: ≈ 1 THz



**J = K**

Selection rules for optical transitions:

- $\Delta \mathbf{v_2}$ = 0 or ±1
- $\Delta$**K** = 0
- $\Delta$**J** = 0 or ±1
- **s←a** and **a←s**

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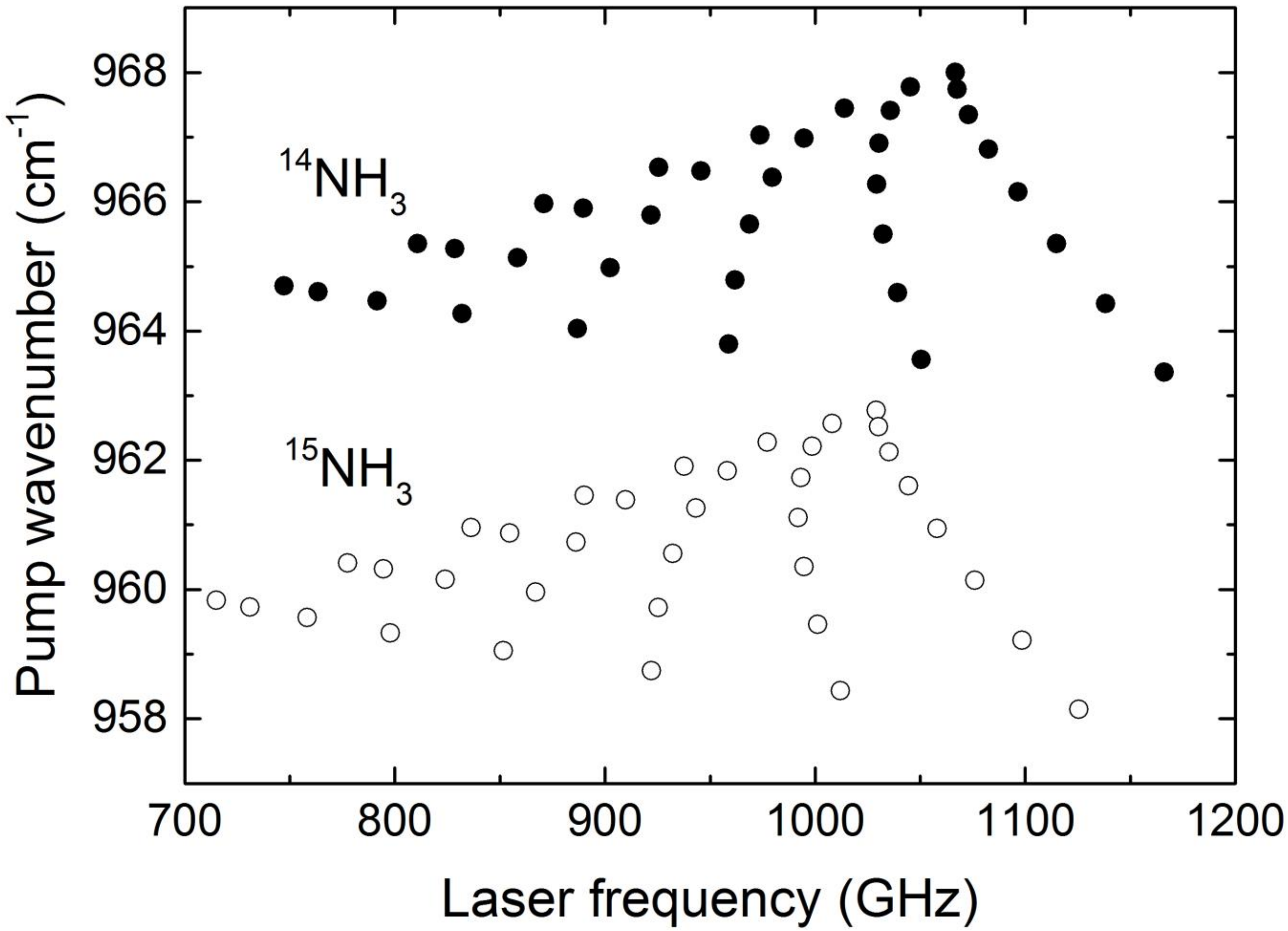
# Pure inversion transitions in $NH_3$

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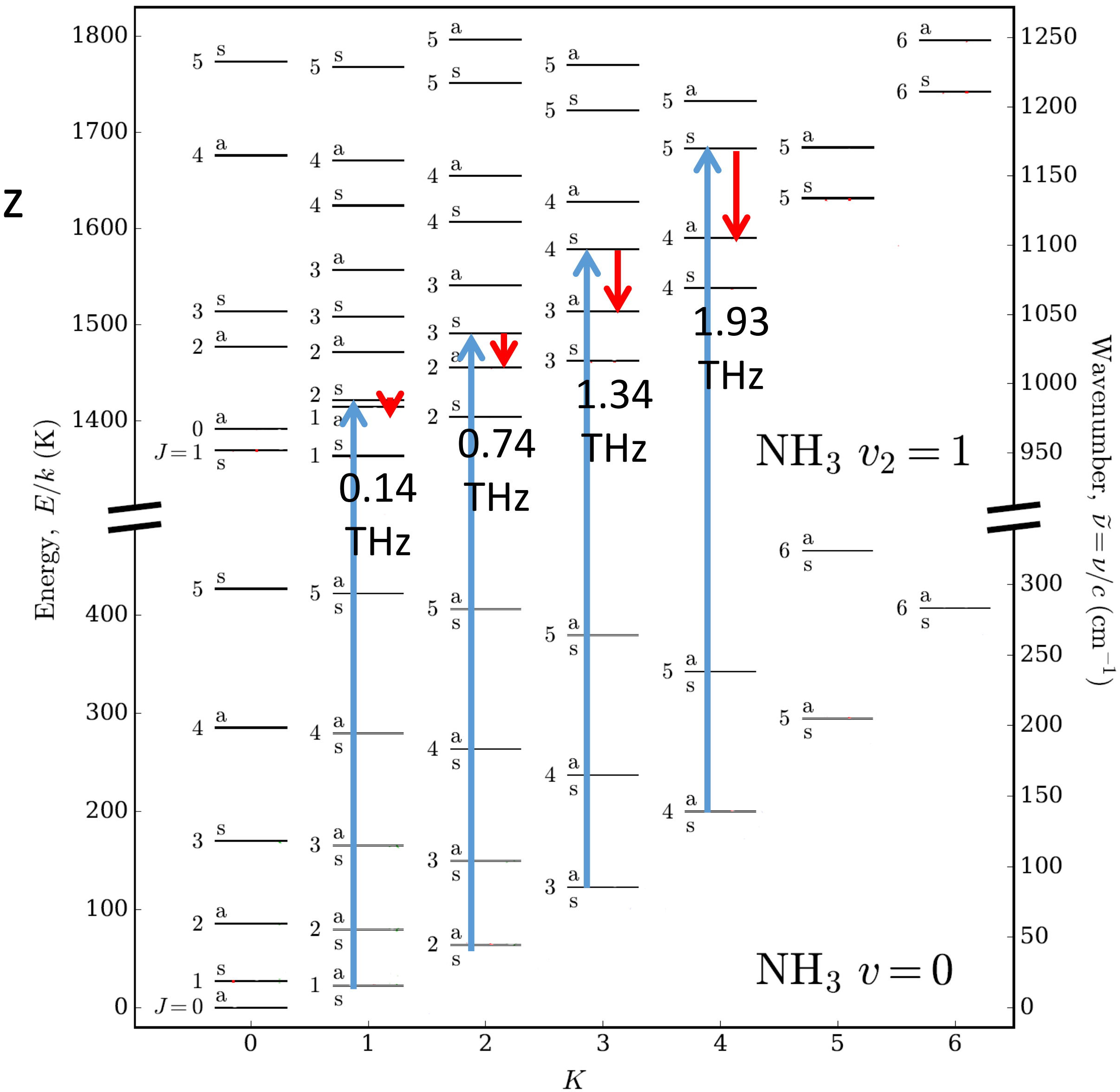
# Rotation-inversion transitions in $NH_3$

Rotations: $2B \approx 0.6$ THz




Selection rules for optical transitions:

- $\Delta v_2$ = 0 or ±1
- $\Delta$**K** = 0
- $\Delta$**J** = 0 or ±1
- **s**←**a** and **a**←**s**

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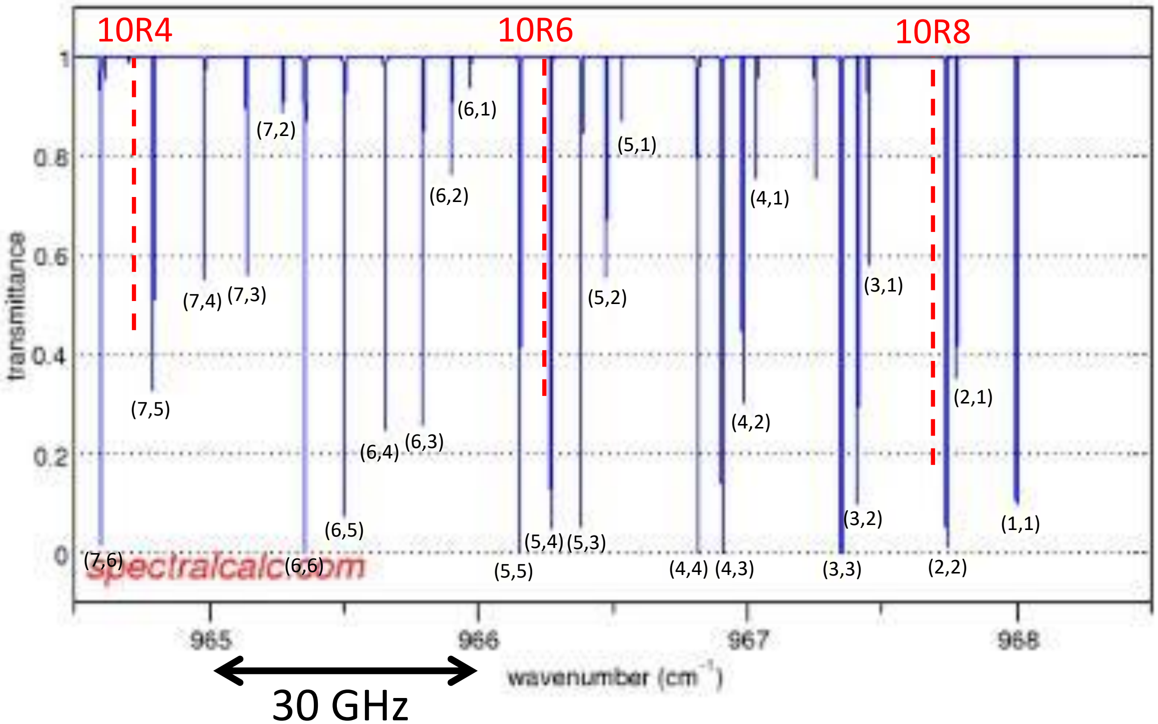
# $NH_3$ MIR pumping with a QCL

MIR absorption spectrum of $NH_3$ around 966 $cm^{-1}$
(simulation)




- Q-branch transitions ($\Delta J = 0$)
- Cell length: 50 cm
- Pressure: 50 µbar

- Dense region
- **Strong lines !**
- FWHM ≈ 80 MHz
- **No coincidence** with $CO_2$ lines !

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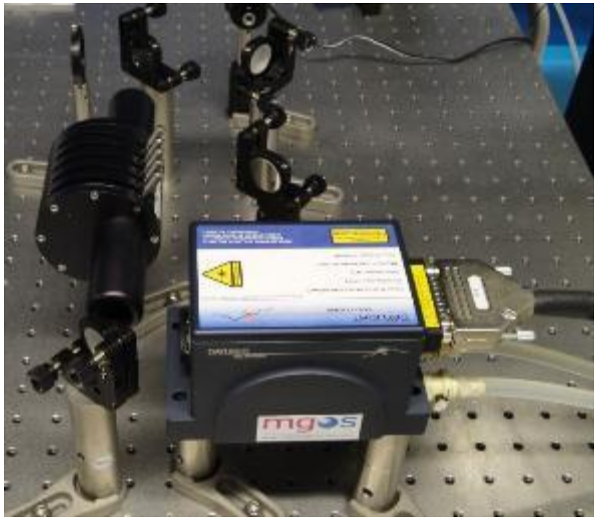
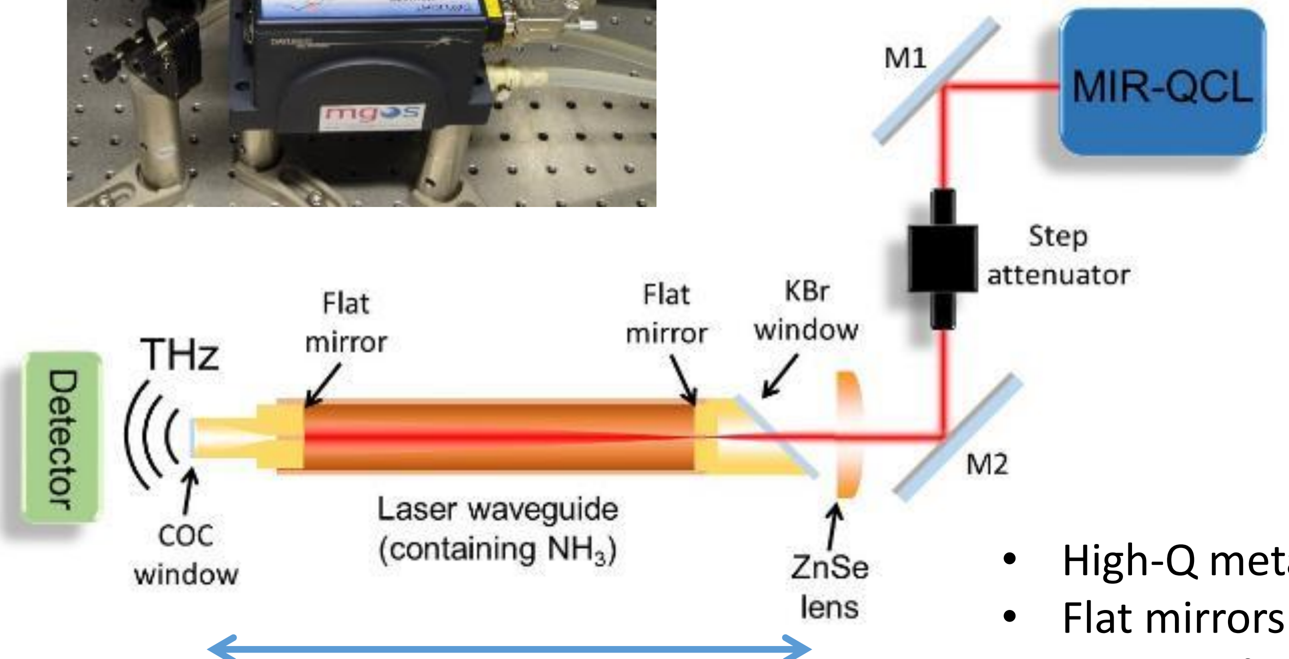
# The first setup



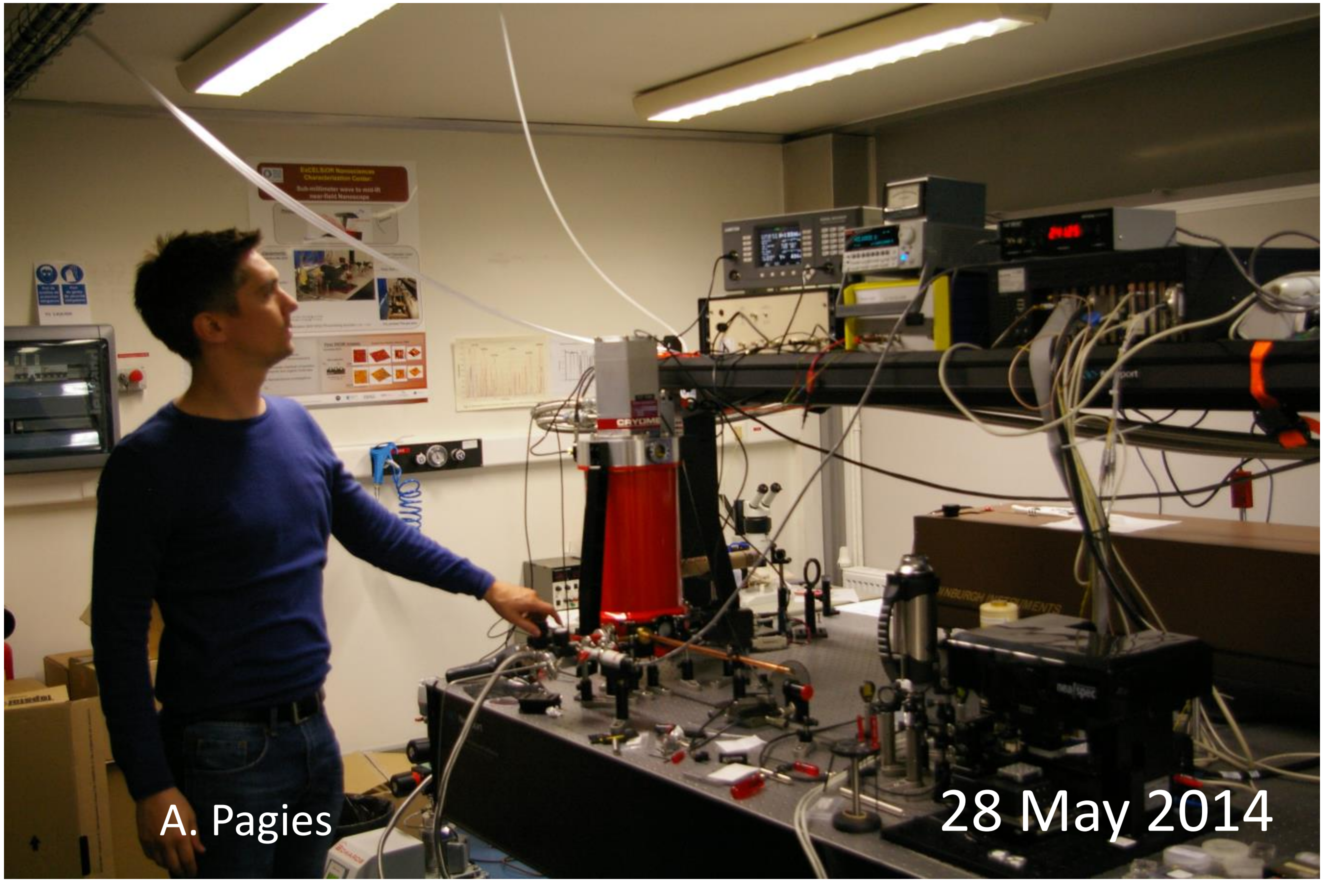
**10-10.5 µm** CW grating external cavity quantum cascade laser (80-100 mW at peak)




50 cm, FSR= 300 MHz

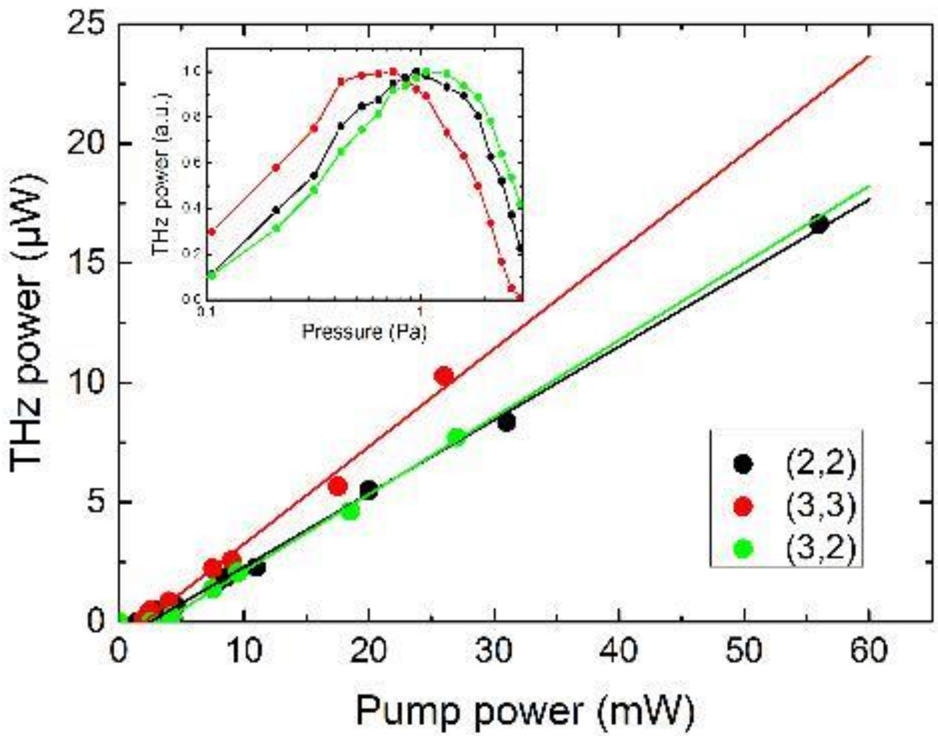
- High-Q metal FP cavity
- Flat mirrors
- **Symmetric** cavity

# The first try




Lasing with 0.1 W pump (instead of 10 W) ???

# First results at 1.073 THz




- Pyro calibrated at PTB
- Threshold is very **low**: 2 mW
- Real power is twice (symmetry) ≈ **40 μW**
- Differential efficiency: 0.8 mW/W (3,3)
- **10 laser lines** have been obtained
- Optimum pressure ≈ 1 Pa

**1st THz generation by a laser pumped by a QCL !**

A. Pagies *et al.*, APL Photonics **1** 031302 (2016)
A. Pagies *et al.*, IRMMW-THz 2016

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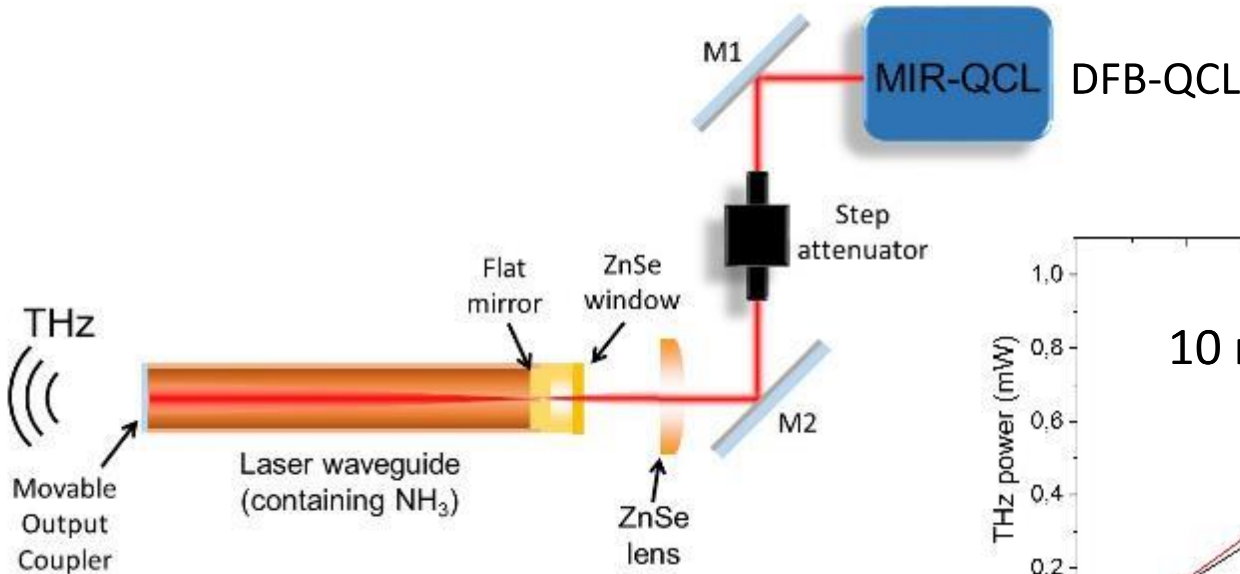
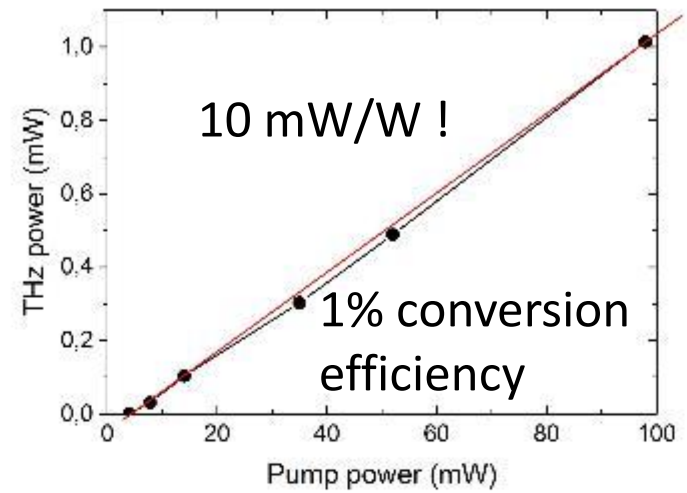
# New design of the cavity




Efficiency for an optimum laser:

$$\eta_{opt} = \frac{1}{2}\frac{\nu_{THz}}{\nu_{pump}}$$

$\nu_{THz}$ = 1 THz and $\lambda_{pump}$ = 10 µm
($\nu_{pump}$ = 30 THz): $\eta_{opt}$ = **1.7 %**

A. Pagies et al., IRMMW-THz 2017
J.-F. Lampin et al., submitted

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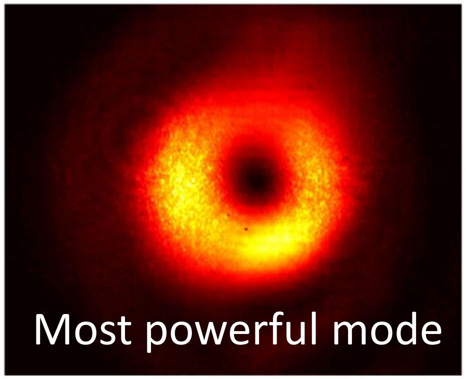
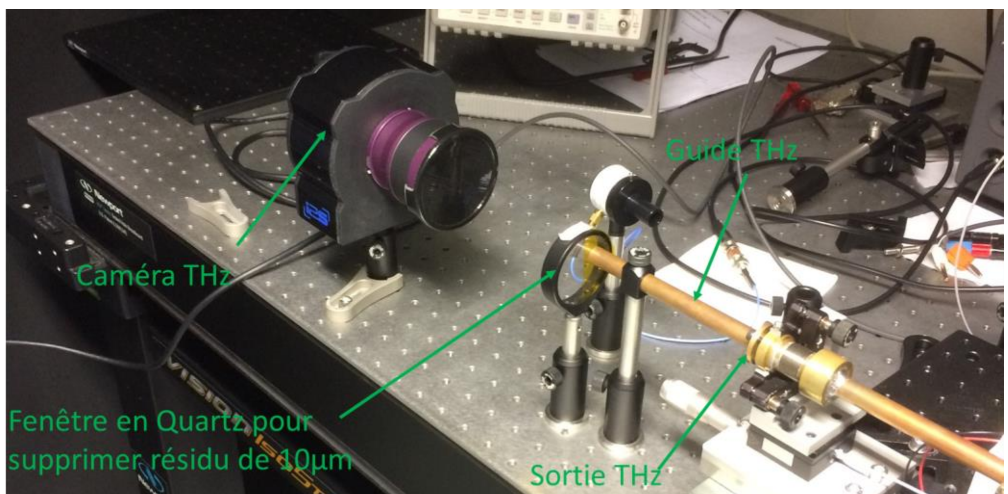
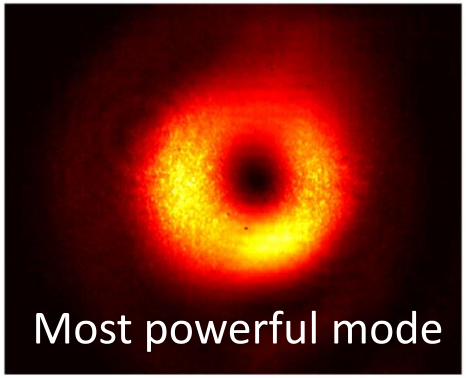
# Mode measurements

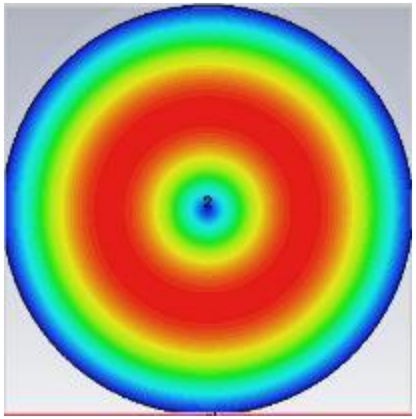



I2S TZcam microbolometer camera



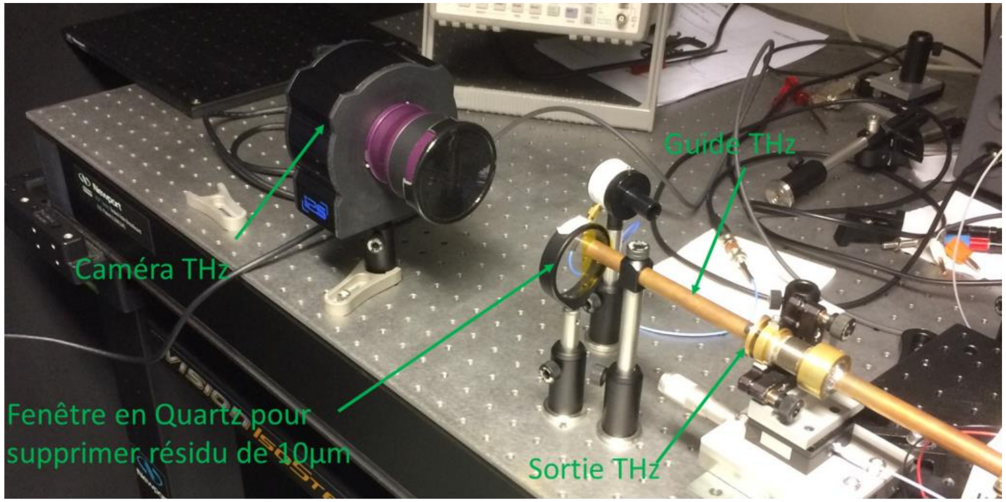
# Mode measurements



$TE_{01}$ mode of the cylindrical metallic waveguide (low loss)

I2S TZcam microbolometer camera

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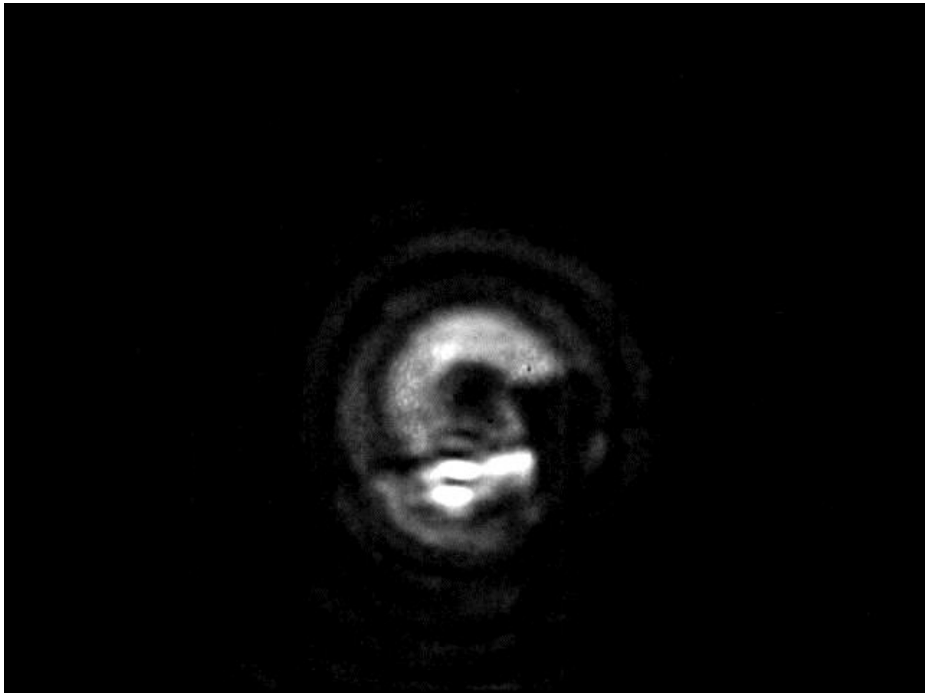
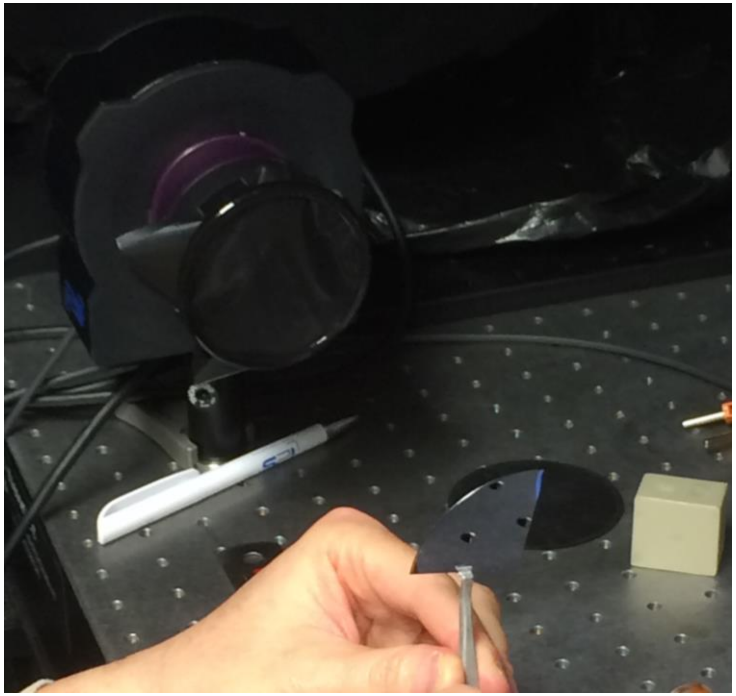
# A THz real-time video





Movie of a HR silicon substrate at 1.073 THz with TZcam
(I2S microbolometer camera)

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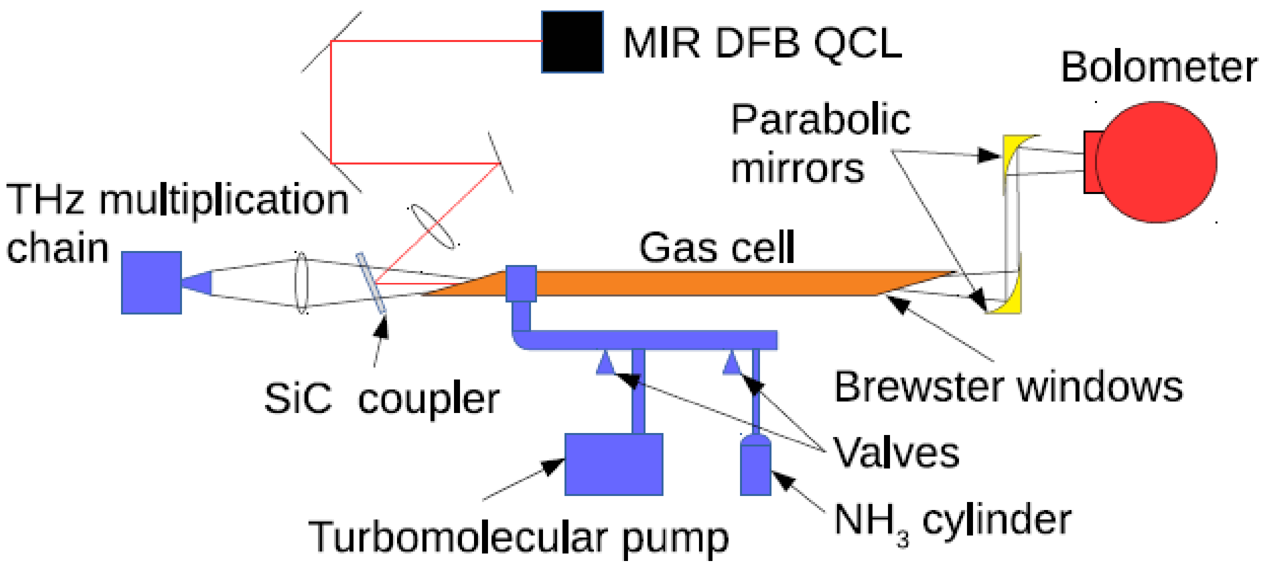
# Gain measurements set-up




10 mm diameter, 50 cm long copper cell with Si Brewster windows

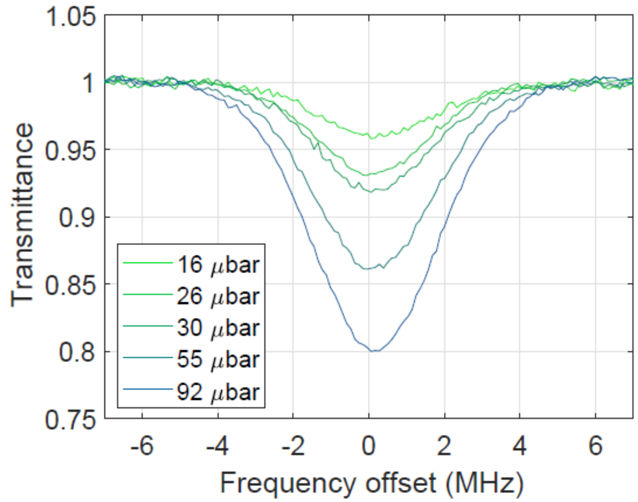
M. Micica et al., IRMMW-THZ 2017
M. Micica *et al.*, Optics Express **26**, 21242 (2018)

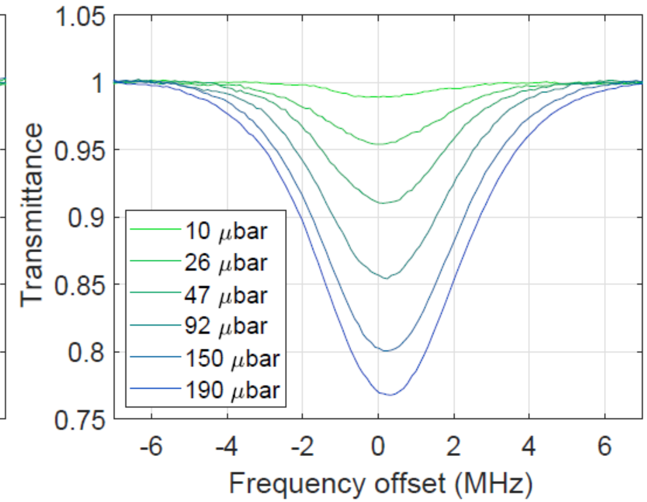
# Gain measurements

(3,3) 1073049.6 MHz

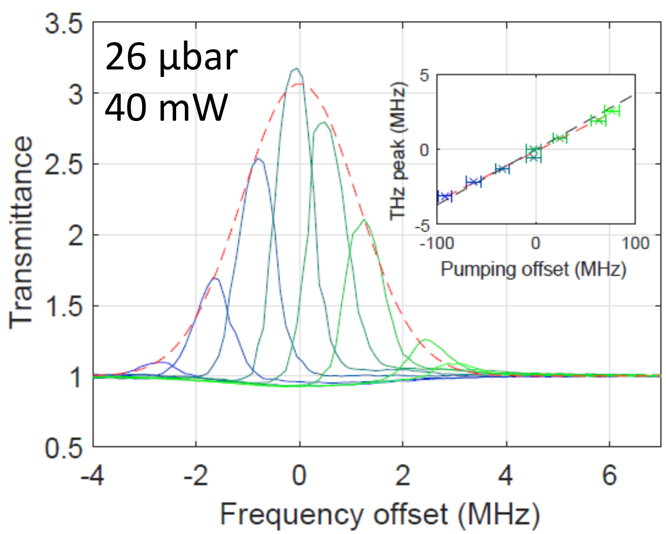
(4,4) 1082592.4 MHz

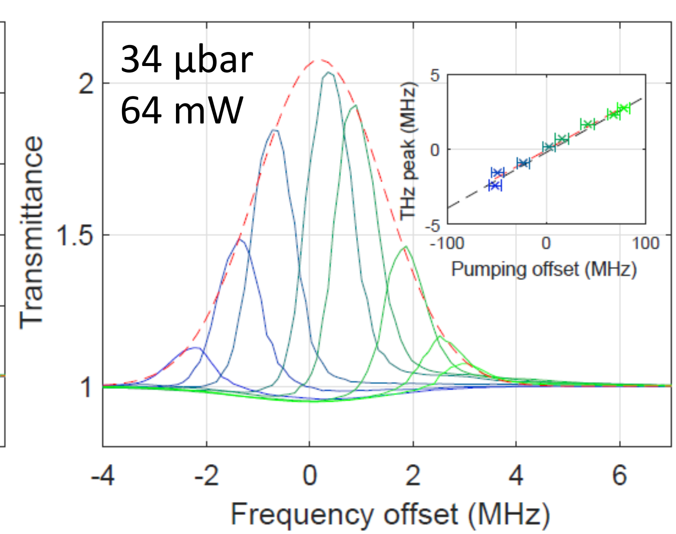
QCL off




QCL on

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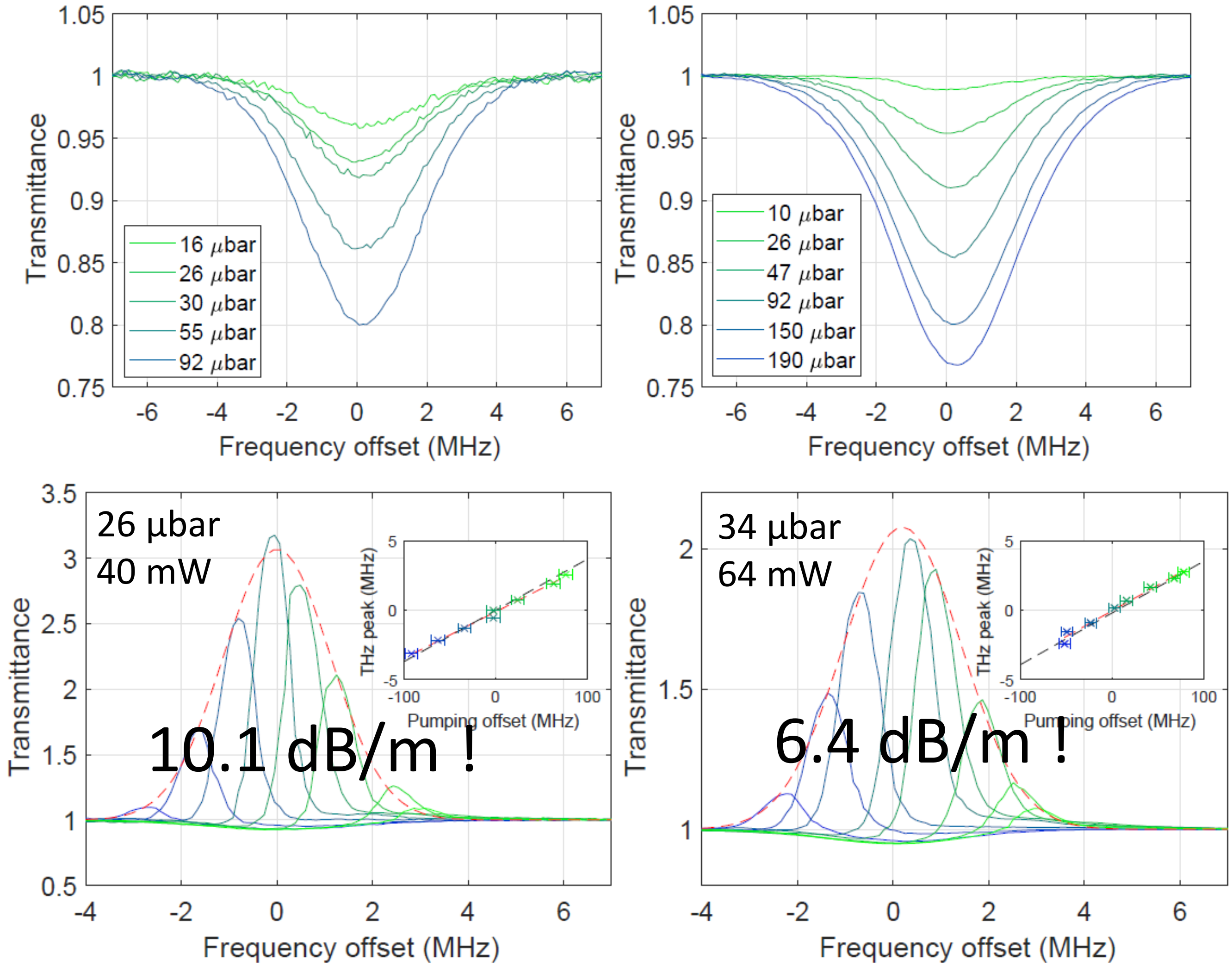
# Gain measurements

(3,3) 1073049.6 MHz

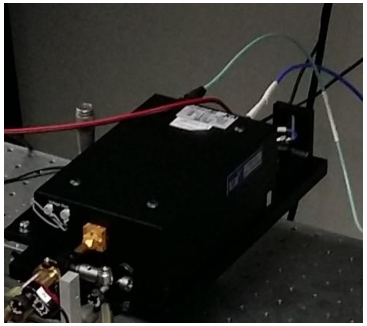
(4,4) 1082592.4 MHz

QCL off

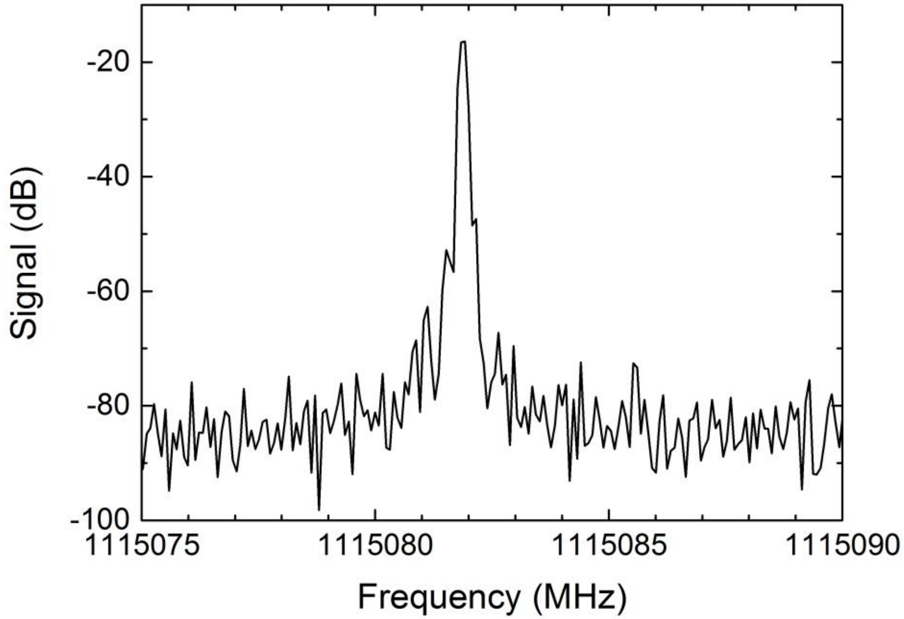
QCL on

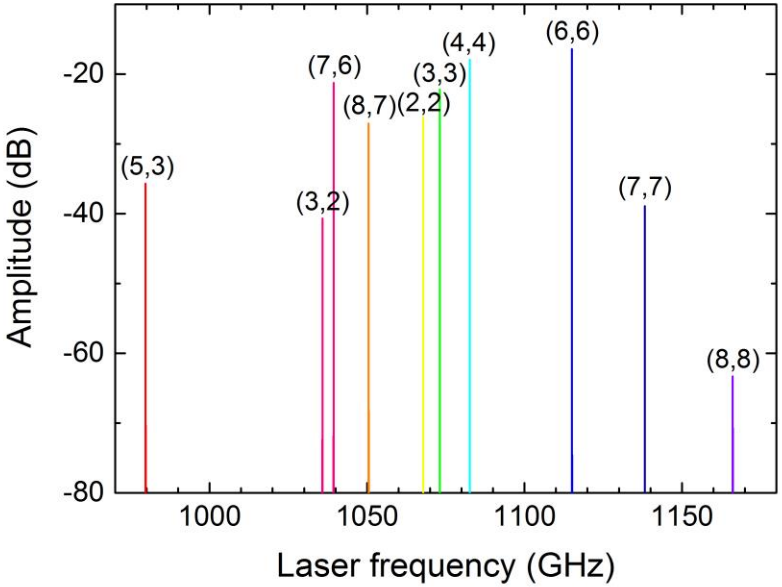


# Other 1 THz inversion laser lines



1 THz VDI subharmonic mixer

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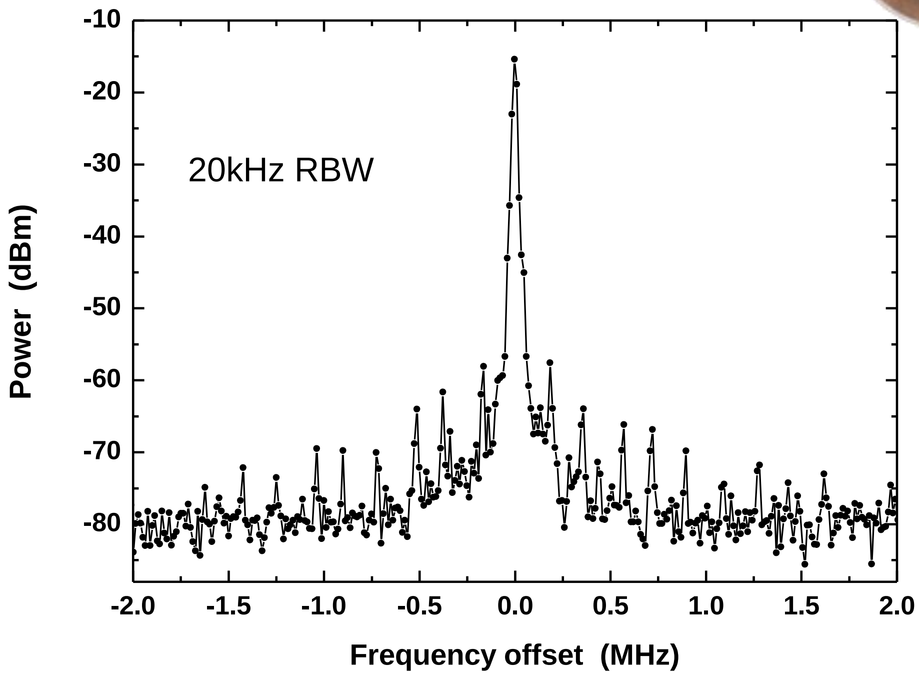
# High-resolution spectra (1.073 THz)

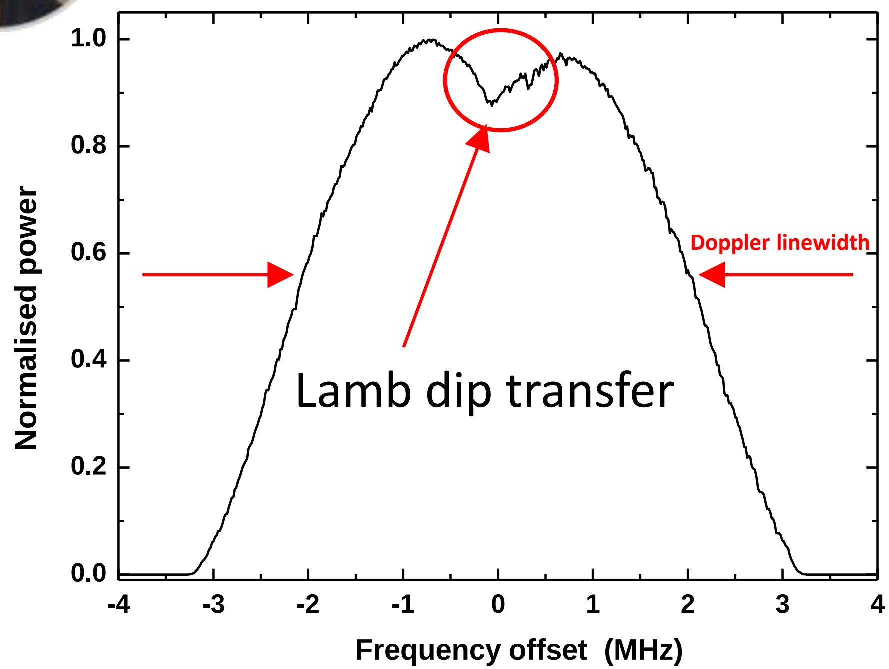
**S. Barbieri *et al.***, Mo-PM2-6-1

**Single-shot**




**Max hold spectrum**

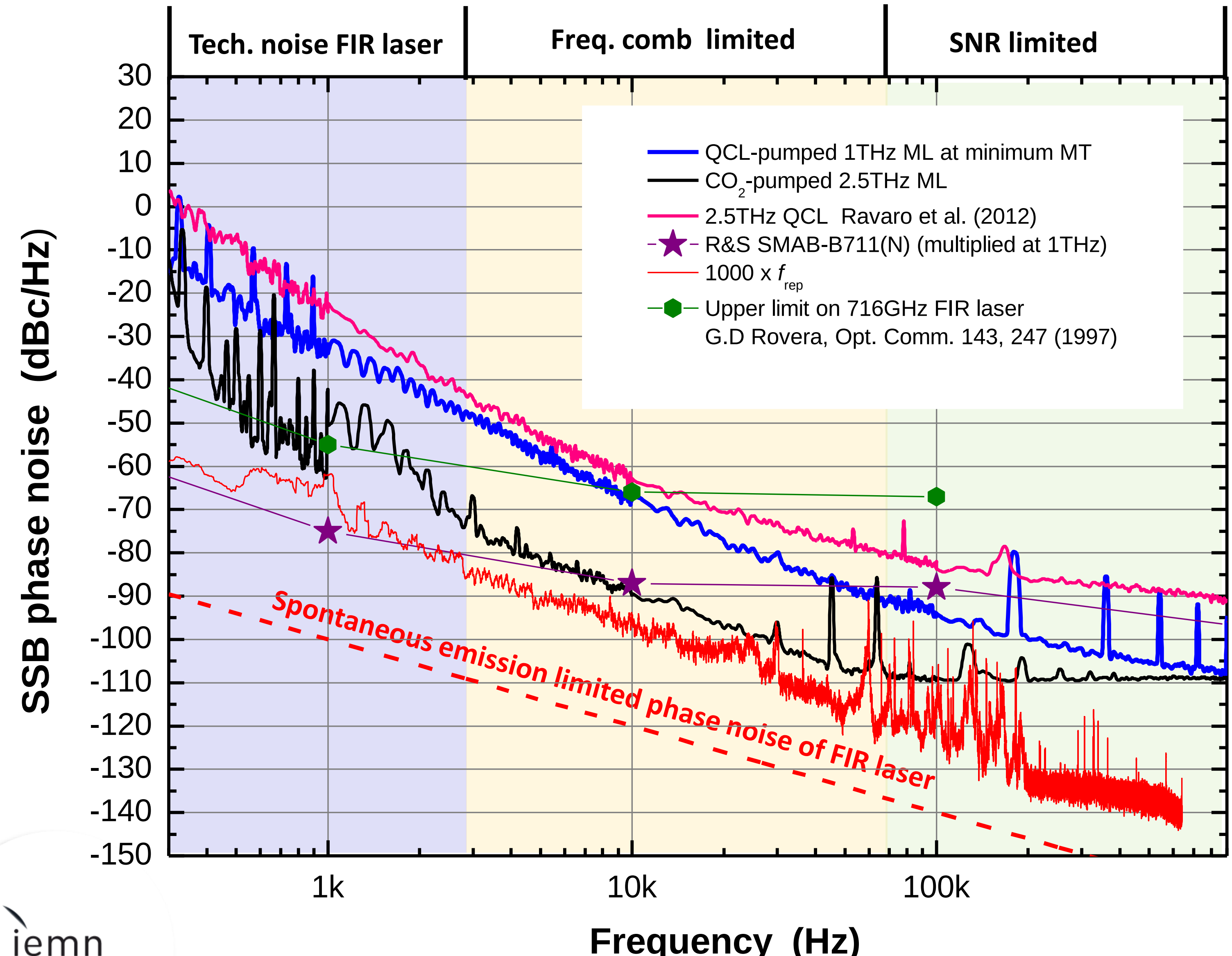
Obtained by scanning the QCL frequency across the pump transition




- <u>Lamb dip transfer</u>: well known effect in THz molecular lasers produced by stronger saturation of the mid-IR transition when the QCL is pumping at the line center

See for example T. A. De Temple *et al.*, J. IR MM Waves, **7**, 1-41 (1983)

# Phase noise

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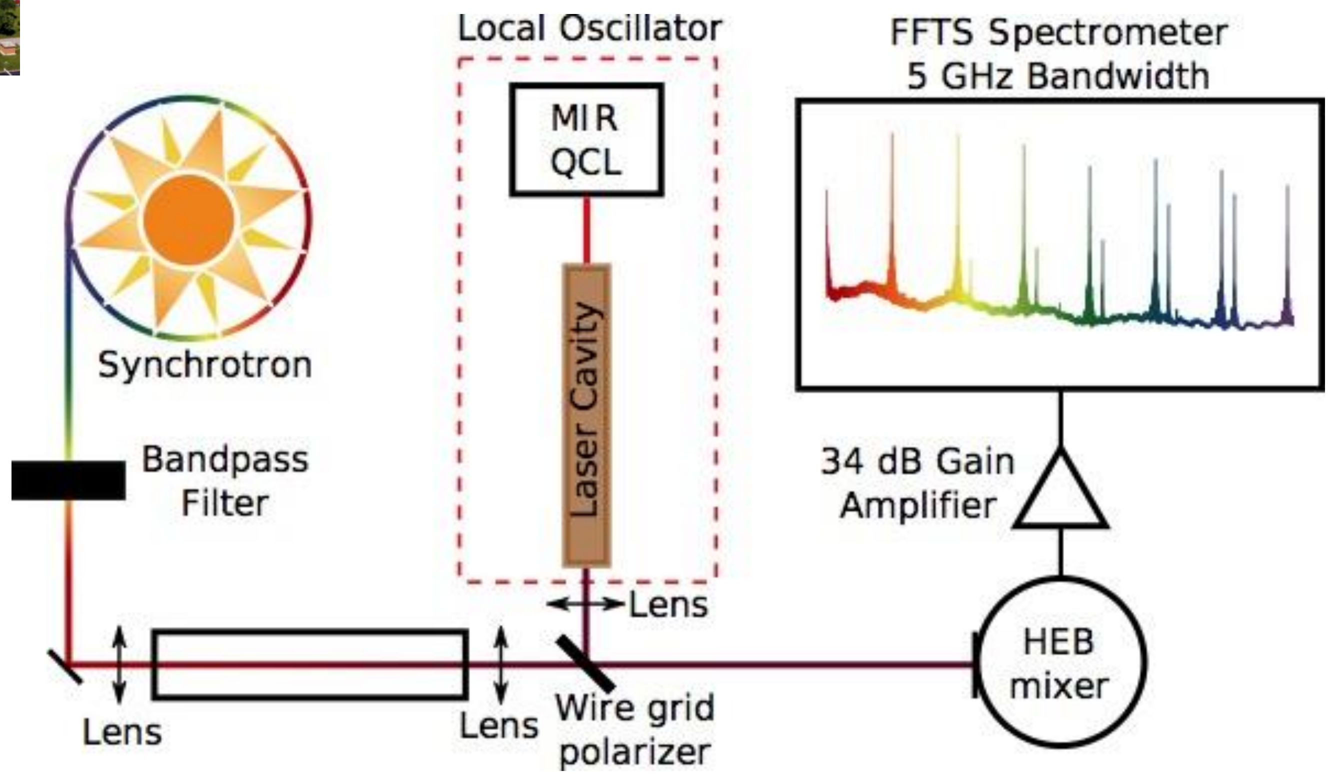
# Local oscillator for heterodyne

SOLEIL synchrotron
FTIR resolution limited to 30 MHz

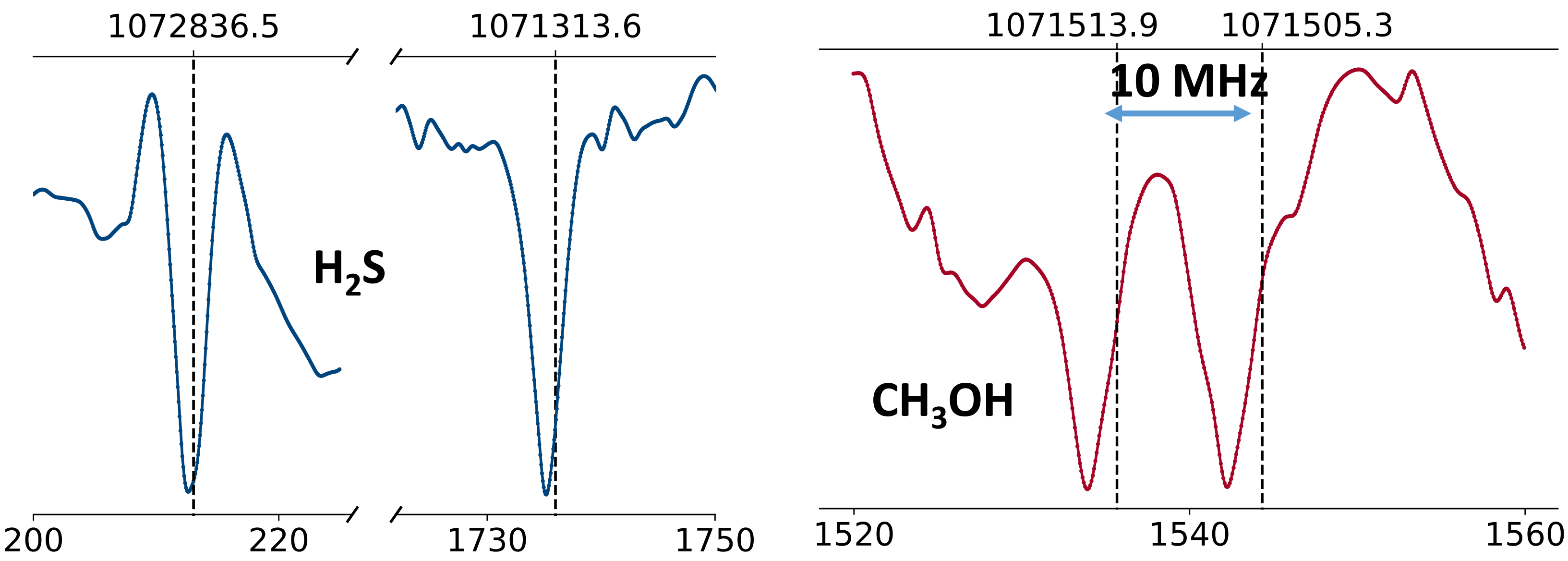
HEROES Project




Heterodyne receiver (**Mouret *et al.***, Mo-AM-7-5)

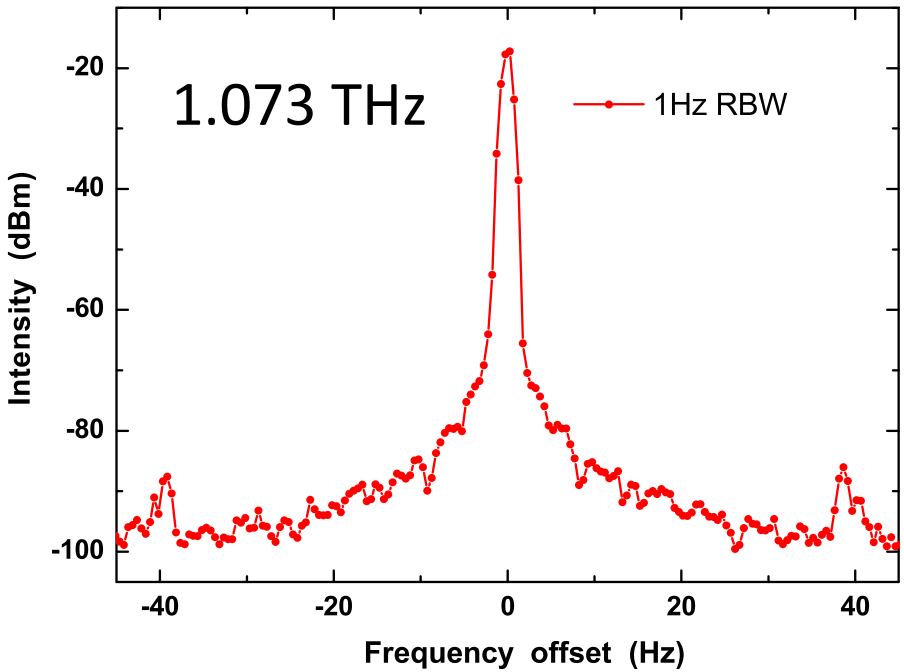
# Experimental molecular spectra

Portions of the IF spectra extracted from the 2.5 GHz full bandwidth

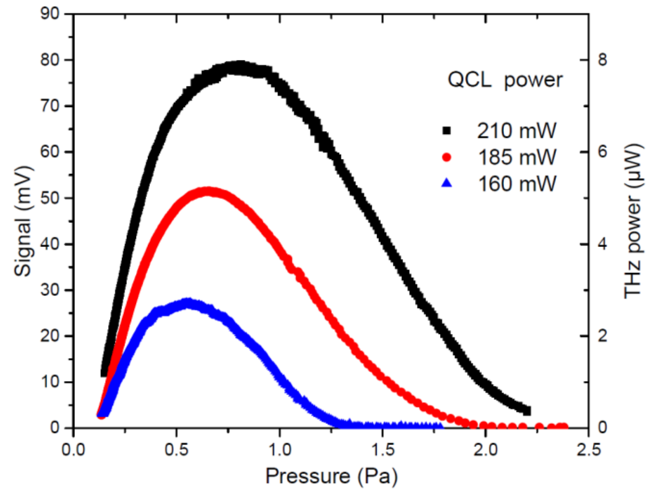



Spectra are recorded in 1 s using the same 1,073,049 MHz LO
The vertical dotted lines indicate the literature frequencies

# Phase lock and rotational lines




July update:
$^{14}NH_3$ **Rotational** laser lines
0.742, 1.34, 3.1 THz




- Phase-locking successful: sub-Hz linewidth
- ~ 0.04rad residual rms phase noise → > 99.5% of BN signal power coherently locked

Rotational line of $^{15}NH_3$ at 4.5 THz
M. Wienold *et al.*
Tu-Po2-104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# Conclusion

- $1^{st}$ THz laser pumped by a MIR QCL & **$1^{st}$ THz molecular laser pumped by a solid-state source**
- **Room temperature, high gain, no high voltage**
- ≈ **30 new laser lines** available around 1 THz with $NH_3$
- 1 mW CW @ 1.073 THz, 10 mW/W, THz WPE = $10^{-4}$
- Rotation-inversion lines: 0.7 – 5 THz

Photomixing: 10 µW @ 1.04 THz, efficiency: $1.6 \times 10^{-5}$

MIR-QCL intracavity DFG: 0.6 mW/$W^2$, CW RT: 14 µW @ 3.4 THz, THz WPE = $0.8 \times 10^{-6}$

**Applications:**

- THz imaging (far- & near-field)
- Local oscillator for heterodyne receivers)
- Metrology (low phase noise, molecular clock ?)
- Magnetospectroscopy
- …


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# THANK YOU !


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Université Polytechnique HAUTS-DE-FRANCE
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centralelille
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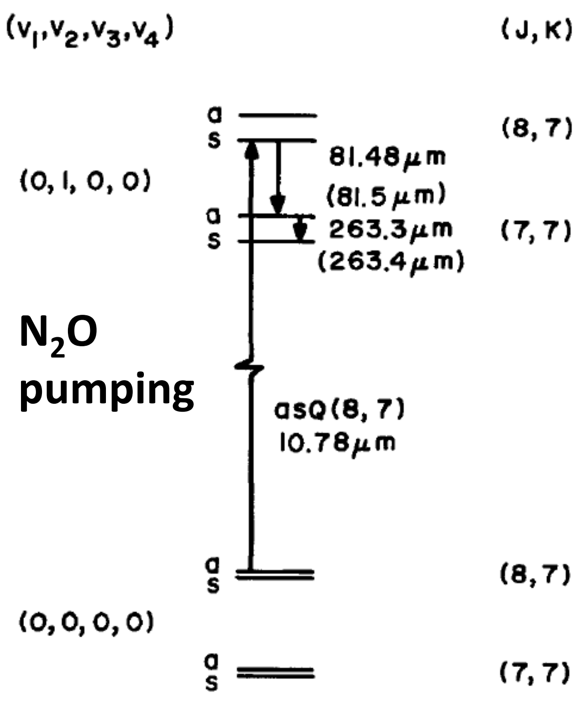
# NH3 was investigated…

Unfortunately **no coincidence** between standard $NH_3$ and standard $CO_2$ lasers !

Chang, Bridges, Burkhardt APL 1970




**$N_2O$ pumping**

**$NH_3$ levels**

- Rotational transition @ 3.7 THz
- Cascade
- $N_2O$ pumping

*Quantum Electronics* **29** (8) 704 – 707 (1999) ©1999 Kvantovaya Elektronika and Turpion Ltd

*CONTROL OF LASER RADIATION PARAMETERS* PACS numbers: 42.55.Lt; 42.62.Eh

**Experimental investigation of a waveguide submillimetre optically pumped laser**

G N Grachev, V F Zakhar'yash, V M Klement'ev, A G Khamoyan

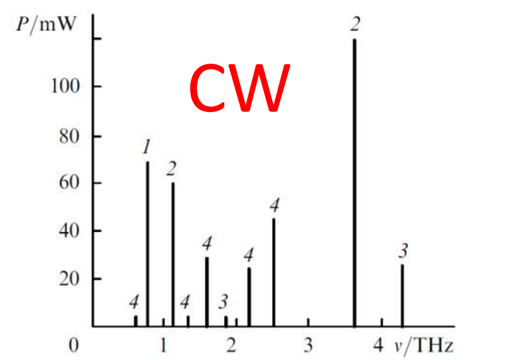



**Figure 3.** Lasing lines obtained by changing the active medium of a laser: (1) HCOOH; (2) $NH_3$; (3) $CH_3OH$; (4) $CH_2F_2$.