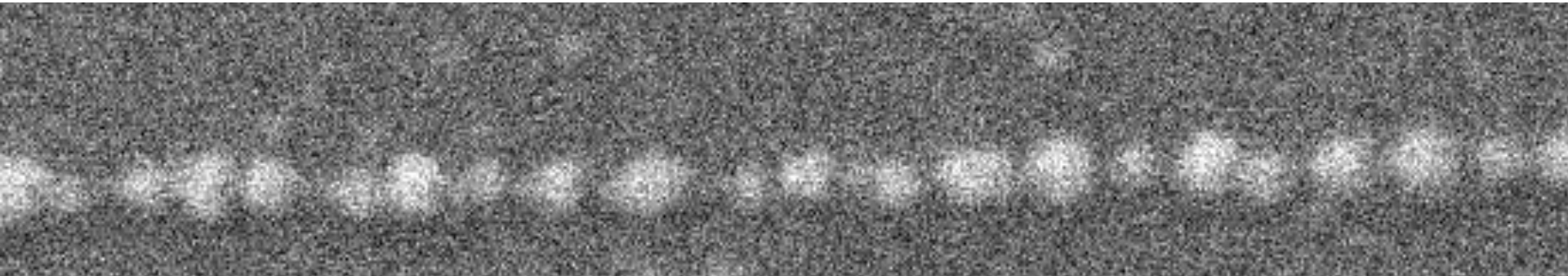


Novel features of plasmon-assisted solid-state lasers at the nanoscale



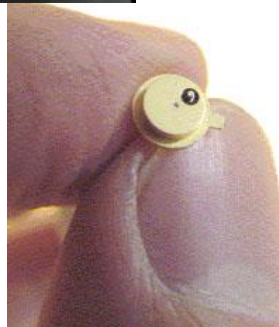
Luisa E. Bausá

*Dpto. Física de Materiales, Instituto Nicolás Cabrera and Condensed Matter Physics Center (IFIMAC)
Universidad Autónoma de Madrid (Spain)*

Miniaturization of lasers



Different gain media, operation modes, designs, sizes...

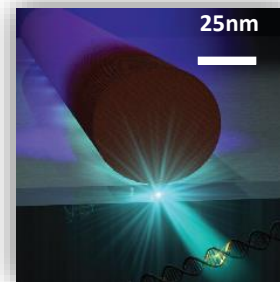


Microlasers
(sub- μm)



Diffraction limit
 $\sim \lambda_0 / 2n$

Plasmon lasers
(nm)



Scaling down of photonic devices

Nanolasers

Plasmon lasers

A new class of optical amplification and laser action, which involves charge density waves at the nanoscale

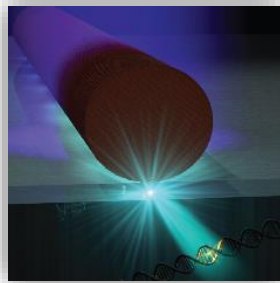
- **High confinement of EM fields** by plasmonic nanostructures



High energy density of coherent light in sub-wavelength confined volumes

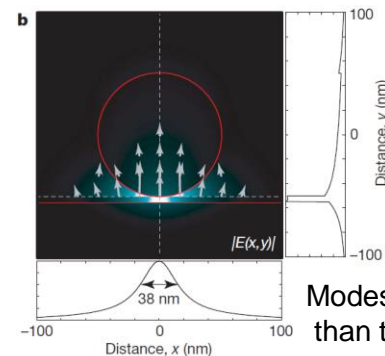
**Laser volumes
below the diffraction limit**

Nanometer-scale plasmonic laser



R. F. Oulton et al. Nature 461, 629 (2009)

Hybrid plasmonic waveguide



Modes hundred times smaller than the diffraction limit


Metallic nanostructures adjacent to a gain medium

GAIN MEDIA:

Semiconductors

&

Organic Dyes

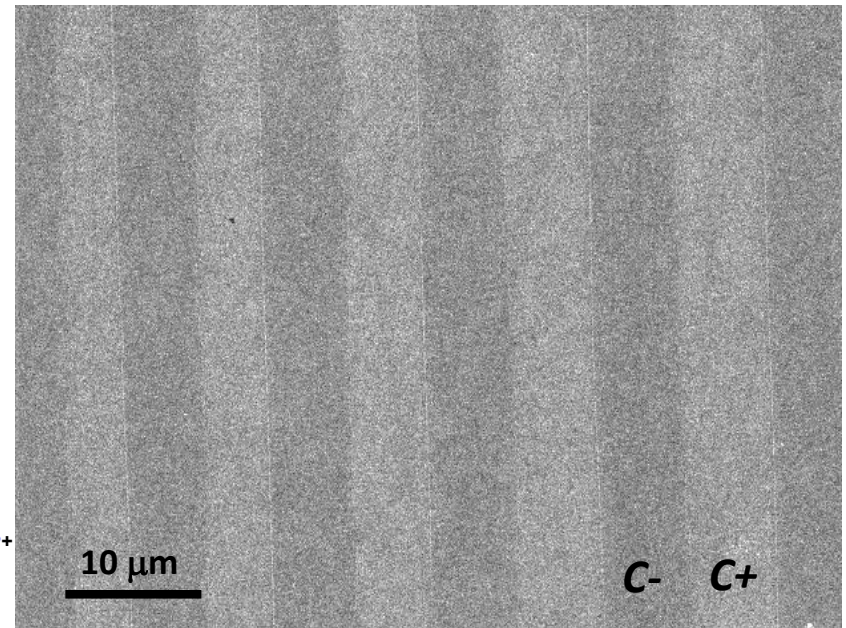
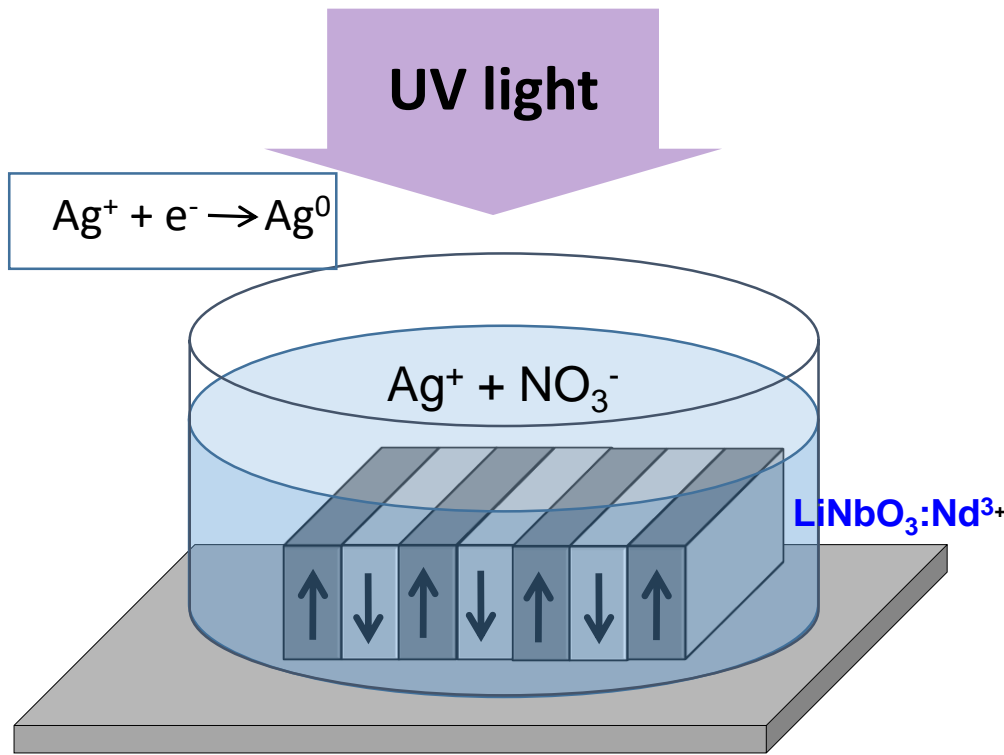
- 
- InGaN nanorods
 - InGaAs layers
 - InAsP quantum Wells
 - CdS nanowires
 - CdSe nanobelts

Fabrication process

Photochemical formation of metallic nanostructures

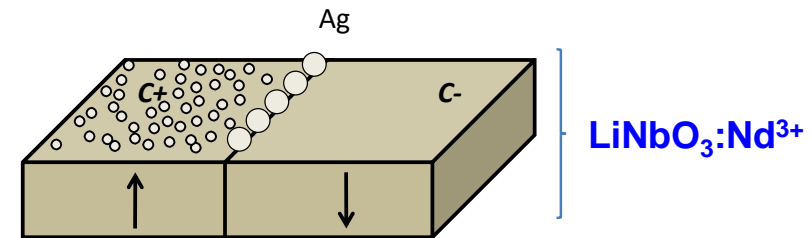
Fabrication process: Photochemical formation of metallic nanostructures

Silver: lowest Ohmic damping in VIS –IR → highest electromagnetic field enhancement



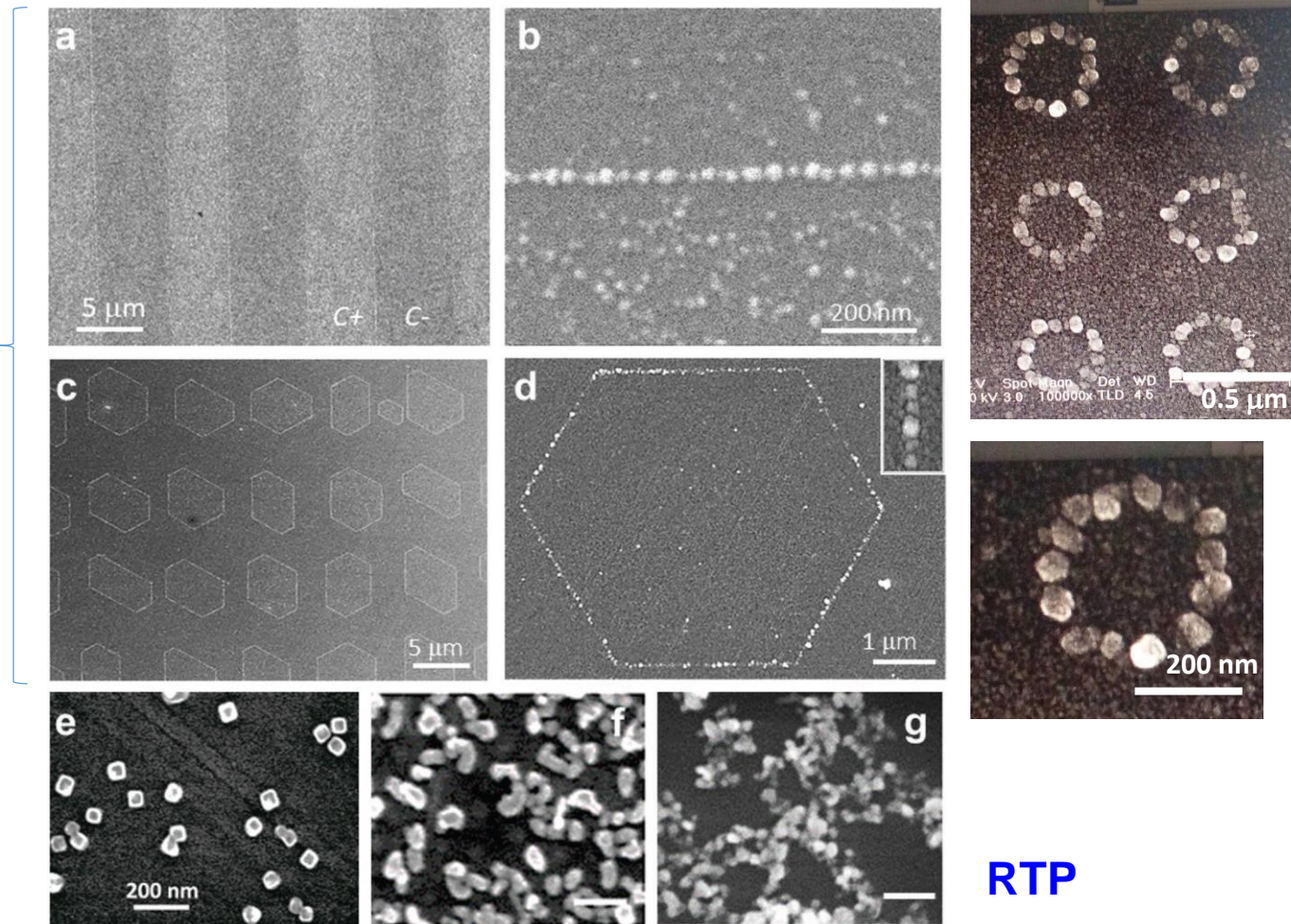
Ag nanoparticle sizes

- ~20 nm (domain surface)
- ~40- 70 nm (domain boundary)



Arrays of Ag NPs on ferroelectrics formed by photo-chemical procedure

LiNbO₃



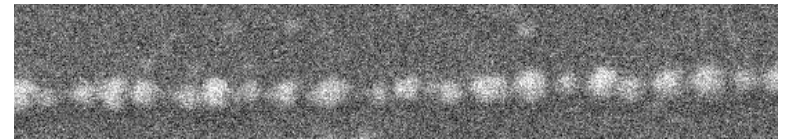
RTP

Plasmonic metasurfaces obtained by a simple and cost effective method

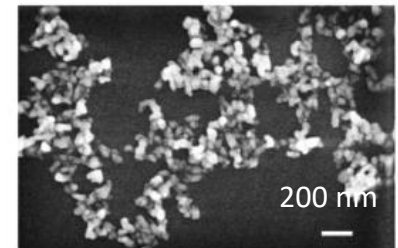
Different types of plasmonic arrangements of NPs

To improve the spectroscopic and lasing properties of Rare Earth ions at the nanoscale

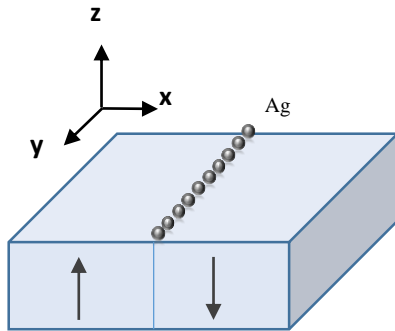
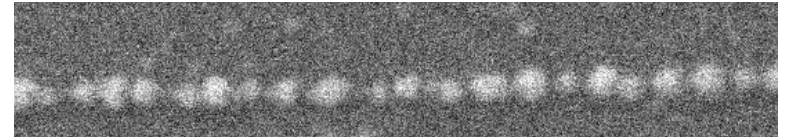
I. Effect of **linear chains** of silver nanoparticles on the properties of a **Nd³⁺** doped SSL



II. Effect of **Disordered Plasmonic Networks (DPNs)** of silver nanoparticles on the optical performance of a **Yb³⁺** doped SSL

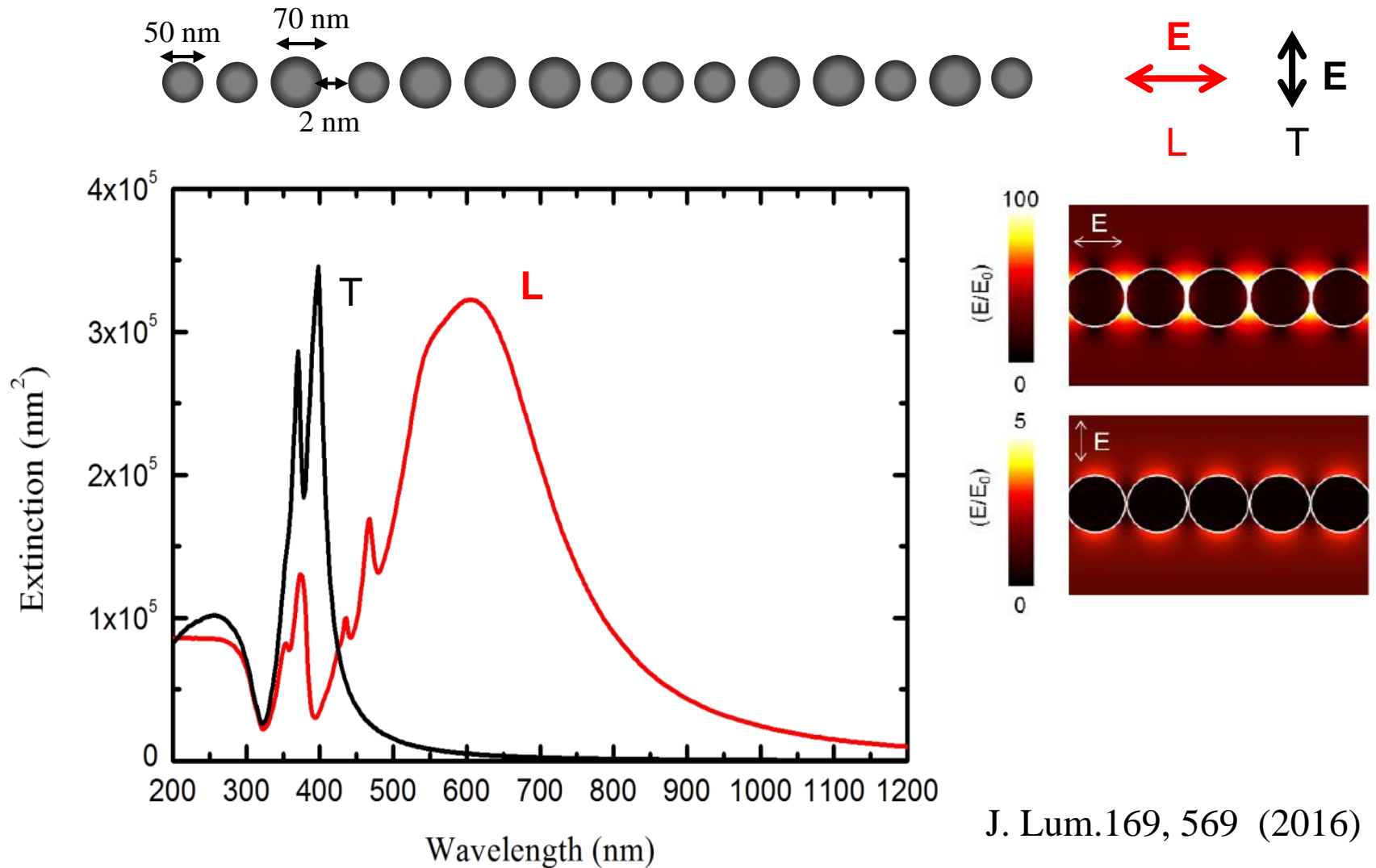


II. Effect of **linear chains** of silver nanoparticles on the properties of a **Nd³⁺** doped SSL



Nd³⁺ doped LiNbO₃ (PPLN) crystal

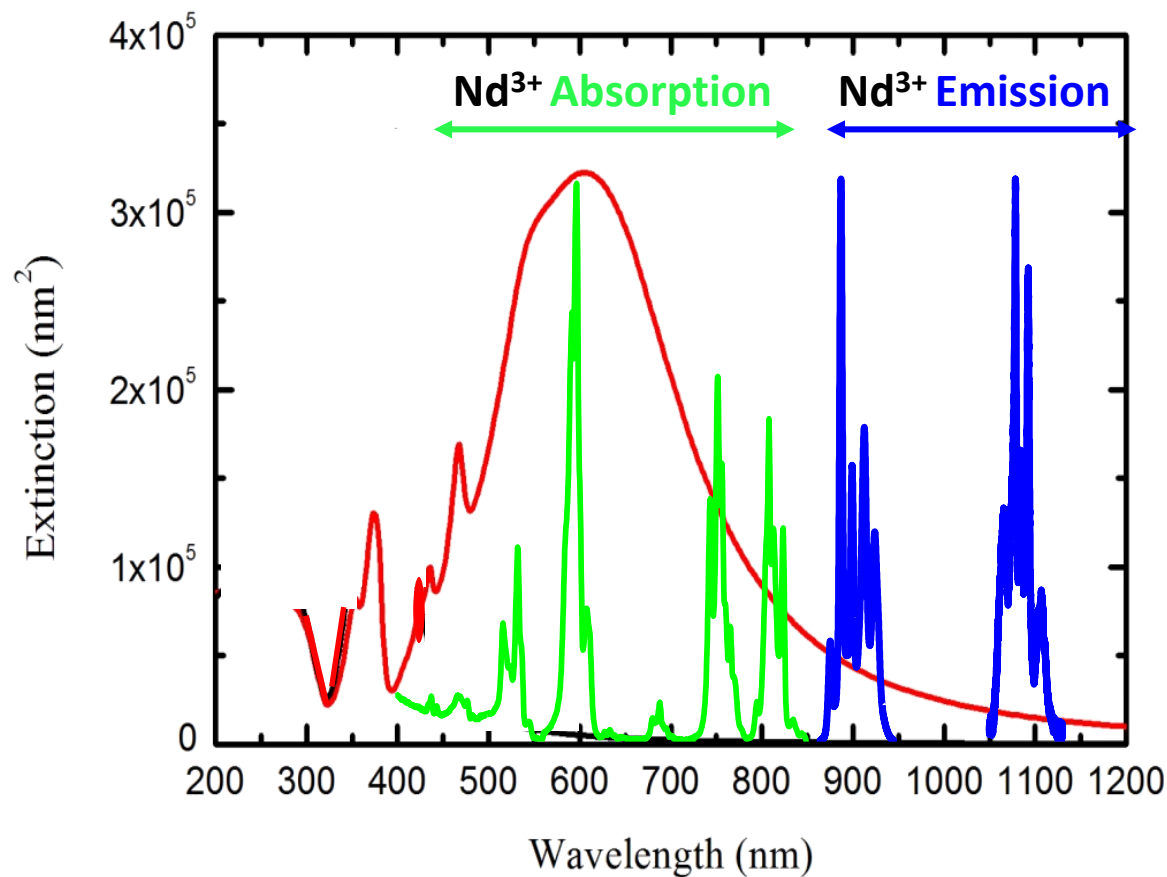
Far-field spectra of the Ag NPs chains



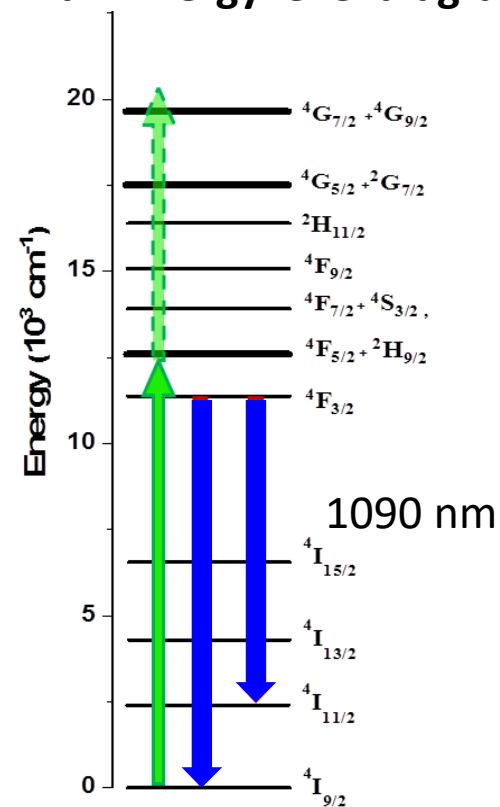
J. Lum.169, 569 (2016)

The long-wavelength mode excited for electric field parallel to the chain (L) is a strongly radiative mode.

L plasmon mode spectral overlaps the optical transition of Nd³⁺ ions

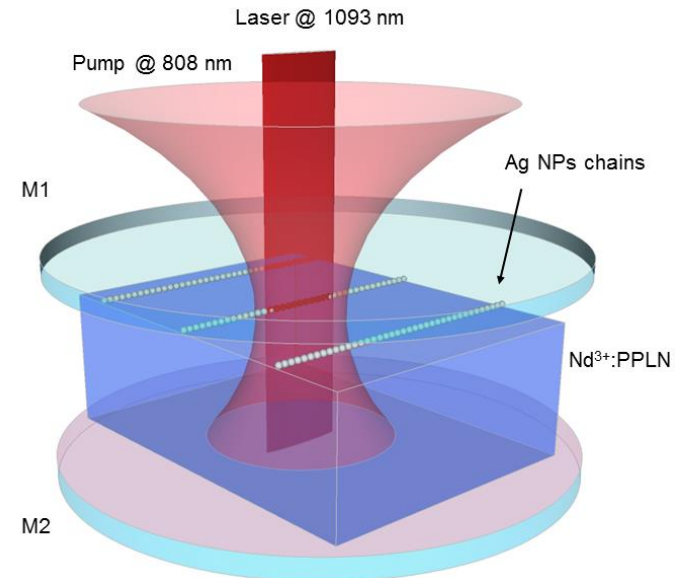
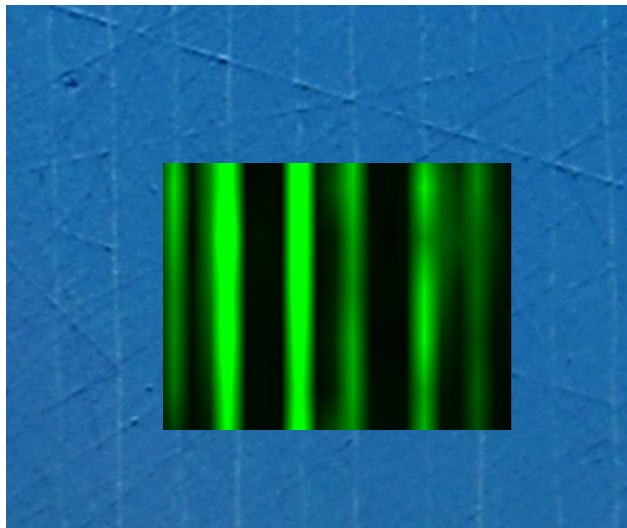
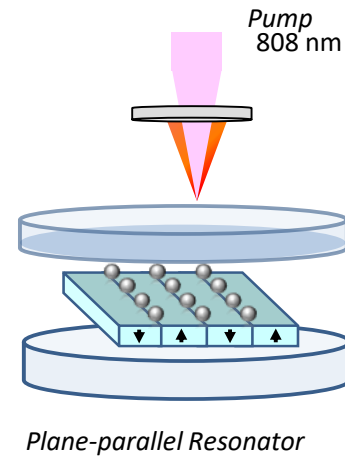
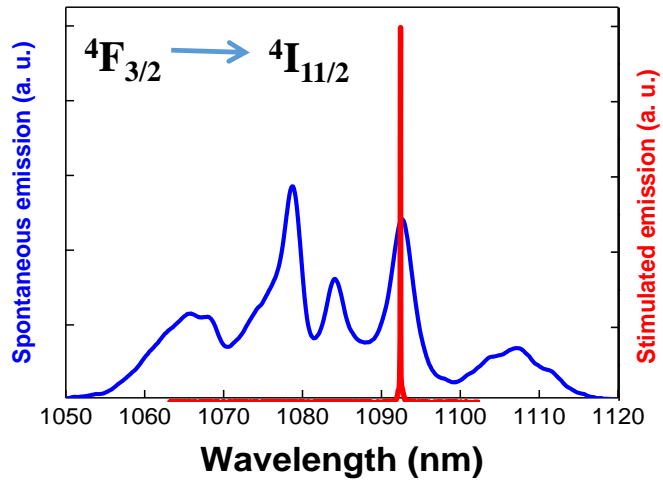


Nd³⁺ Energy level diagram

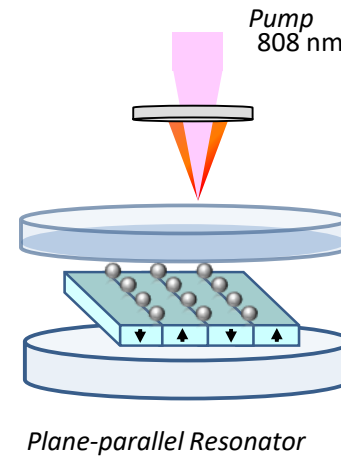
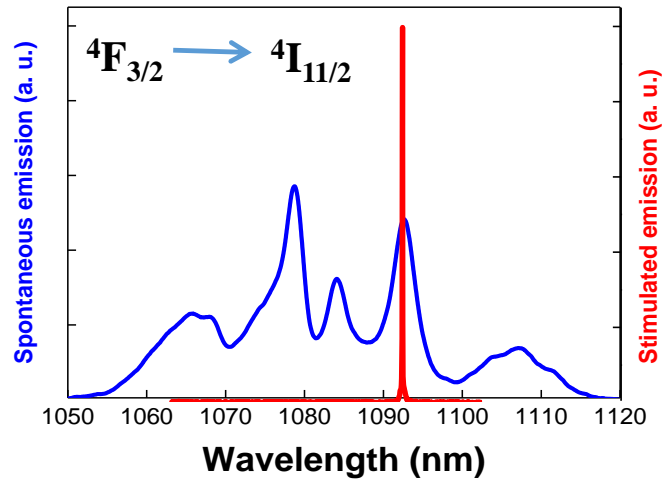


Matching between Ag NPs plasmon and Nd³⁺ absorption/emission bands

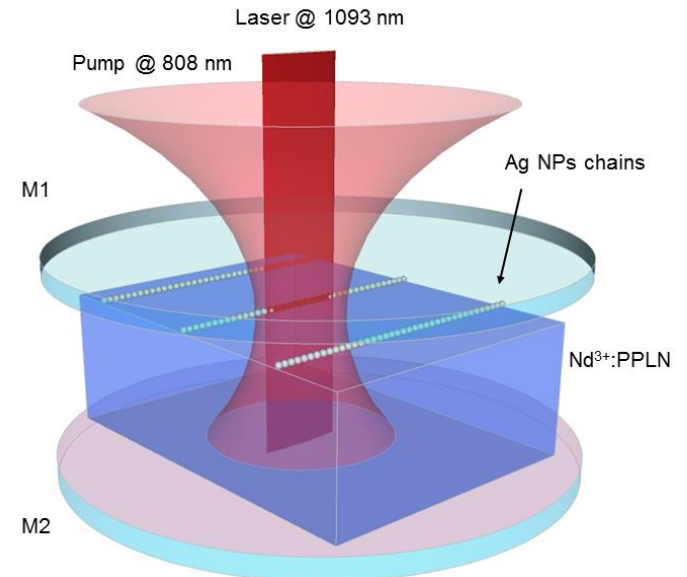
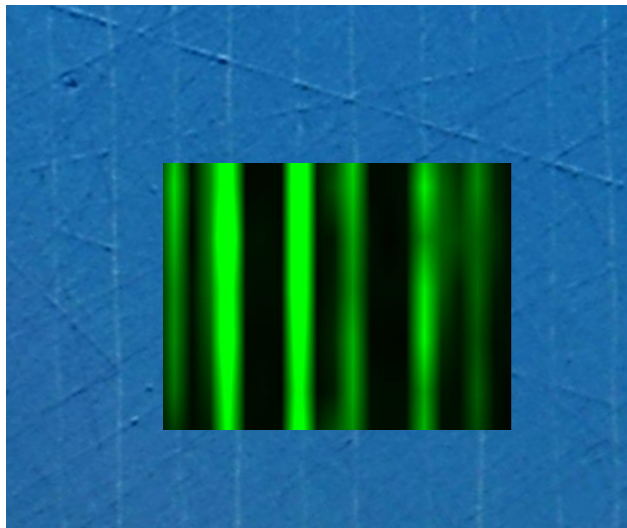
Lasing around the Ag NPs chains on Nd³⁺:LNB



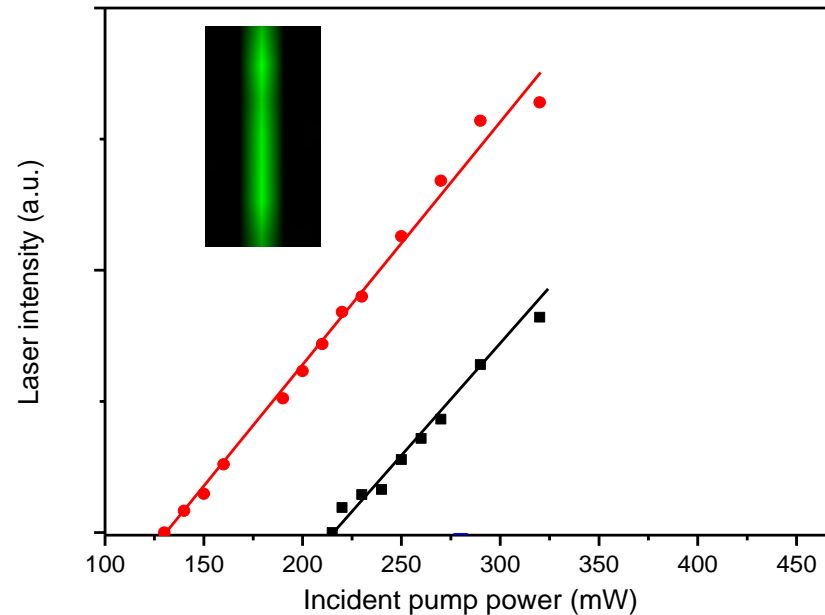
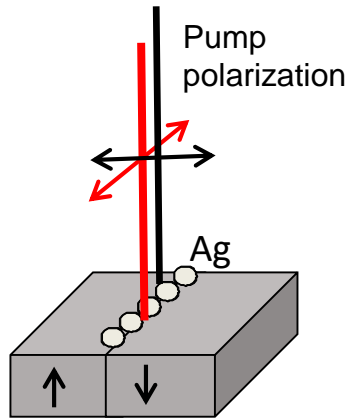
Lasing around the Ag NPs chains on $\text{Nd}^{3+}:\text{LNB}$



Room temperature
CW laser action at
the nanoscale



Laser performance at room temperature

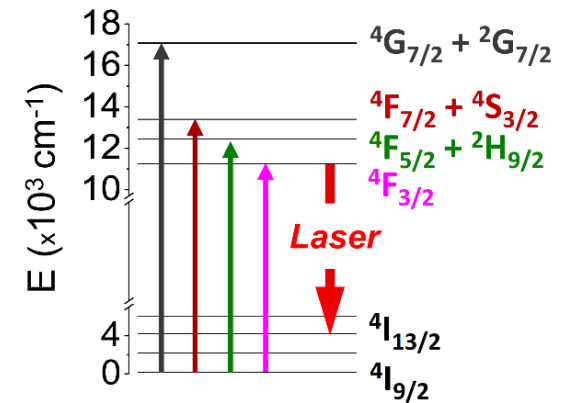
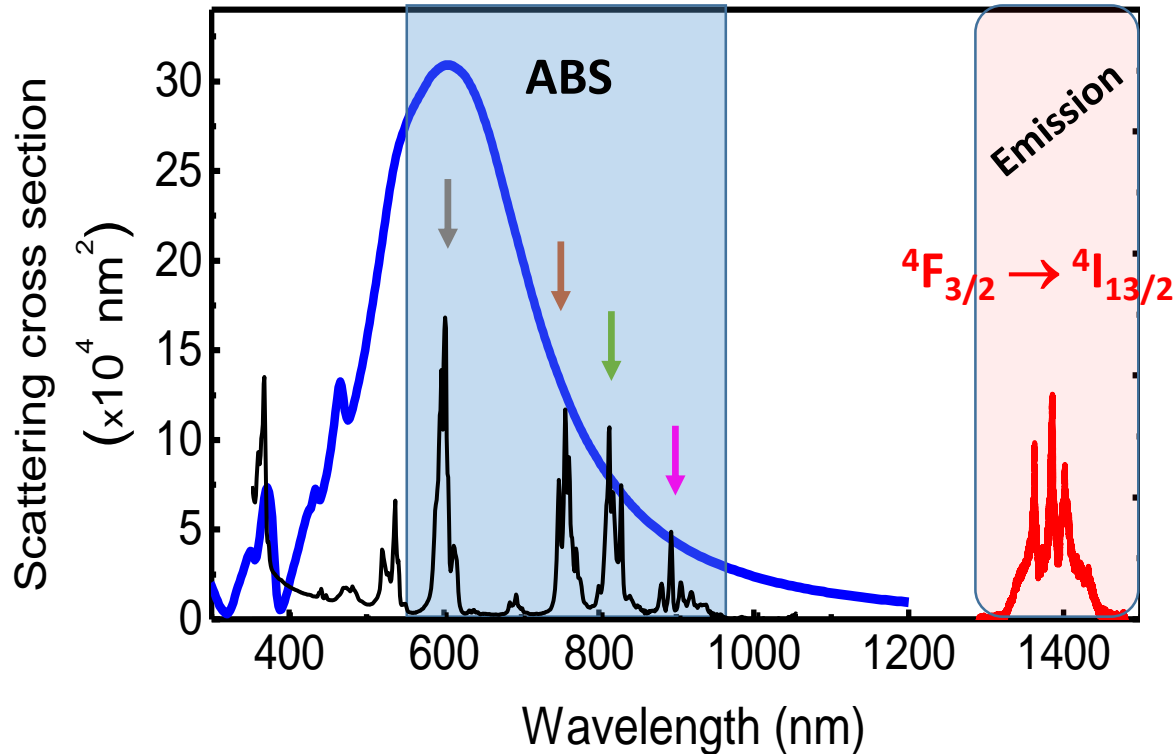


RT & CW Laser performance along the c axis at 1093 nm, pumping at 808 nm

→ The pump power at threshold is half the value of the bulk mode operation

→ The efficiency increases in a factor of 15

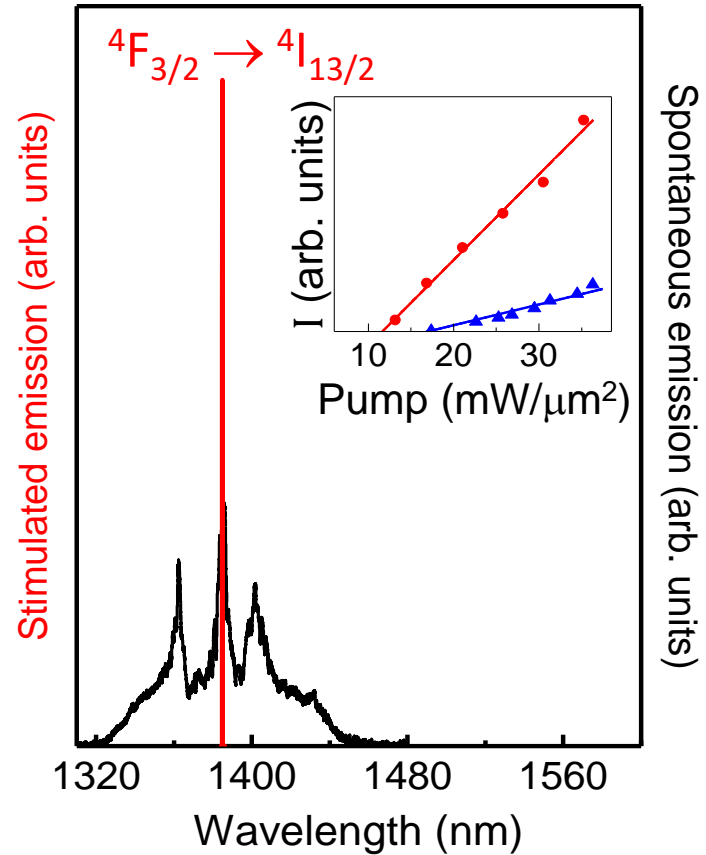
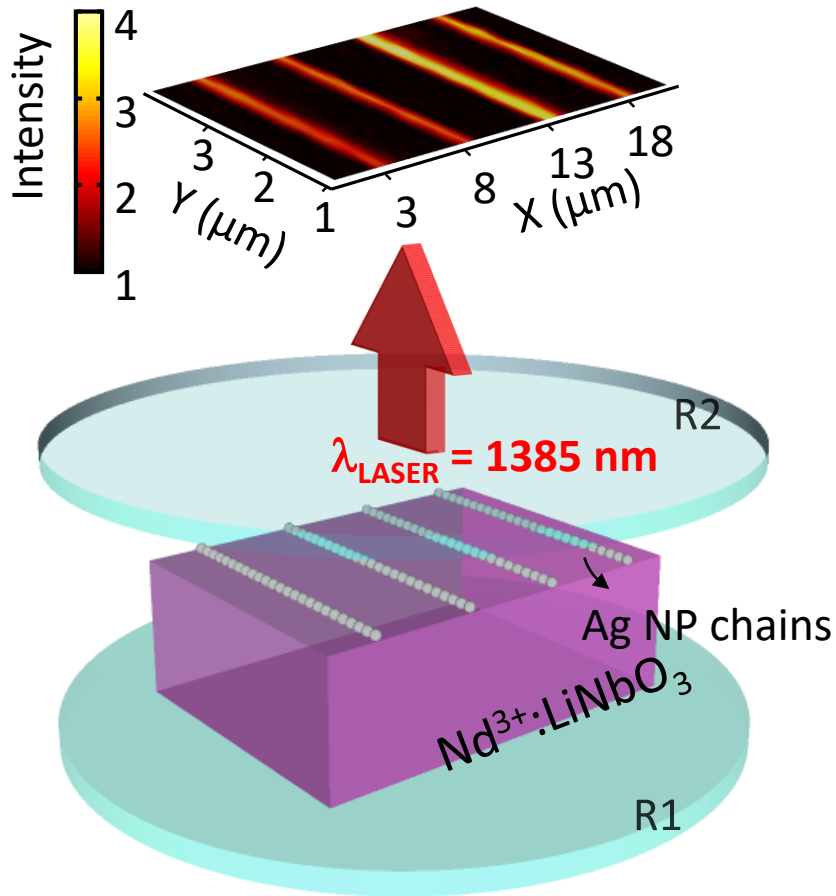
Plasmon-induced spectral Narrowing in a Subwavelength Solid-State Laser



Effect of the absorption enhancement on the laser emission linewidth

Nd^{3+} emission at 1385 nm is located well outside the spectral region of the LSP

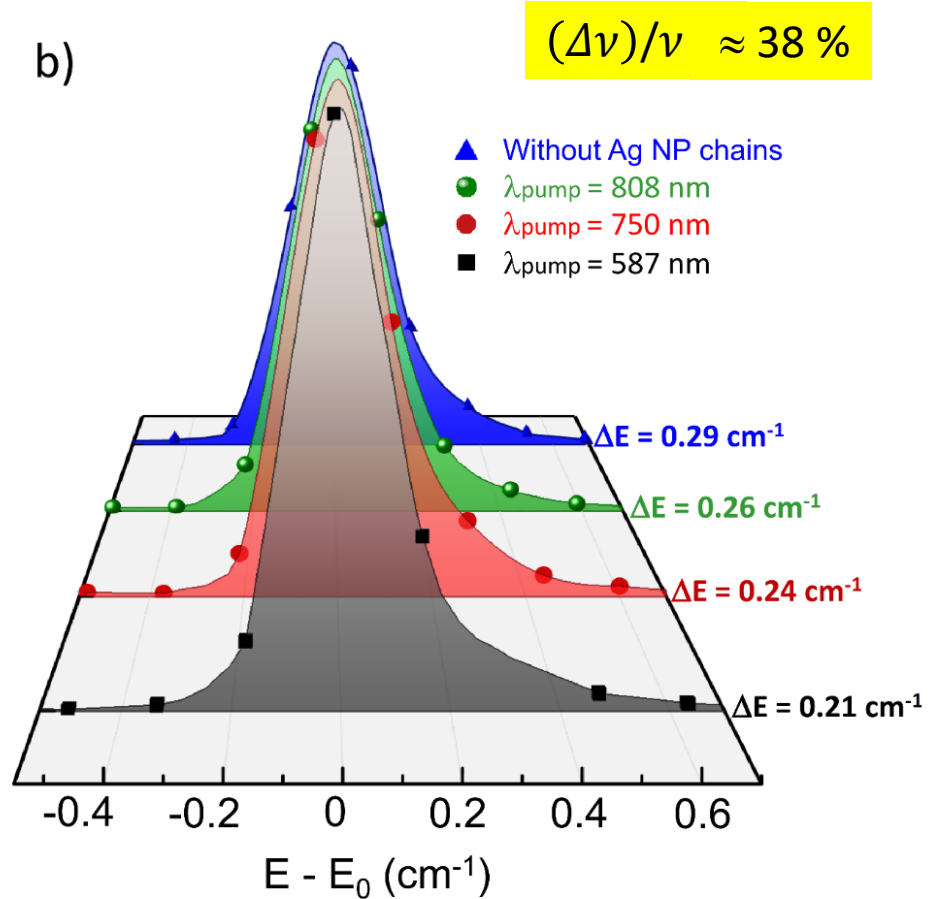
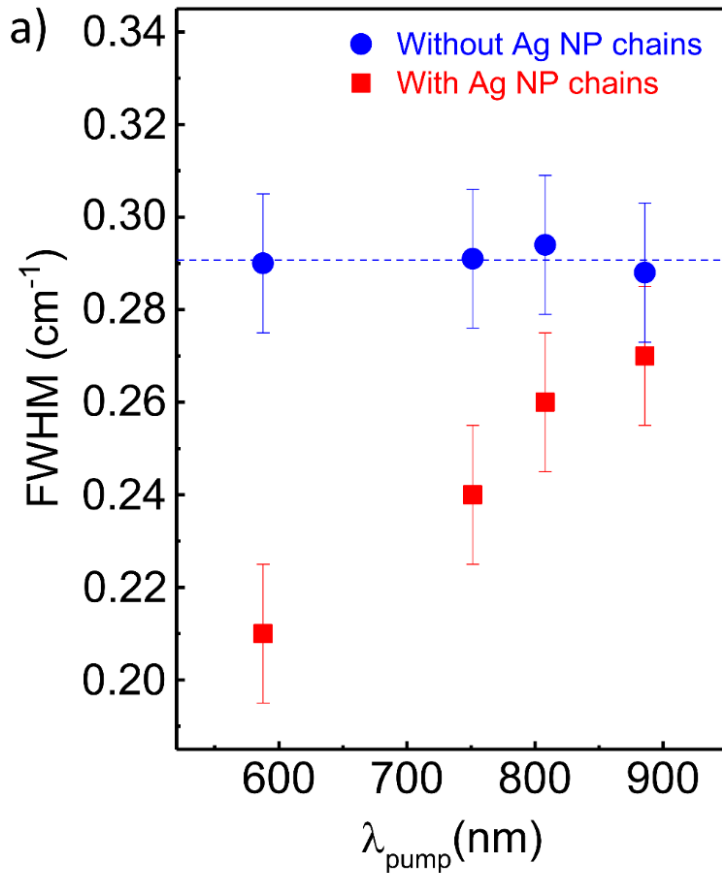
Lasing performance in the vicinities of the Ag NP chains at 1.38 μm



Lasing at the nanoscale in the TELECOM optical E-band

A significant **threshold reduction** & **increase in the slope efficiency** were achieved in the vicinities of the Ag NPs

Effect of the LSP on the monochromaticity of the laser emission



A systematic decrease of the laser linewidth is observed as the pump wavelength approaches the maximum of the plasmonic response

Microscopical origin of the laser linewidth narrowing

Semianalytical theory based on a rate equations system

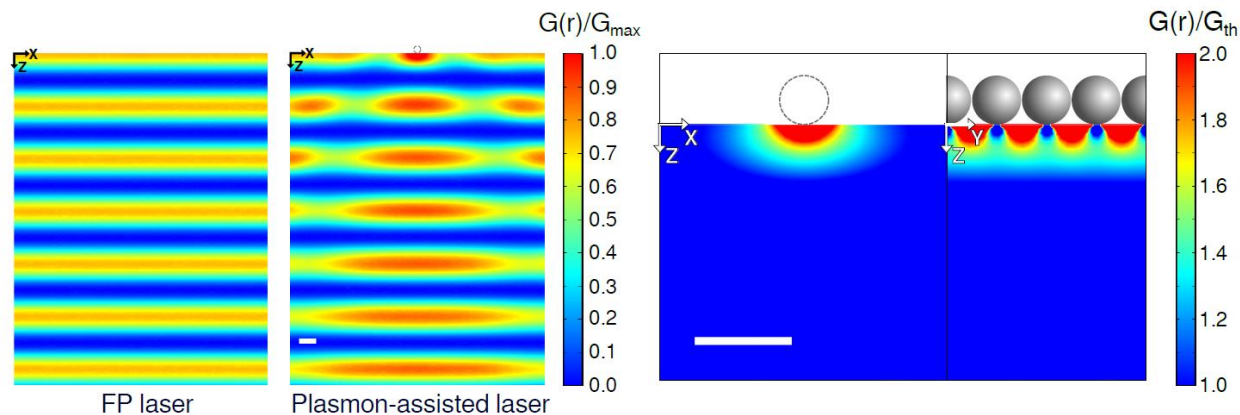
(J. Cuerda et al. *Phys. Rev. B* **91** (2015))

$$(\Delta\nu)_{laser} = \frac{\xi}{Q^2 \Gamma R_p}$$

The pump rate that is effectively transferred (via the active medium) to the lasing mode is given by

$$R_p = K_a \int_{V_a} d^3\mathbf{r} |\mathbf{E}_a(\mathbf{r})|^2 |\mathbf{E}_e(\mathbf{r})|^2$$

$G(\mathbf{r}) = |\mathbf{E}_a(\mathbf{r})|^2 |\mathbf{E}_e(\mathbf{r})|^2$ optical gain distribution inside the active medium



Highly localized gain distribution

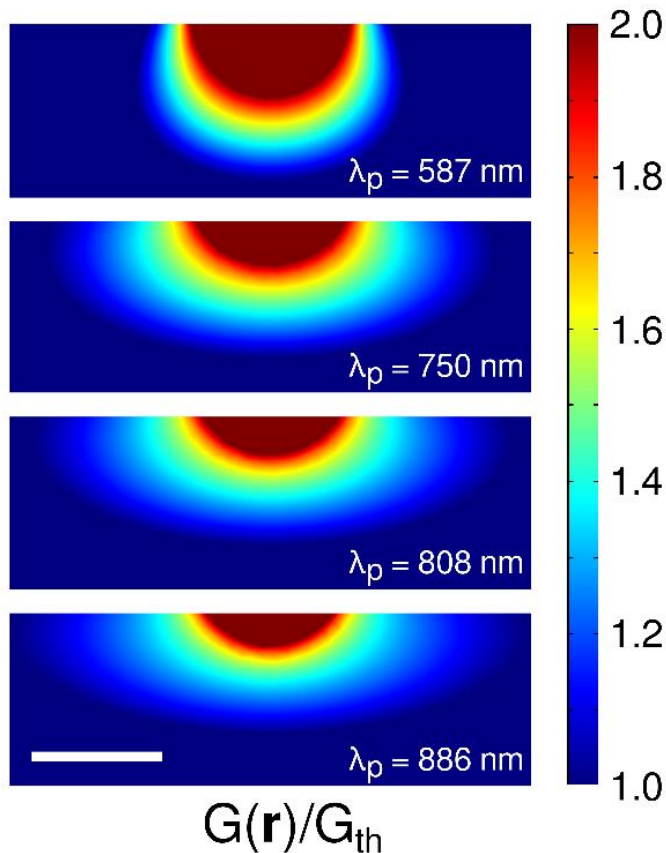
Regular emission pattern

FP cavity mode + Uniform profile of the pump field

Complex pattern

Efficient excitation of the longitudinal LSP of the chain

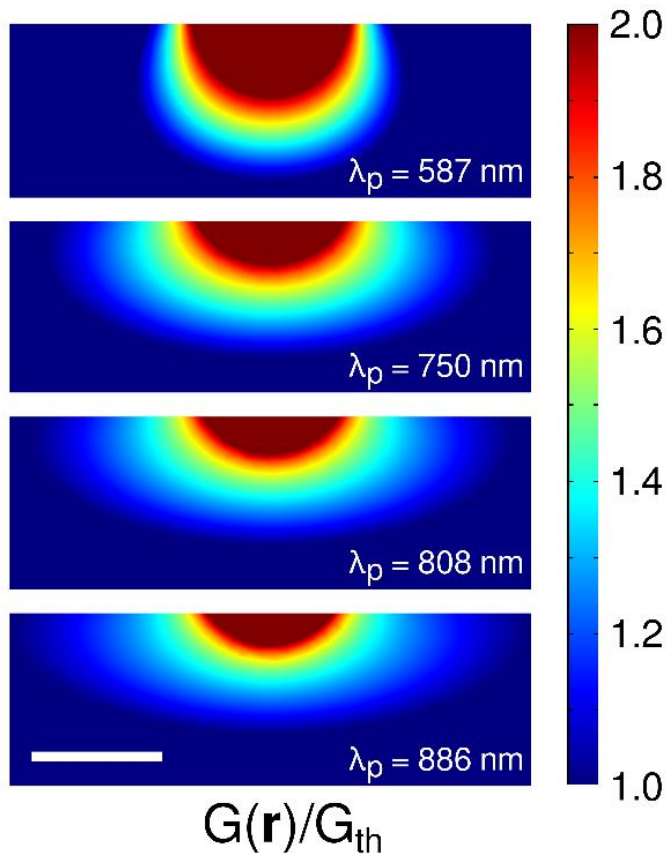
Optical gain profile near the crystal surface for different pump wavelengths



Further enhancement of the optical gain in the vicinities of the plasmonic nanostructures

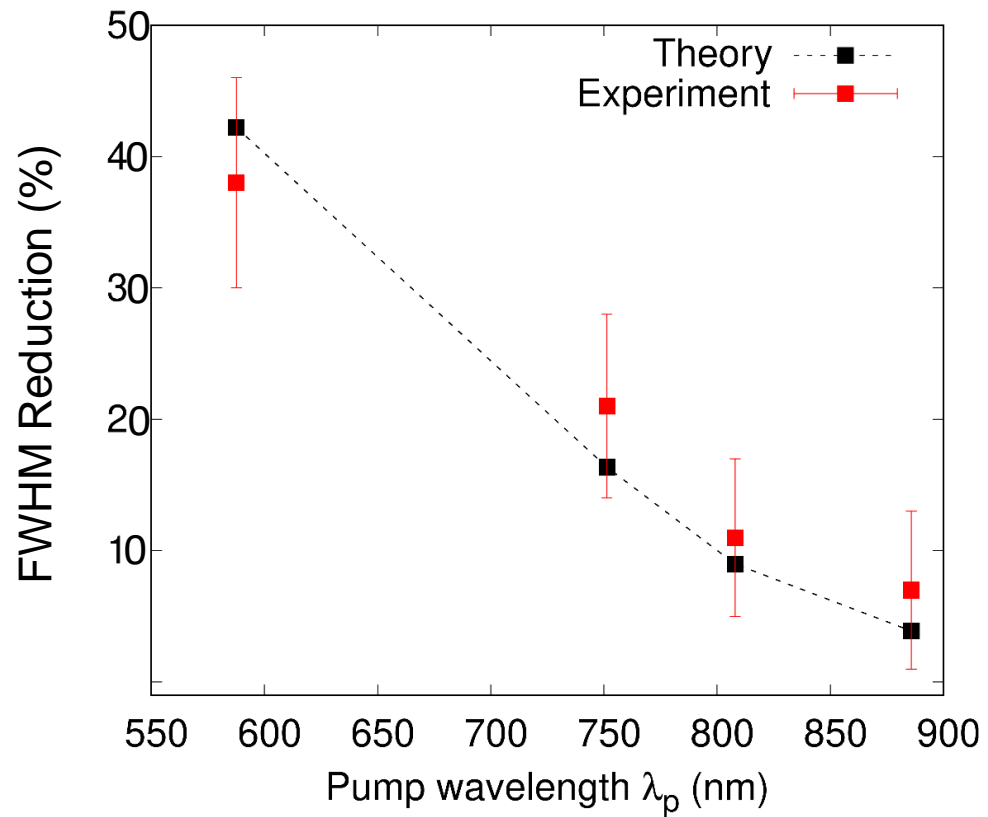
Higher degree of confinement, leading to nanolaser emission from an increasingly ultra-small volume

Laser linewidth narrowing as a function of the pump wavelength



$$(\Delta\nu)_{laser} = \frac{\xi}{Q^2 \Gamma R_p}$$

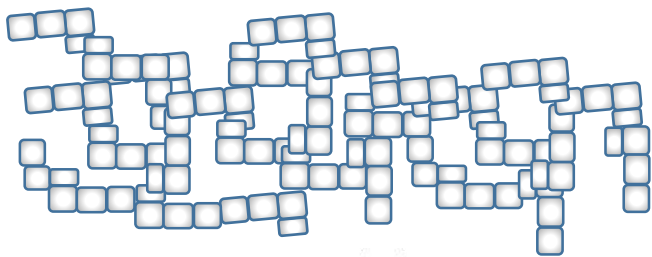
$$(\Delta\nu)_{pl}/(\Delta\nu)_b \approx R_p^b/R_p^{pl}$$



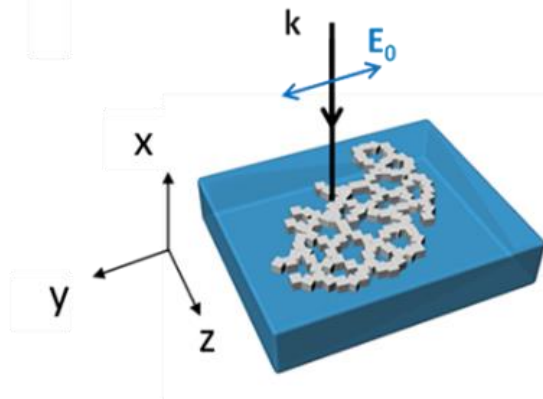
II. Effect **Disorder Plasmonic Networks (DPNs)** of silver nanoparticles on the optical performance of **Yb³⁺** doped RTP

Ag NP connected forming Disorder Plasmonic Networks (DPNs)

- Spectrally broad field amplification which is extended over large areas
- Spectral shift of the plasmonic resonance from the visible region to the near-IR



VIS → NIR

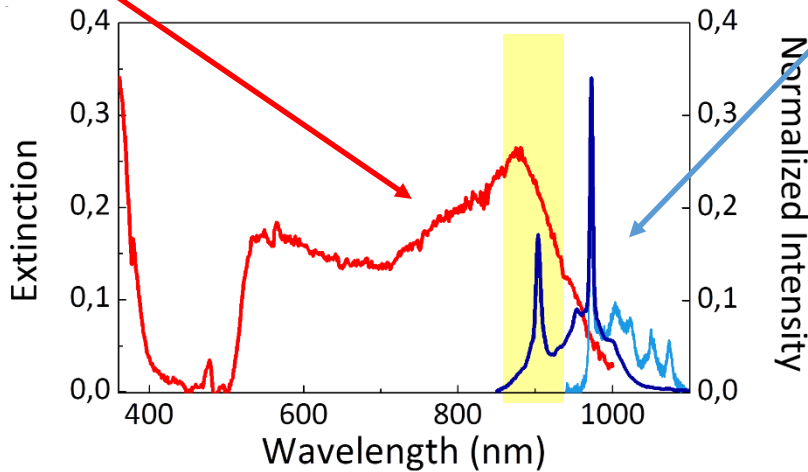
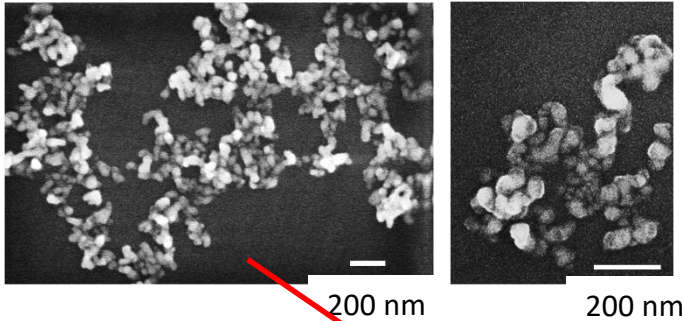


Yb³⁺ doped RbTiOPO₄ (RTP) crystal

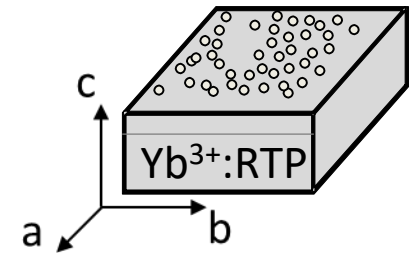
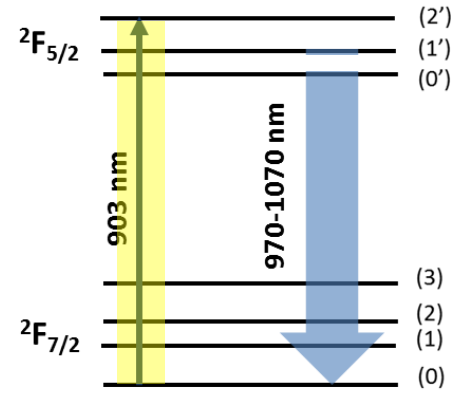
Spectral response of DPNs + Yb^{3+}

Disordered plasmonic networks (DPNs)

Formed by Ag NPs with sizes in the range 60-90 nm connected in a disordered network-like arrangement



Yb^{3+} :RTP



Spectral overlap between the plasmonic response of the Ag DPN and the optical absorption of Yb^{3+}

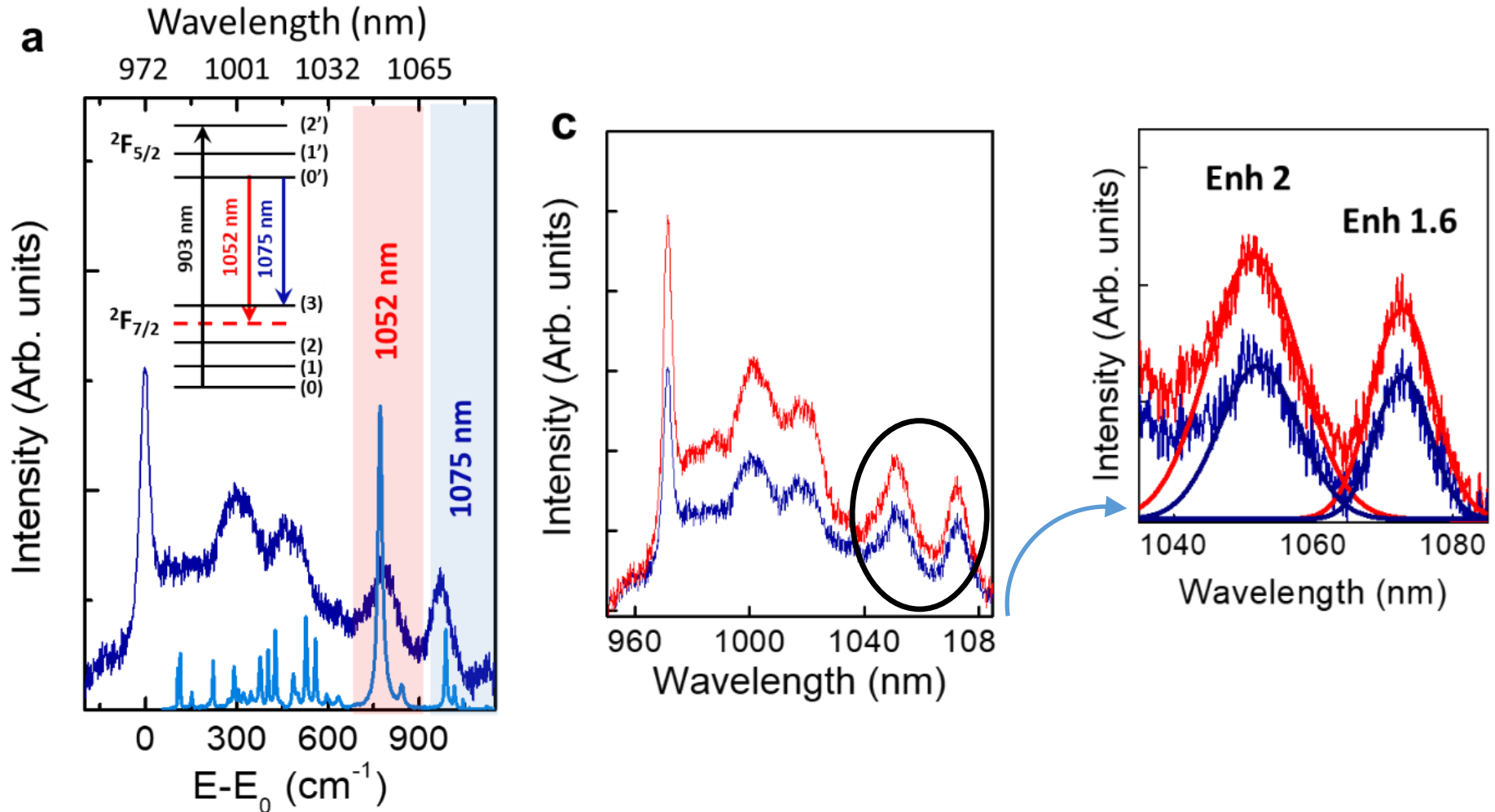
Disordered plasmonic networks induce drastic modifications in the optical properties of Yb^{3+} :RTP

→ INCREASE THE EXCITATION RATE OF Yb^{3+} ions and remarkable enhancement of the photoluminescence

→ PLASMON-INDUCED DUAL WAVELENGTH LASER

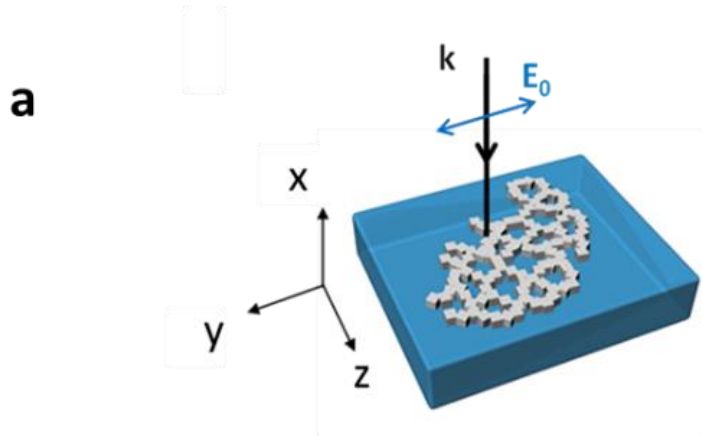
Simultaneous oscillation of two different laser lines due to the effect of the DPNs deposited on the surface of the Yb^{3+} doped RTP laser crystal

Plasmon-induced dual wavelength Yb³⁺ laser



The emission at 1052 with a strong vibronic character is preferentially enhanced by the DPNs about 20% larger than that of the emission at 1075 nm

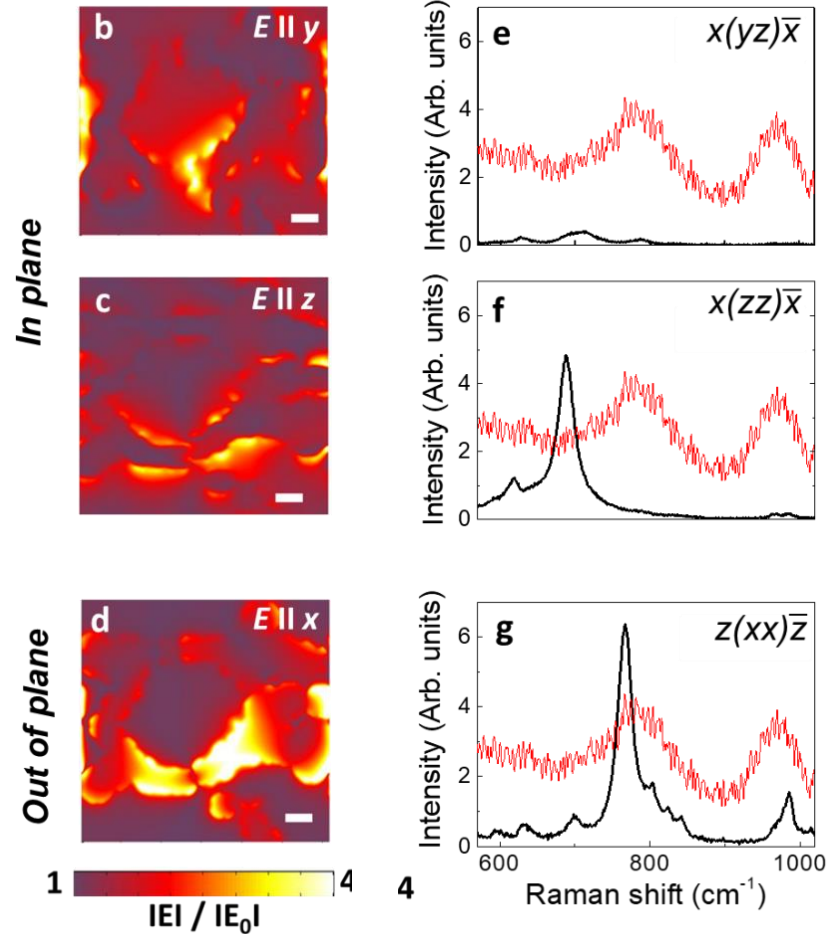
Selective enhancement of Yb^{3+} vibronic transition by Ag DPNs



Geometric configuration of the RTP crystal used in the lasing experiments

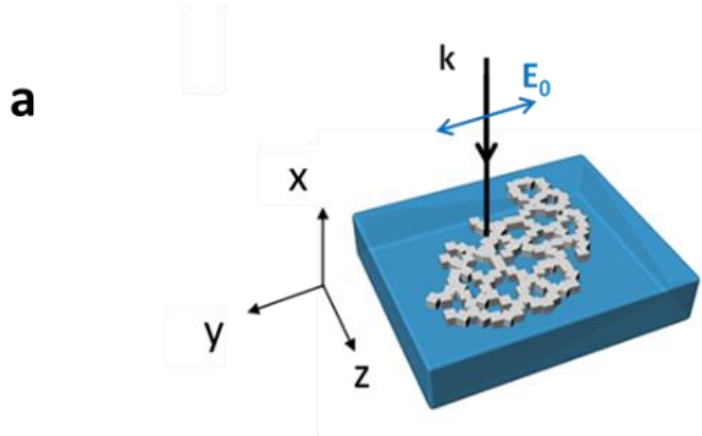
Presence of an **out-of-plane polarization** component **additional** to those of the incident field

The **largest values of the local field enhancement** correspond to the **out-plane field component**



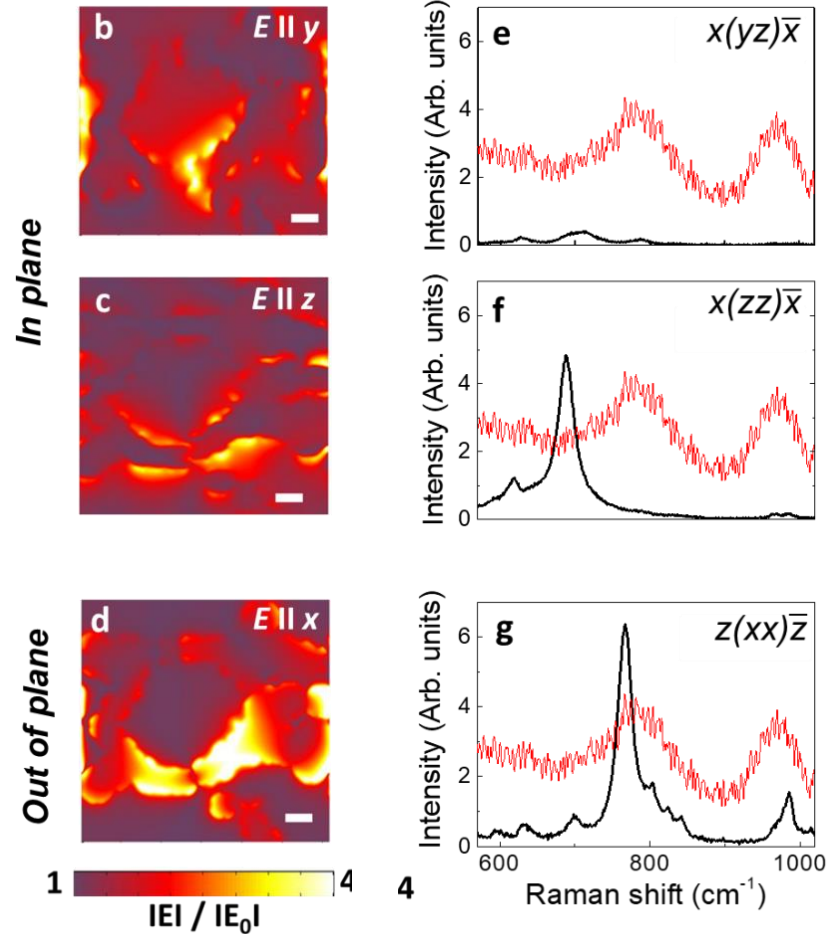
-----Emission spectrum of $\text{Yb}^{3+}:\text{RTP}$ in the lasing spectral region
 -----Raman spectra

Selective enhancement of Yb^{3+} vibronic transition by Ag DPNs



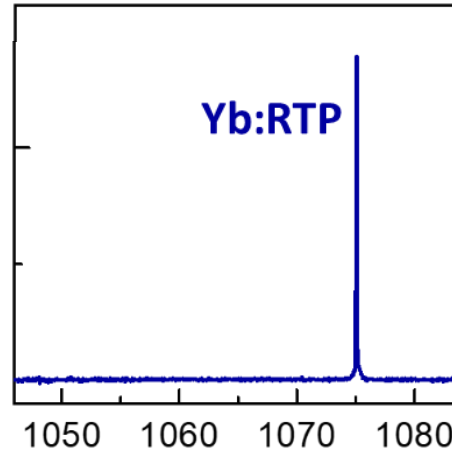
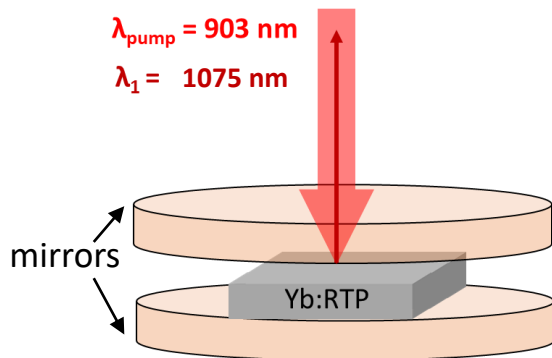
Geometric configuration of the RTP crystal used in the lasing experiments

This out-of-plane component allows the excitation of the vibrational modes, not accessible without the presence of DPNs, with the subsequent enhancement of the phonon-terminated transition at 1052 nm.

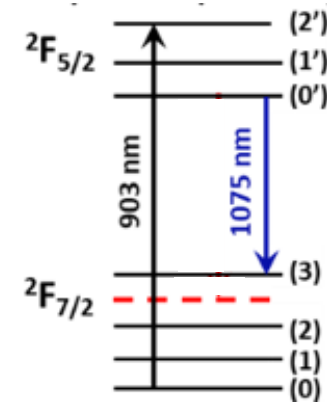


-----Emission spectrum of $\text{Yb}^{3+}:\text{RTP}$ in the lasing spectral region
 -----Raman spectra

Plasmon-induced dual wavelength Yb³⁺ laser



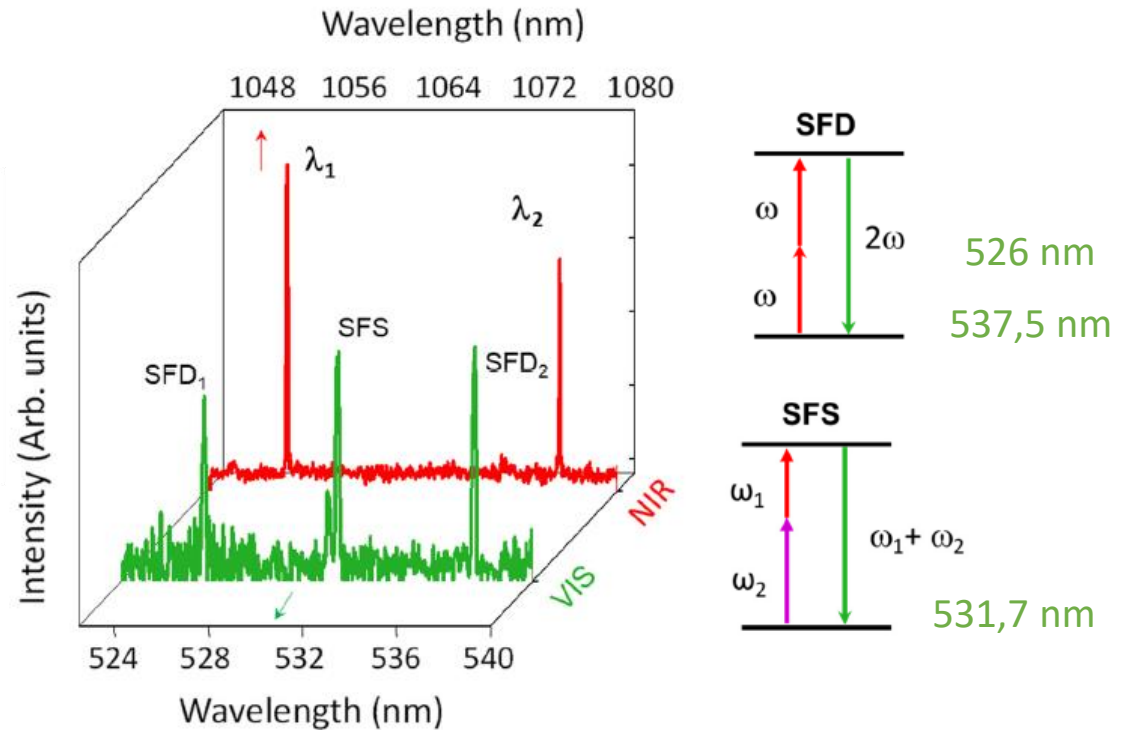
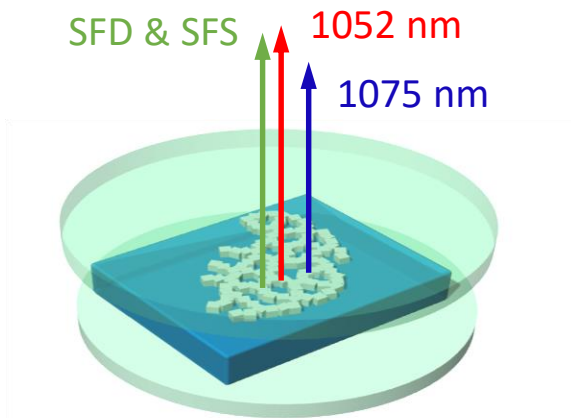
Yb³⁺ → quasi 3-level laser



Dual wavelength laser behavior induced by the plasmonic resonances of Ag DPNs

Multiline visible radiation in the vicinities of DPNs

RTP matrix: large $\chi^{(2)}$ coefficients



Multiline green radiation is achieved via SFS and SFD processes involving the plasmon-mediated laser line

Summary

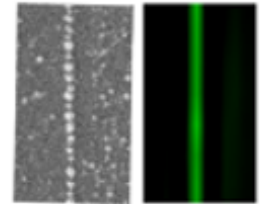
Effects of arrays of Ag NPs on nonlinear solid-state gain media

1. Demonstration of plasmon-assisted solid state lasers at the nanoscale

➤ **Improved performance** with respect to conventional bulk laser operation

→ Threshold reduction and gain increase

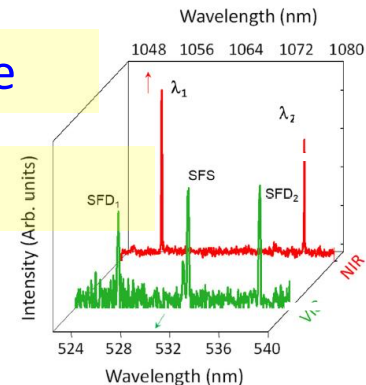
→ Laser linewidth reduction



➤ **New functionalities** → Dual wavelength lasing operation

2. Enhancement of frequency conversion processes at the nanoscale

Multiline operation from a single plasmonic nanolaser



The results can be extended to the large diversity of solid-state lasers with the subsequent impact on the applications

