



"Quantum Cascade Lasers: past, present and future"

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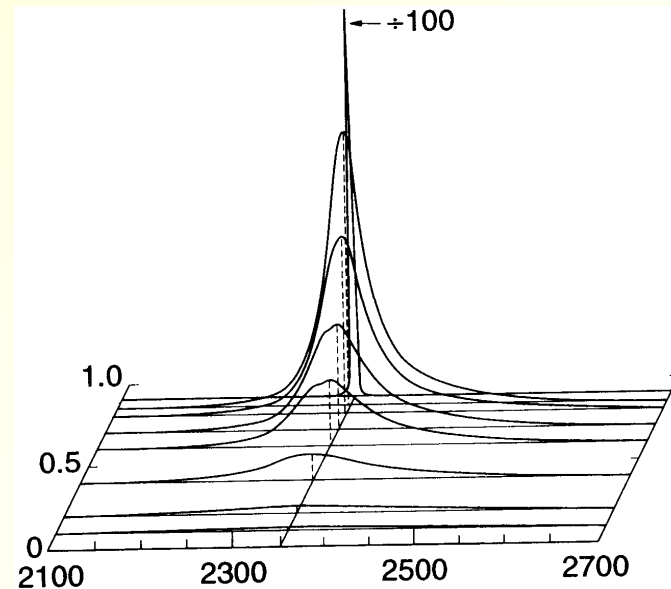
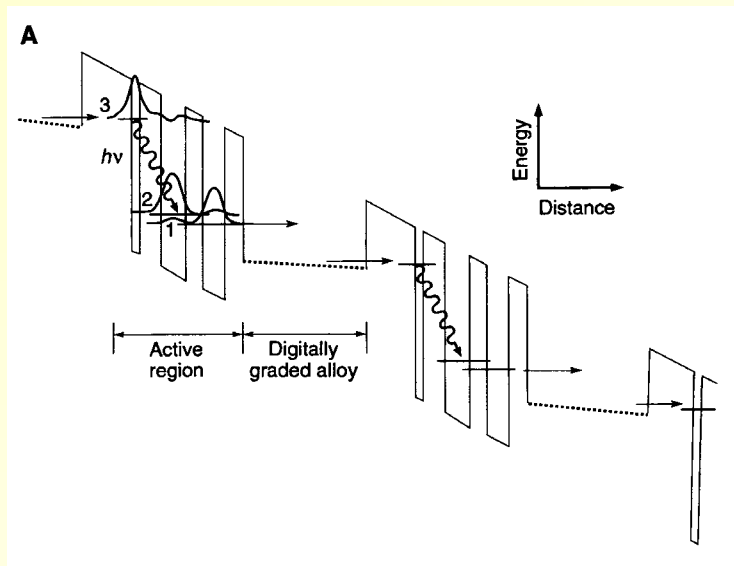
**School of Engineering and Applied Sciences
Harvard University**

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NSF Nanoscale Science & Engineering Center @ Harvard

Quantum Cascade Laser

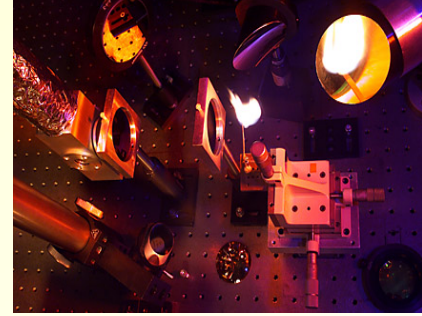
1994: Bell Labs

$T_{\max} = 125\text{K}$ (pulsed), $P_{\max} = 10\text{mW}$, $\lambda = 4.26\mu\text{m}$



J. Faist, F. Capasso, D. L. Sivco, C. Sirtori, A.L. Hutchinson, A. Y. Cho, *Science* **264**, 553 (1994)

QC Laser Highlights



Wavelength agility

- layer thickness determines emission wavelength: 3.4 to 24 μm (AlInAs/GaInAs) and 60 to 250 μm (AlGaAs/GaAs); shortest $\lambda=2.9 \mu\text{m}$ (in Sb based QCLs)
- Multi-wavelength and ultrabroadband operation
- Intracavity nonlinear optics gives further flexibility (e.g. THz DFG)
- Broad single mode tuning: broad gain spectrum and external cavity; DFB QCL Chip

High optical power at RT: several W pulsed, 1.5 W cw with 10% WPE; record WPE 20%

- cascading re-uses electrons

Excellent temperature operation: high T_0 due to parallel subbands and optical phonon limited lifetime

Narrow linewidth (ideally Schawlow-Townes limited) < 100 kHz; stabilized < 10 kHz

Ultrafast: high speed modulation/cut off frequency due to absence of relaxation oscillations

Reliability, reproducibility, long-term stability

Applications: trace gas analysis, combustion & medical diagnostics, environmental monitoring, military and law enforcement

Commercialization: Alcatel-Thales, Hamamatsu, Daylight Solutions, Pranalytica, Alpes Lasers, Maxion, Laser Components, Nanoplus, Cascade Technologies, Q-MACS, Fraunhofer Institute, PSI, Aerodyne, etc. and a growing number of foundries growing QCL wafers

Trends in QCLs



- Band-structure engineering and waveguide design as a driving forces for major performance improvements will have diminishing returns since QCLs are a maturing technology at least in the mid-infrared

- Major improvements can still be expected in areas such as wall plug-efficiency, temperature performance (for THz QCLs), high power single mode operation and broadband single mode tunability.

They will mostly emerge from a combination of smart engineering solutions, incremental design improvement, increased physical understanding at the level of transport, improved modeling

- Ultimately market penetration beyond niche areas will be the measure of QCL success: there are encouraging signs as larger companies (Hamamatsu, Corning, etc) are getting into the act. Killer application is needed (breadth analysis?) with prices down to <1\$ K per unit

- In this talk I will explore research opportunities other than the above related to fairly unexplored areas in materials, physics, devices, systems, atmospheric sciences and climate change

Challenges and Frontier topics

- ◆ **Materials:** *High performance nitride based QCLs for the near infrared and the THz?*
- ◆ **Physics:**
 - QED effect on electronic transport?*
 - Fundamental understanding of short time scale non linear dynamics of QCLs: are ultrashort pulse modelocked mid-ir lasers and optical combs possible?*
- ◆ **Devices:** *Beam-Engineering of QCLs: increasing the functionality with plasmonics and metamaterials?*
- ◆ **Systems:**
 - New high brightness spectrometers*
 - Instrumentation for climate change studies*



High temperature nitride based QCLs for the near infrared?

- Fighting low oscillator strength, broad transitions and short lifetimes with low loss to achieve reasonable thresholds
- Take advantage of low free carrier loss and high LO phonon energy (90 meV) for high slope efficiency and high T_0

$$J_{th} = \frac{\epsilon_0}{4\pi q_0} \frac{1}{\tau_3 \left(1 - \frac{\tau_2}{\tau_{32}}\right)} \left(\frac{\lambda n L_p 2\gamma}{\Gamma z_{ij}^2} (\alpha_m + \alpha_w) \right)$$

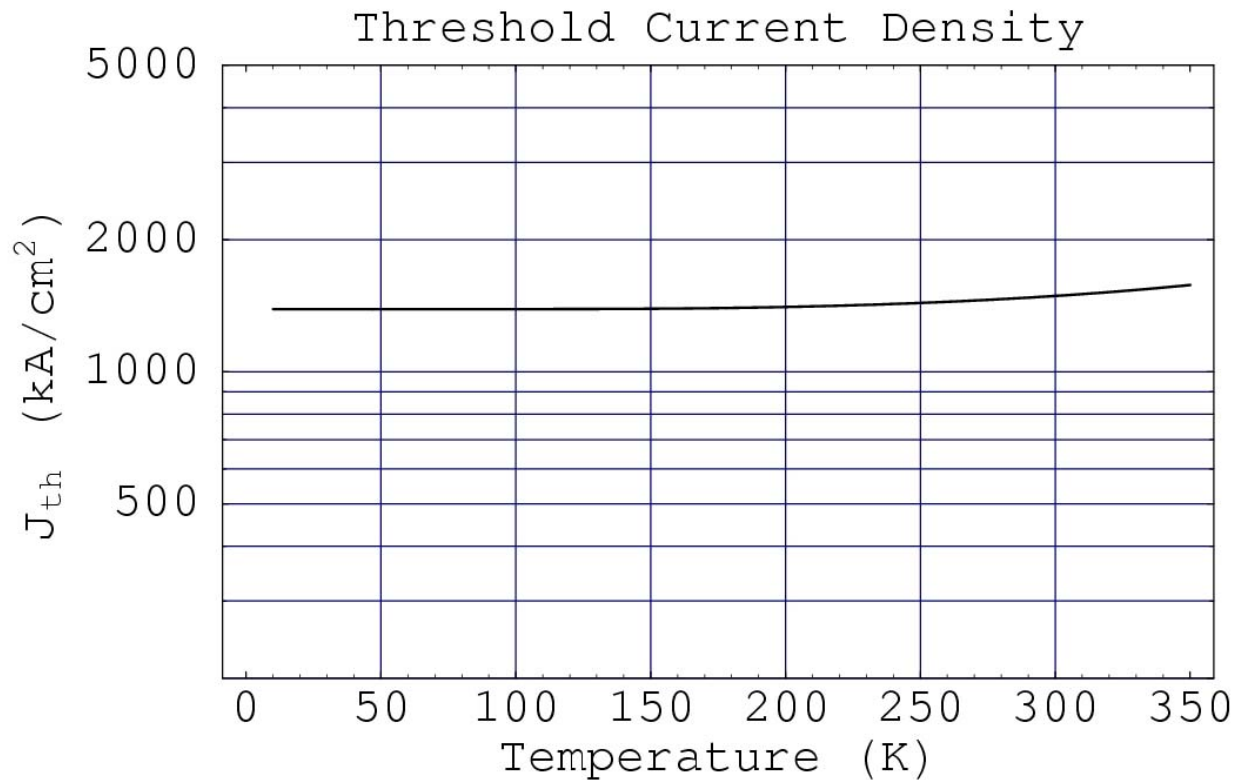
$$\frac{z_{ij}^2}{\lambda} \sim f \sim \frac{1}{m^*}$$

	InGaAs	GaN
m^*	0.043	0.2

$$\alpha_{fc} = \frac{q_0^2}{\epsilon_0 c n_{refr} m^*} \frac{N}{\omega^2 \tau_{//}}$$

Jerome Faist

Temperature performance of $\lambda = 1.5 \mu\text{m}$ Nitride QCL



$$\alpha_w = 0.33 \text{cm}^{-1}$$

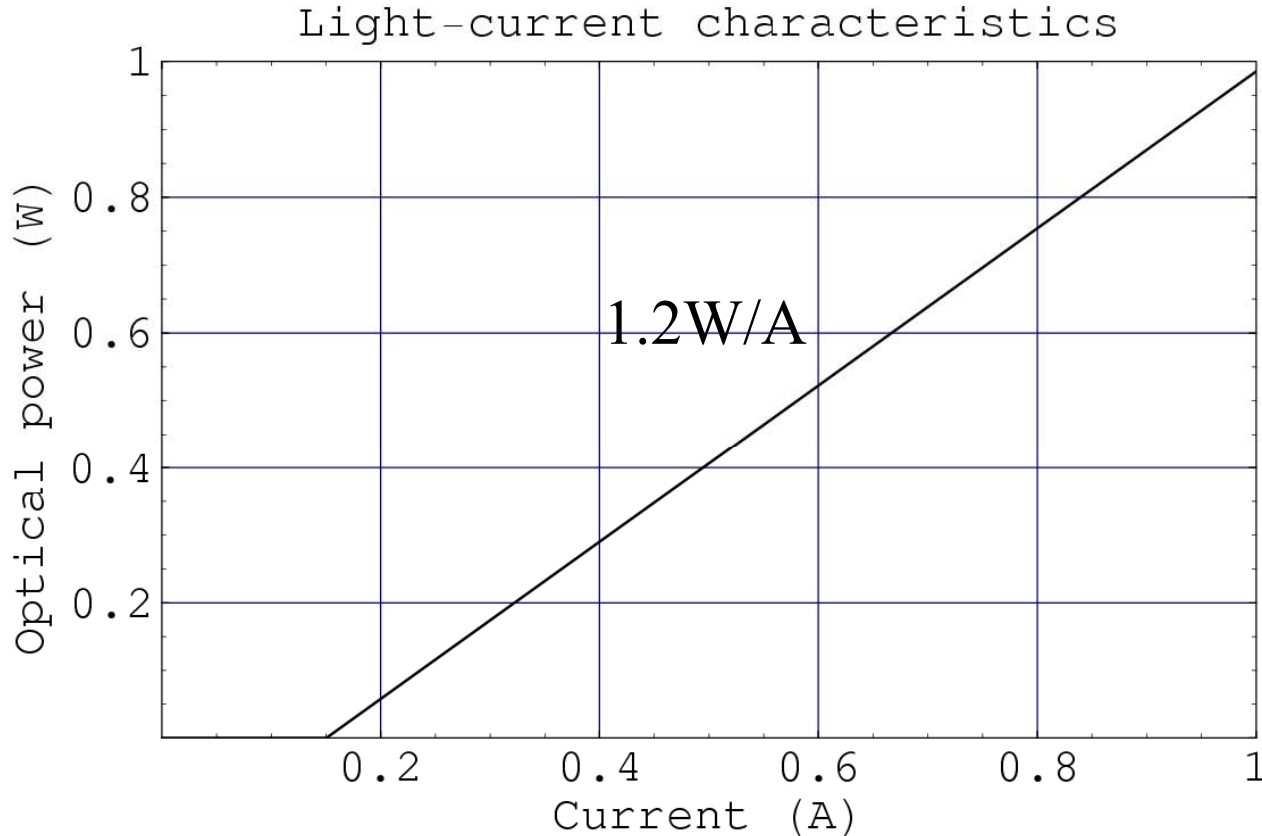
$$L_{cav} = 1 \text{cm}$$

$$R1 = 0.98; R2 = 0.8$$

$$\alpha_m = 0.11 \text{cm}^{-1}$$

Jerome Faist

Light-versus current characteristics of 1.5 μm wavelength Nitride Lasers



$$2\gamma = 48\text{meV}$$
$$\tau_{up}(0K) = 200\text{fs}$$
$$N_p = 8$$
$$\Gamma = 0.5$$

High power and temperature independent performance due to low free carrier loss, large phonon energy

J. Faist

Ultrafast laser? No!

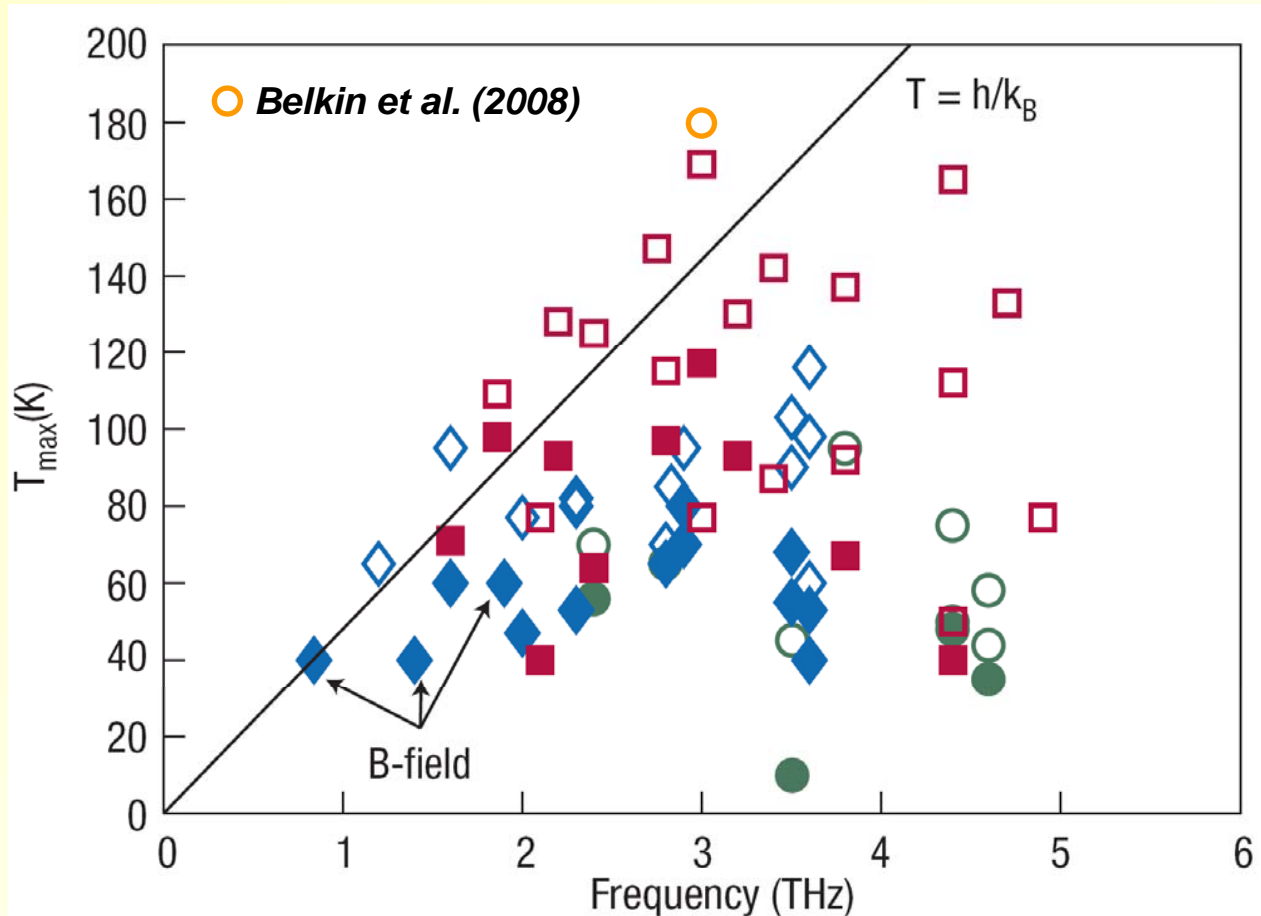
- Inversion quenched very fast (< 200fs)
- However, lifetime of photon in the cavity is very long because of very low losses

$$\tau_{phot}^{-1} = \frac{c}{n} \alpha_{tot}$$

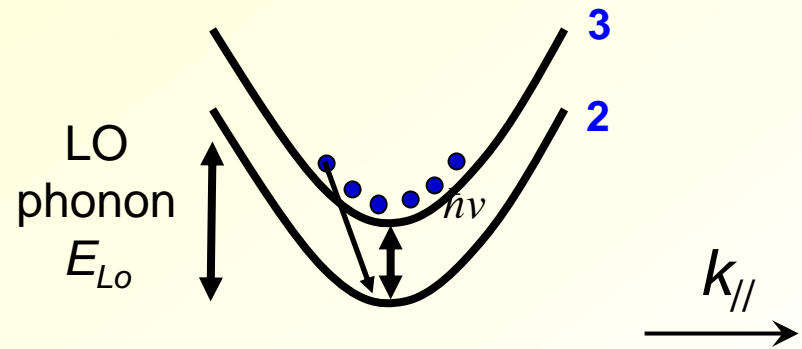
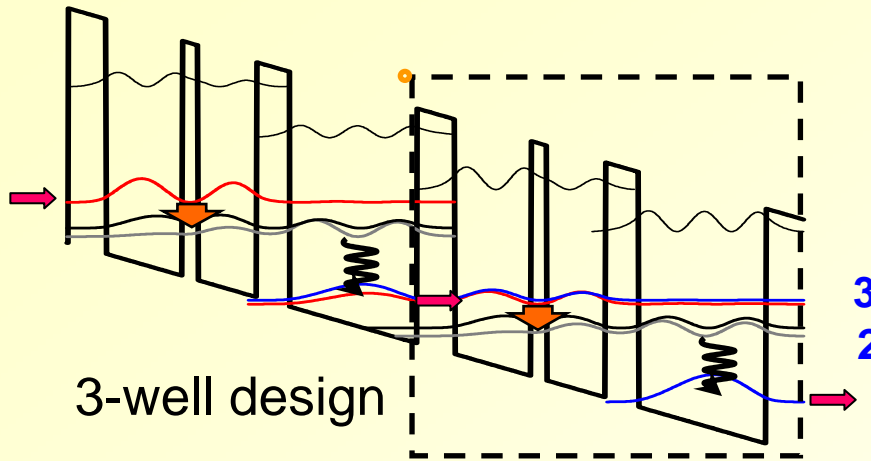
$$\tau_{phot} = 200ps$$

THz QCL temperature performance

THz QCL summary: 2002-2008

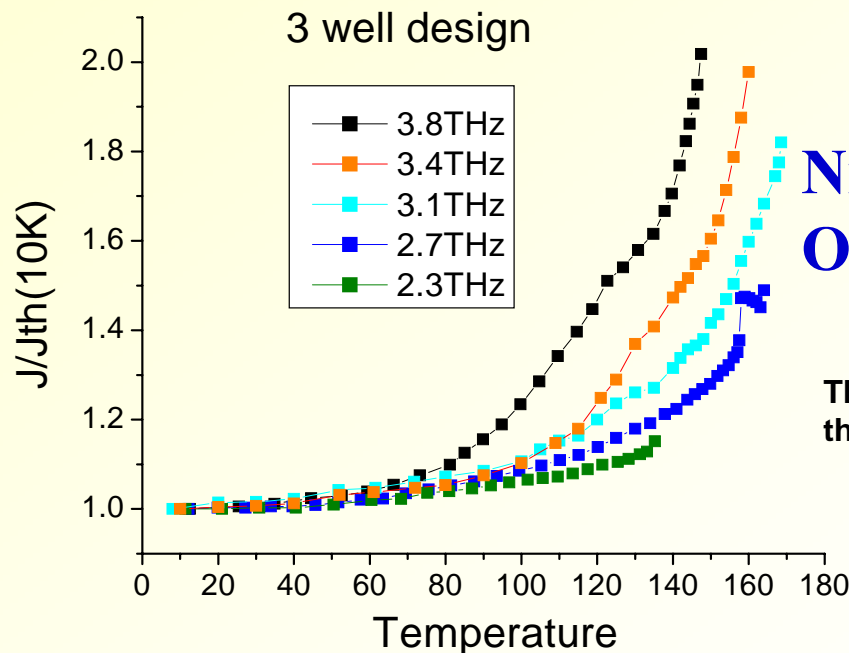


Temperature dependence of threshold in THz QCLs: evidence of thermally assisted LO-phonon emission



$$\exp(-(E_{LO} - h\nu) / kT)$$

H. Luo, APL, **90**, 041112, 2007



Nitride based THz QCL: High Operating temperature, high T_0 !

The data were normalized to the threshold at 10K

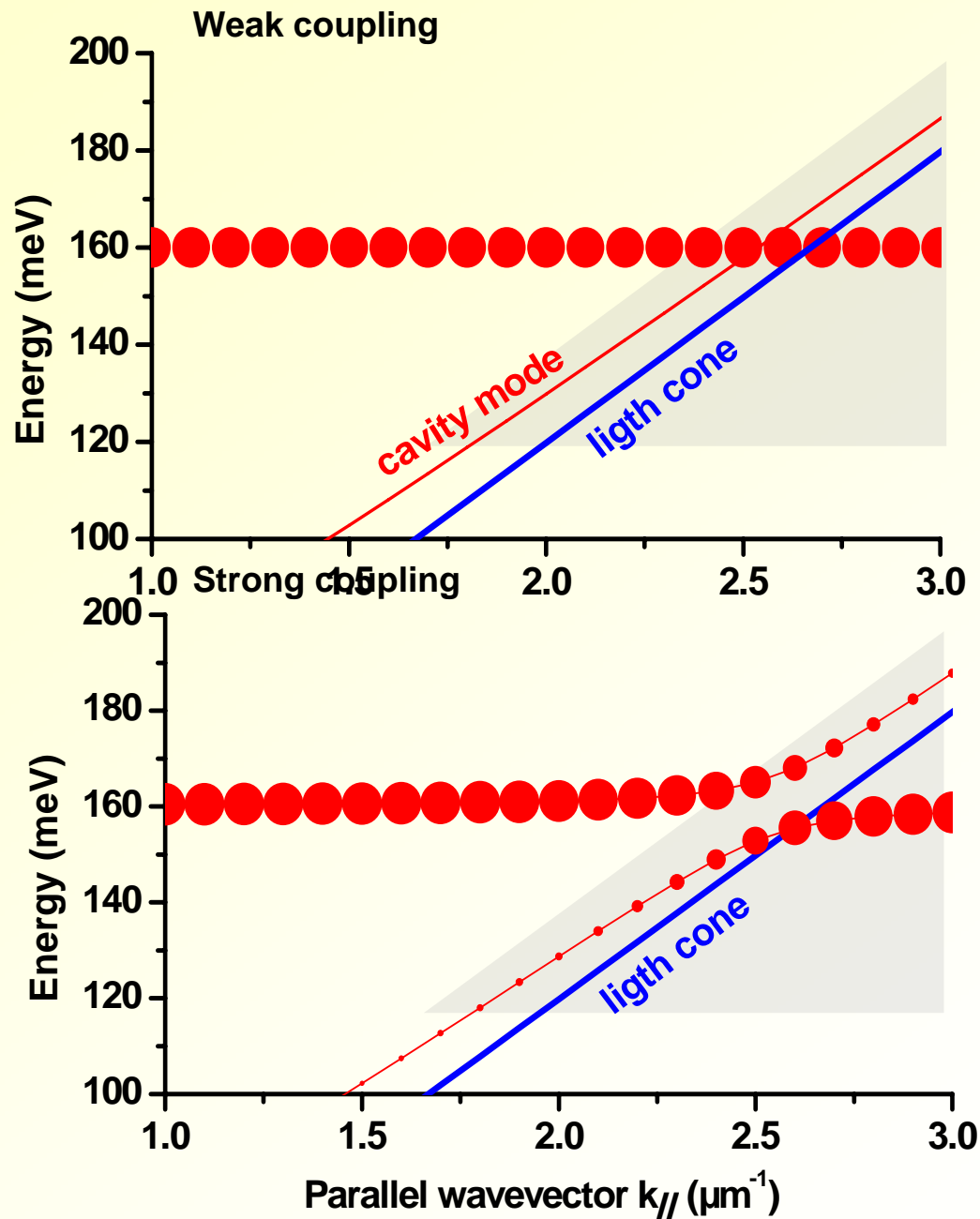
B. S. Williams, Nature Photonics, **1**, 517, 2007

R. Colombelli's group, Harvard group, and E. Linfield's group collaboration

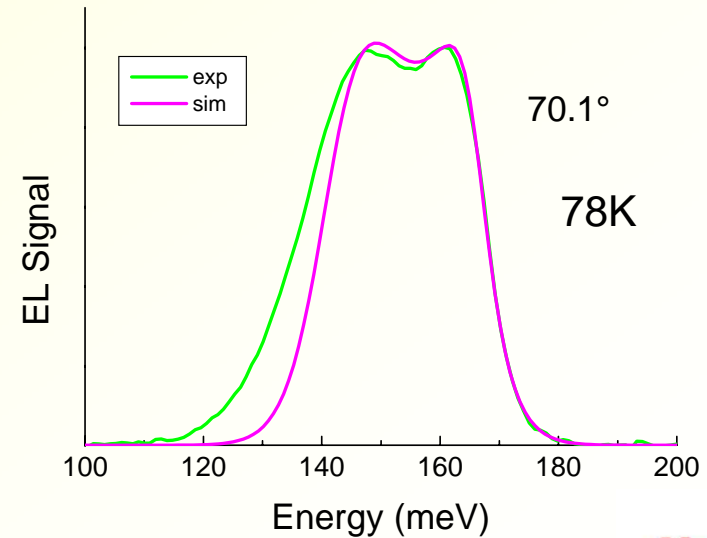
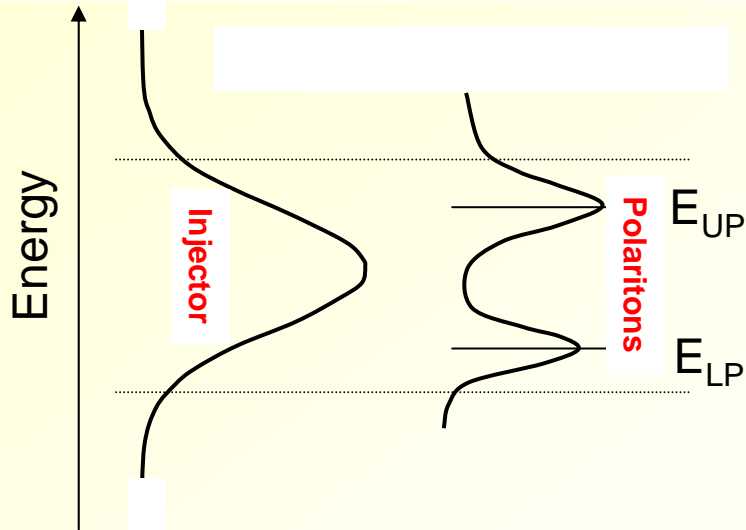
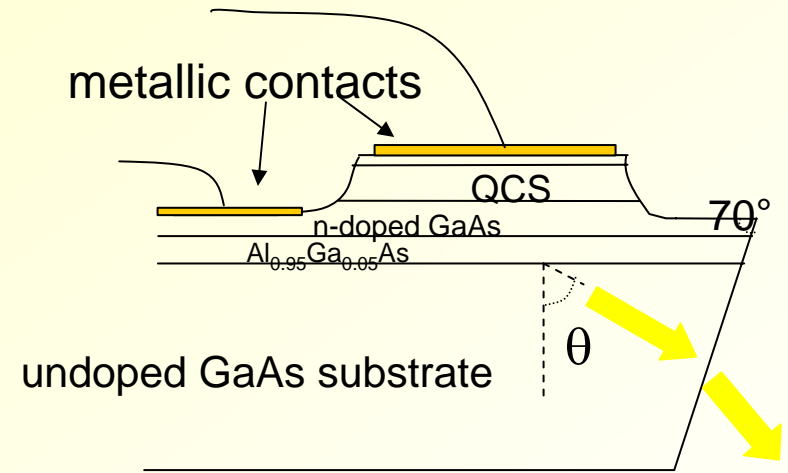
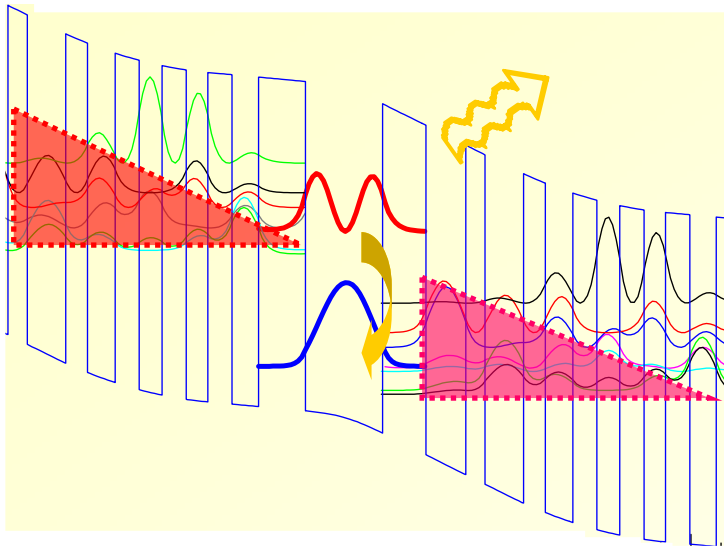
Frontiers of QCL Physics

- ◆ ***Tunneling, QED effects and transport: are QED effects observable in transport ?***
- ◆ ***Fundamental understanding of short time scale dynamics of QCLs: are ultrashort pulse modelocked mid-ir lasers and optical combs possible?***

Cavity QED with intersubband polaritons



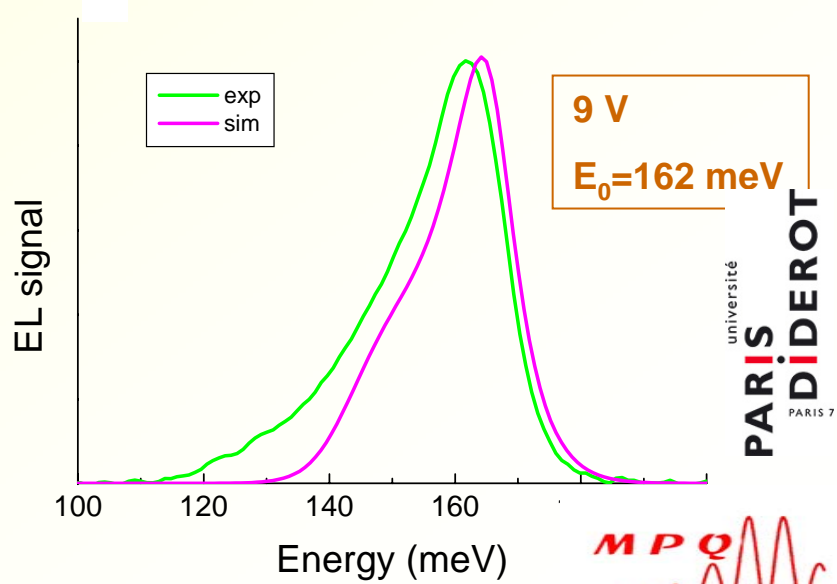
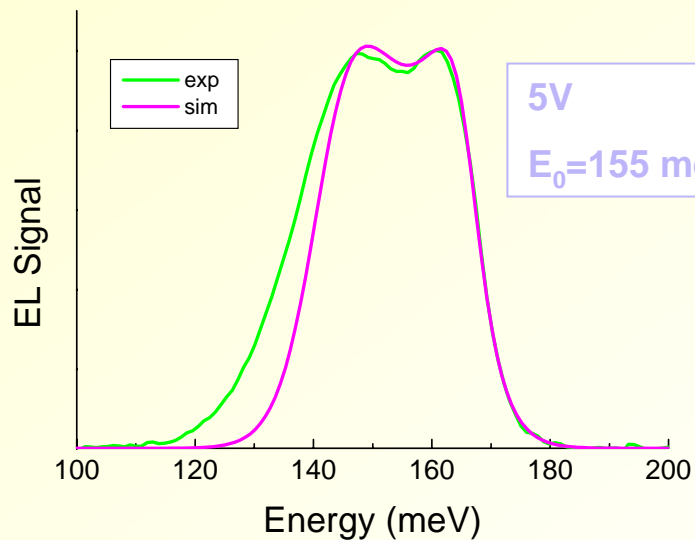
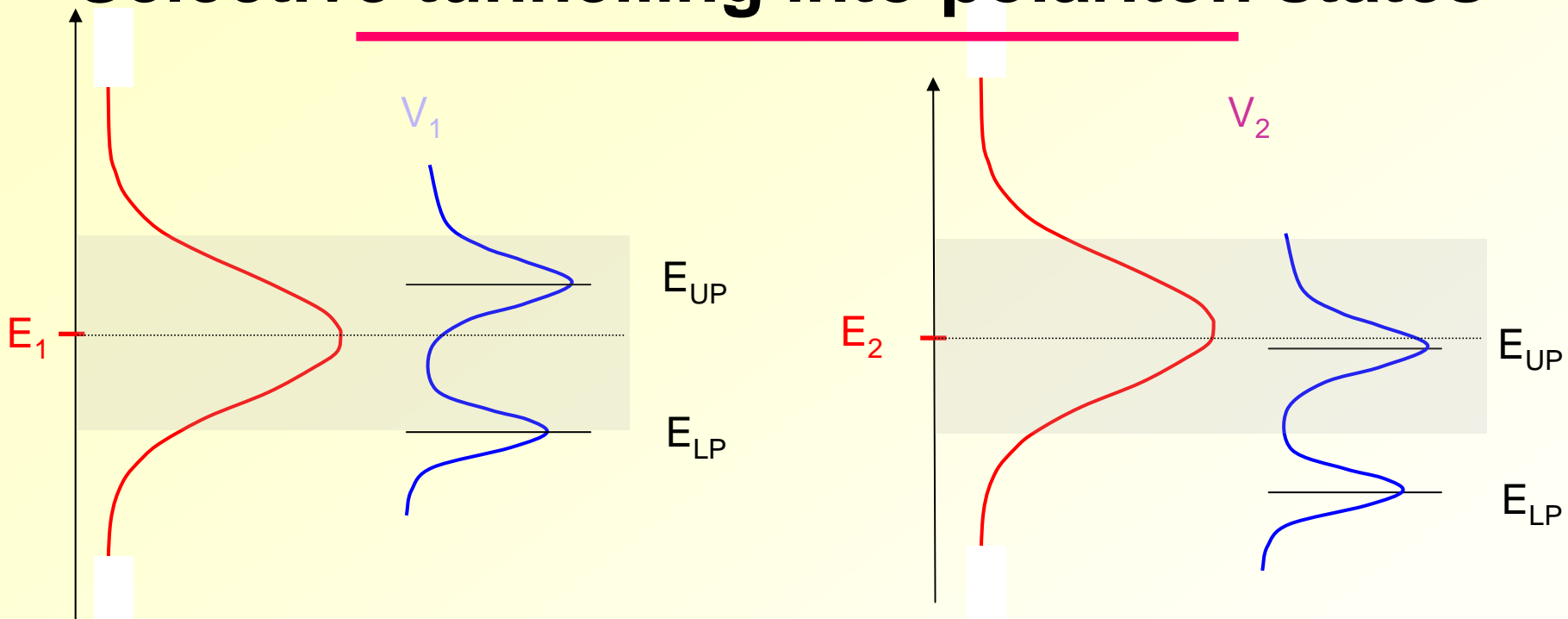
Tunnelling into cavity polariton states



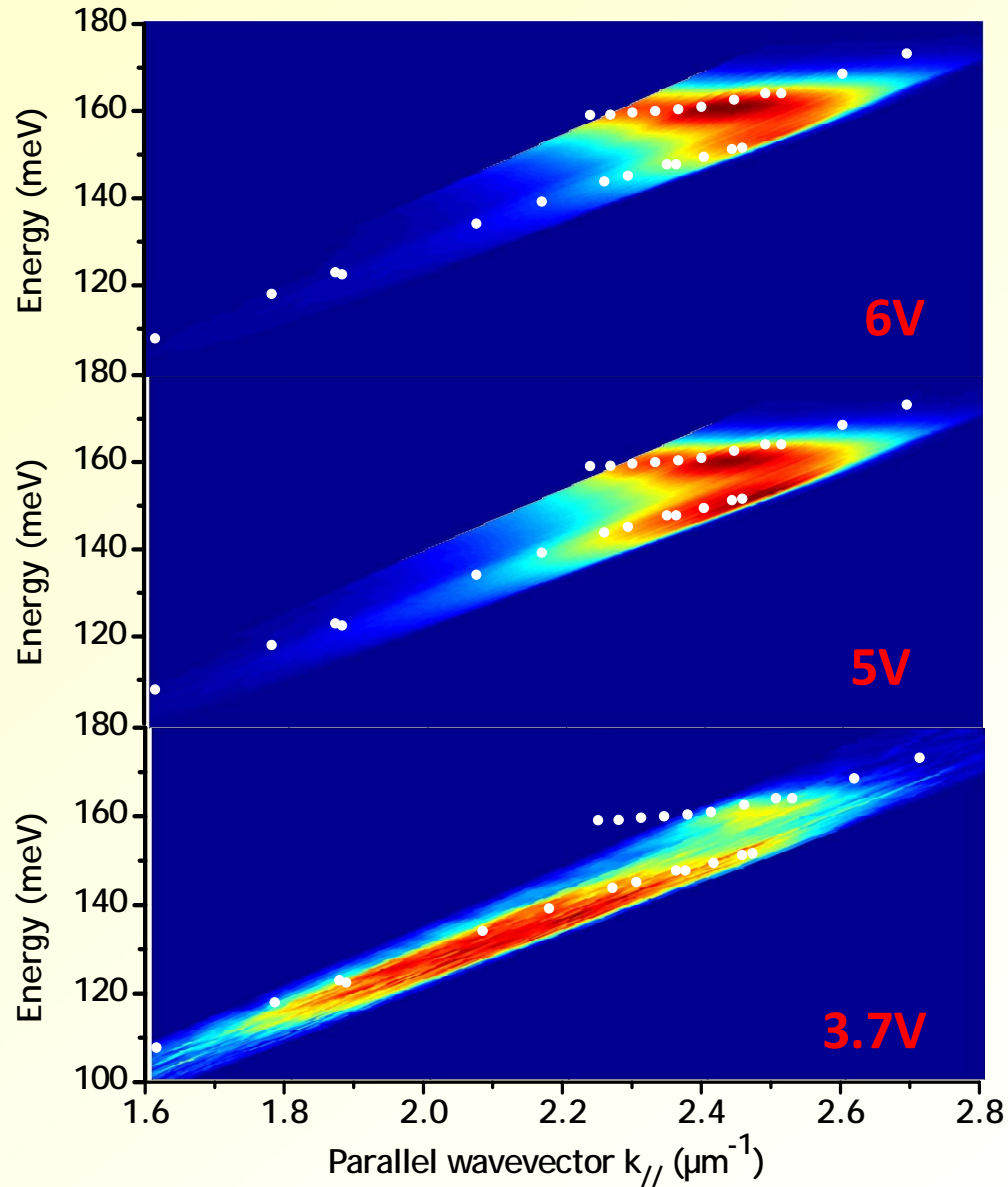
L. Sapienza et al., PRL **100**, 136806 (2008)

L. Sapienza et al., APL **90**, 201101 (2007)

Selective tunnelling into polariton states



Electric field tuning of polariton luminescence



Coherent instabilities in a semiconductor laser with fast gain recovery

Christine Y. Wang,¹ L. Diehl,² A. Gordon,³ C. Jirauschek,³ F. X. Kärtner,^{3,*} A. Belyanin,⁴ D. Bour,⁵ S. Corzine,⁵ G. Höfler,⁵ M. Troccoli,² J. Faist,⁶ and Federico Capasso^{2,†}

¹*Department of Physics, Harvard University, Cambridge, Massachusetts 02138, USA*

²*School of Engineering and Applied Science, Harvard University, Cambridge, Massachusetts 02138, USA*

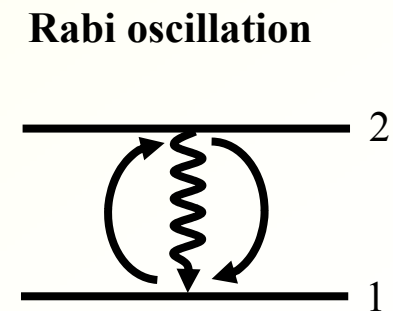
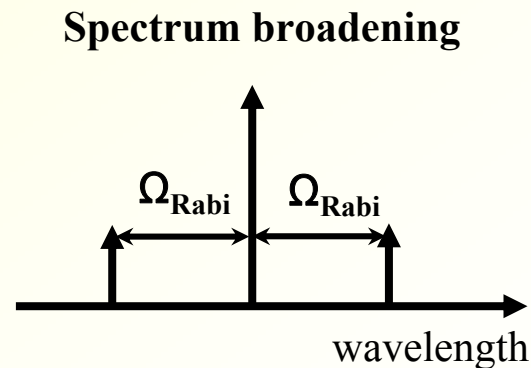
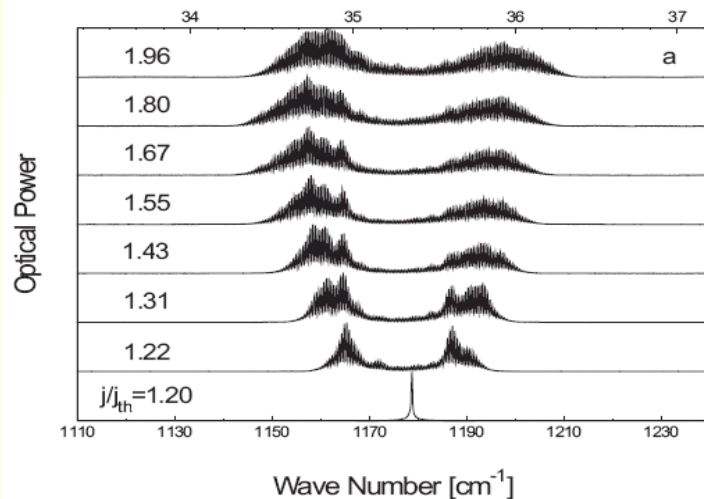
³*Department of Electrical Engineering and Computer Science and Research Laboratory of Electronics, Massachusetts Institute of Technology, 77 Massachusetts Avenue, Cambridge, Massachusetts 02139, USA*

⁴*Department of Physics, Texas A & M University, College Station, Texas 77843, USA*

⁵*Agilent Technologies, Palo Alto, California 94306, USA*

⁶*Institute of Physics, University of Neuchâtel, CH-2000 Neuchâtel, Switzerland*

arXiv:0703006; published 30 March 2007

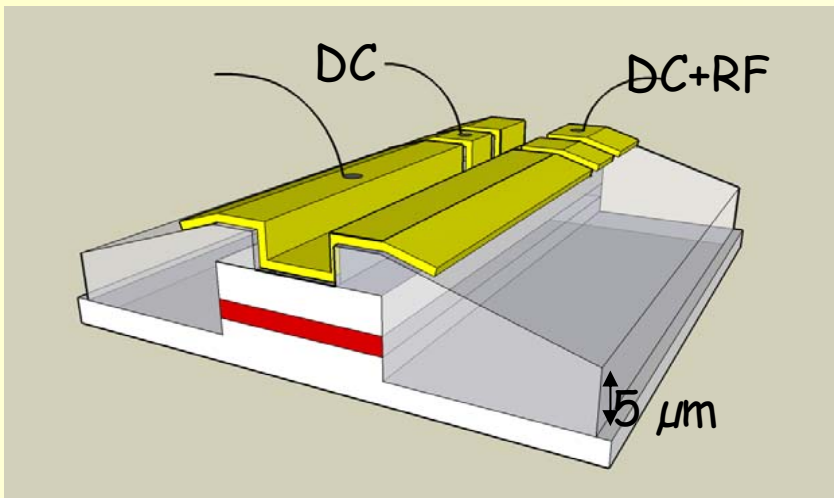


Active Mode Locking of QCLs

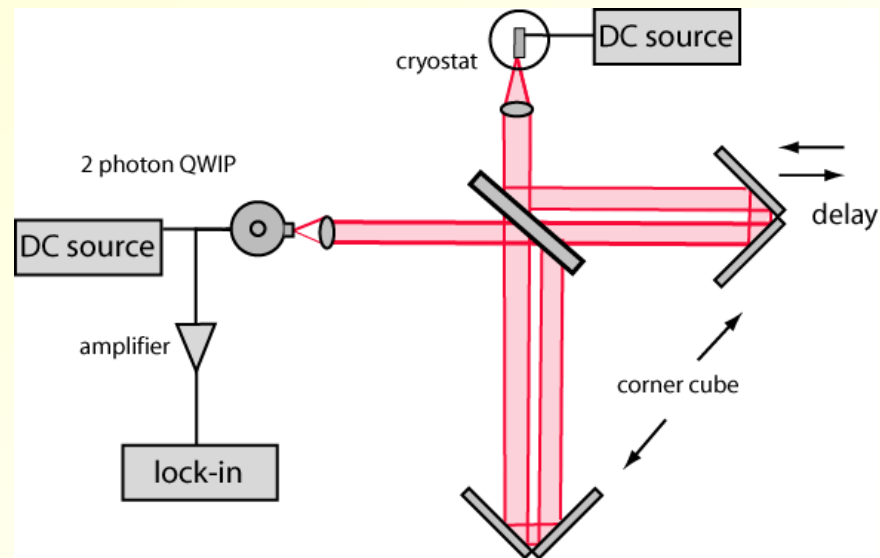
(Harvard, MIT, H.C. Liu)



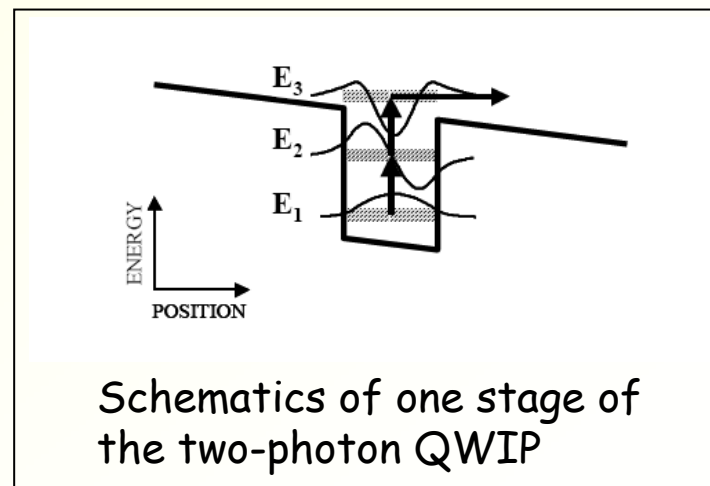
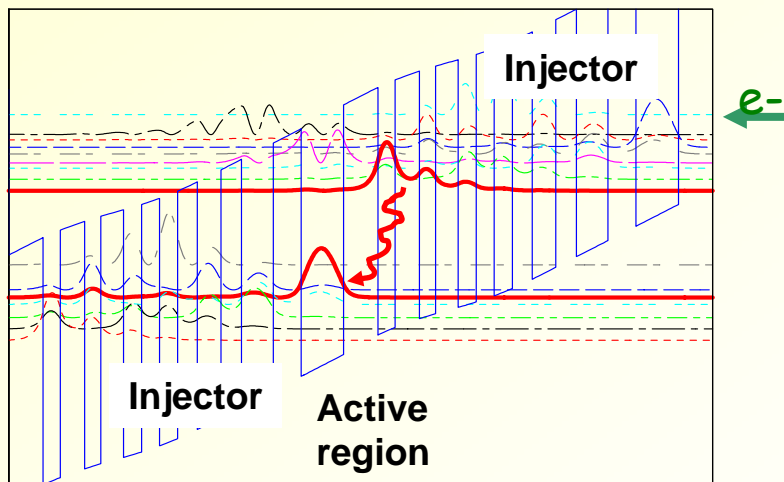
Modulation scheme



Pulse characterization



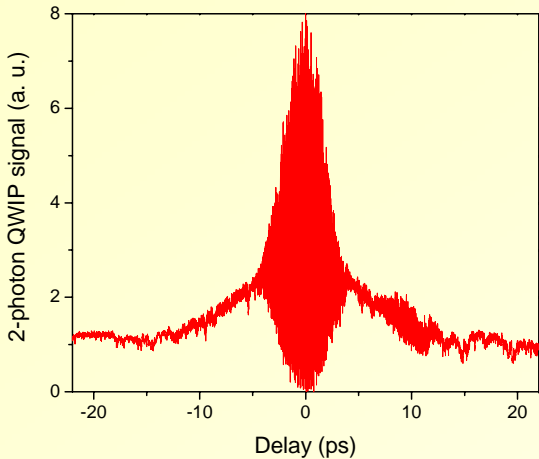
Active region design



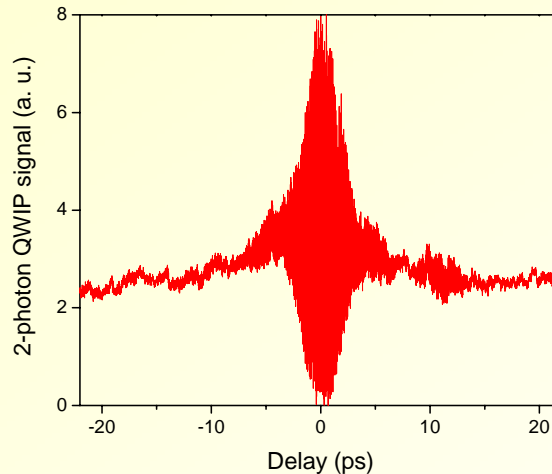
Schematics of one stage of the two-photon QWIP

Active Mode Locking in QCLs

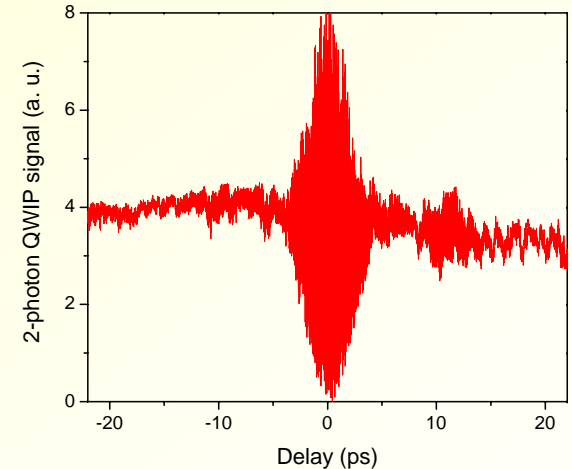
340mA ($I/I_{th}=1.1$)



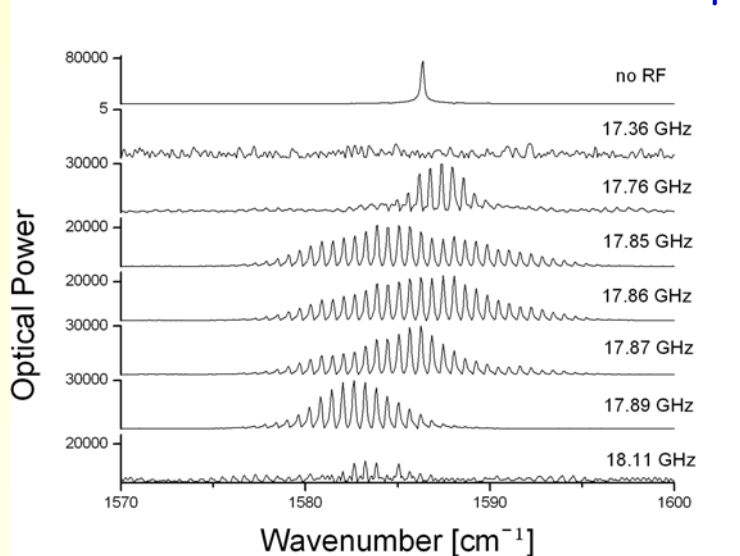
400mA ($I/I_{th}=1.45$)



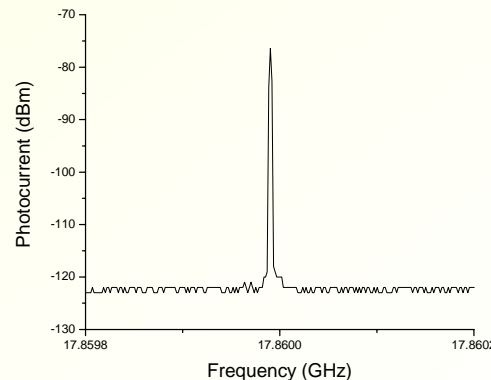
500mA ($I/I_{th}=1.61$)



Spectra at 340mA with various RF frequencies



Beatnote at 340mA
FWHM \sim 3kHz



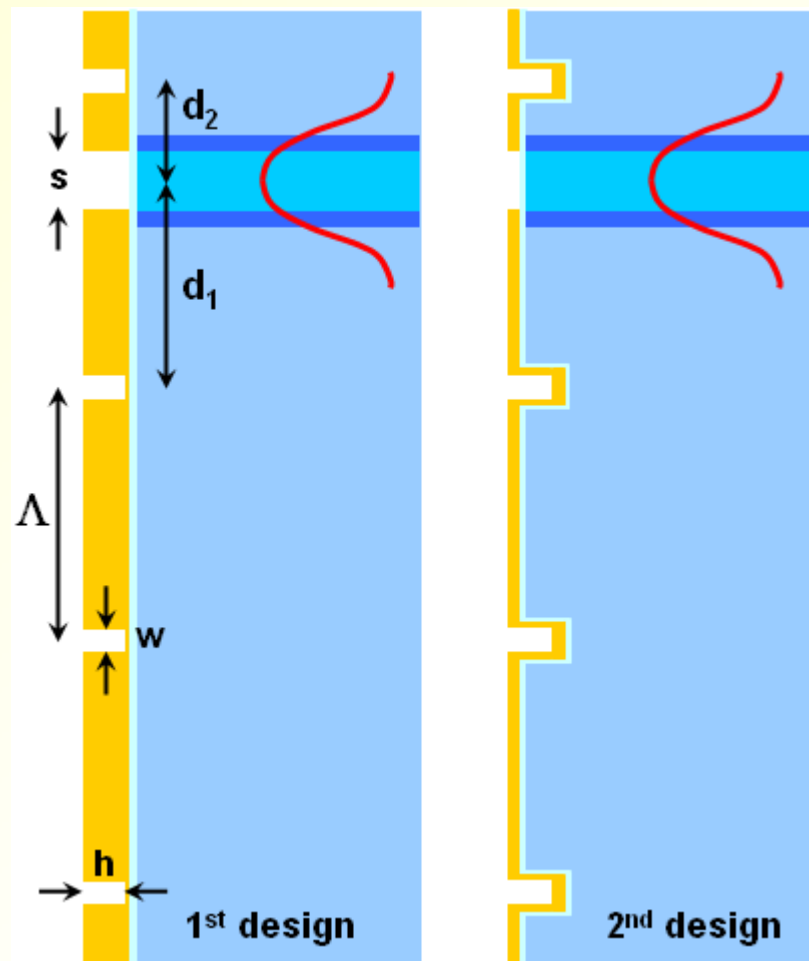
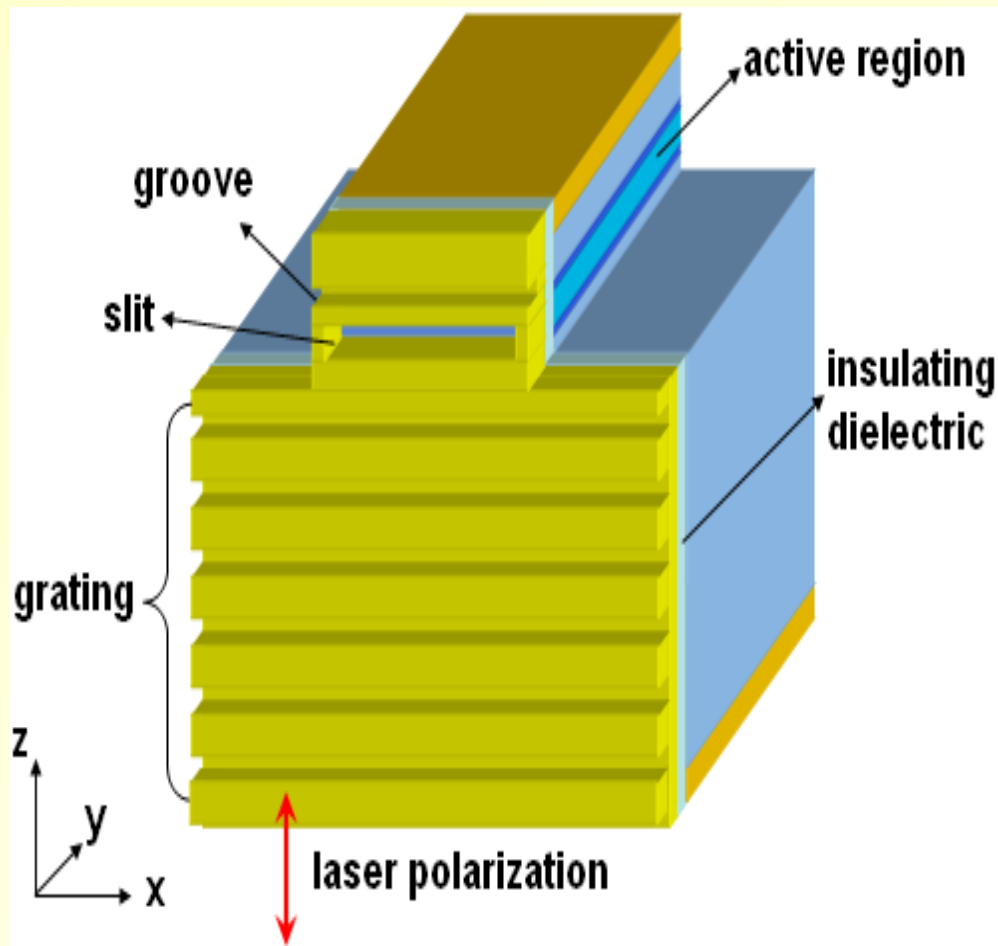
At 340mA,
peak:background=8:1
in the 2nd order
autocorrelation trace
→ Stable mode-locked
pulse train!
The RF modulation
opens a stable mode-
locking window in the
vicinity of the
laser threshold.

Subwavelength photonics with semiconductor lasers (edge emitting and surface emitting)



- ◆ Can we use sub-wavelength photonics (plasmonics and metamaterials) to advance the state of the art of semiconductor lasers? Solve some major problems in laser technology? new lasers sources?
- ◆ Facet engineering to achieve enhanced performance and/or new functionalities in the near- and far-field? Can we design semiconductor lasers with “arbitrary” wavefront (beam engineering: high collimation, super focusing, “cork screw beams). control of polarization, beam steering with a single laser?
- ◆ Low divergence semiconductor lasers, Polarization control
- ◆ New fabrication techniques for large area patterning
- ◆ We have used QCLs as a platform to demonstrate these concepts

Plasmonic collimator for low divergence edge emitting lasers (N. Yu et al; Hamamatsu Photonics)

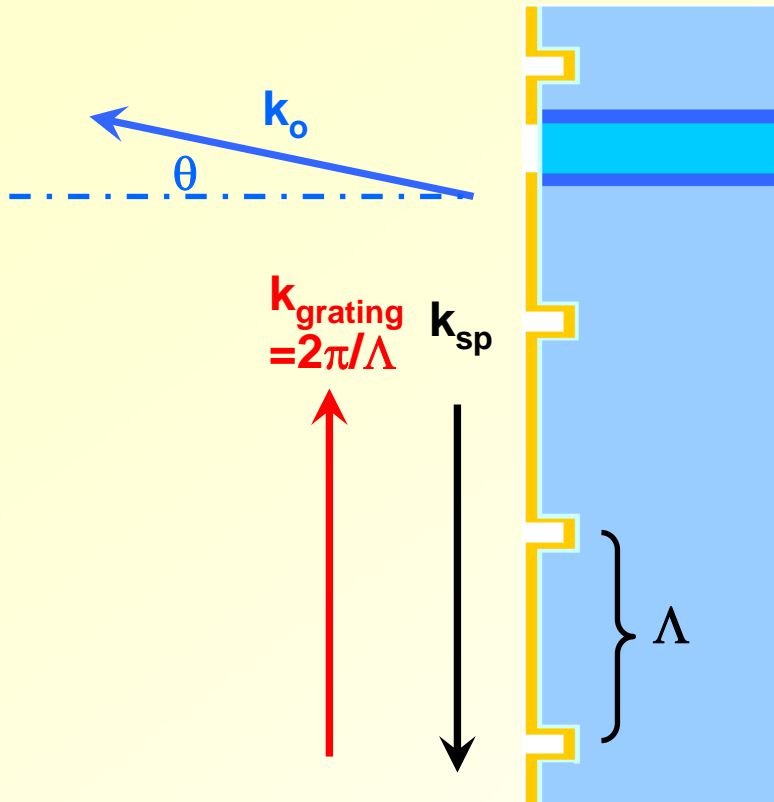


All the parameters optimized for beam collimation: s , Λ , w , h , d_1 , and d_2 .
N. Yu et al. Nature Photonics, September 2008

Physics of plasmonic collimator



Second order grating



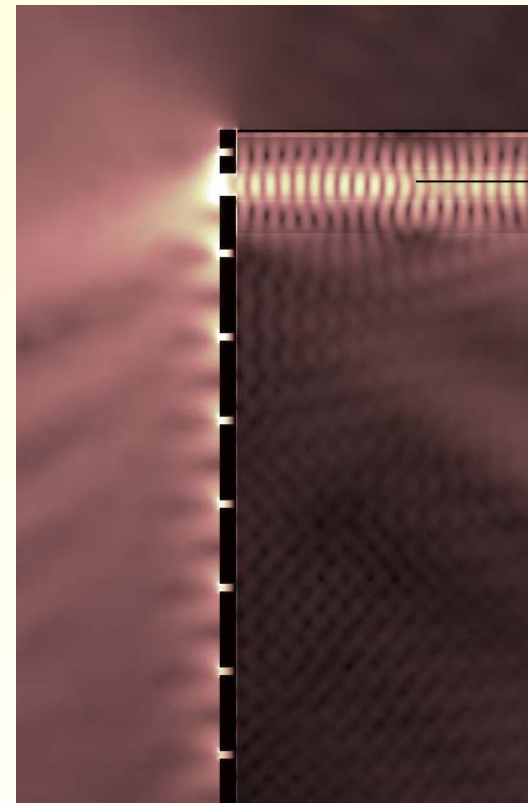
$$k_{\text{grating}} = 2\pi/\Lambda$$

$$k_{\text{sp}}$$

$$\Lambda$$

$$k_0 \sin \theta = k_{\text{sp}} - k_{\text{grating}} = 0$$

SP modes of the grating structure, shown is the distribution of $|E\text{-field}|$



QC laser active region

Second order grating ensures emission normal to the facet

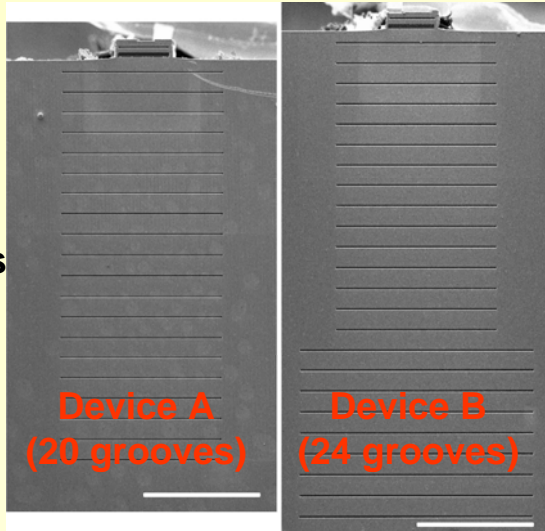
Aperture designed for maximum coupling of radiation to surface plasmons

Small divergence arises from coherent scattering (interference) by the grating grooves into the far field (beaming or antenna effect)

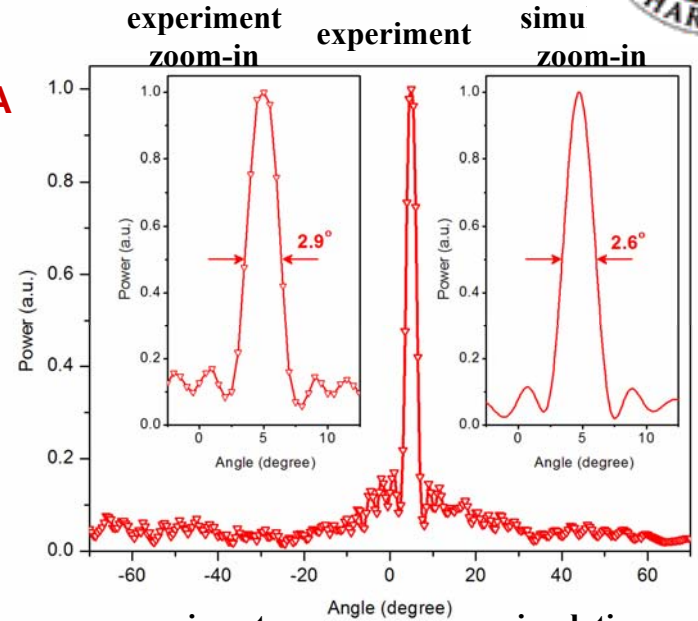
Experimental results: device with grooves cut into the semiconductor



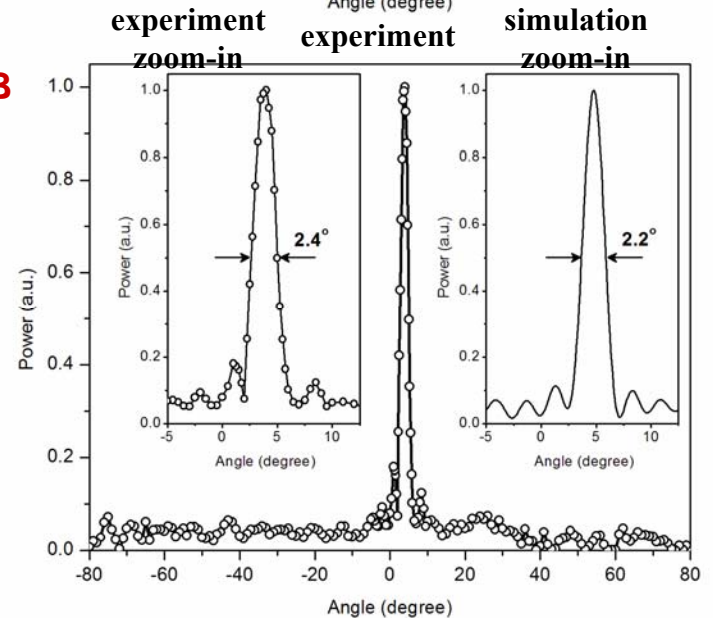
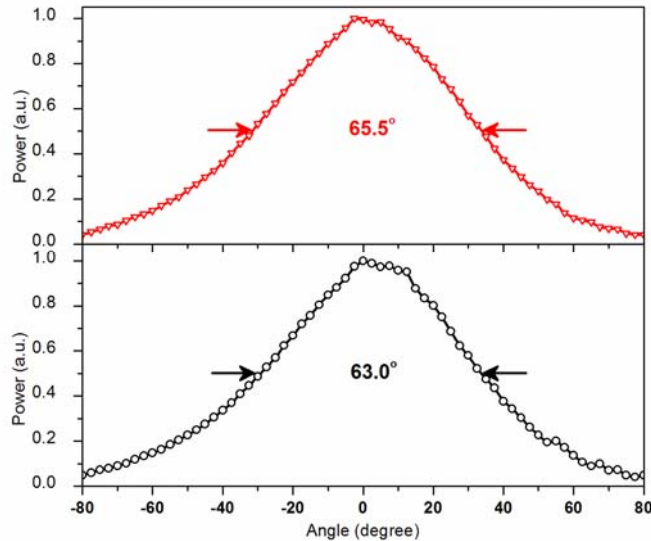
$\lambda=9.9 \mu\text{m}$ QCLs
Scale bars
50 μm



Device A

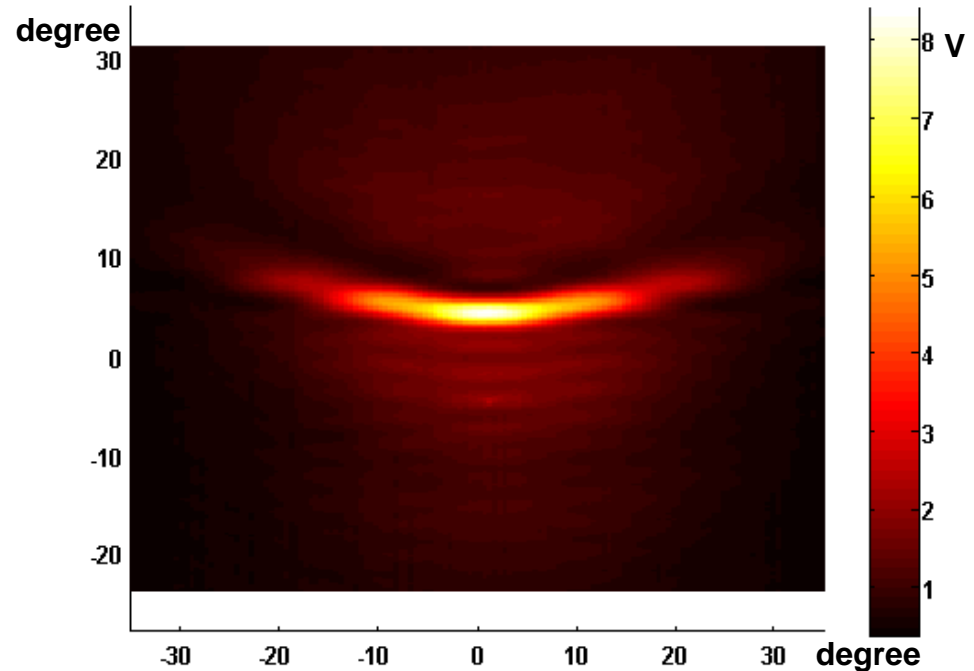
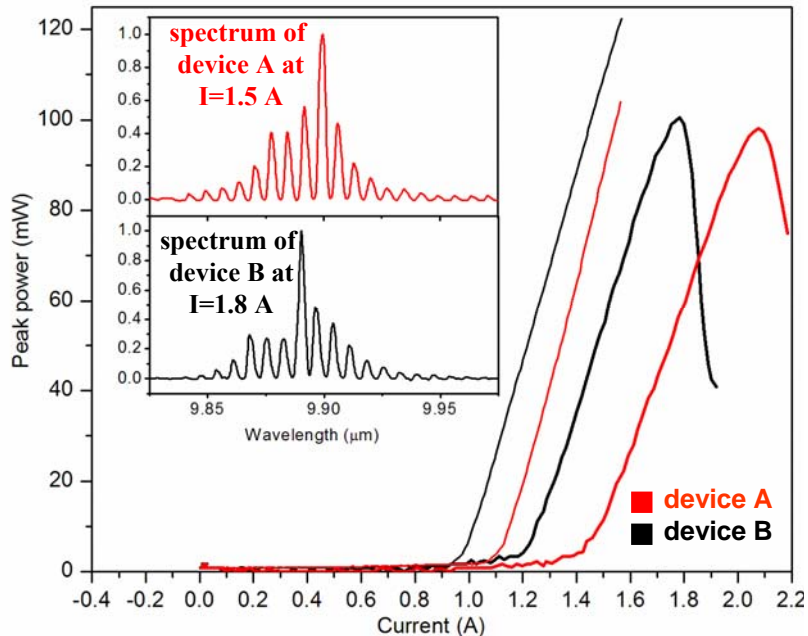


Device B



vertical far-field power distribution of the unpatterned lasers

Low vertical divergence QCLs



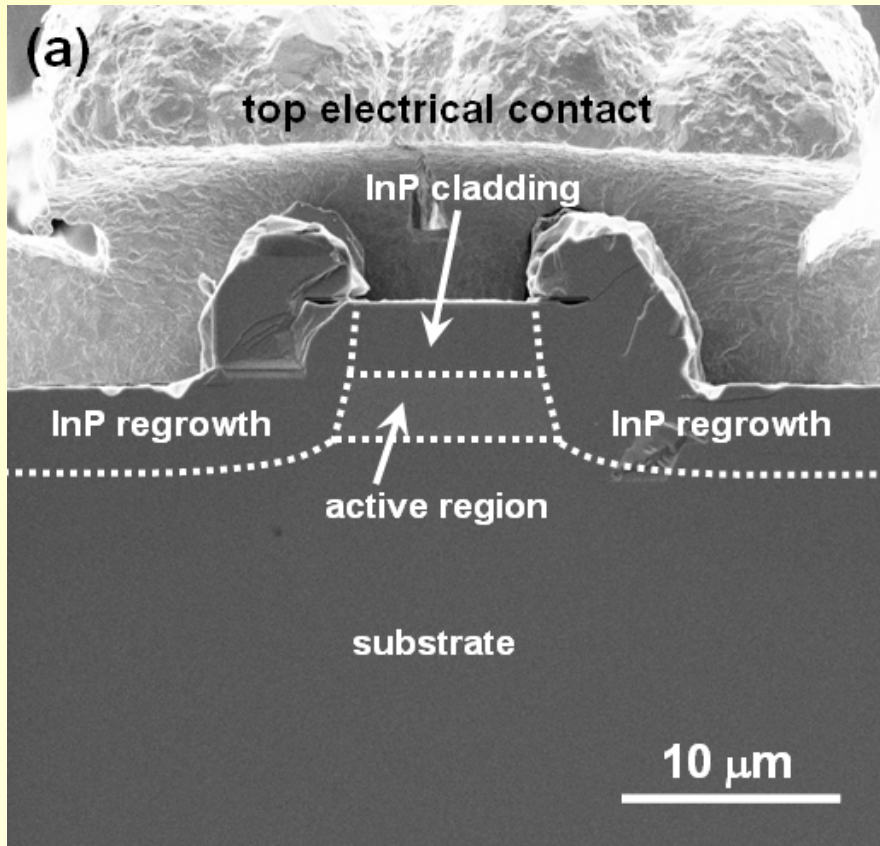
L-I curves taken before and after defining the slit-groove structure. The thin and the thick curves are for the unpatterned devices and the ones with the plasmonic collimator, respectively.

Measured 2D far-field power distribution of device A.

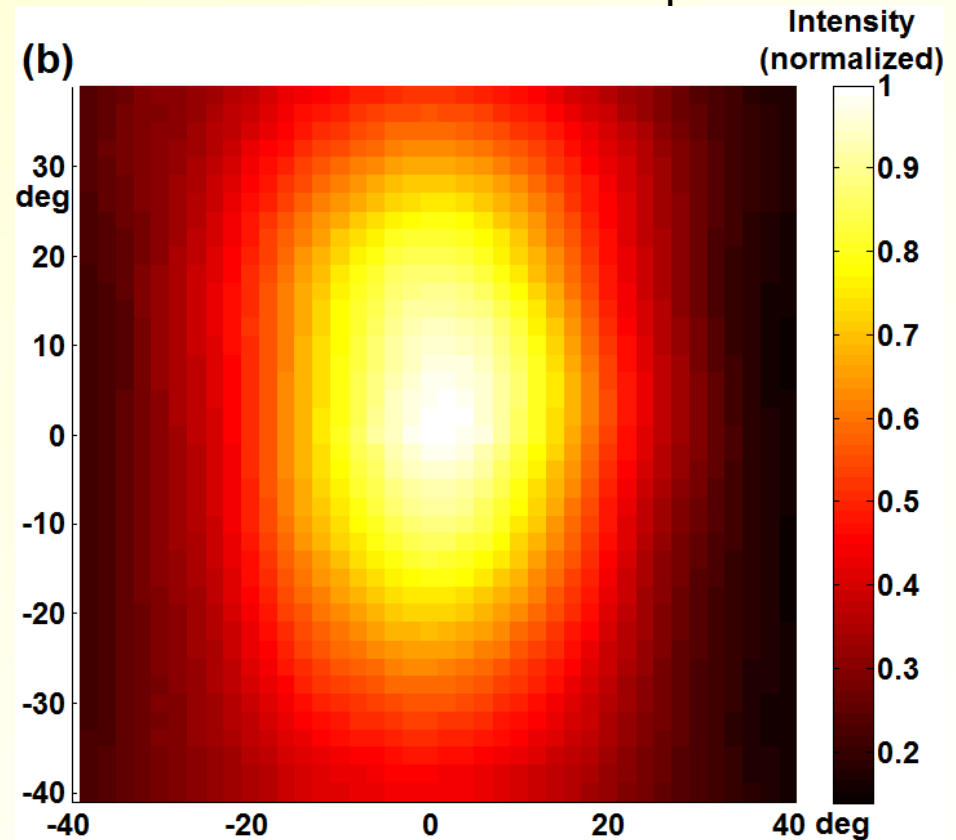
Vertical divergence reduced by a factor > 25 !

2D plasmonic collimation: original device

SEM image of the laser facet



Measured far-field mode profile



Hamamatsu MOCVD-grown BHT device
 $\lambda = 8.06 \mu\text{m}$

FWHM divergence angles:

$$\theta_{\perp} = 74^{\circ}$$

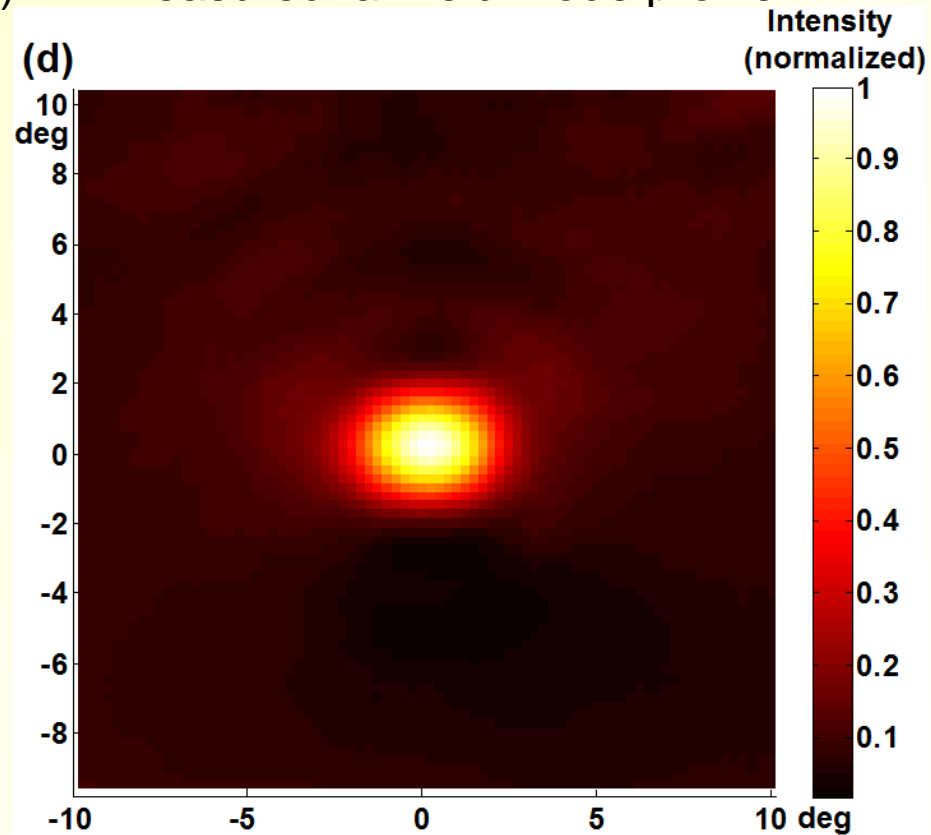
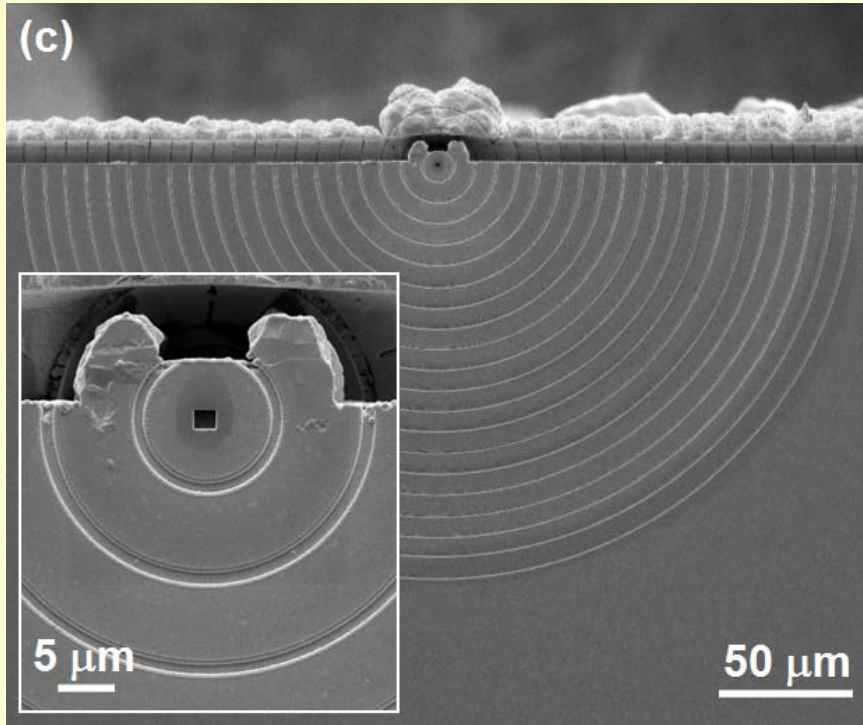
$$\theta_{\parallel} = 42^{\circ}$$

(N. Yu et al; Hamamatsu Photonics)

2D plasmonic collimation: far-field mode profile

SEM image of a fabricated device ($\lambda=8.06 \mu\text{m}$)

Measured far-field mode profile



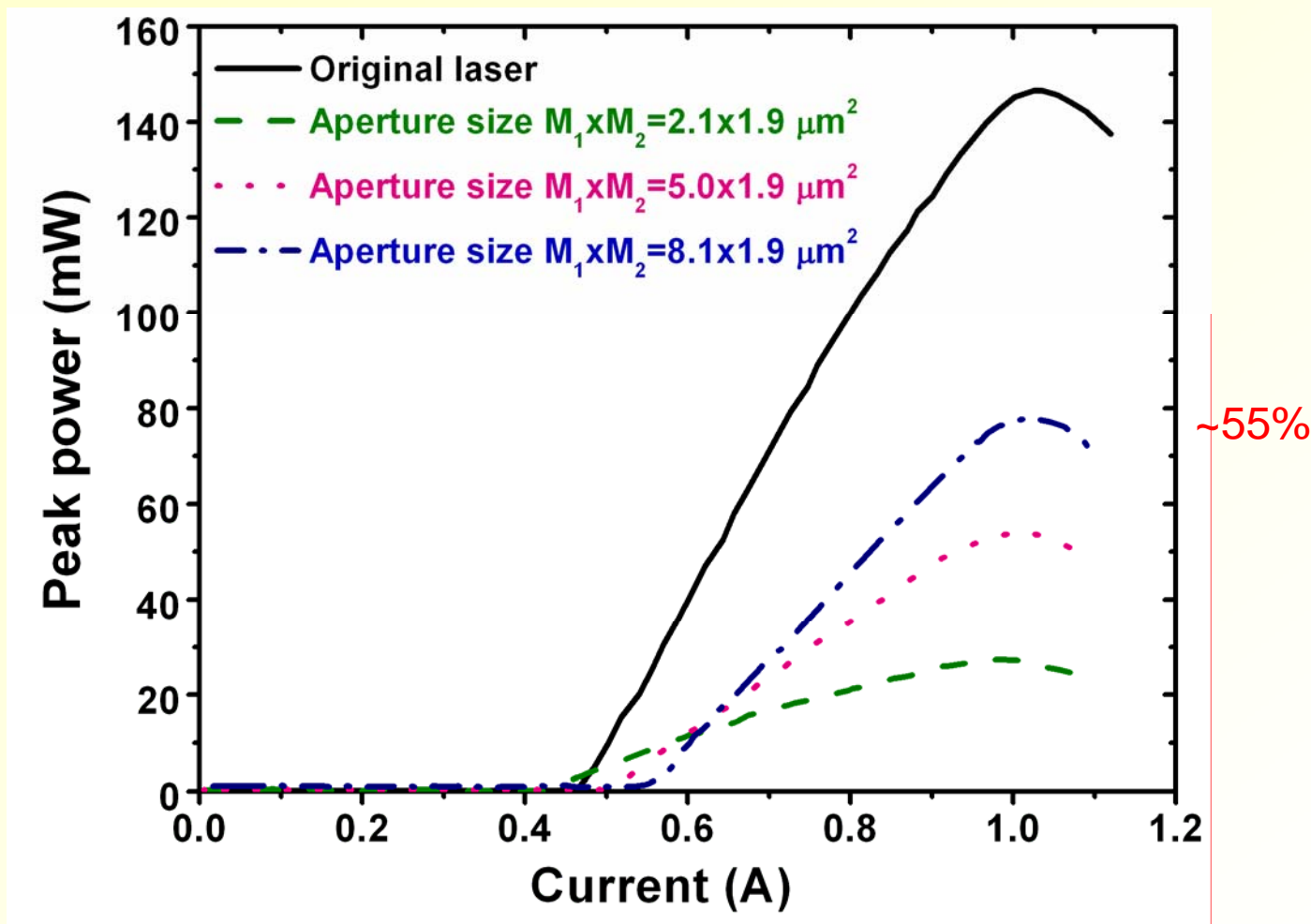
Aperture size $w_1 \times w_2$ (μm^2)	grating period Λ (μm)	groove width w (μm)	groove depth d (μm)	radius of the first groove r_1 (μm)
2.1×1.9	7.8	0.6	1.0	6.0

FWHM divergence angles:

$\theta_{\perp} = 2.7^\circ$ (reduction by a factor of ~ 30)

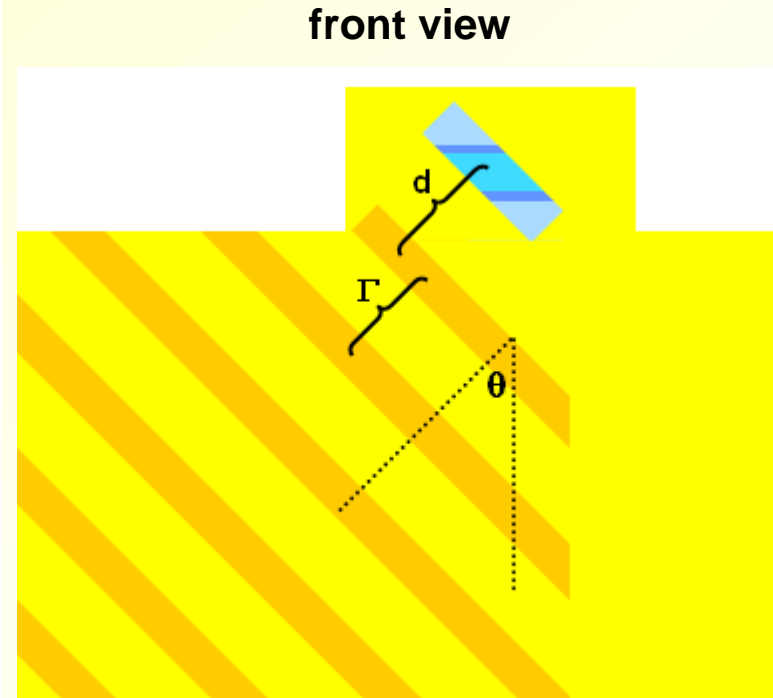
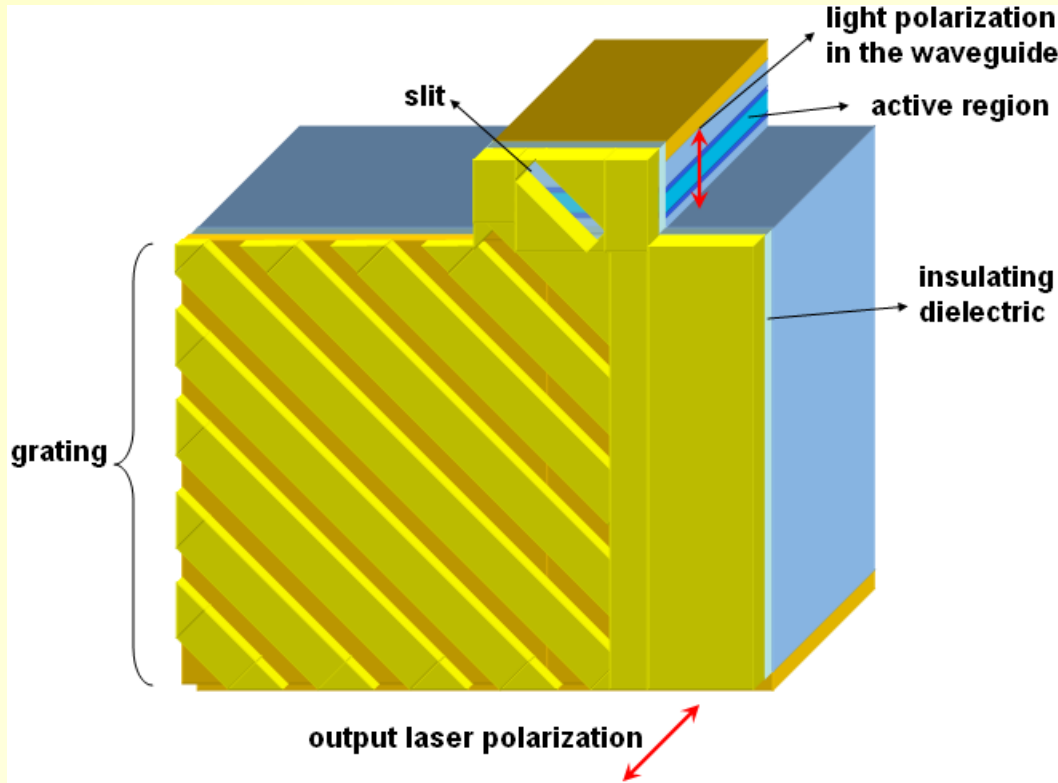
$\theta_{\parallel} = 3.7^\circ$ (reduction by a factor of ~ 10)

2D plasmonic collimation: LI characteristics at different aperture size



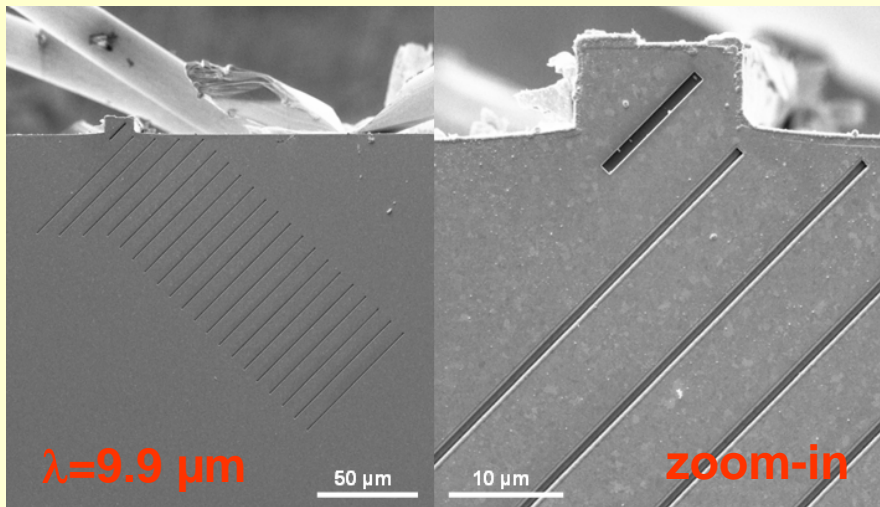
Larger aperture \rightarrow larger mirror loss \rightarrow
larger slope efficiency \rightarrow larger maximum power output

Rotating the linear polarization of a QCL

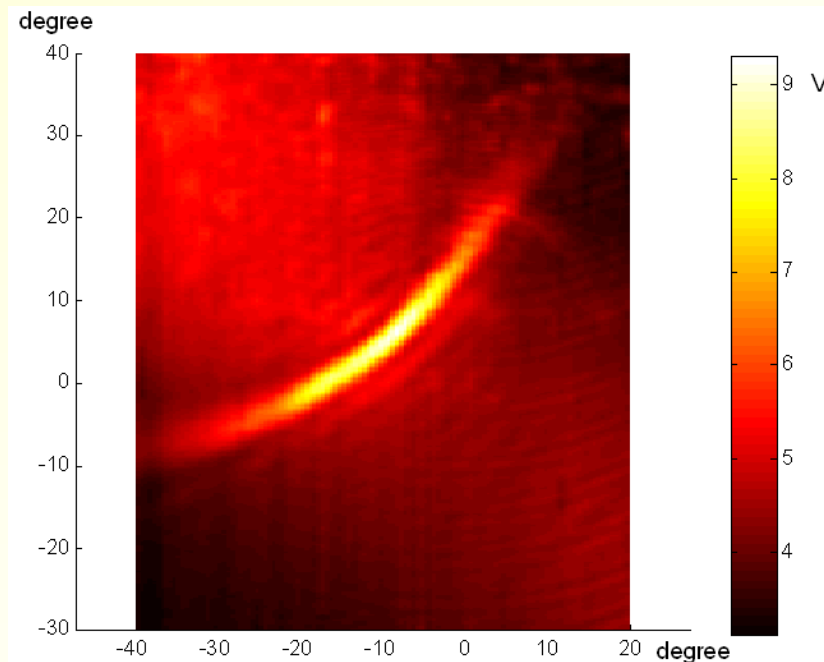


- The plasmonic polarizer can rotate the linear polarization direction of the original laser to an arbitrary angle, determined by the orientation of the grating θ .
- Γ satisfying the condition for second order grating.

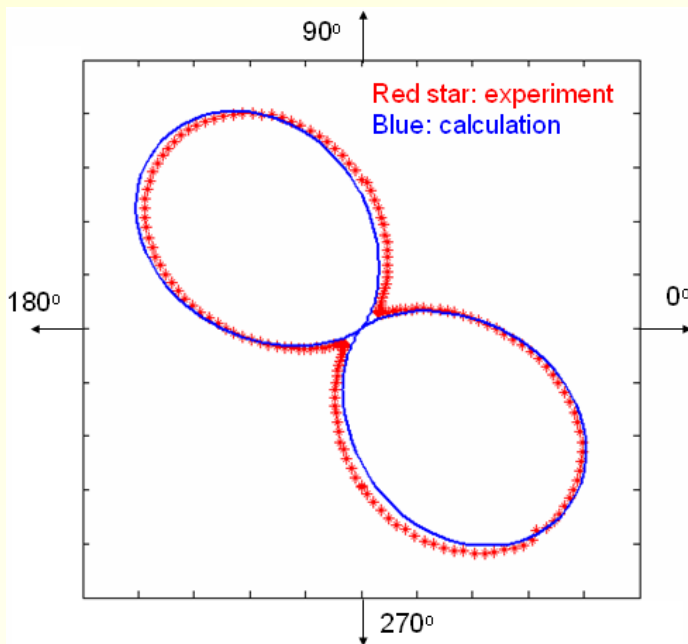
Initial experimental results: 45° linear polarization



Measured 2D far-field power distribution of the device



45-degree linear polarization
a mid-infrared detector detects the power of the main lobe of the far-field mode profile while a polarizer is rotated in front of the detector.

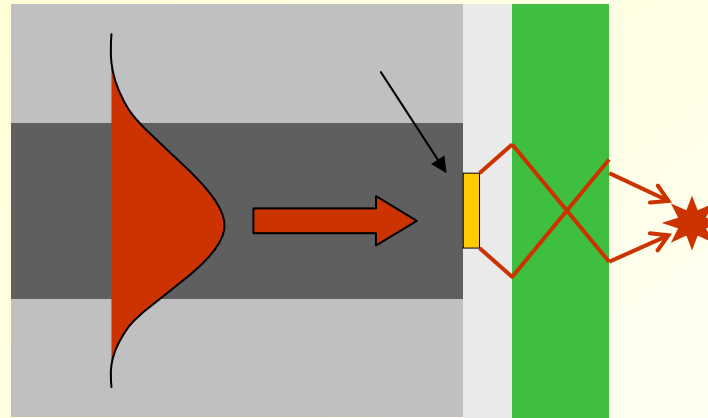


Challenge: make a circularly Polarized QCL

Design of laser facets with Metamaterials



Self-assembled negative index layer



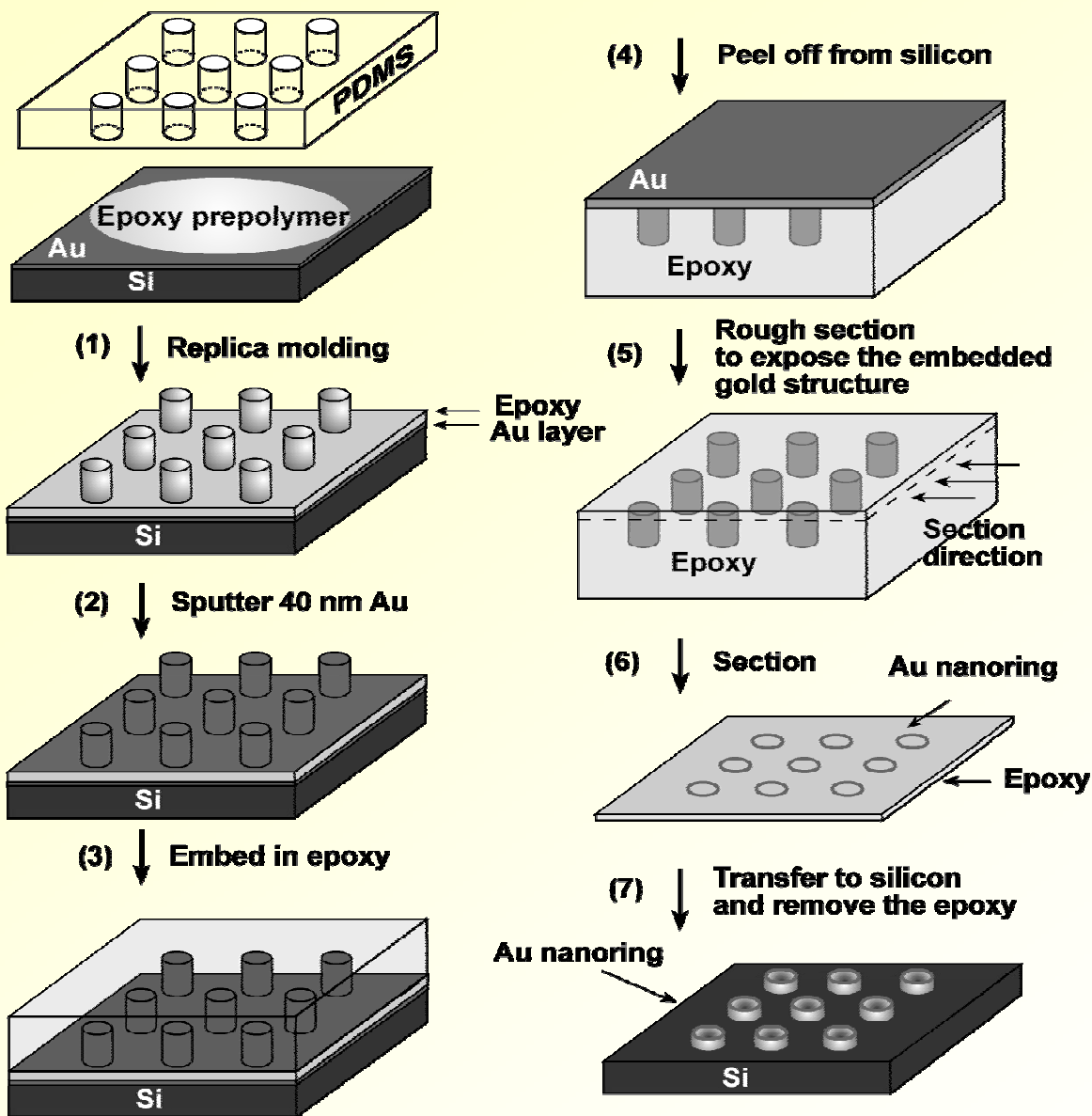
Negative index or hyperbolic metal-dielectric materials may be directly applied to a laser facet to get super-focusing of output light

New patterning techniques needed to create “Smart Surfaces”

Nanoskiving of Plasmonic Nanostructures

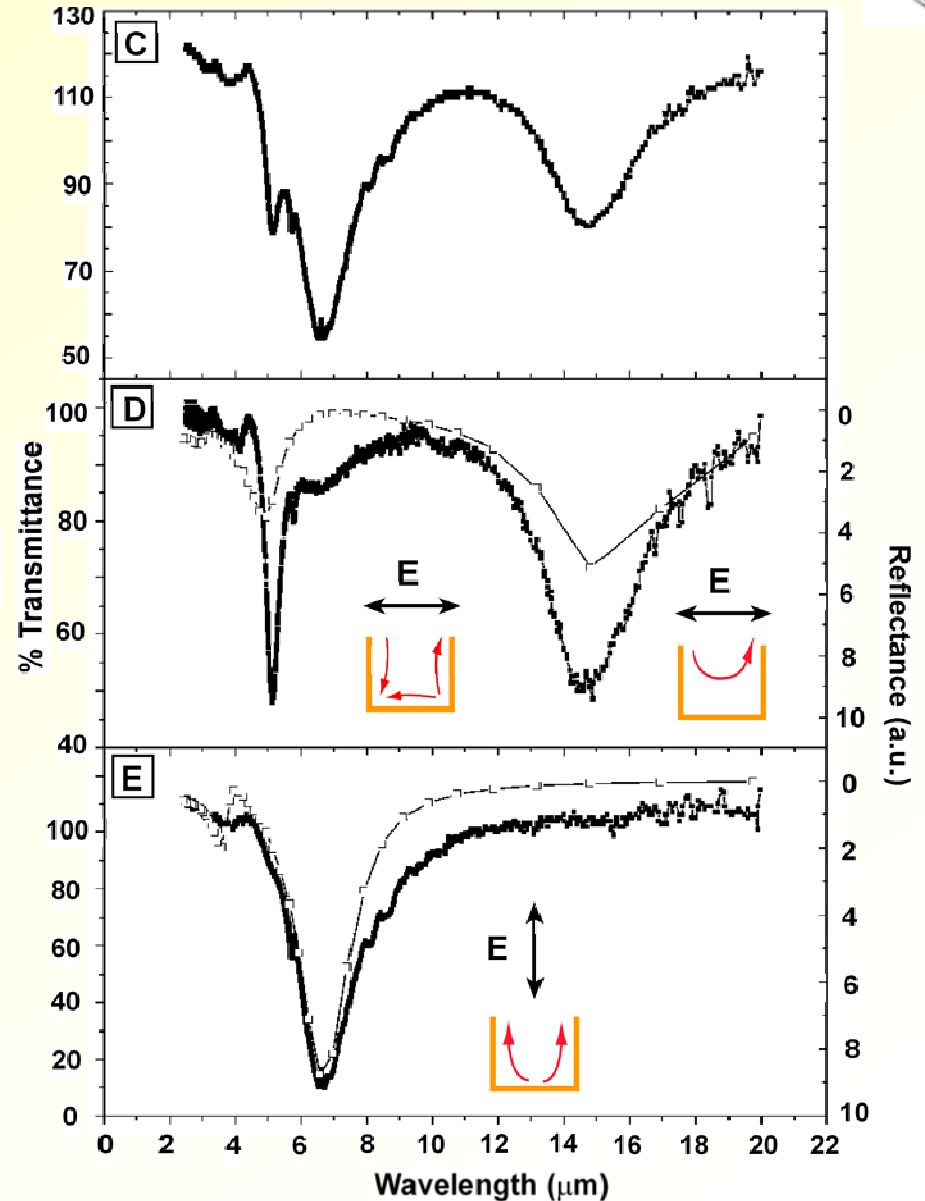
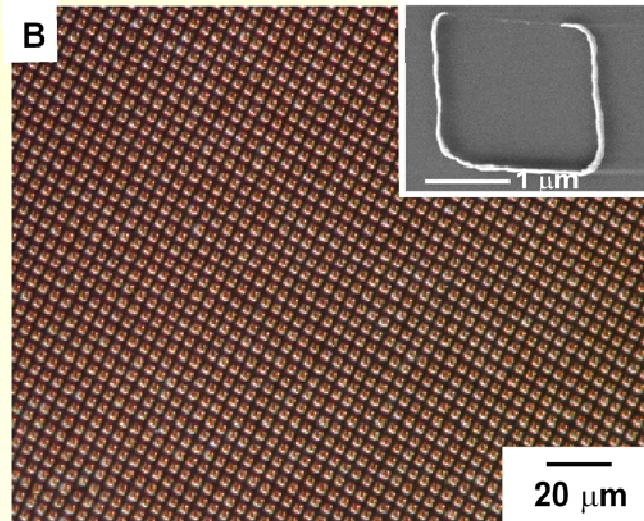
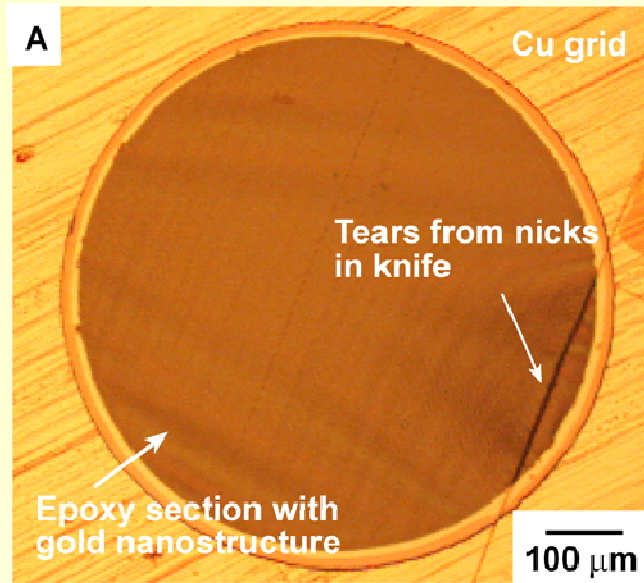


With George Whitesides Group (Harvard)



Large Area Patterning!

Transmission of U-shaped Nanostructure Array



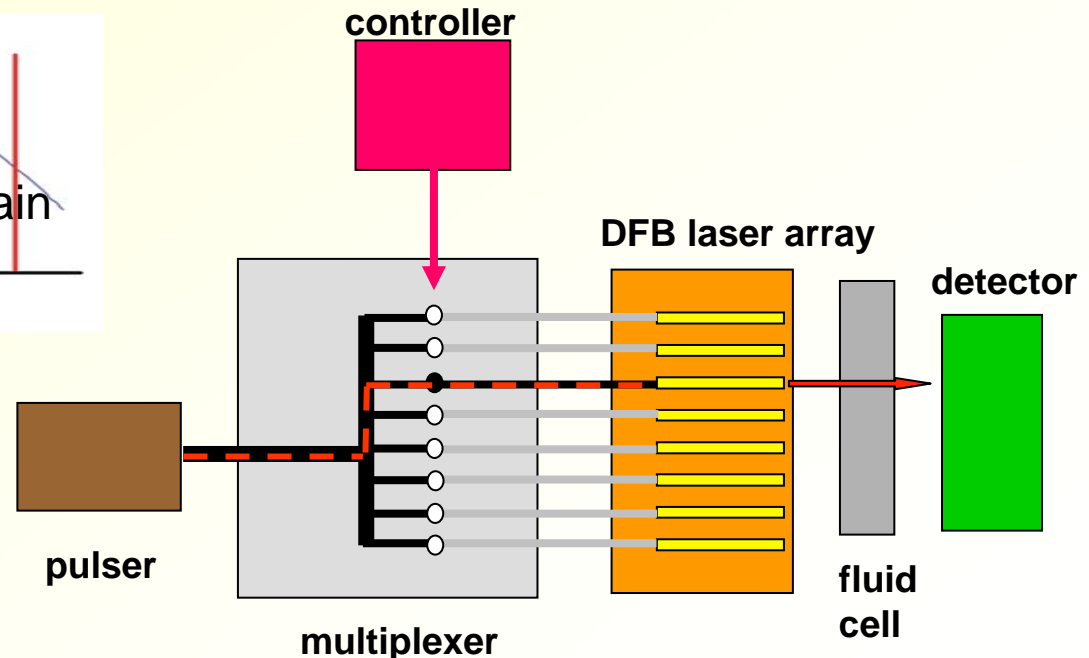
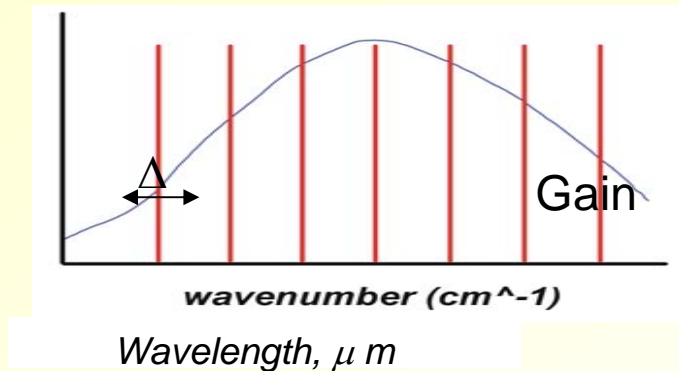
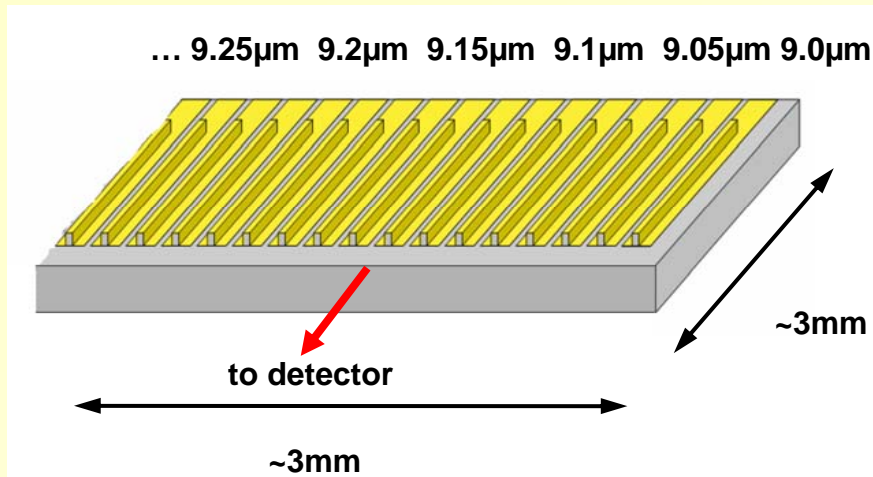
System Challenges

- ◆ *New broadband, portable QCL spectrometers that will outperform FTIR spectrometers in brightness and resolution*
- ◆ *Killer application in sensing?*
- ◆ *Instrumentation for climate change studies*

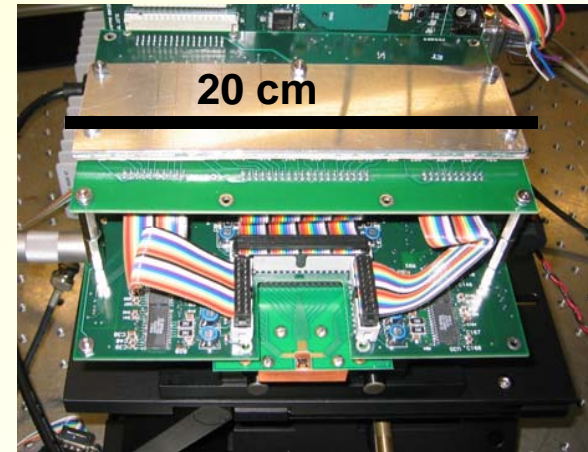
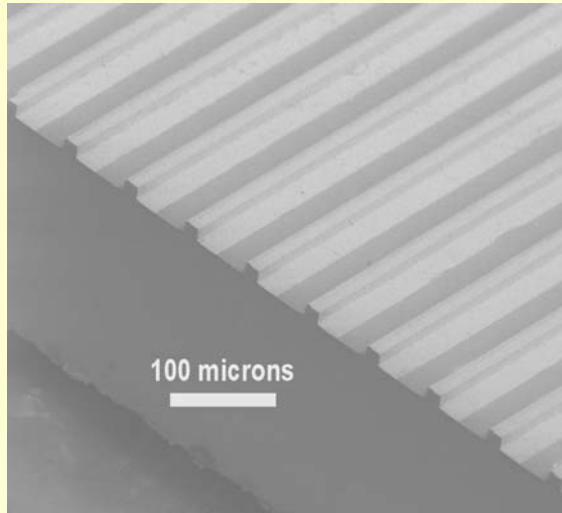
Broadband QCL Spectrometer on a Chip

Technical Approach

- *Broadband mid-IR QCL material (8-10 μm)*
- *Distributed feedback (DFB) laser array spanning laser gain curve*
- *Temperature tuning for continuous spectral coverage*
- *Computer control*

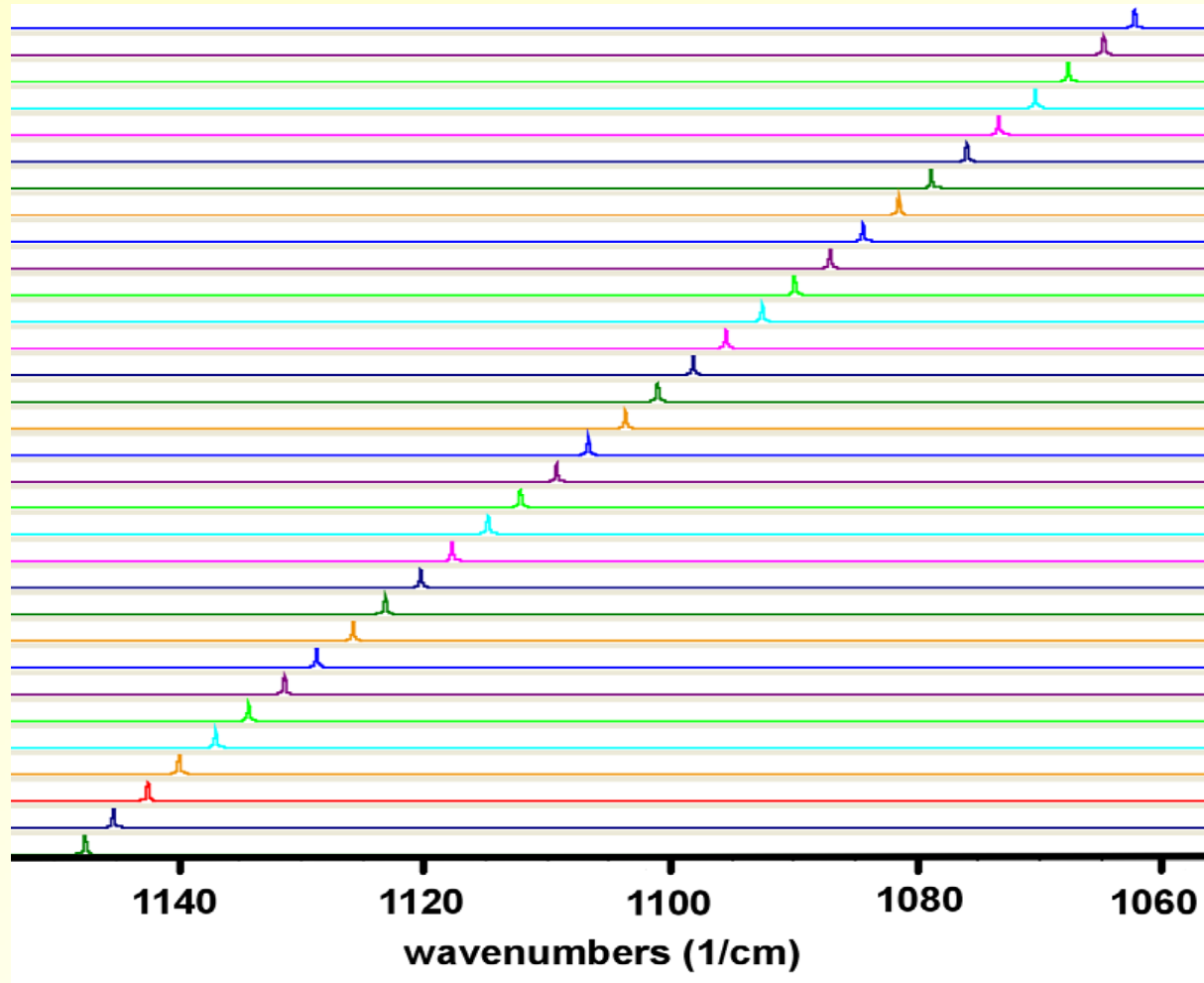


Broadband QCL Spectrometer on a Chip



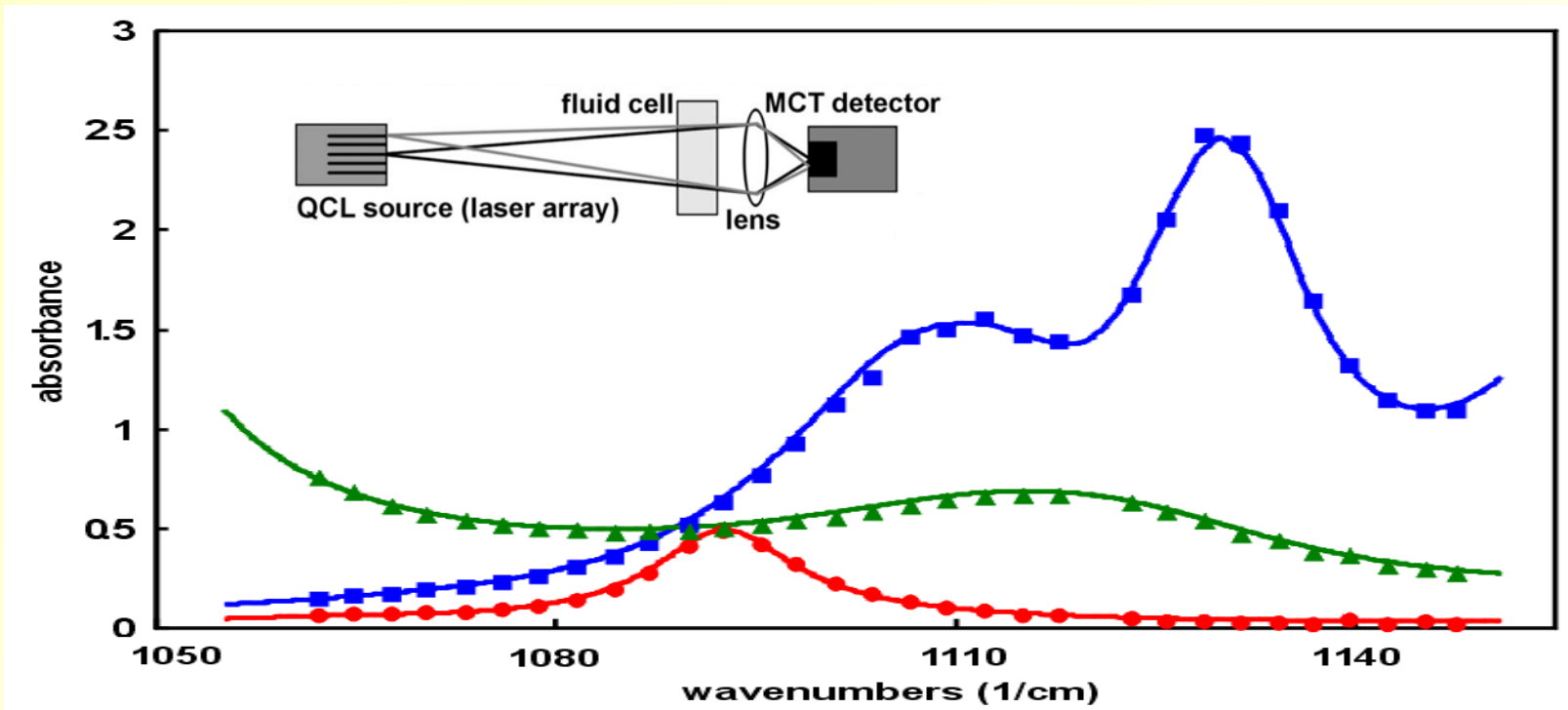
- DFB laser devices fabricated
- array of 32 DFB lasers on a single chip
- computer control and selective firing of lasers in array using custom-built electronics board

Spectrometer wavelength coverage



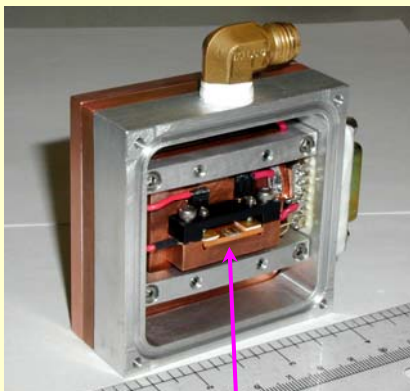
32 wavelengths from 1060 to 1150 cm^{-1} in array
Pulsed operation (80kHz, 50ns) at room temperature

Application: Absorption Spectroscopy



- absorption of isopropanol (blue), methanol (green) and acetone (red) measured with QCL array (points) and conventional FTIR (solid line)

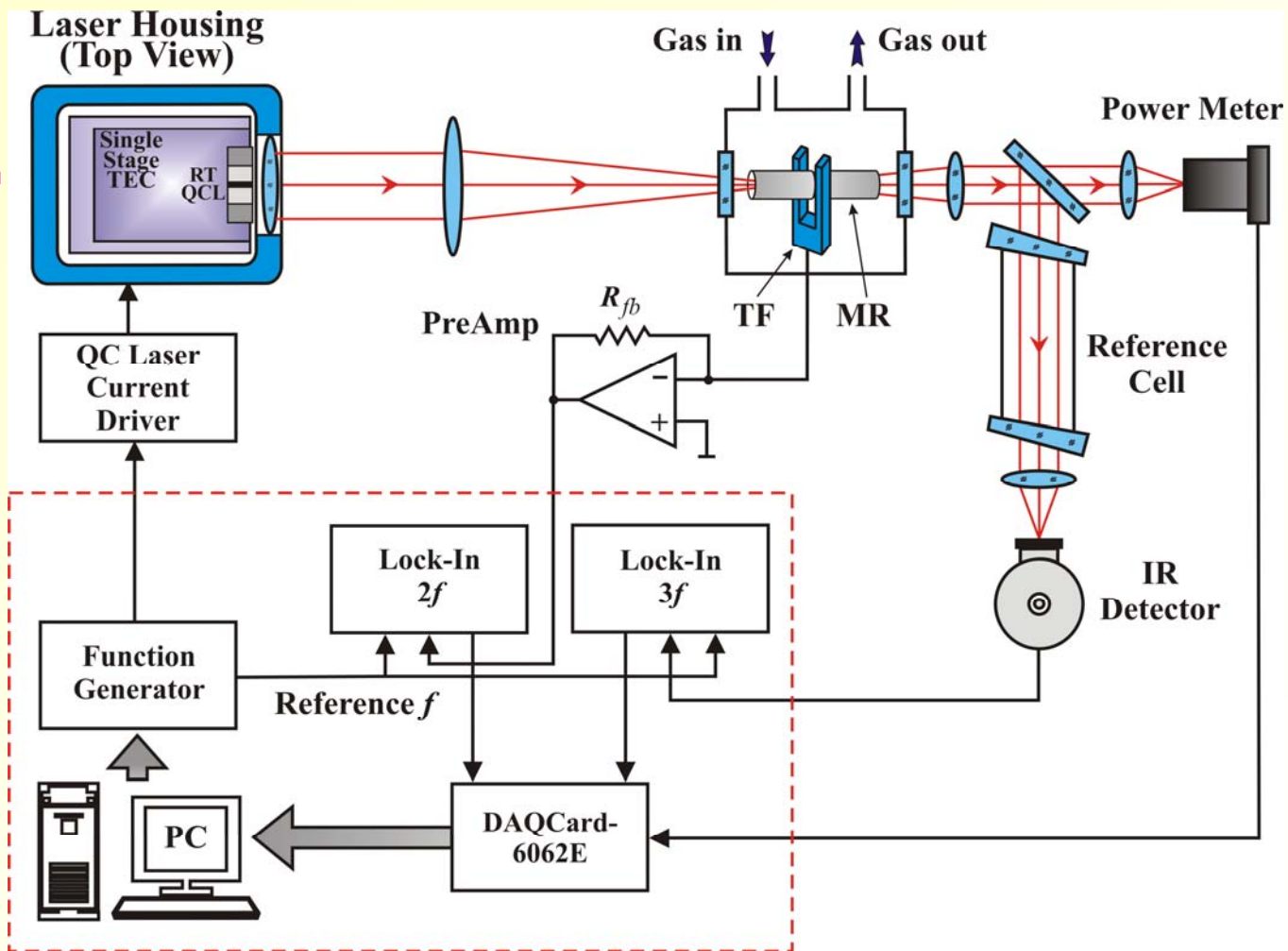
Mid-IR QEPAS based NH₃ Gas Sensor Architecture



MAXION CW DFB
QCL DQ9-M532P



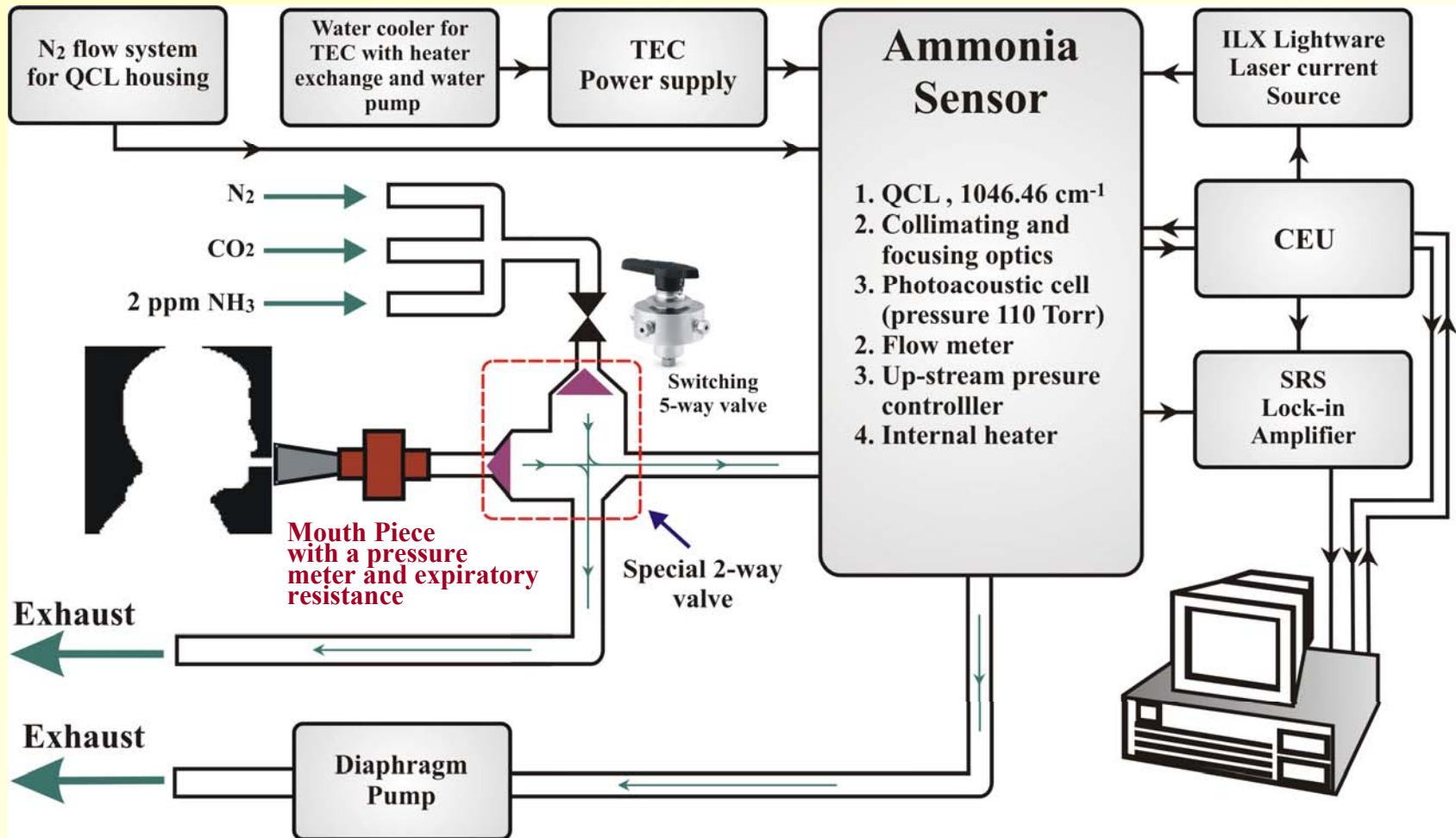
QEPAS Driver



Noise-equivalent concentration (NEC)
for $t=1$ s time constant is 6 ppb for 20mW
excitation power at 1046.4 cm^{-1} (110 Torr)

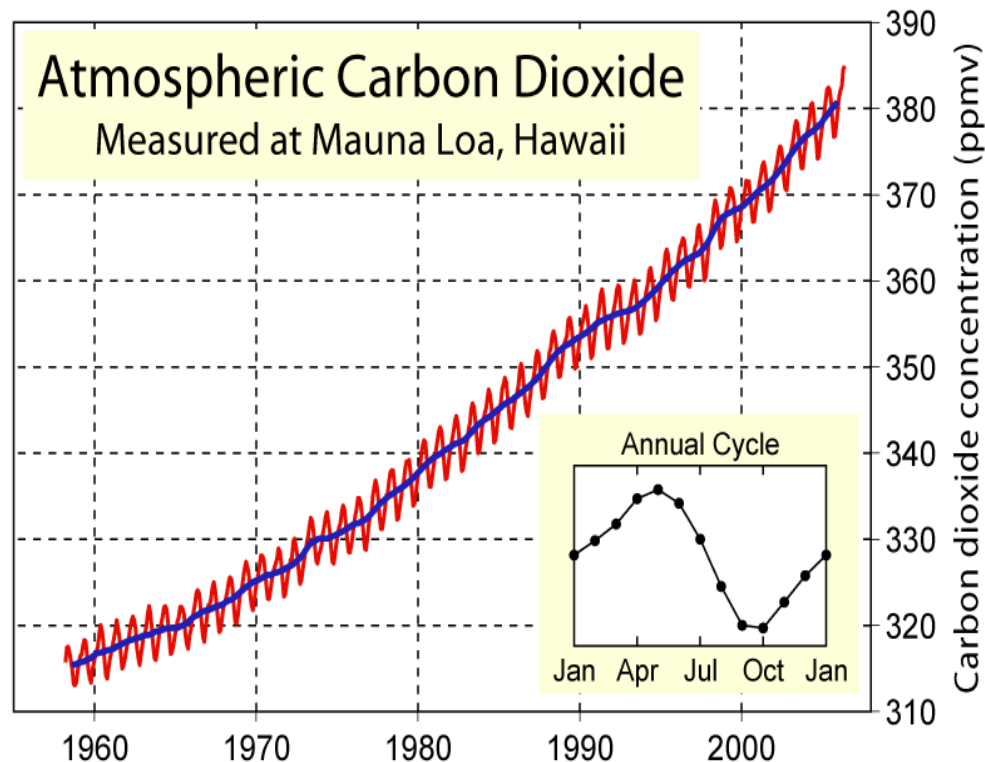


Interface for Real-time Breath NH_3 Analyzer



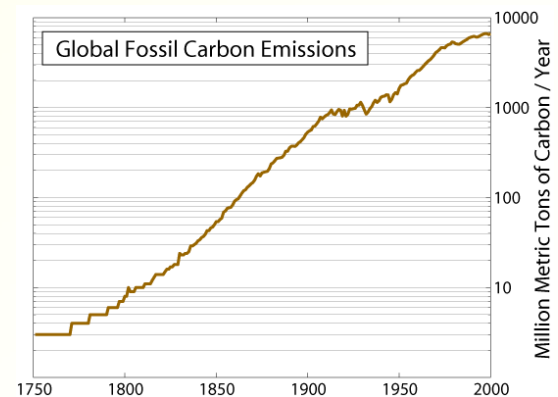
Climate change: can we help?

(Good reading material: the Revenge of Gaia by J. Lovelock)



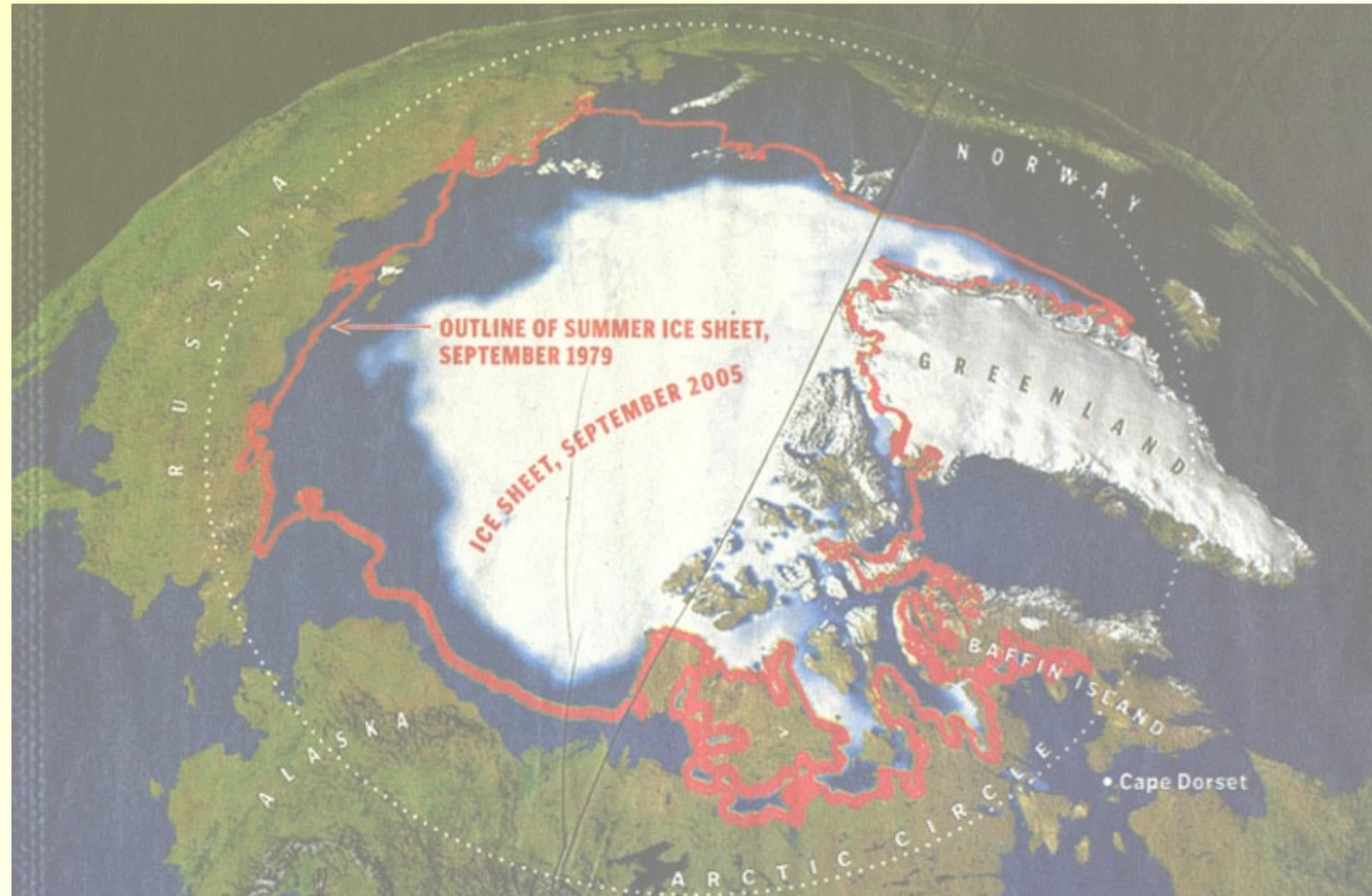
http://en.wikipedia.org/wiki/Image:Mauna_Loa_Carbon_Dioxide.png

QCL workshop CO2 emissions:
4 Tonnes per participant from US
= yearly personal quota
at current unsustainable rate



http://upload.wikimedia.org/wikipedia/en/5/50/Global_co2_emissions_graph.png

Global Warming



Effects of Sea level rise

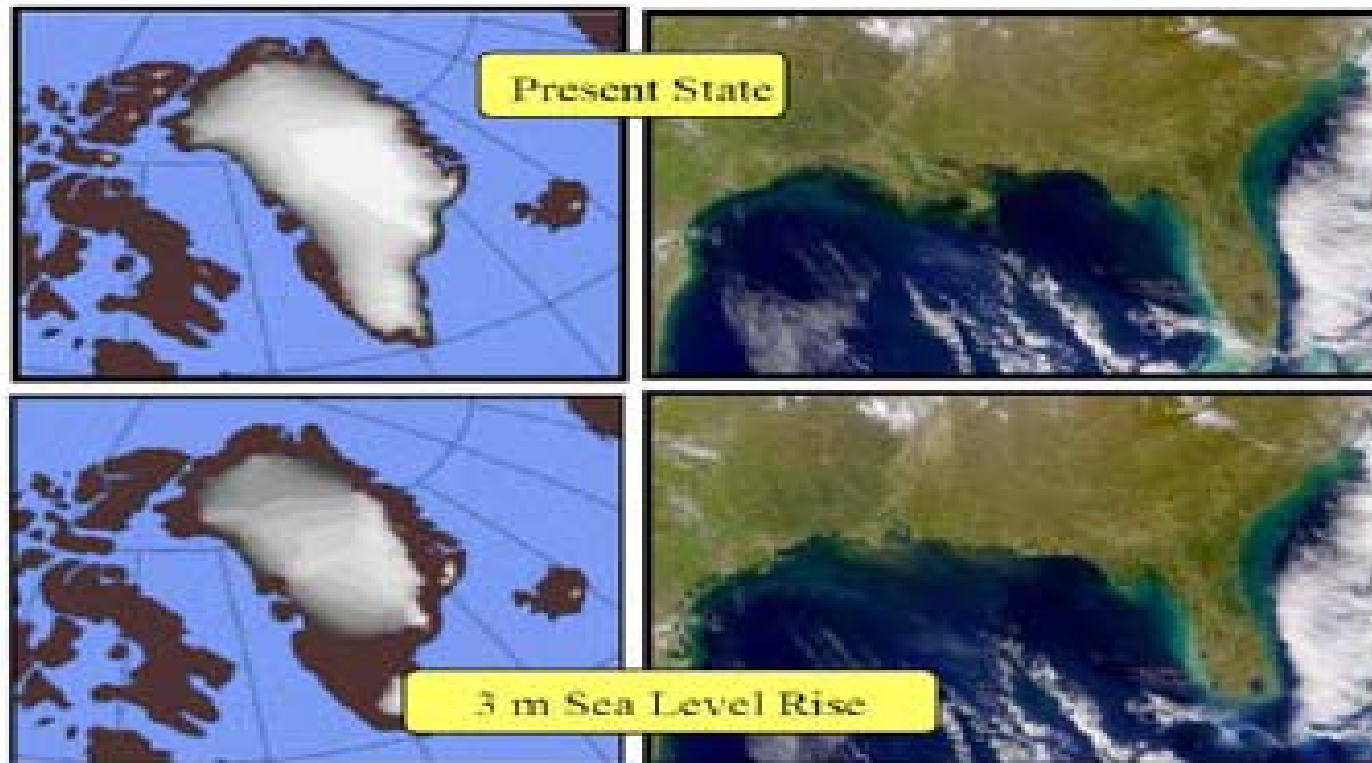
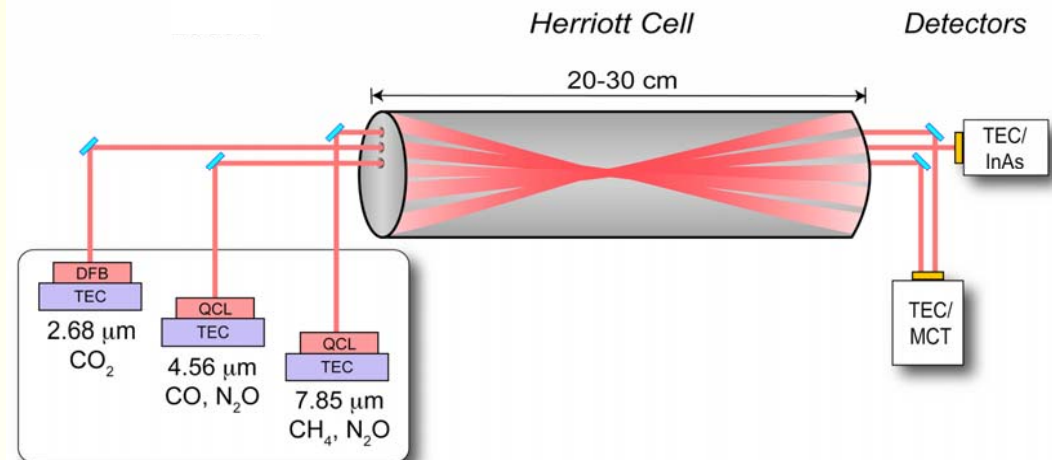


Figure 1 – The Gulf coast of the United States shown in its present state (top panels) and with 3 m of sea level rise (bottom panels). Preventing a catastrophe of this magnitude will require disciplined synergy between science, technology, and public policy.

One Example of QCLs in the Field: Measuring Tracers on a UAV

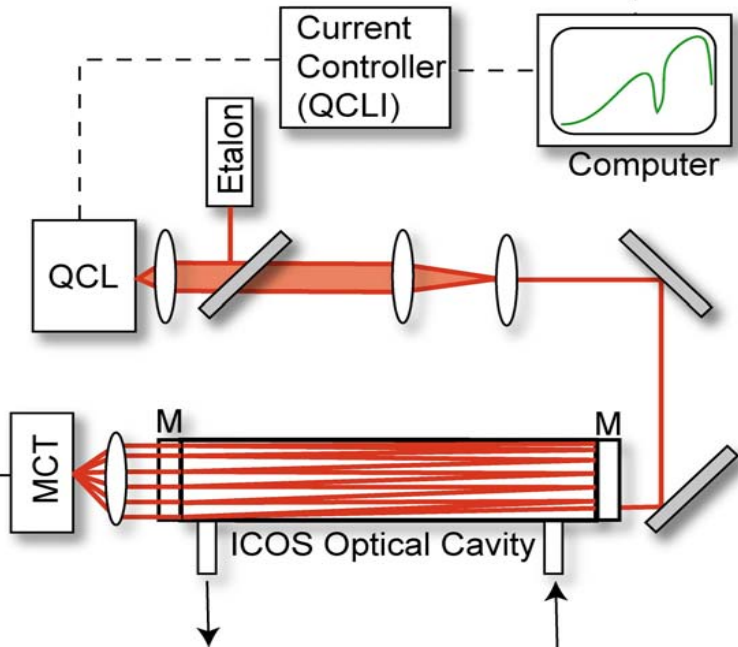


In order to make *in situ* measurements of trace gases over a broad area, UAVs could be used. Because of their size, stability, and low power consumption, **QCLs are ideally suited to UAV environments.**



Anderson group, Harvard

Integrated Cavity Output Spectroscopy (ICOS) in the Field



Light from a CW QCL is coupled into a high finesse cavity where it makes thousands of trips before escaping. Our ICOS instrument achieves a pathlength of 4 km using mirrors that are just 1 m apart.

Anderson group, Harvard

The instrument is loaded into the belly of NASA's WB-57 each morning. The plane flies through the upper troposphere and lower stratosphere.

The pilot need only flip a switch to turn on the ICOS instrument. All control circuits and data acquisition are automated.

Probing aerosols with LIDAR

Atmospheric aerosols are important for several reasons:

- ◆ They transport nonvolatile material such as soot and dust, thereby affecting tropospheric air quality
- ◆ They serve as cloud condensation nuclei, strongly influencing the atmospheric chemistry and radiative balance of the planet
- ◆ NASA mission will harness recent developments in Light Detection and Ranging (LIDAR) to examine the aerosol-cloud system from space.
- ◆ Although the strong absorption of aerosol particles in the mid-IR makes this spectral region an attractive choice for aerosol LIDAR from ground stations, UAVs, and satellites, **the current lack of high power single mode and collimated light sources has so far been a limiting factor. While ready laser collimation is desired for any application, it is especially important in LIDAR applications where propagation distances of hundreds of kilometers are common.**

Proposed Altair Payload

Cloud/Particle
LIDAR

Pressure
Temperature
Telemetry/GPS

Tracer Measurements

Methane (DFG/Herriott)
N₂O (QC/Herriott)
Carbon Dioxide (DFG/Herriott)
Carbon Monoxide (QC/Herriott)
Formaldehyde (Fiber/CRDS)
Formaldehyde (Fiber/LIF)
Water (NIR DFB/Herriott)
Total Water (NIR DFB/Herriott)
Water (Lyman Alpha)
Total Water (Lyman Alpha)
Ozone (UV Absorption)

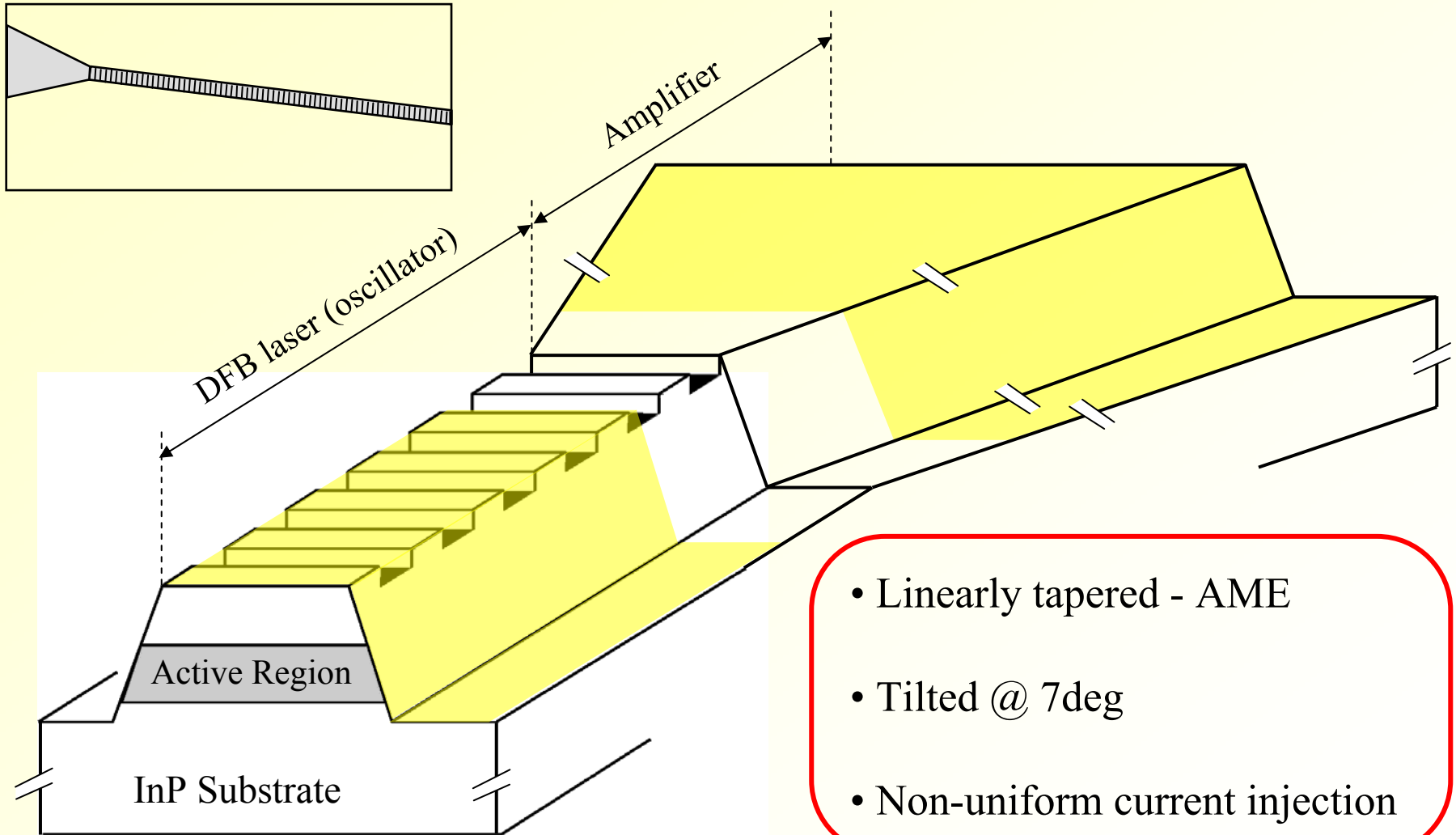
Radical Measurements

OH (Ti:Saph/LIF)
HO₂ (Ti:Saph/LIF)
NO₂ (Ti:Saph/LIF)
ClO (RF)
BrO (RF)
IO (Ti:Saph/LIF)
NO (Ti:Saph/LIF)

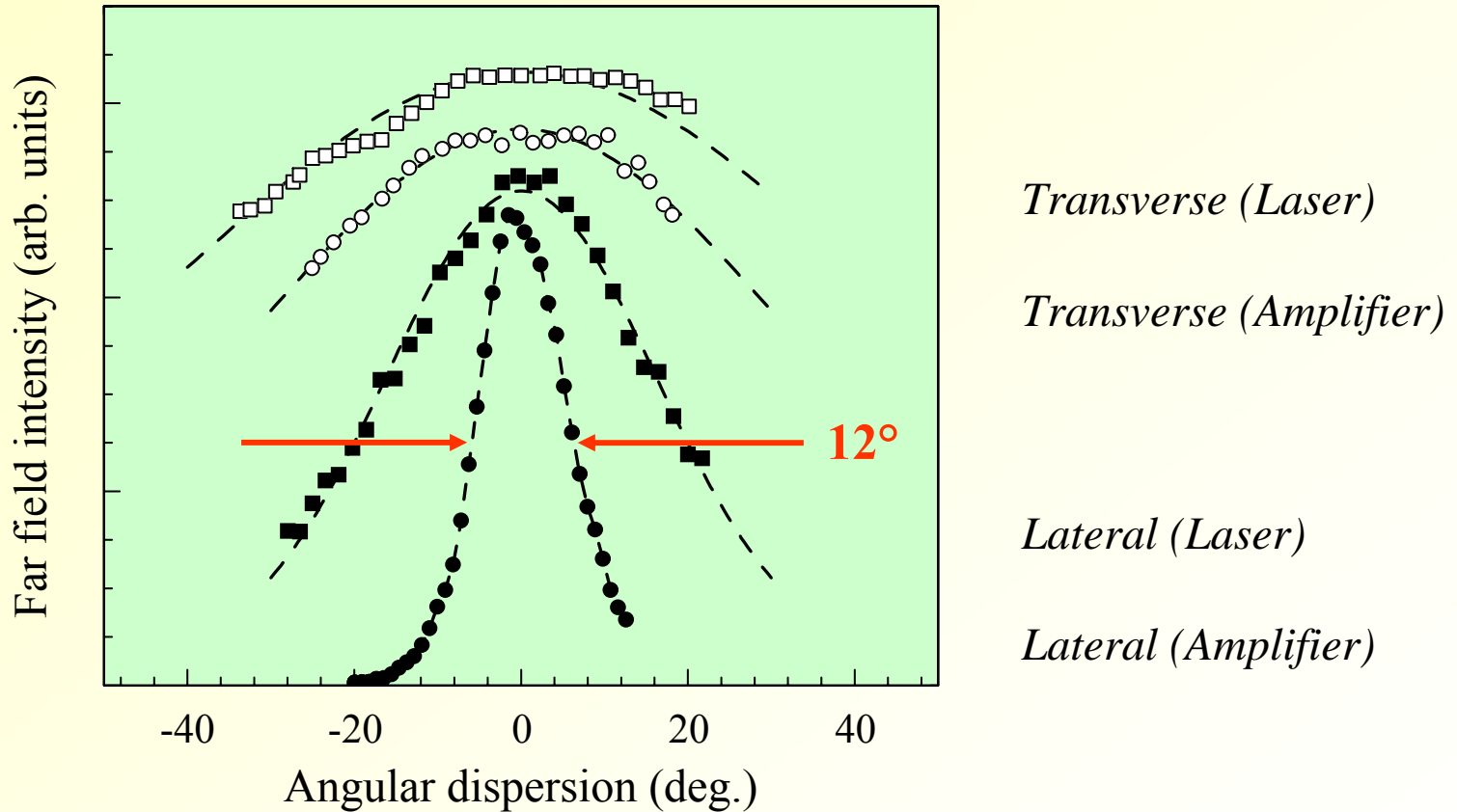
Requirements

- high (pulsed) output power
- single mode
- high directional laser beam preferred

Master Oscillator Power Amplifier for high power single mode emission



Far field intensity of MOPA



M. Troccoli et al. Appl. Phys. Lett. 80,4103 (2002)

Integration of plasmonic collimator on facet for highly directional beams
High peak power

Acknowledgments

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