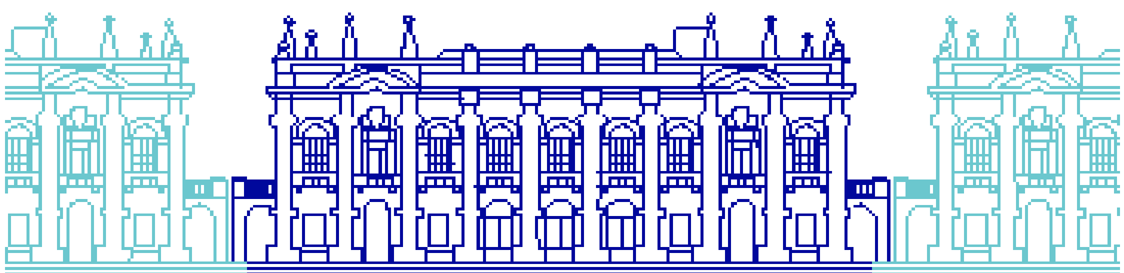
# ORGANIC LASERS

Margherita Zavelani-Rossi



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# OUTLINE

INTRODUCTION

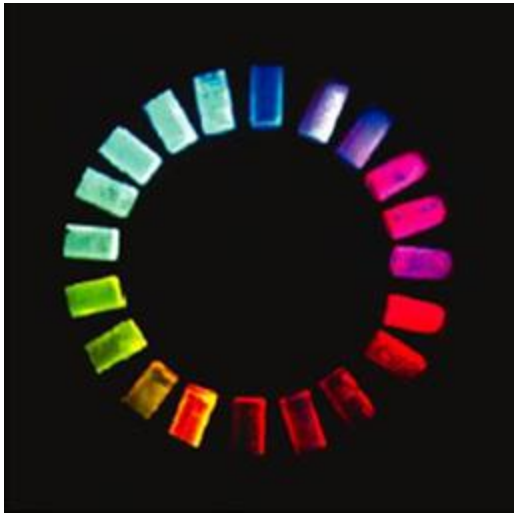
The LASER
- active material
- optical resonator

O- LASERS EXAMPLES
- external resonator
- compact micro-cavity
- DFB resonator

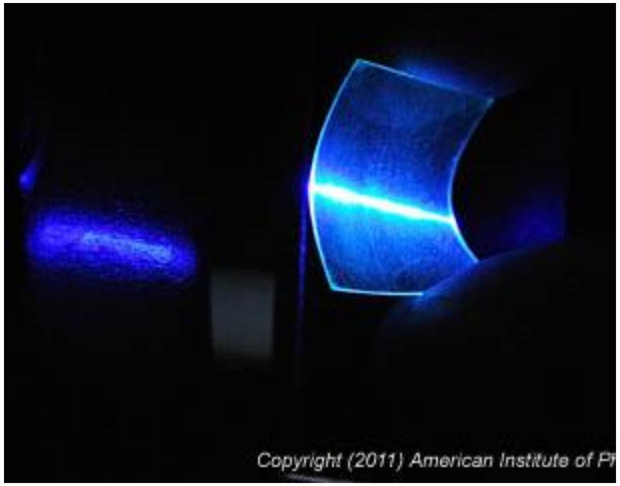
Further developments
- electrical pumping
- diode pumping
- LED pumping
- photonic applications

CONCLUSIONS

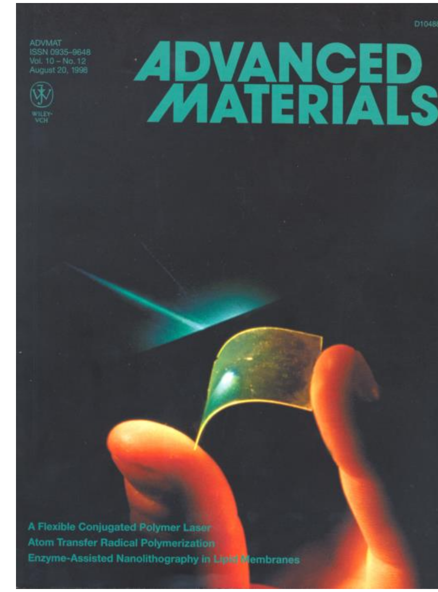
# ORGANIC MATERIALS for lasers



- high photo- and electro- luminescence efficiency
- tunable emission all over the visible, by chemical synthesis
- ease of processing (solution) - low cost technology
- appealing mechanical properties (flexibility)
- large active areas
- large optical absorption cross section (~$10^{-15}$ $cm^2$)



# ORGANIC LASERS




Organic lasers have been shown with:

- various materials
- various optical cavities
- optical pumping

**advantages**:

- small devices
- compatible for integrated optics (integration to planar waveguides)
- connection to plastic optical fibers (POF) ('last mile' applications)

- by soft lithography technique
- compact cheap flexible sources

# the LASER

*"Light Amplification by Stimulated Emission of Radiation"*

- OBTAINED with:
  1) active material
  2) resonator
  3) pump / external excitation source

✓ PROPERTIES of laser beams :
- directionality
- brightness
- monochromaticity
- coherence

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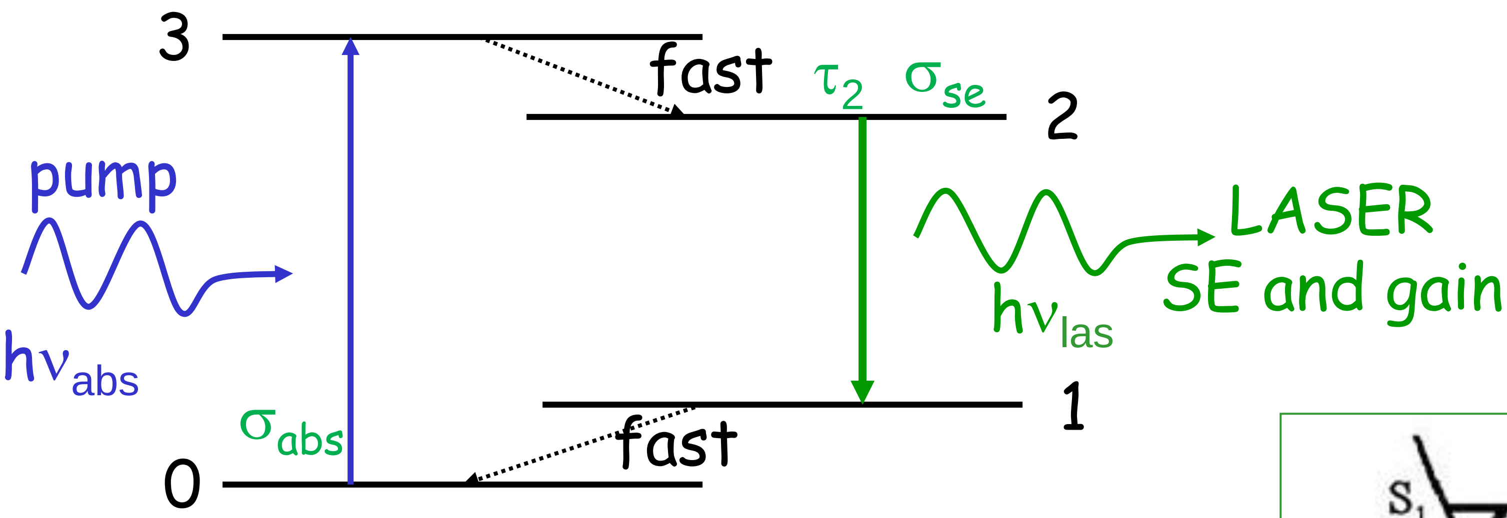
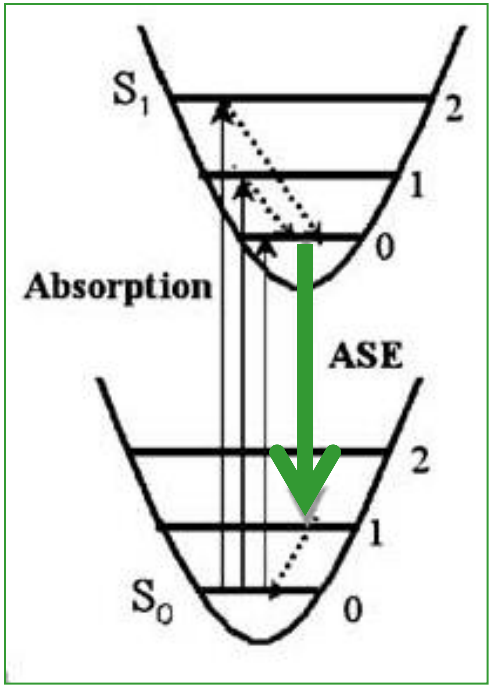
# 4-LEVEL SYSTEM




- $\sigma_{abs}$ absorption cross section $\Rightarrow$ **big**
- $\sigma_{se}$ stimulated emission cross section $\Rightarrow$ **big**
- $\tau_2$ lifetime laser upper state $\Rightarrow$ **long**
  - $1/\tau = 1/\tau_{spontaneous} + 1/\tau_{non\ radiative}$
  - population inversion $(N_2\text{-}N_1)>0$
- gain > losses

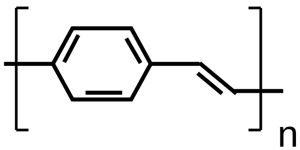
# ACTIVE MATERIAL REQUIREMENTS

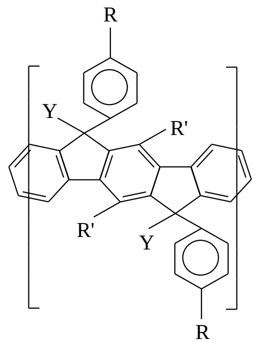
- large $\sigma \cdot \tau$ product ⇒ low threshold ( $\propto$ loss / $\sigma \cdot \tau \cdot l_{active\ material}$ )
  ⇒ high efficiency (slope eff $\propto$ loss / $\sigma \cdot \tau \cdot P_{th}$ )
  $\sigma \cdot \tau = 10^{-26} - 10^{-27}\ cm^2 \cdot s$ ($\sigma \sim 10^{-16} cm^2$ - $\tau \sim 10^{-10} s$ ) 😐
- long laser level lifetime $\tau$ ☹ (pump with fs-ns pulses, short cavities)
- good quality ⇒ small scattering losses ☺
- no absorption of SE
  - from ground state ☺
  - from triplet states 😐
  - from charge excited states ☹
- broad gain bandwidth ⇒ tunability ☺
- stability under high excitation ⇒ long device lifetime 😐
- good charge injection and transport ⇒ electrical pumping ☹

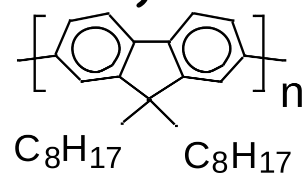
# ORGANIC ACTIVE MATERIALS

Two approaches:

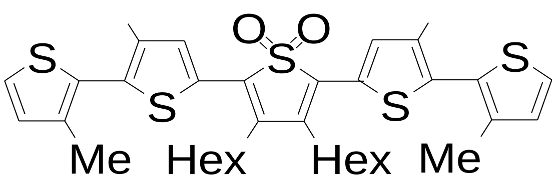
- **Polymers** (amorphous, ease of processing, soluble, good solid state emission, low inter-chain interactions)
  ( PPV, m-LPPP, PFO, ... )

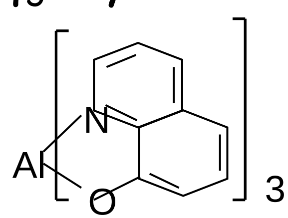


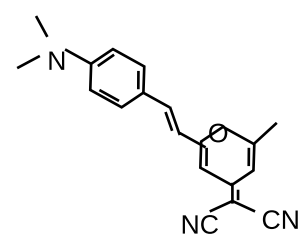



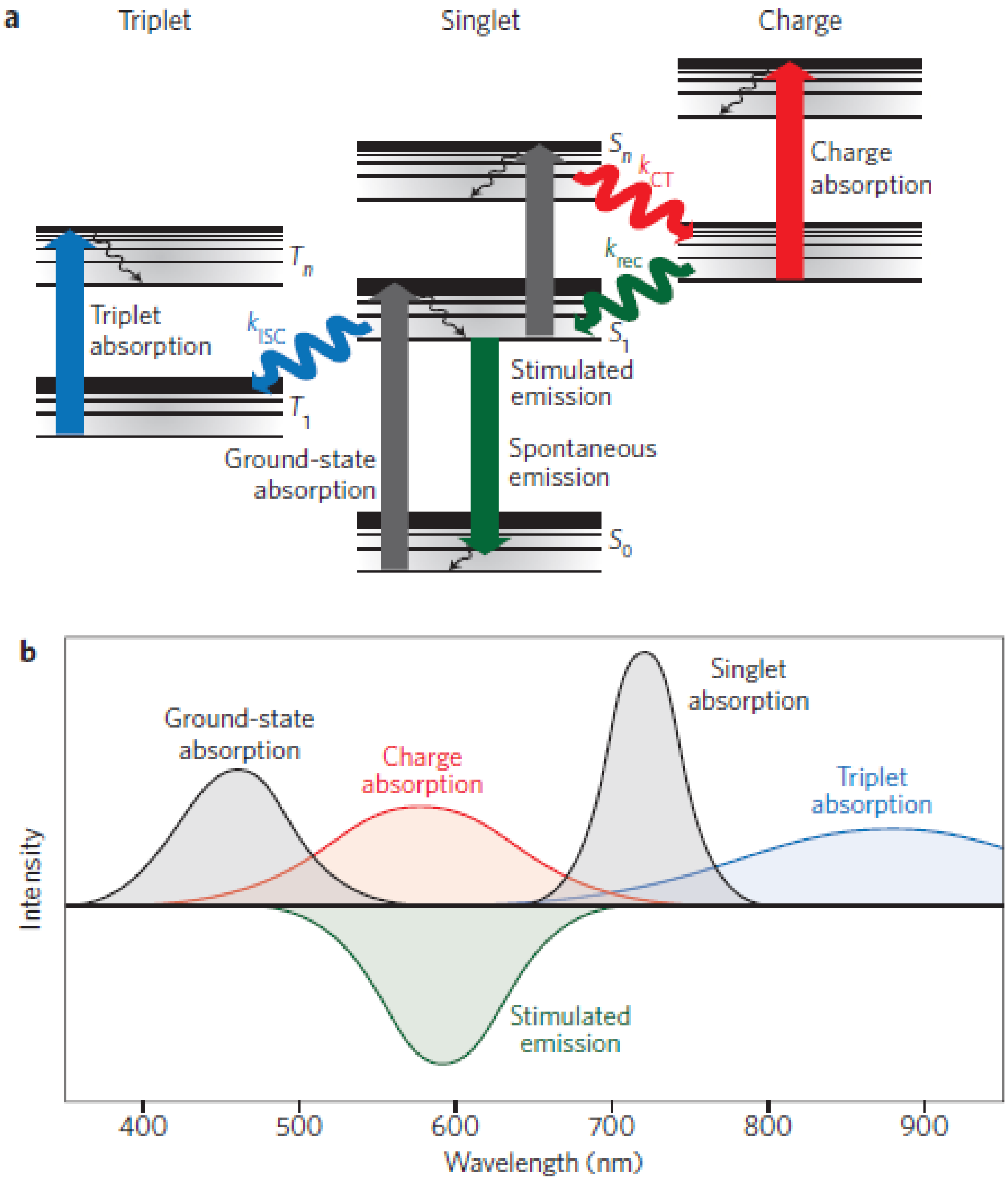

- **Small molecules** (crystalline, high mobility, larger inter-chain interactions, good control on morphology)
  ( thiophenes, $Alq_3$+dye , ... )



# ORGANIC ACTIVE MATERIALS

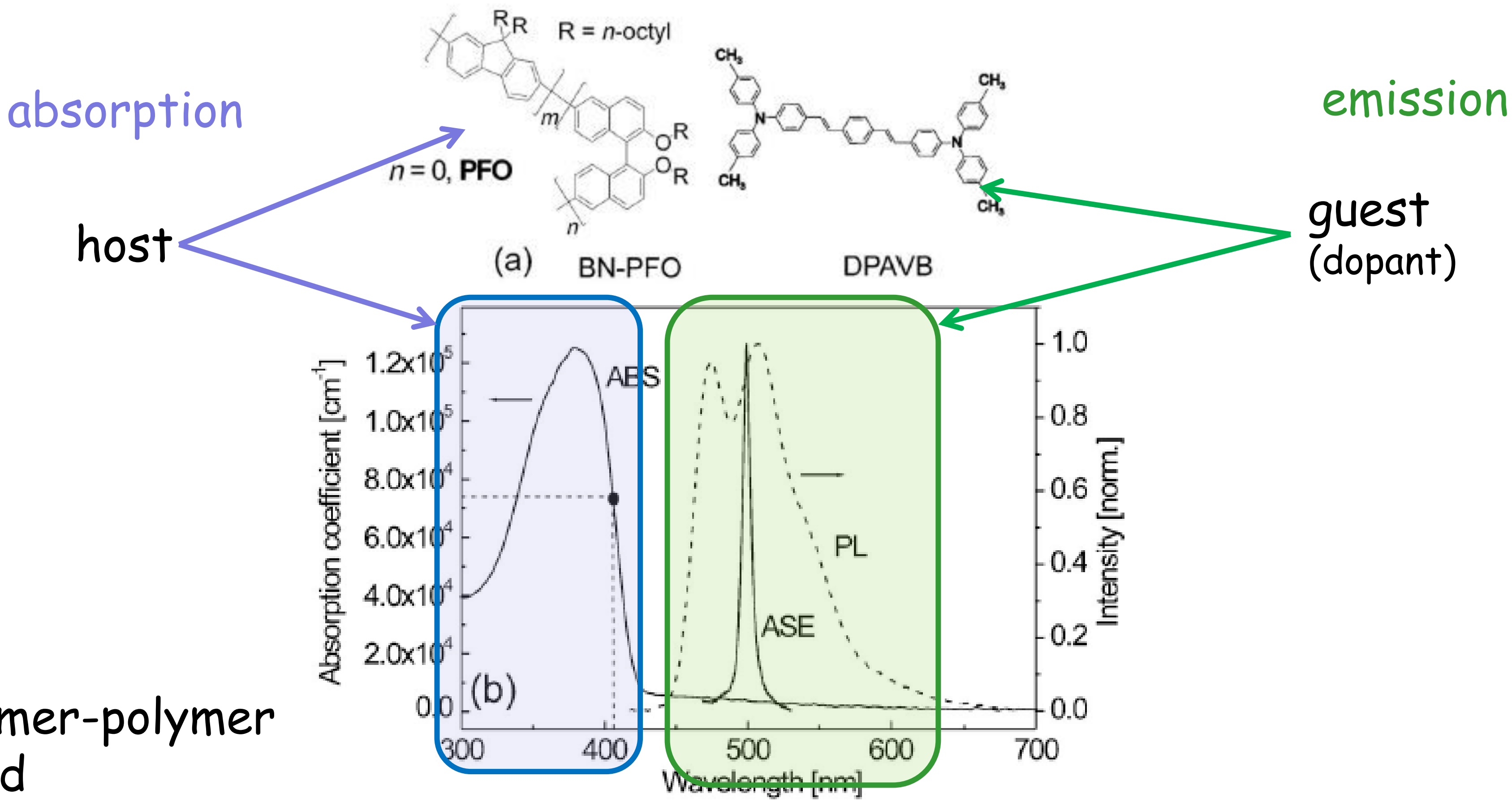



- singlet-singlet annihilation
- exciton dissociation
- ground state self-absorption
- absorption from triplet states
- absorption from charge states

J. Clark and Guglielmo Lanzani, Nature Photonics 4, 438 (2010)

# LOW SELF-ABSORPTION
## guest-host systems

- blends
- host fluorescence @ guest absorption
- non-radiative energy transfer
- increase the separation between optical absorption and laser emission

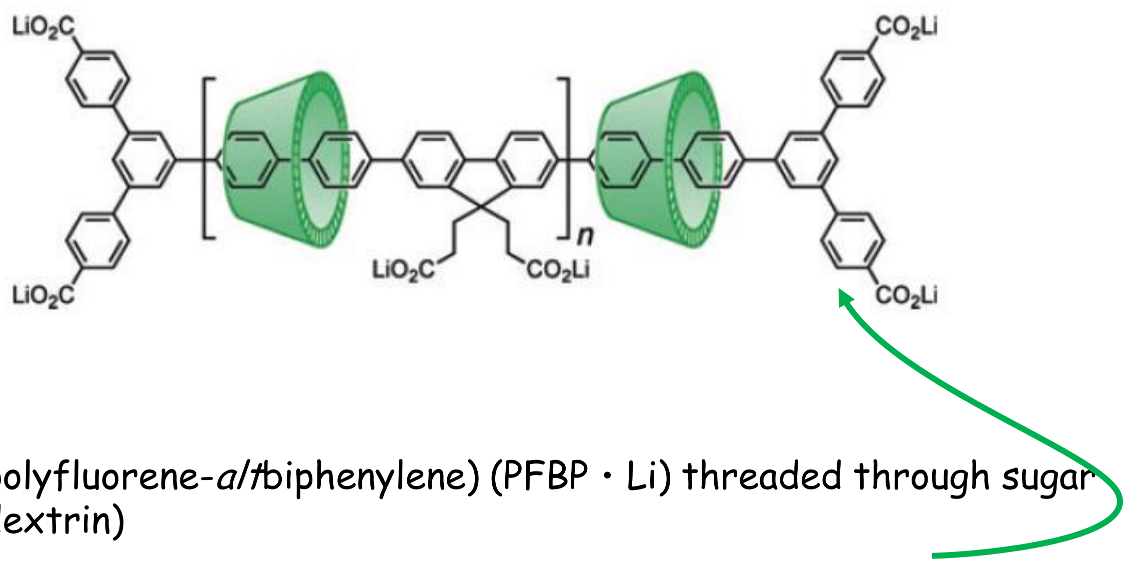



Polymer-polymer blend

T. Riedl *et al.*, Appl. Phys. Lett. 88, 241116 (2006)

# LOW CHARGE-ABSORPTION

- low charge generation
- low chain-chain (inter-chain) interactions
  - suitably design the sidechains of the polymer backbone
  - introduce "spacers"




poly-rotaxane:

conjugated polymer (polyfluorene-*alt*-biphenylene) (PFBP • Li) threaded through sugar macrocycles (β-cyclodextrin)

- insulated molecular wires

M. M. Mroz, et al., Appl. Phys. Lett. 95, 031108 (2009).

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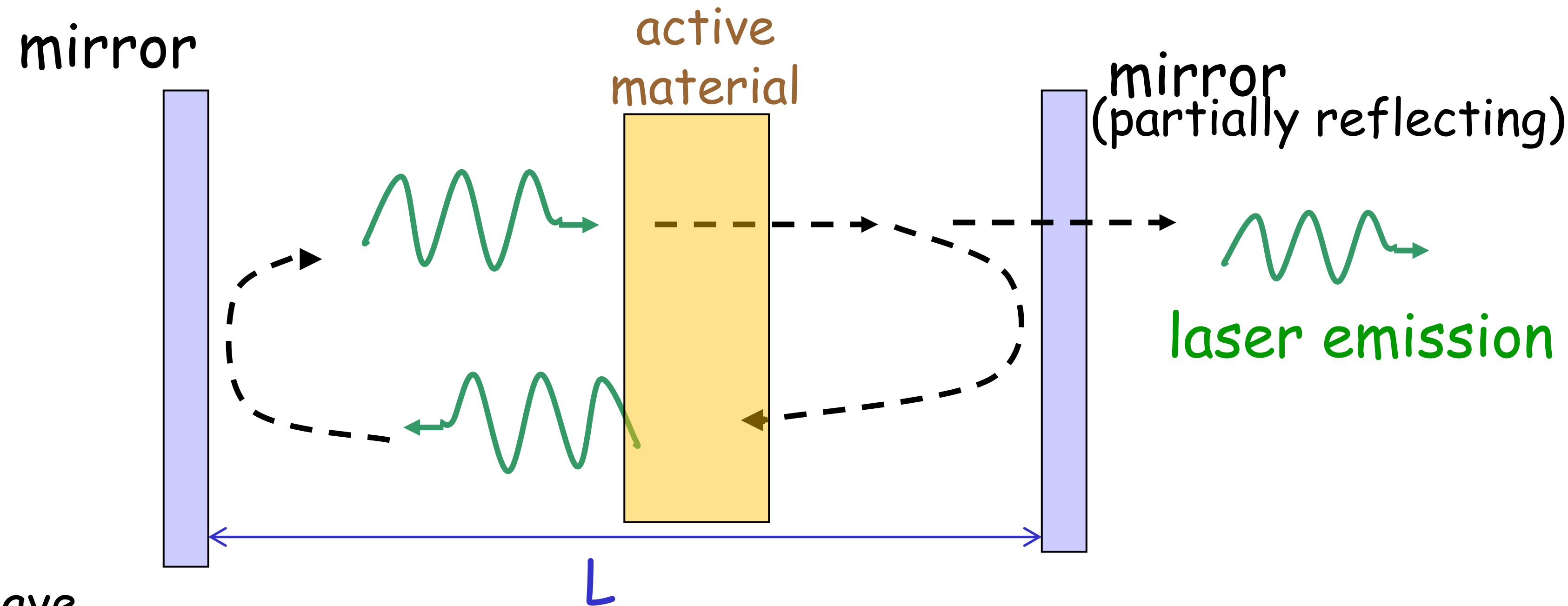
## CONCLUSIONS

# LASER OPTICAL RESONATOR




em wave

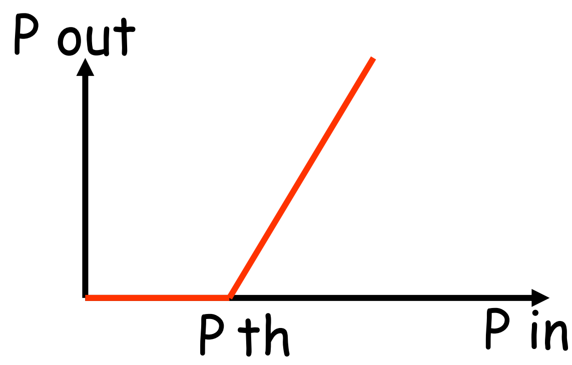
- phase condition $k\, 2L = 2\pi/\lambda\, 2L = 2n\pi$
- constructive interference
- em mode

-feedback

-threshold condition : gain = losses

# LASER ACTION

To unambiguously identify laser action:




1) clear threshold in in-out characteristic
   Threshold $\propto$ loss / $\sigma \cdot \tau \cdot l_{active\ material}$

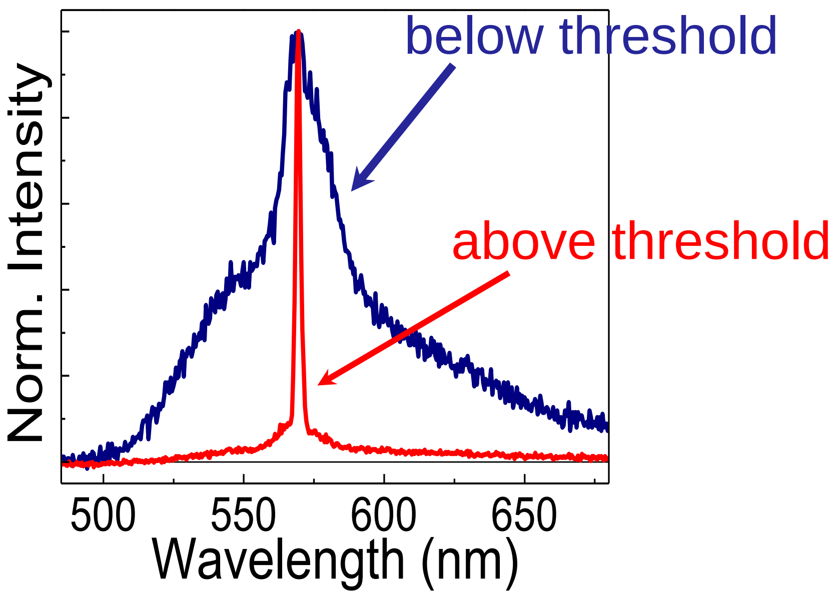
2) linear in-out characteristic

3) directionality - diffraction limited divergence

4) significant spectral narrowing

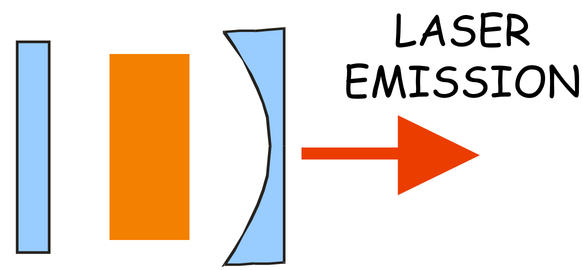
5) existence of laser modes

6) coherence

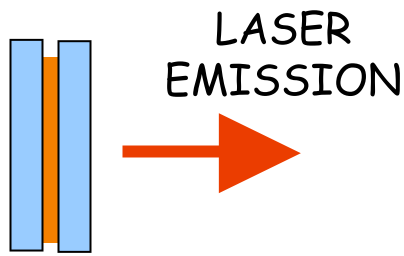


# RESONATORS

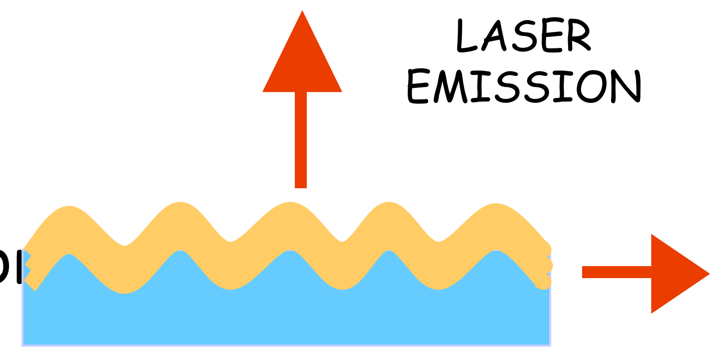
- external resonator/cavity




- microcavity




- Distributed FeedBack (DFB) resonator




- others

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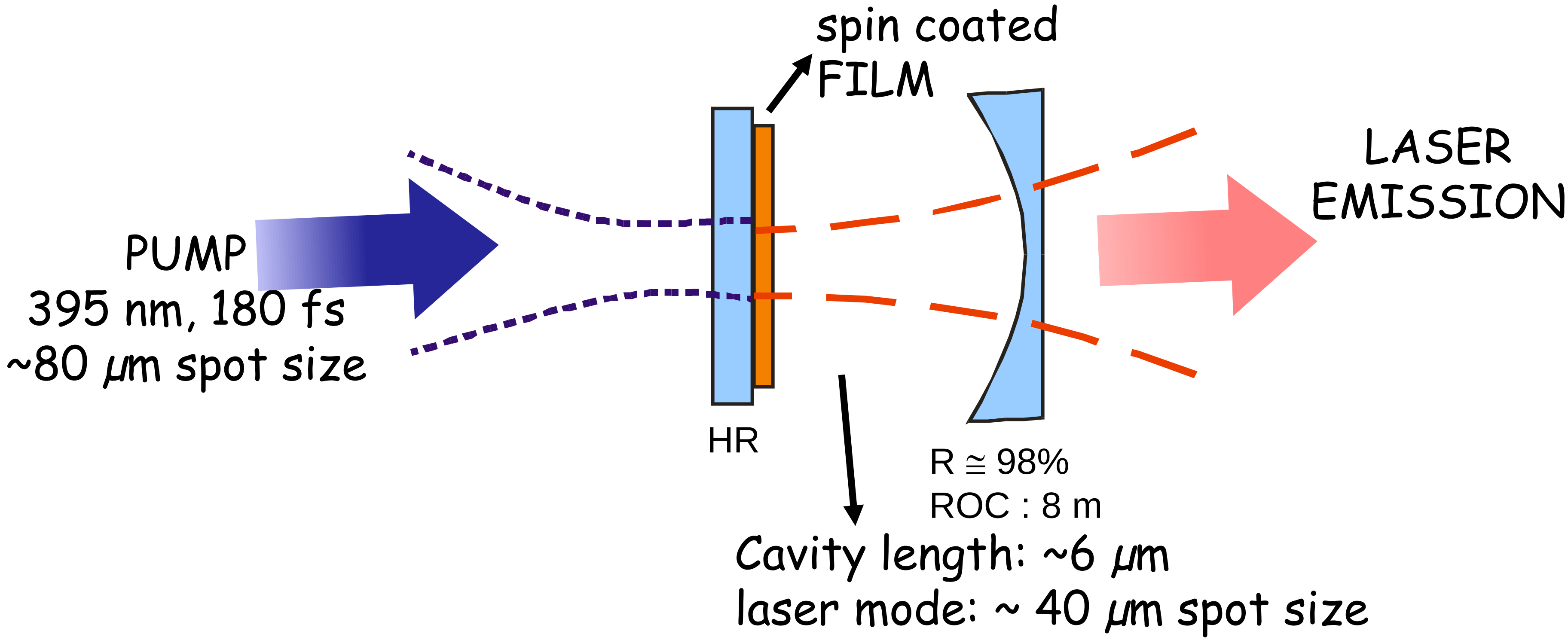
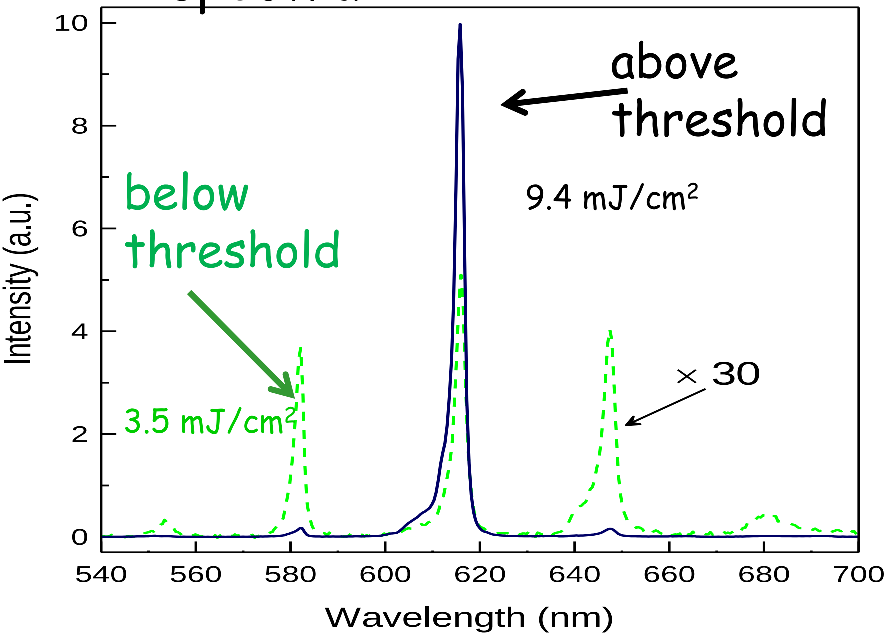
# THE CAVITY



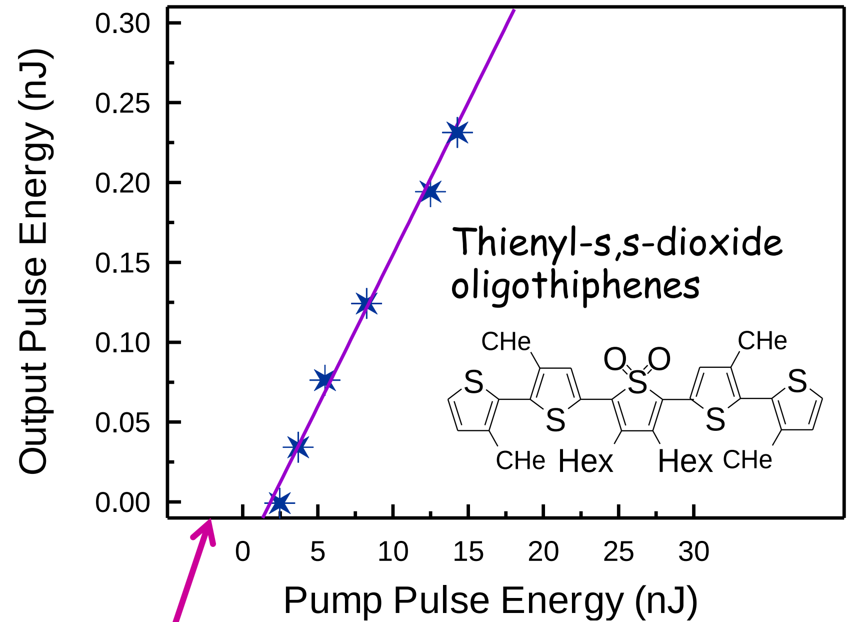

- plane-concave resonator
  - stable resonator
  - good pump-mode overlap
- single longitudinal mode
- tunability (by changing the cavity length $k2L = 2n\pi$)
- spincoated organic film

# LASER EMISSION

spectra




input-output characteristics



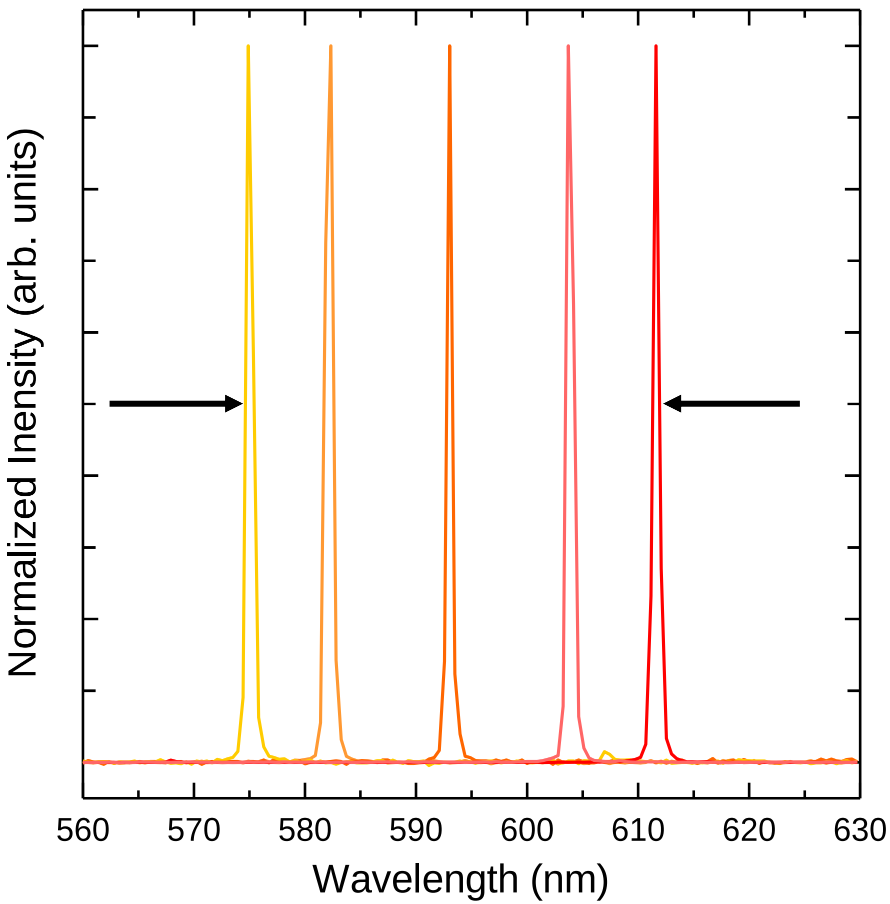

- Below threshold
  ⇒ modulated fluorescence
- Above threshold
  ⇒ single laser mode
  - Δλ ~ 2 nm (FWHM)

Laser threshold:
2 nJ pump energy (10 μJ/cm$^2$ )

**efficiency 1.7 %**


*G. Barbarella et al., Adv. Mater. 10, 551 (1998)*
*M. Anni, et al. , Appl. Phys. Lett. 78, 2679 (2001)*
*M. Zavelani-Rossi, et al. Appl. Phys. Lett. 79, 4082 (2001)*

# LASER EMISSION TUNABILITY




- Tunable wavelength (by changing cavity length)
- Tunability on 37 nm (from 575 to 612 nm)
- $\Delta\lambda$ < 1. 2 nm (FWHM)

M. Zavelani-Rossi et al, Synt. Met. 139 (2003)

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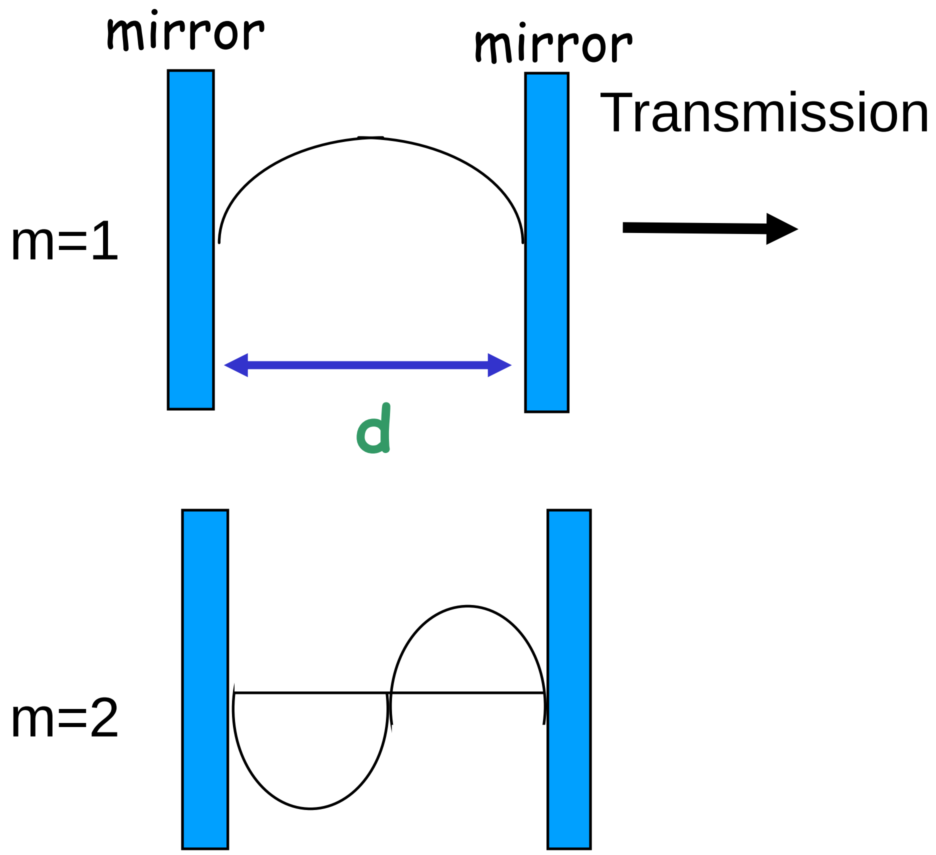
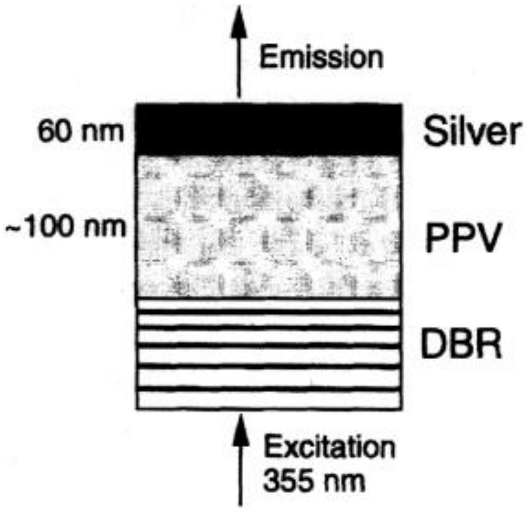
# THE PLANAR MICRO-CAVITY



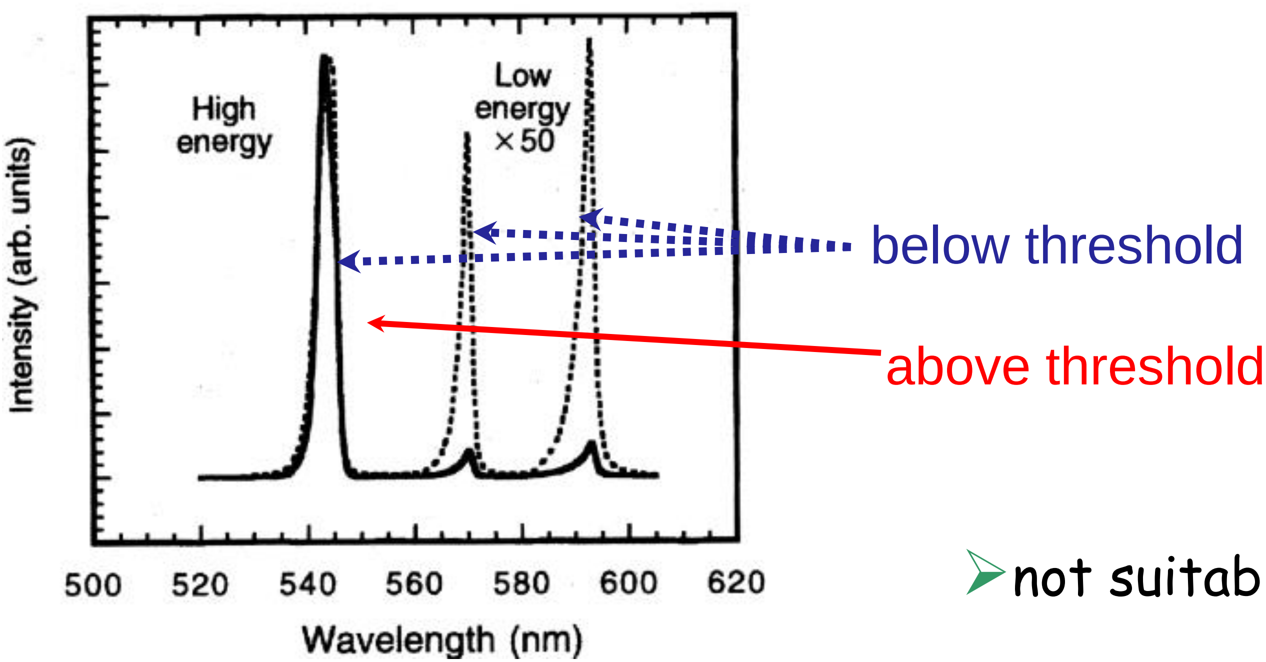

- Fabry-Perot resonator : $m\lambda/2=nd$ (eigen solution)
- cavity modes selected by d
- for m=1 or m=2 $\lambda$ is in the visible spectral region (d≈200nm)
- the active material fills the cavity

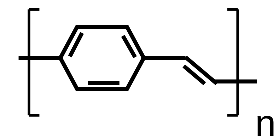
# FIRST PLANAR MICRO-CAVITY




- Commercial DBR
  + spincoated PPV (100 nm)
  + deposited silver

- threshold 200 $\mu J/cm^2$
  (100 nJ)



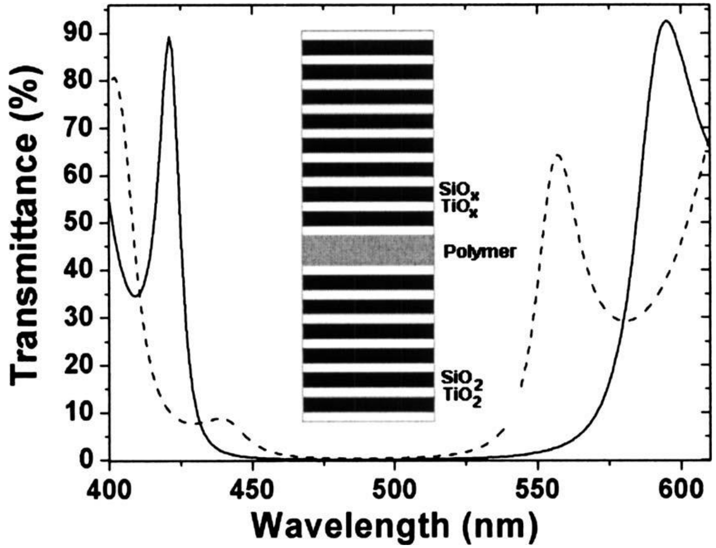




PPV
poly(*para-phenylene vinylene)*

- not suitable for integration

N. Tessler, G. J. Denton, and R. H. Friend, Nature 382, 695 (1996)

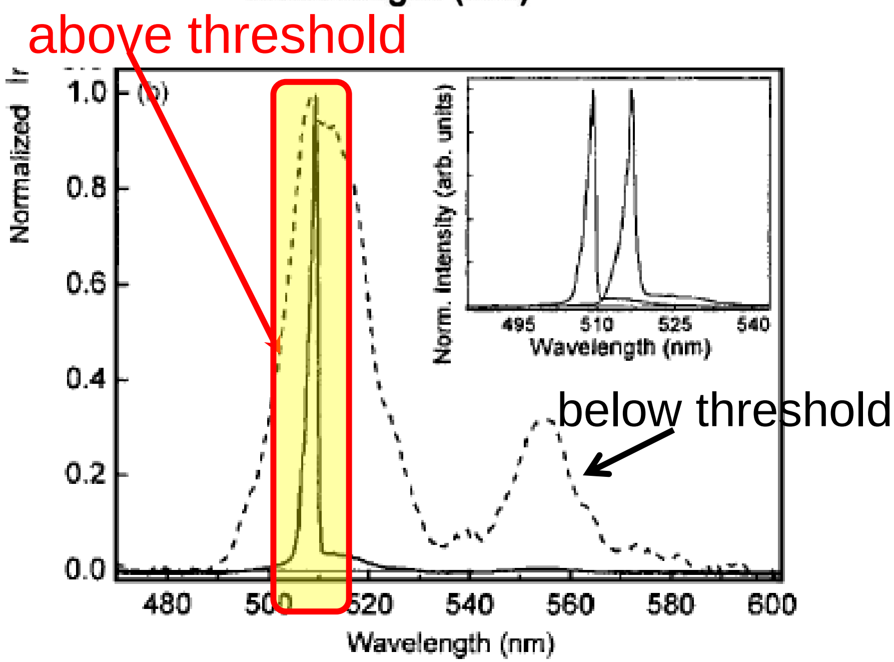
# MULTILAYERED MICRO-CAVITY

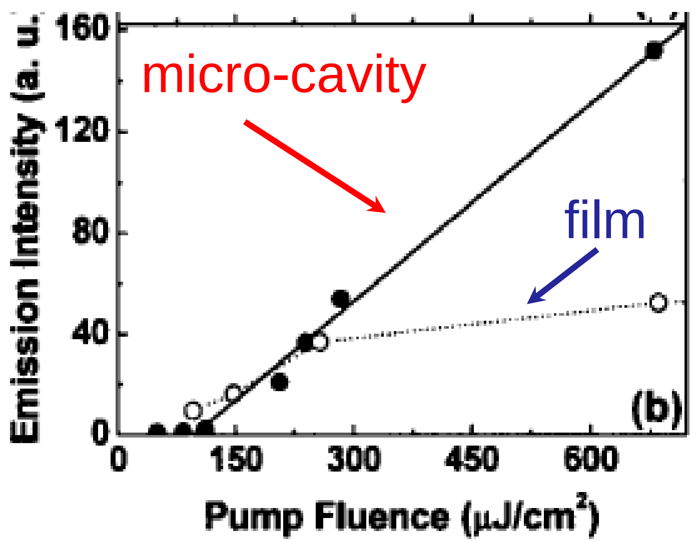



- multilayer (n-bilayers)
- low-T electron-beam evaporation
- $TiO_2$ / $SiO_2$ films
  + spincast polymer (155-nm thick)

- threshold 84 $\mu J/cm^2$

- suitable for integration

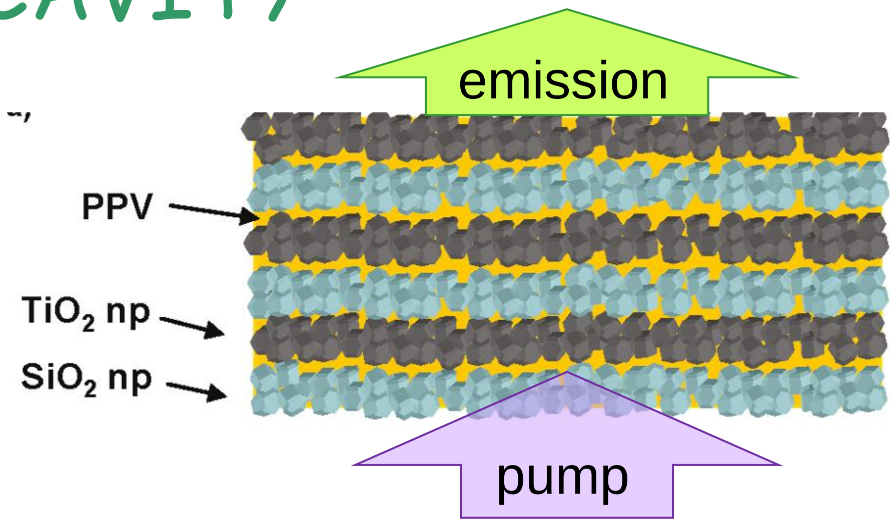


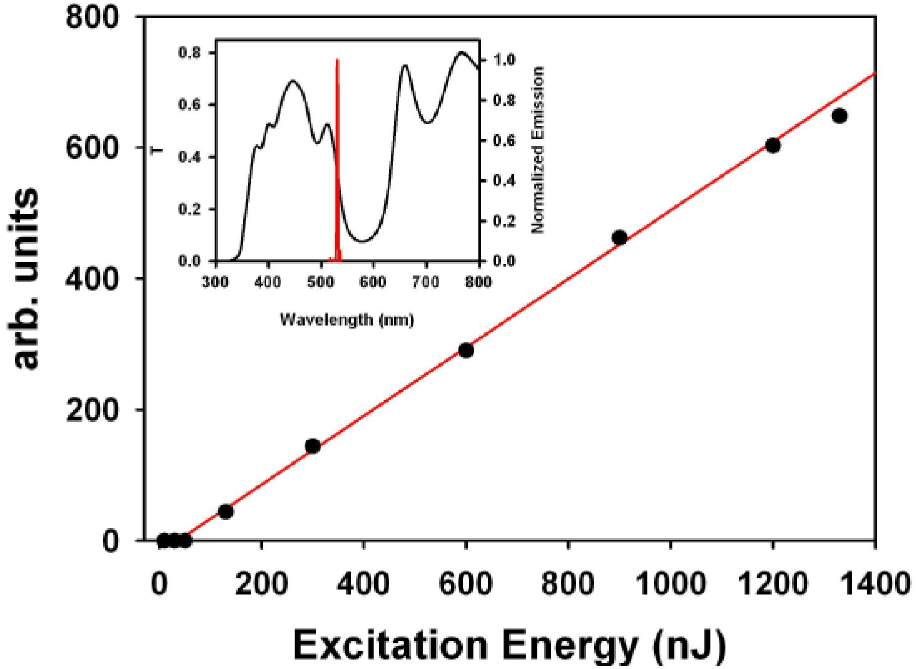



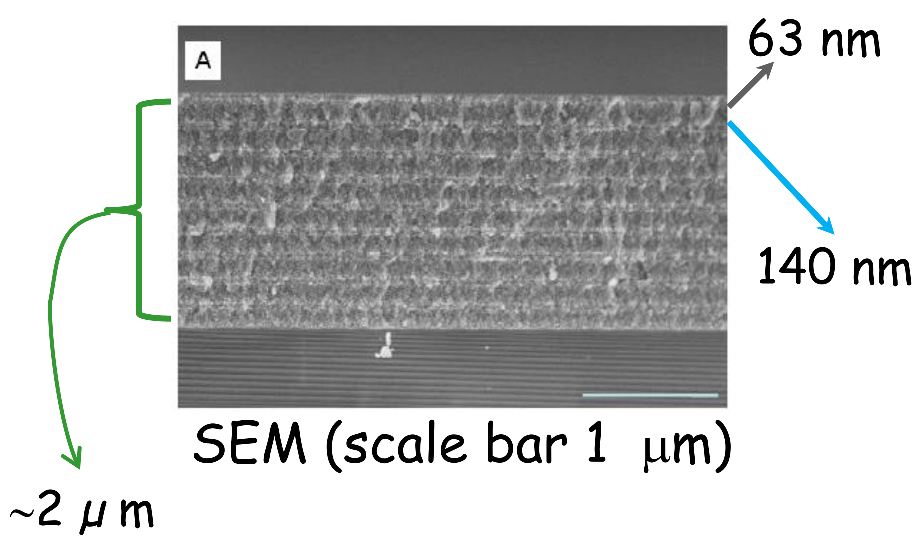

L. Persano, et al., Appl. Phys. Lett. 88, 121110 (2006)

# MULTILAYERED INFILTRATED MICRO-CAVITY

- multilayer by spin coating (11 bilayers)
- polymer (PPV) infiltrated




SEM (scale bar 1 μm)

D.P. Puzzo, et al., NanoLetters 9, 4273 (2009)

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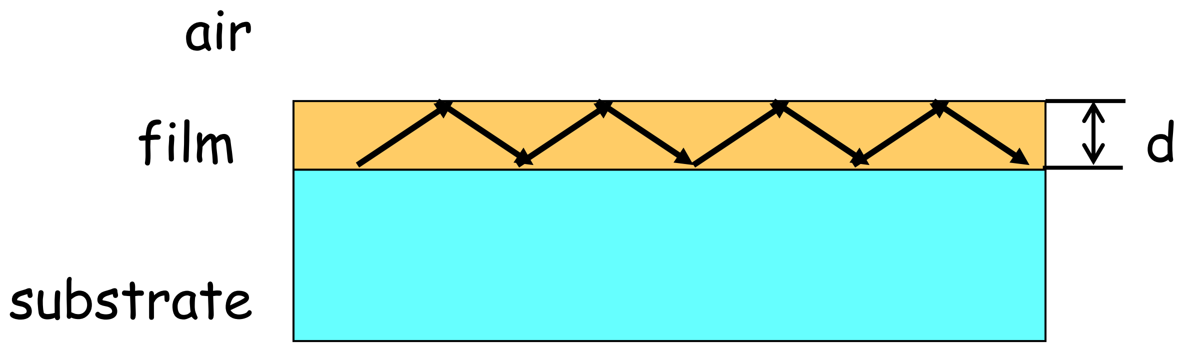
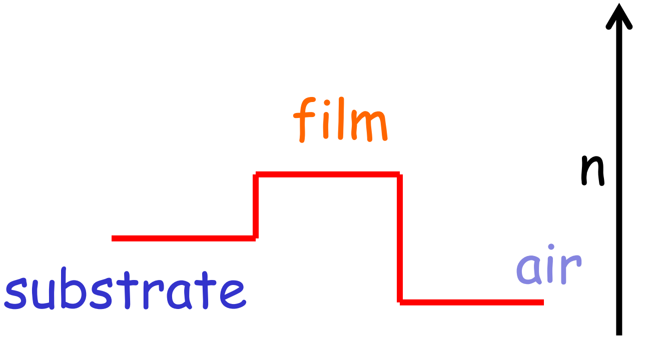
# PLANAR WAVEGUIDE



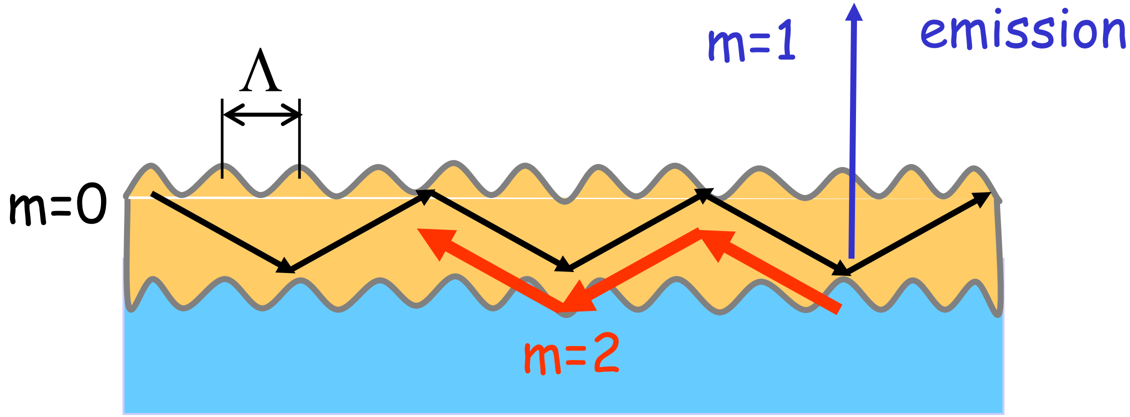

- guided modes require

  - condition on the index of refraction n (internal reflection)




  - cutoff thickness of the film d ⇒ wavelength discrimination (phase condition) small d, short $\lambda$

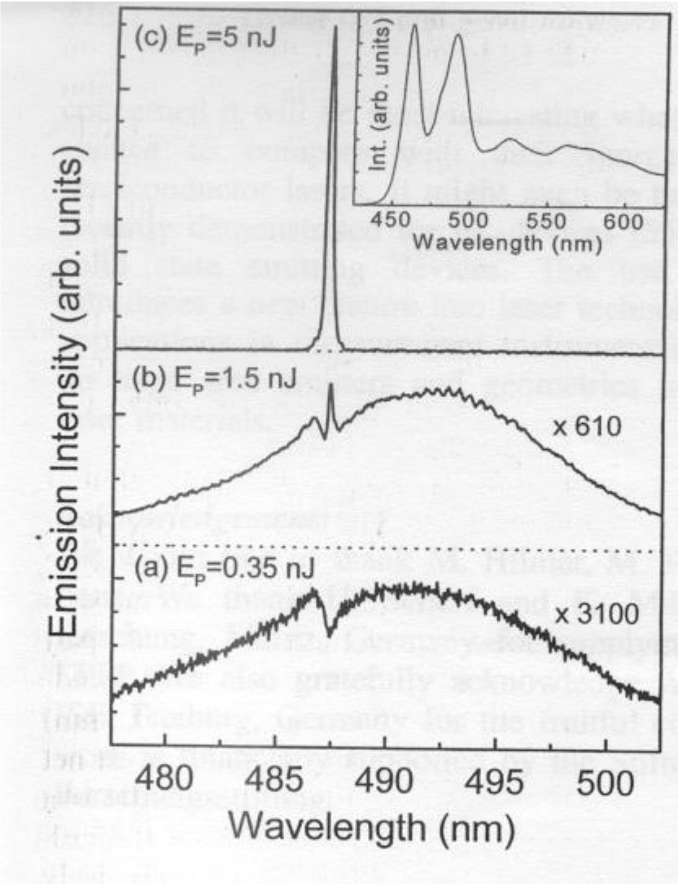
# DISTRIBUTED FEEDBACK

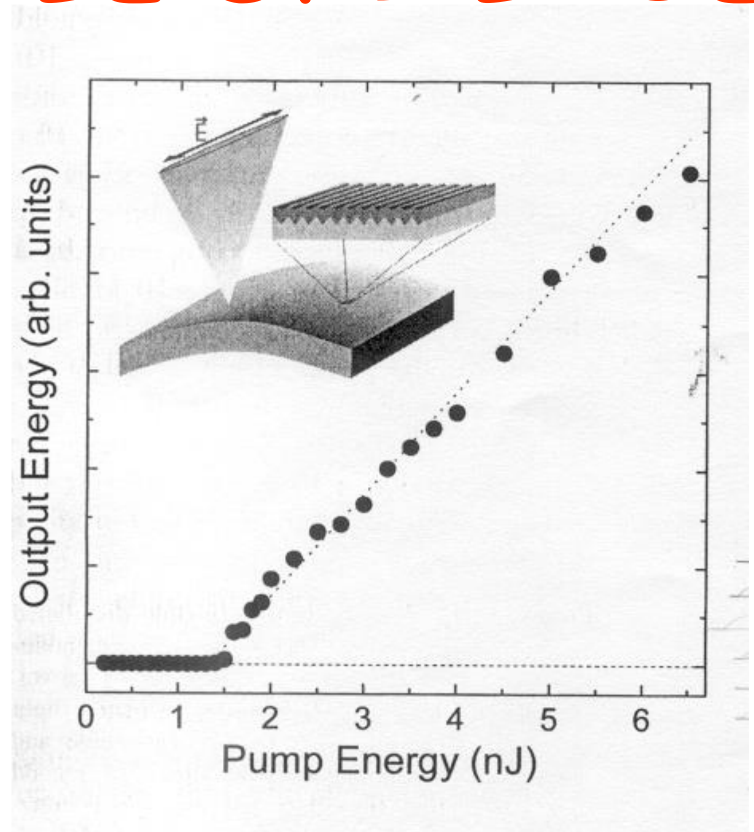



- Periodic modulation of the refractive index, gain and/or film thickness (=> Bragg reflection)
- diffraction grating
- m=1 first order diffraction => coupled out
- m=2 second order diffraction => fed into the counter-propagating wave => optical feedback

$$m \lambda_{Bragg} = 2 n_{eff} \Lambda$$

# FIRST FLEXIBLE DFB LASER



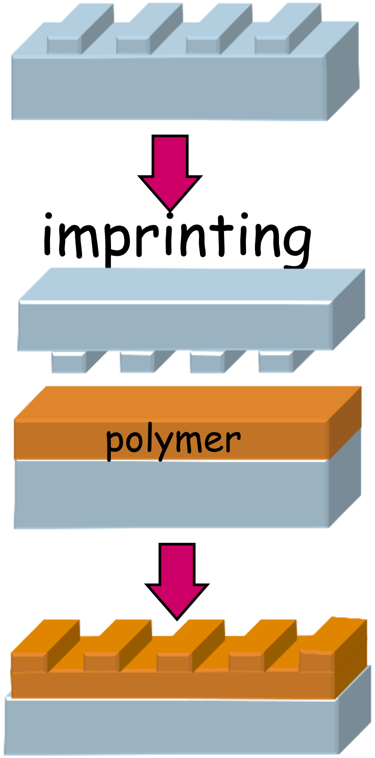

- Poly(ethylene terphthalate) (PET) with acrylic coating substrate
- UV embossing ⇒ modulation pitch 300nm, depth 10 nm
- LPPP by spincoating (440-nm thick)
- laser emission (single mode, polarized)

C. Kallinger et al. Adv.Mater. 10, 920 (1998)

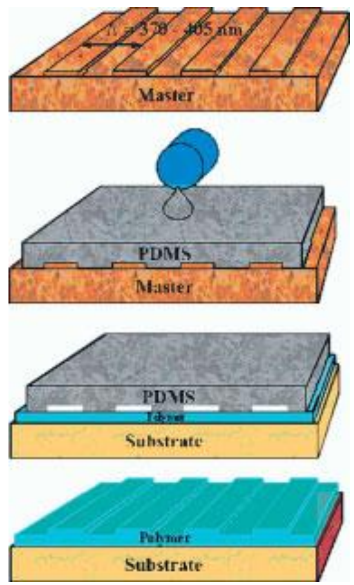
# IMPRINTED DFB LASER

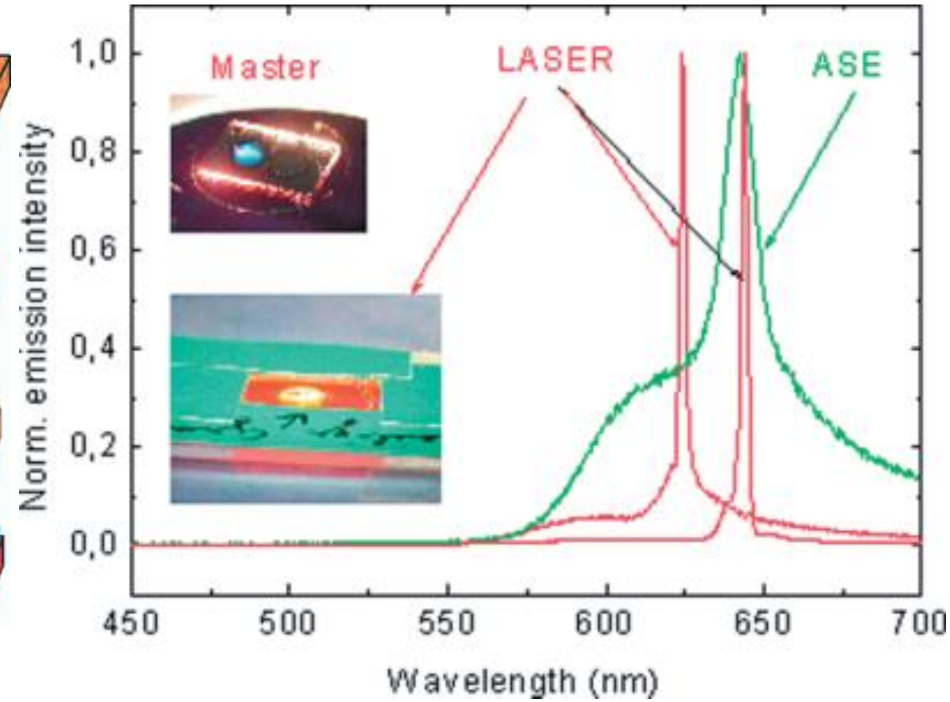
master
(Electron Beam Lithography)

imprinting




"imprinted"

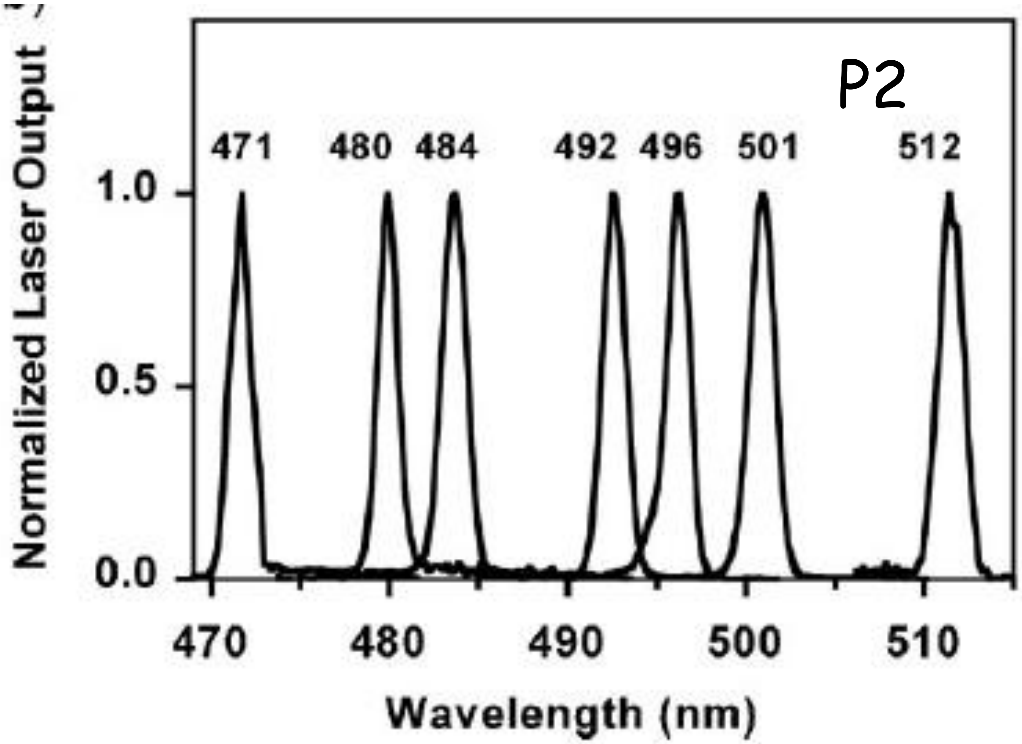


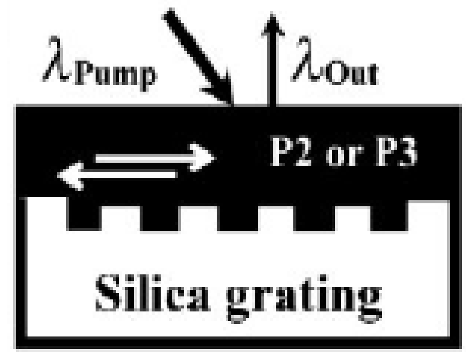

- simple low cost soft-lithographic technique
- modulation directly on the polymer (MEH-PPV)

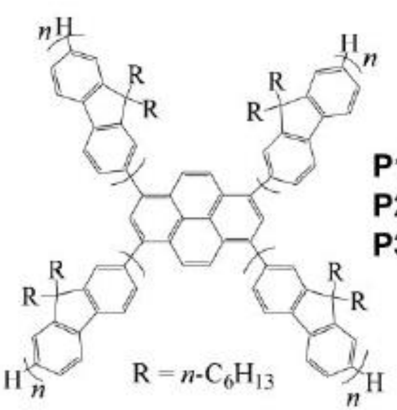
M. Gaal, et al., Adv. Mater. 15, 1165 (2003)

# TUNABILITY




Changing the film thickness
⇒ 41 nm range



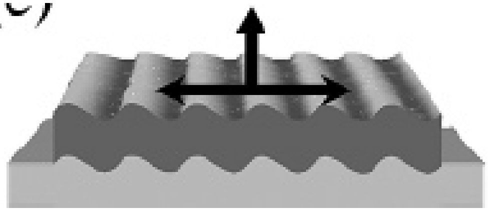

**P1**: $n$ = 1
**P2**: $n$ = 2
**P3**: $n$ = 3

Pyrene-cored
9,9 dihexylfluorne starbust

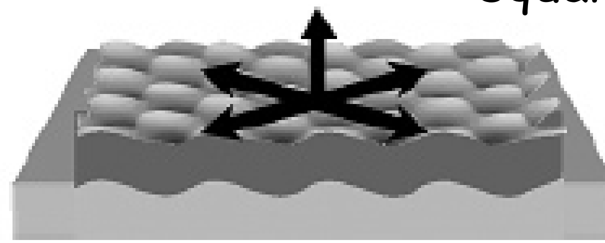
R. Xia, et al., Adv. Funct. Mater. 19, 2844 (2009)

# DFB GEOMETRIES

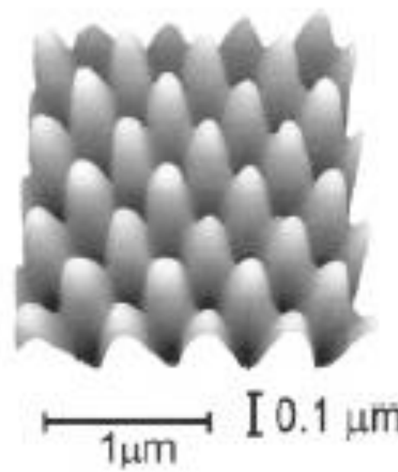
-standard
(1D)



- square array
2D



-concentric
circular



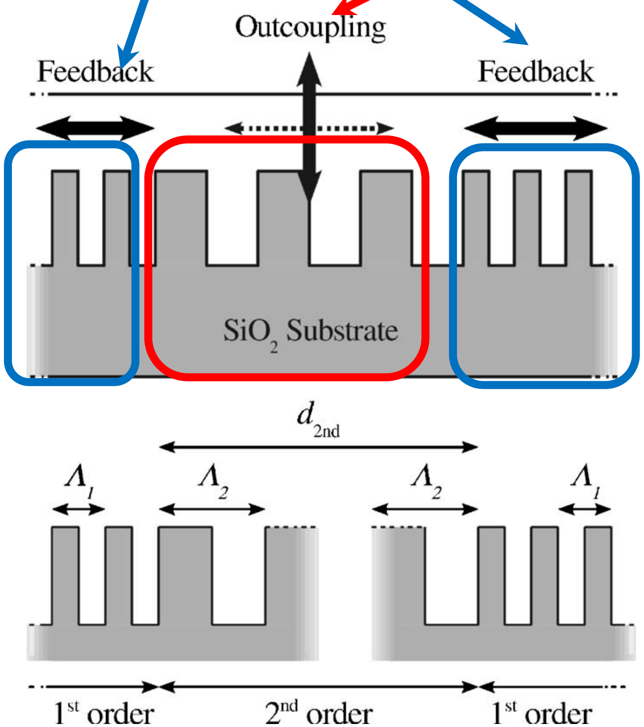

- hexagonal lattice
- honeycomb lattice

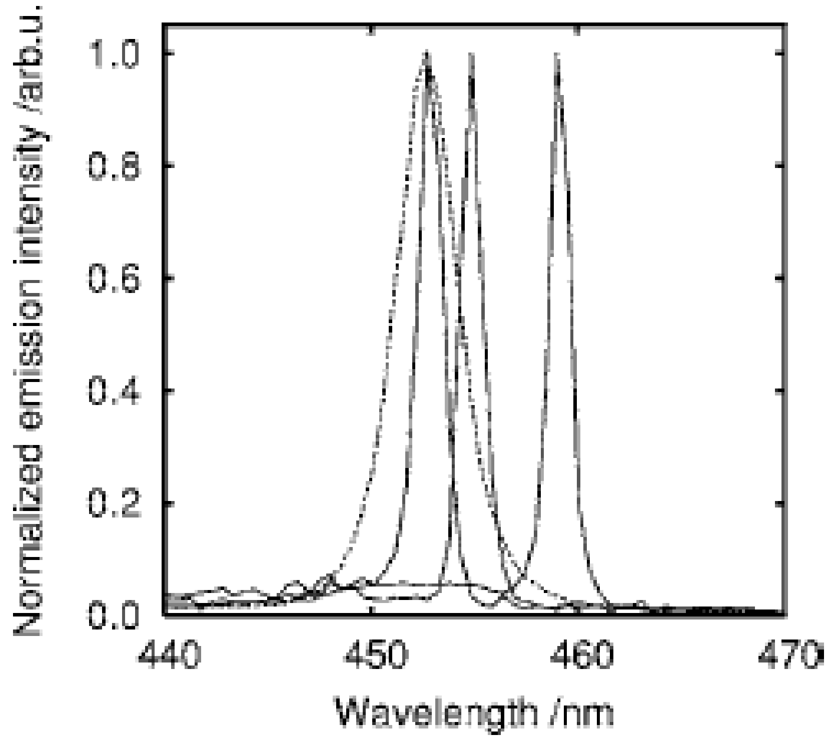
⇒increasing the feedback
⇒lowering the threshold

# MIXED-ORDER DFB LASER

1° order ⇔ feedback

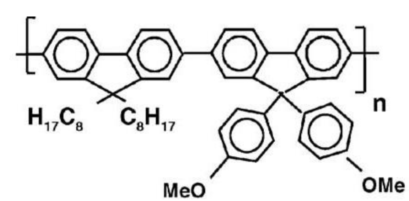
2° order ⇔ coupling



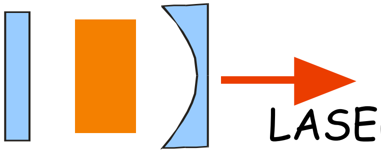

- improved resonator
- **very low laser threshold** (~36 nJ/cm²)
- tunability by film thickness

F8DP




C. Karnutsch, et al., Appl. Phys. Lett. 90, 131104 (2007)

# EXTERNAL CAVITY




- ☺ easy setting up
- ☺ study of the material (tunability, lifetime, gain parameters)
- ☺ efficient
- ☹ Not suitable for integration in devices

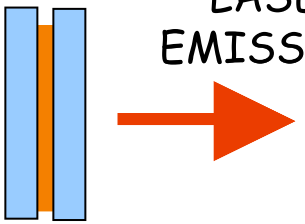
✦ PPV - $\lambda = 480\text{-}510$ nm - $F_{thresh} \sim 10\ \mu J/cm^2$ - **$\eta$~10%**

*N. Deepak Kumar et al. APL 71, 999 (1997)*

✦ PMMA+Rodhmine - $\lambda$=660 nm - $F_{thresh}$ ~6 $mJ/cm^2$ - **$\eta$~43%**

*Hadi Rabbani-Haghighi, et al. Opt.Lett 35, 1968 (2010)*

# MICRO CAVITY




- ☺ easy fabrication (evaporation, spin coating)
- ☺ no alignment required
- ☺ can be integrated ( 😐 in some cases)
- 😐 electrical pumping ?

- ✦ Polythiophene - $\lambda$=655 nm - fs pump - $F_{thresh}$ ~ 0,17$\mu$J/cm$^2$ - external mirrors

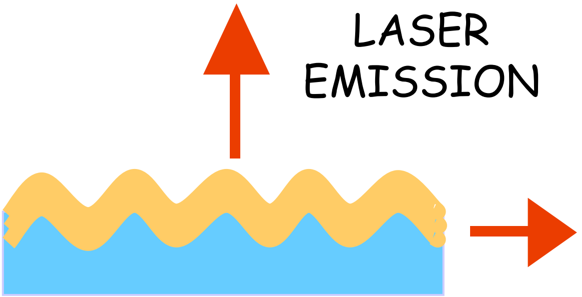
  *T. Granlund et al. Chem. Phys. Lett. 288, 879 (1998)*

- ✦ green copolymer - $\lambda$=509 nm - fs pump - $F_{thresh}$ ~ 84 $\mu$J/cm$^2$ - $TiO_2/SiO_2$ bilayers by evaporation

  *L. Persano et al. Appl. Phys. Lett. 88, 121110 (2006)*

# DFB CAVITY

- ☺ large area geometries
- ☺ low threshold
- ☺ no alignment
- 😐 easy and low cost (spin coating)
- ☺ mechanical flexibility (all plastic device)
- ☺ can be integrated
- 😐 efficiency
- 😐 electrical pumping ?




- ✦ LPPP - $\lambda$=487 nm - fs pump - $F_{thresh}$ ~ 3,7$\mu$J/cm$^2$

  *C. Kallinger et al. Adv.Mat 10, 920 (1998)*

- ✦ Truxene-cored 9,9-dialkylfluorene - $\lambda$=470-510nm - ns pump - **$F_{thresh}$ ~ 160 nJ/cm$^2$**

  *R. Xia et al. Adv. Funct. Mater. 19, 2844 (2009 )*

- ✦ F8DO - $\lambda$ = 455 nm- ns pump-mixed grating- **$F_{thresh}$ ~ 36 nJ/cm$^2$**

  *C. Karnutsch et al. Appl. Phys. Lett. 90, 131104 (2007)*

# OUTLINE

INTRODUCTION

The LASER
- active material
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O- LASERS EXAMPLES
- external resonator
- compact micro-cavity
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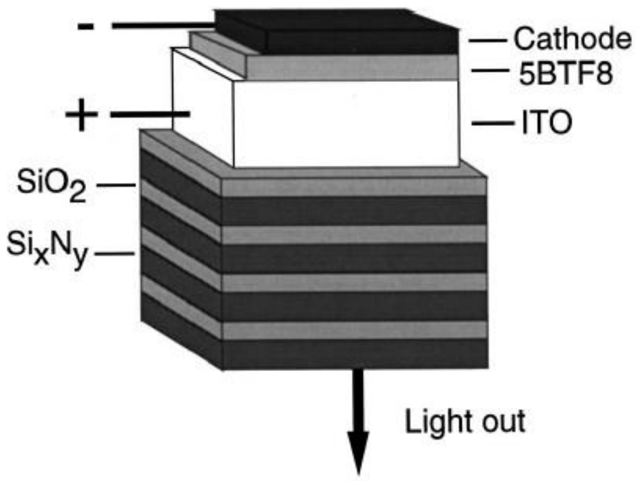
Further developments
- electrical pumping
- diode pumping
- LED pumping
- photonic applications

CONCLUSIONS

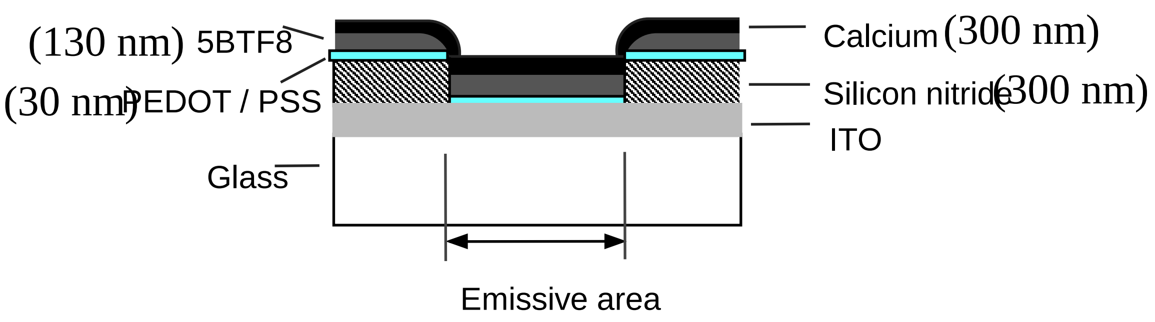
# FROM LED TO LASER ?

resonant microcavity




No lasing

small area device (50 μm)



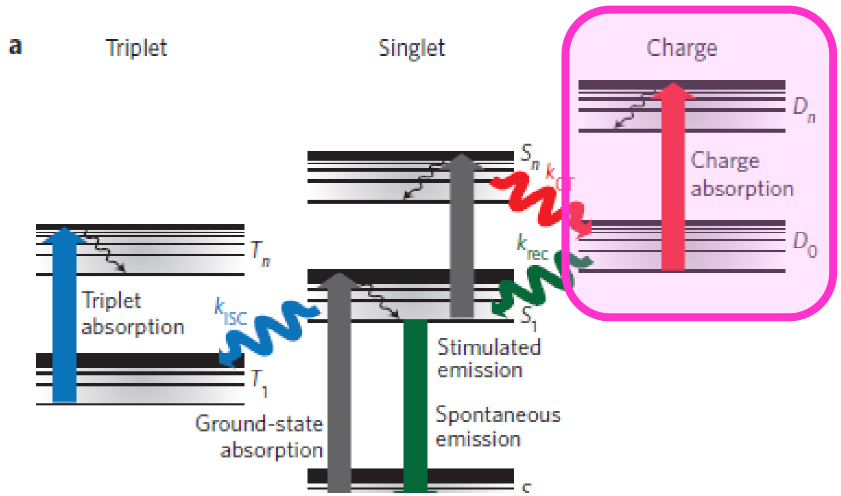

No lasing

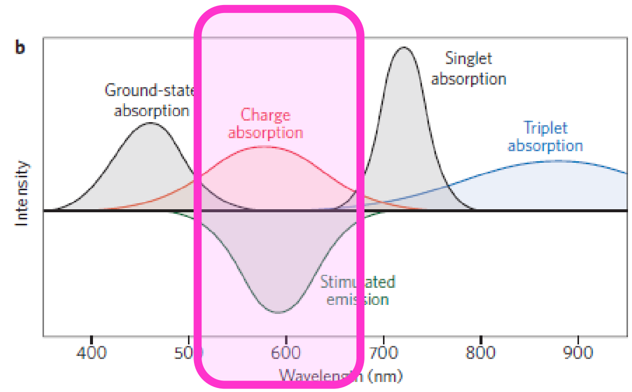
even with improved design
(eg ITO / PEDOT / 5BTF8 / CAO / AL)


D.J. Pinner et al. Synth. Met. 111-112, 257 (2000)
C.I.Wilkinson et al. Appl. Phys. Lett. 79, 171 (2001)
R.B.Fletcher et al. Appl. Phys. Lett. 77, 1262 (2000)

# ELECTRICAL PUMPING ?

- large stimulated emission + high charge carrier mobility ☹
- charges absorption ☹




## Possible solutions:

⇒ emitting, recombination and transport zones separated ☺
⇒ cw pumping
⇒ indirect pumping (diode lasers, LEDs)

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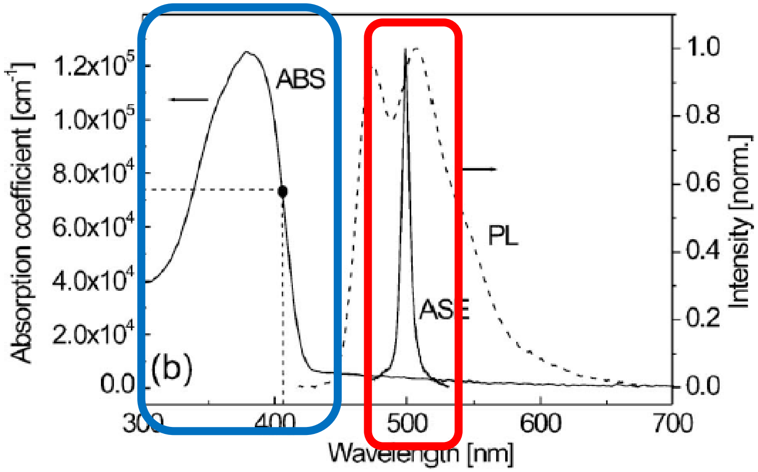
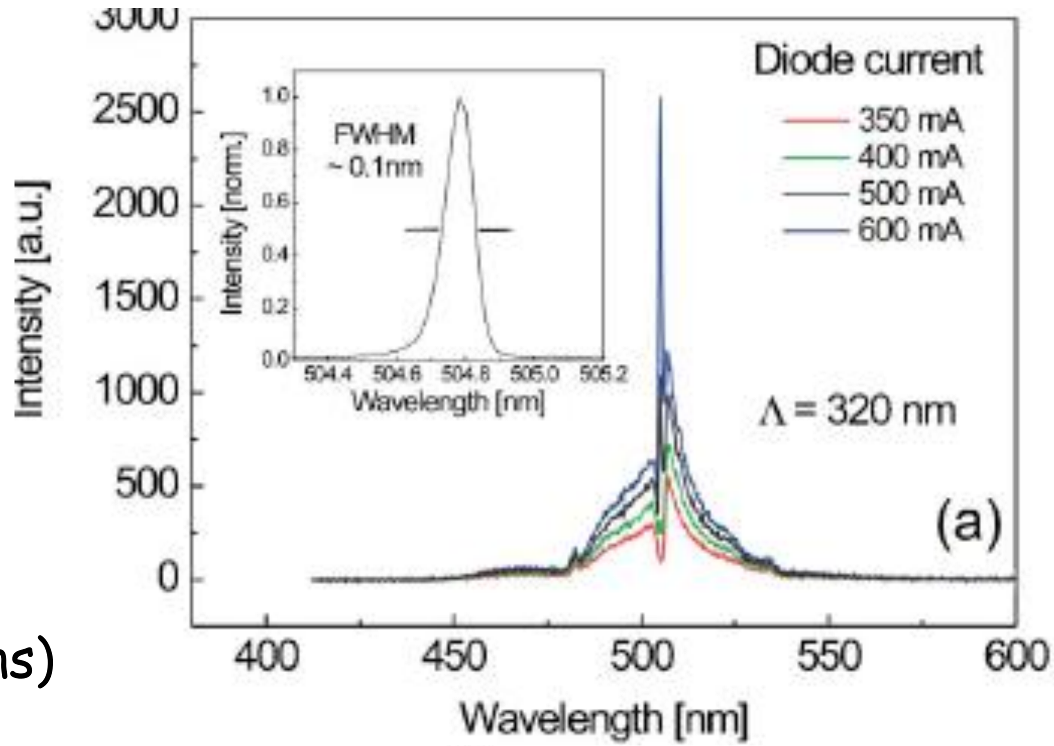
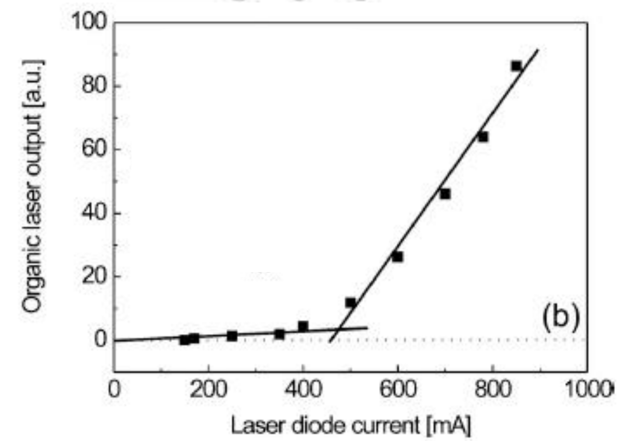
# DIODE laser PUMPED DFB o-laser



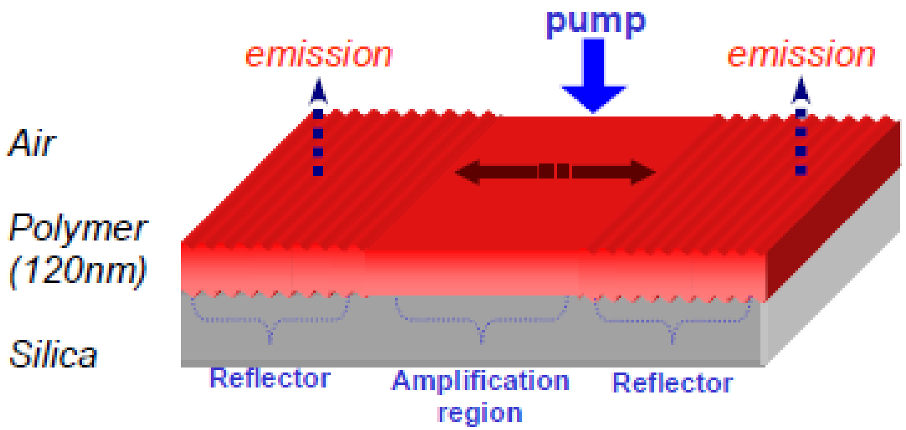




- ✓ guest-host (BN-PFO+DPAVB)
- ✓ InGaN violet laser diode (50 ns)
- ✓ 2nd order DFB laser
- ✓ threshold <1μJ/cm$^2$
- ✓ same behaviour as with ns laser pump
- ✓ compact
- \- expensive pulse generator



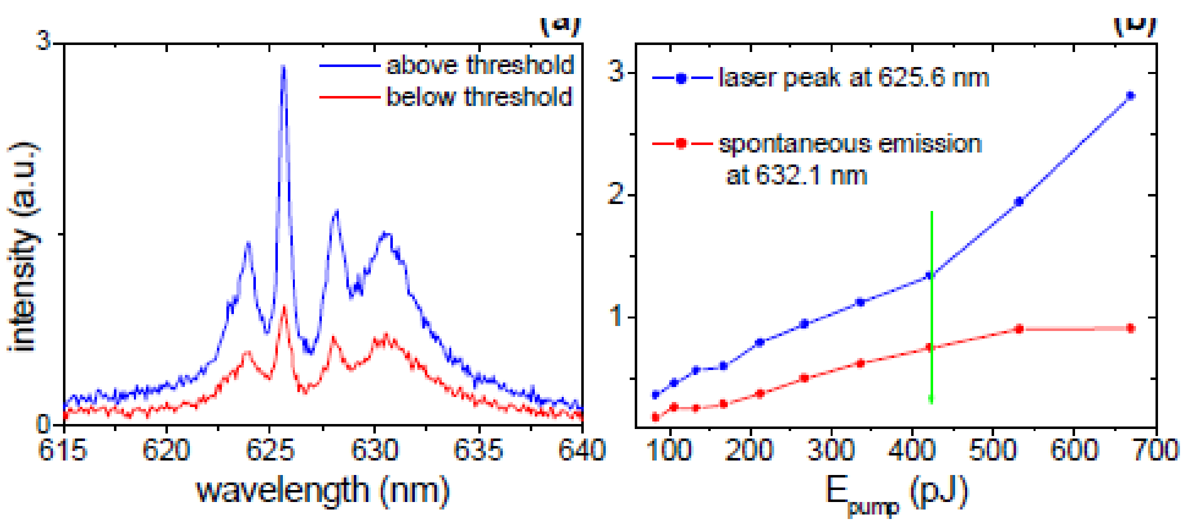

T. Riedl, et al., Appl. Phys. Lett. 88, 241116 (2006)

# DIODE laser PUMPED DBR o-laser




✓ improved resonator quality (EBL + lateral etching)

✓ guest-host (improved pump light harvesting)




- GaN diode laser (1 ns)
- threshold 0,42 nJ/pulse

A. E. Vasdekis, et al., *Optics Express* 14, 9211 (2006)

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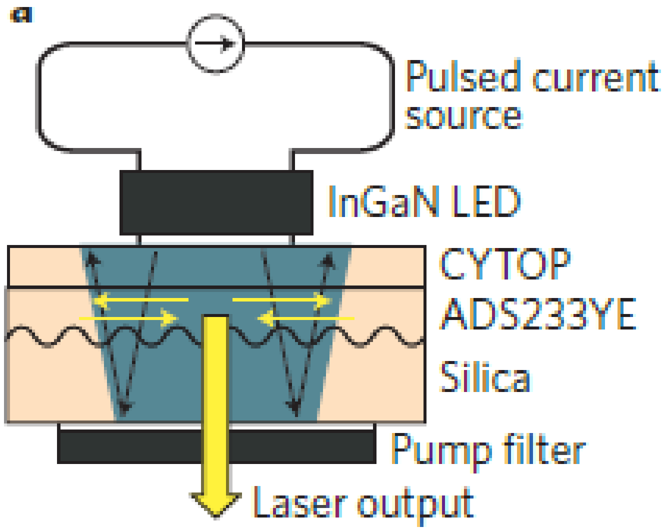
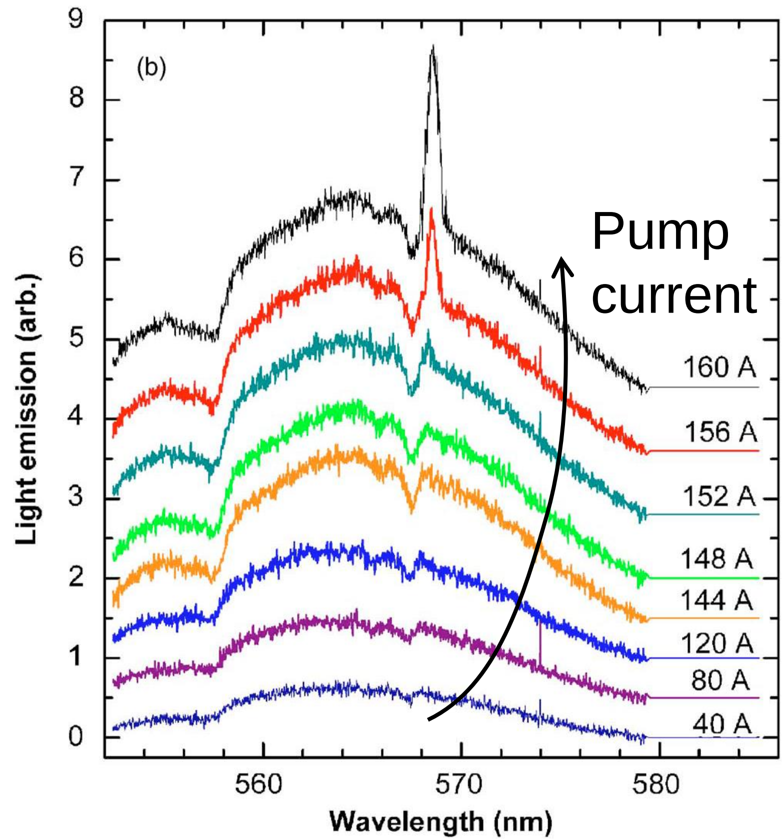
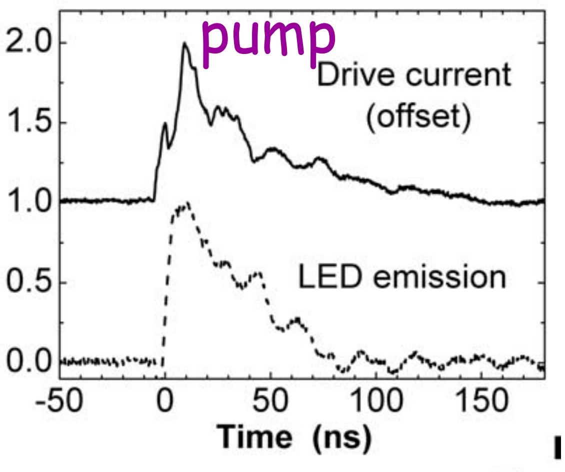
# LED PUMPED DFB O-LASER




- LED light: high divergence
- ✓ engineered configuration
- ✓ high power InGaN violet LED (~50 ns, @450 nm)
- ✓ compact

A. Y. Yang, G. A. Turnbull, I. D. W. Samuel, Appl. Phys. Lett. 92, 163306 (2008)

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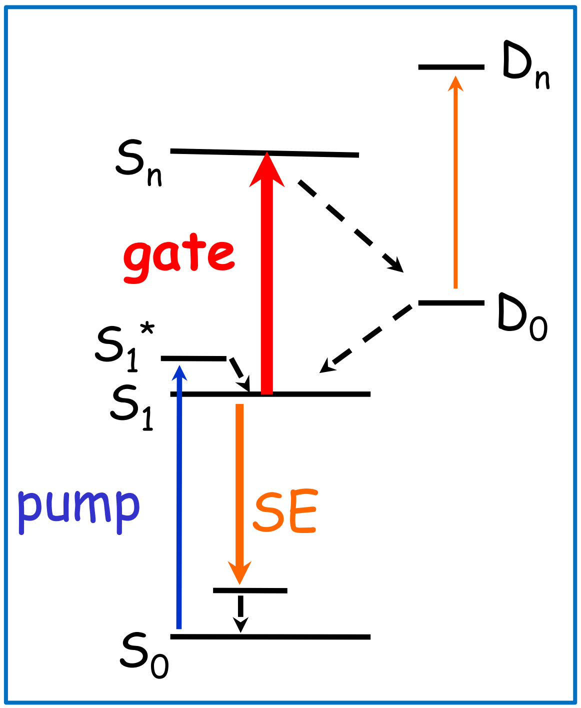
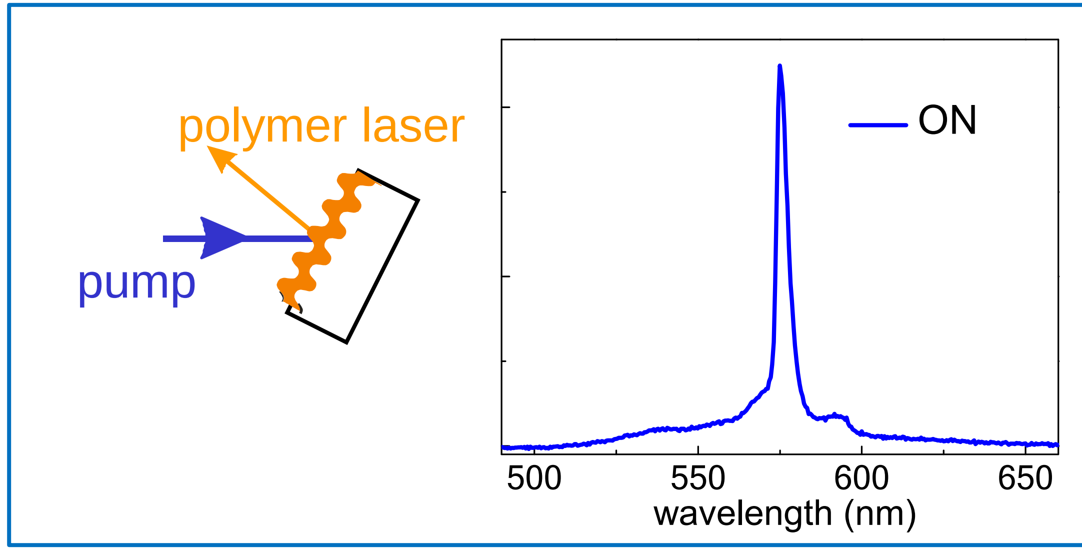
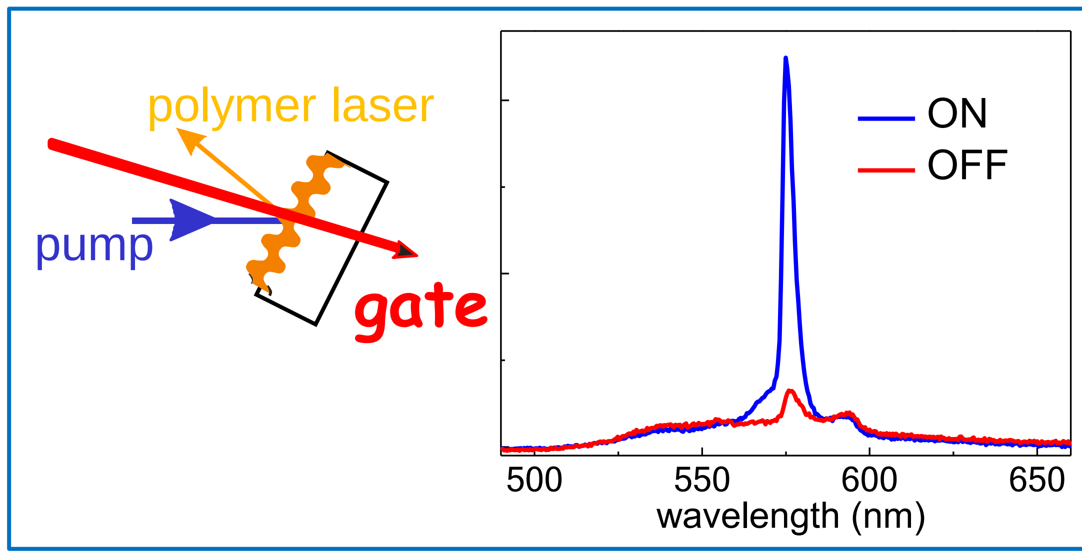
# OPTICAL SWITCHING

- -DFB resonator
- -F8BT polymer
- ✓charge generation
  ⇒switching
- ✓high repetition rate

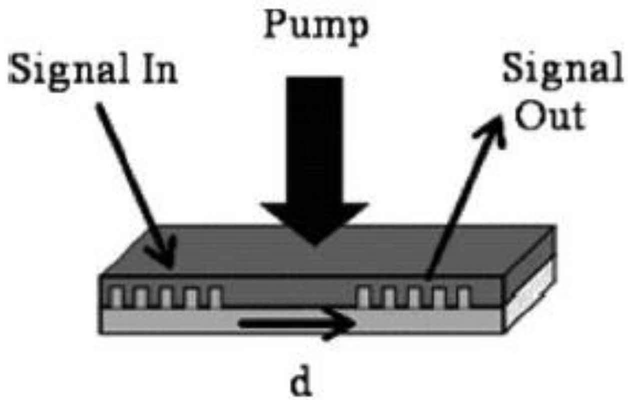


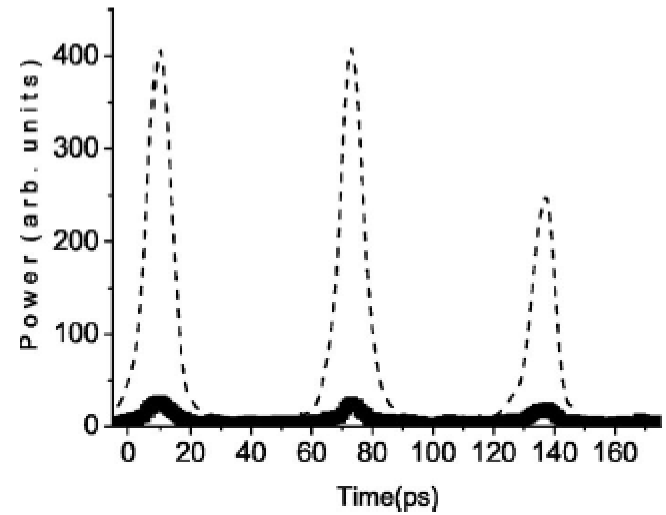



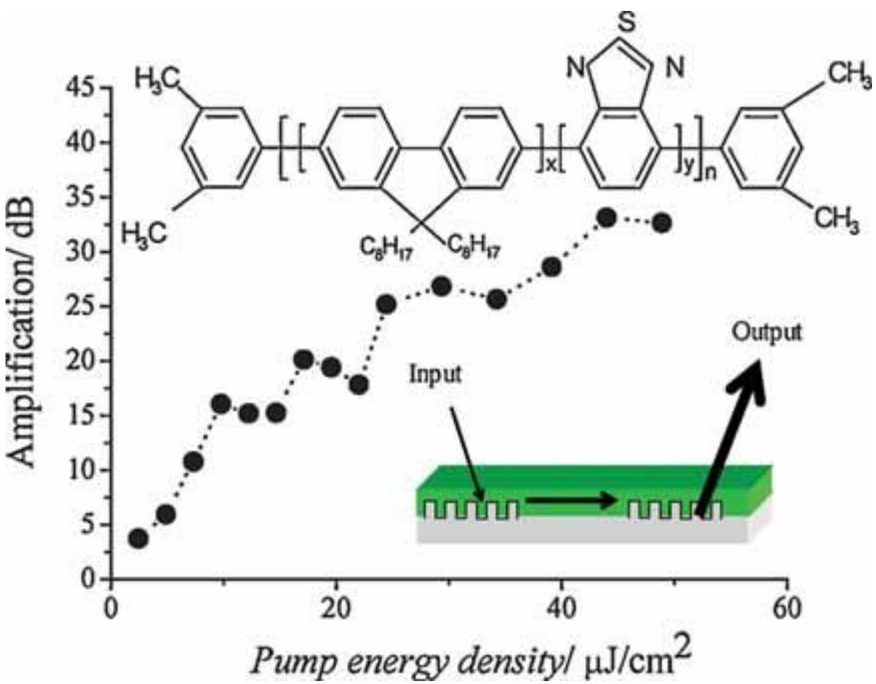

S. Perissinotto et al., Appl. Phys. Lett. 91, 191108 (2007)

# OPTICAL AMPLIFIER




F8BT




$$G(\mathrm{dB}) = 10 \log \left[ \frac{P_{on} - P_b}{P_{off}} \right]$$

- pulses: 140 ps, 5 kHz

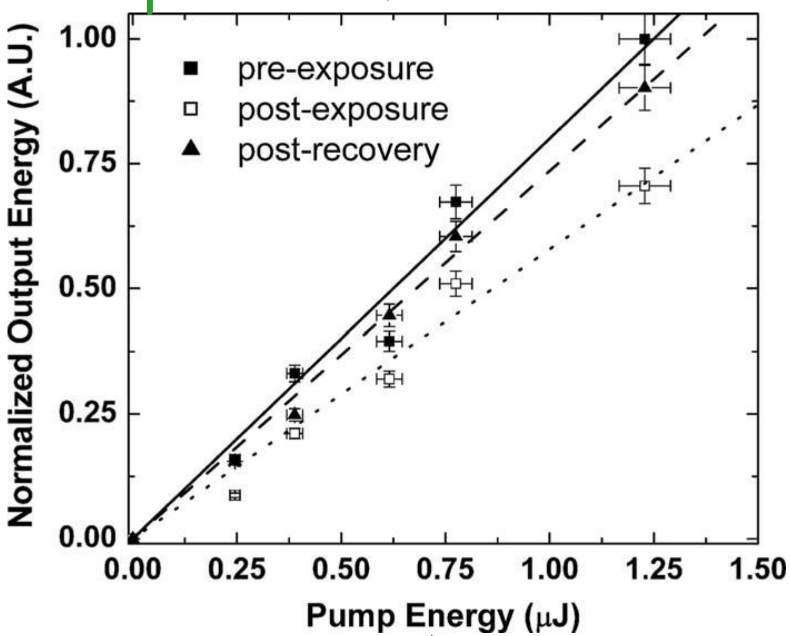
✓ 32 dB gain

D. Amarasinghe, et al., Adv. Mater. 21, 107 (2009)

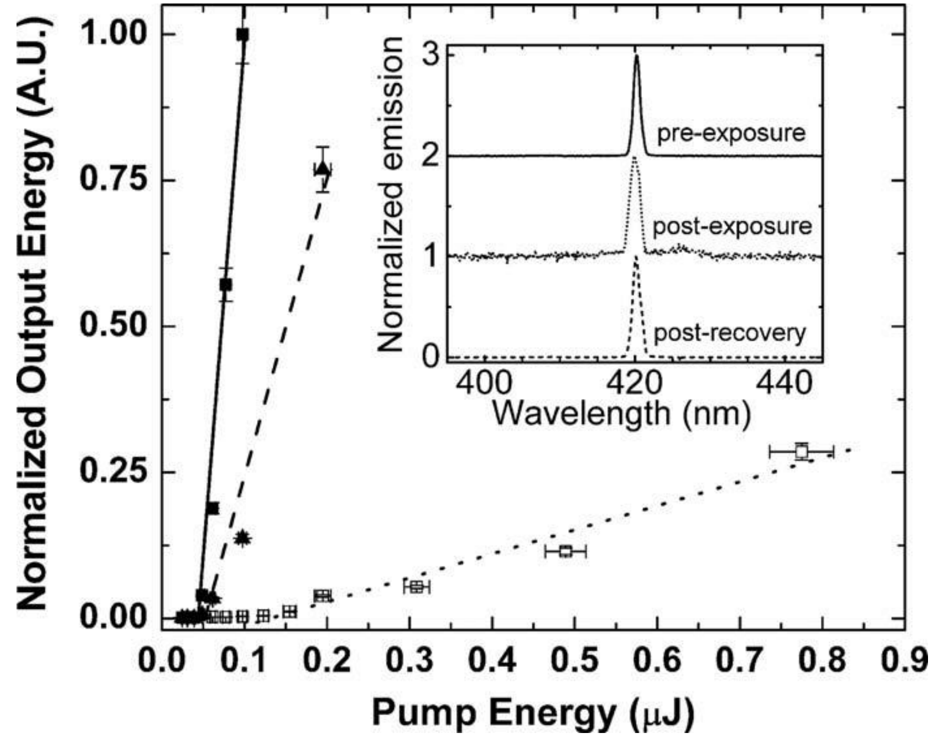
# CHEMOSENSING

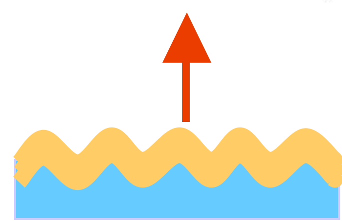
-dendrimer
-DNT analyte (TNT)

## photoluminescence




## laser emission



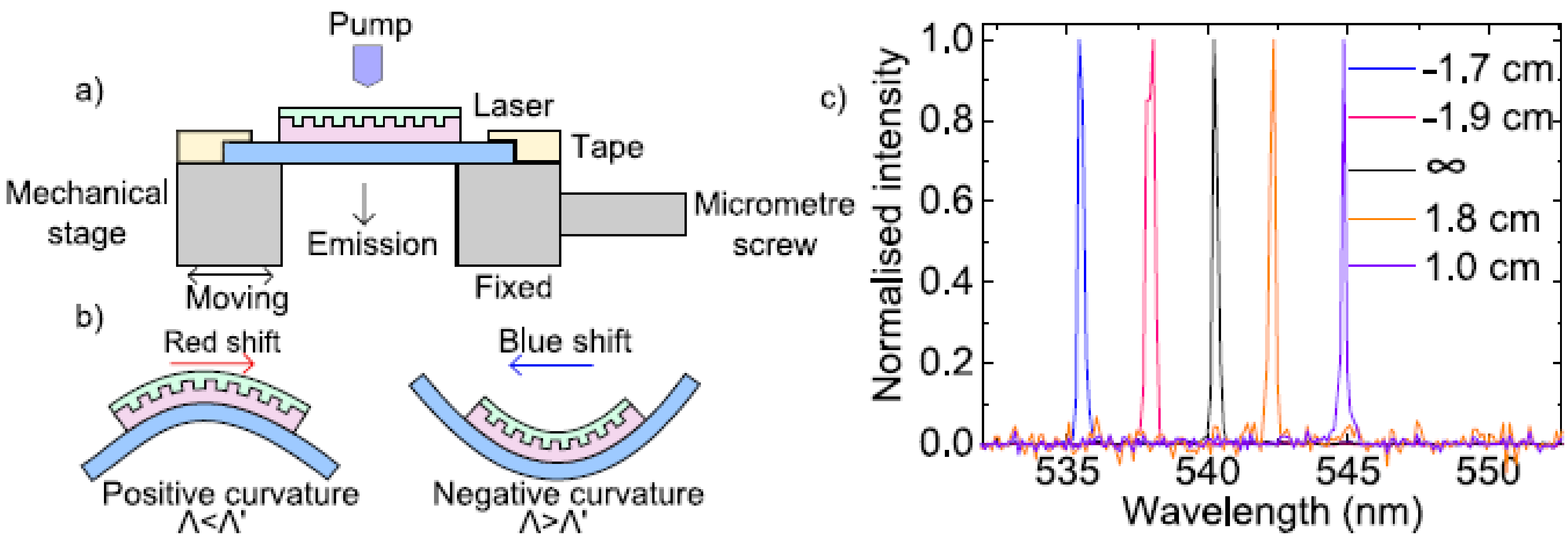



-DFB resonator
✓ **high sensitivity**

S. Richardson et al., Appl. Phys. Lett. 95 (2009)

# MECHANICALLY FLEXIBLE

laser emission

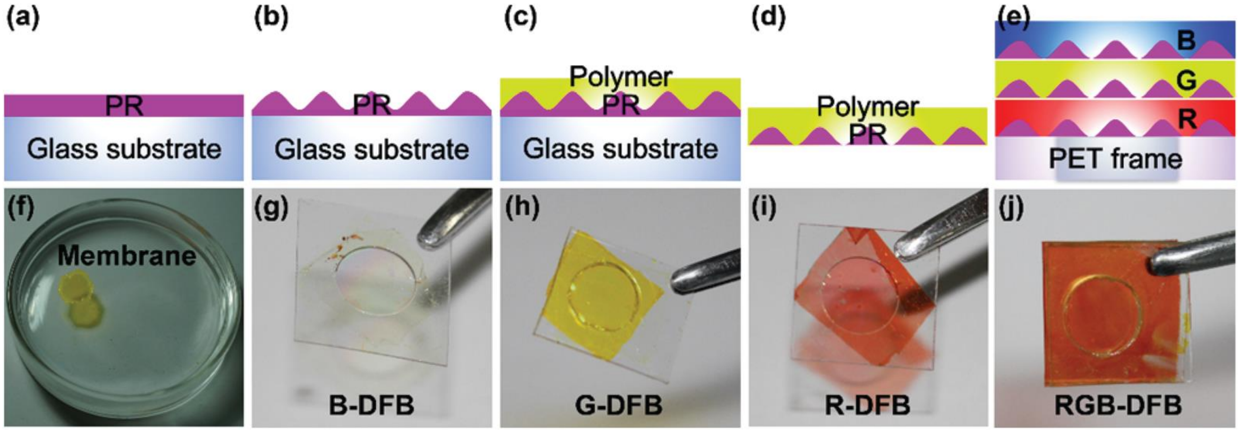



- ✓ long lifetime
- ✓ (bio)-sensing, medical diagnostic, spectroscopy

-DFB resonator
-encapsulated
-diode pumped

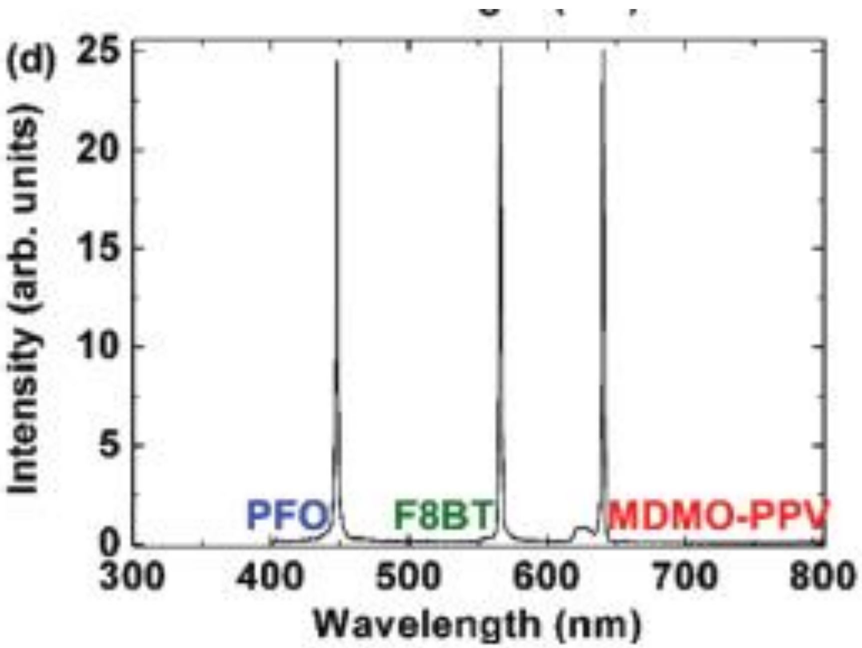
*C. Foucher et al., Opt. Express 22 (2014)*

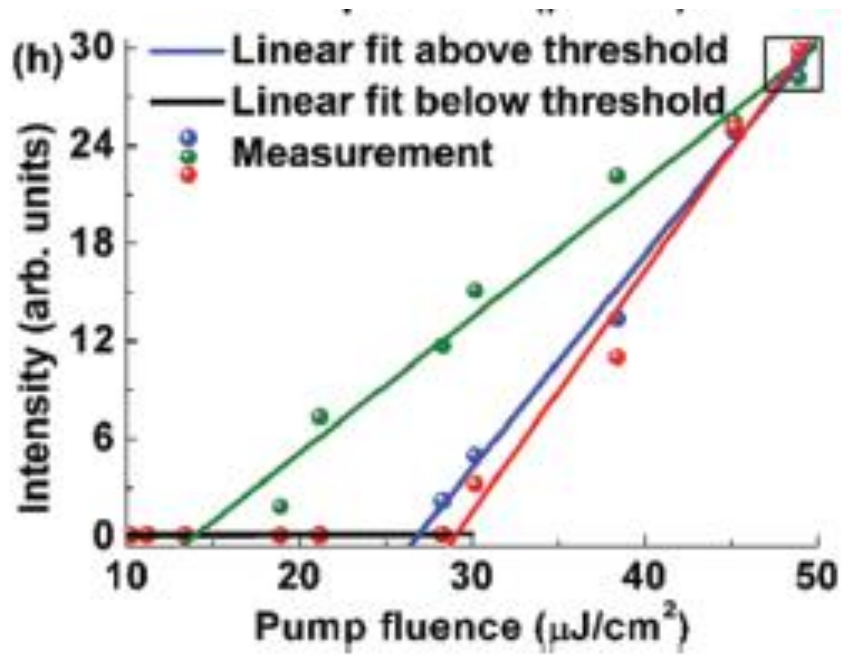
# RGB LASER EMISSION




Free-standing membrane DFB cascade

⇒ RGB emission




T. Zhai, et al., Nanoscale, accepted (2015)

# CONCLUSIONS

lasing with organic materials has been achieved
with optical pumping, different materials,
cavities, geometries, thresholds, efficiencies, ...

Chemical synthesis → material intrinsic properties

photophysics → material intrinsic properties

**material intrinsic properties**
- high $\sigma \cdot \tau$ product
- low charge-induced absorption
- long lifetime $\tau$
- ...

device engineering →
- low threshold
- indirect pumping
- ...

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J. Herrnsdorf *et al*.
**Micro-LED pumped polymer Laser: A Discussion of Future Pump Sources for Organic Lasers**
Laser & Photonics Reviews (2013)


thank you for your attention