Interaction between localized surface plasmon and nonlinear solid state gain media

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http://spectroscopygroup.com

Lead by Prof. L.E Bausá

- 3 permanent professor
- 1 assistant professor
- 3 PhD Students

❖ "Background"

Multifunctional Solid State Lasers

- Optical Spectroscopy
- Laser gain experiments
- Ferroelectric domain engineering (1D & 2D)
- Nonlinear frequency conversion processes
- Assembling metallic nanostructures on polar surfaces

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❖ "Current activity"

- **Multifunctional coherent sources at the nanoscale**

- **Control the optical properties of luminescence ions at the nanoscale**
- **Plasmon assisted solid state nanolasers**
- **Enhancement of frequency conversion processes at sub-wavelength scales**

Hybrid Plasmonic-Ferroelectric systems

FERROELECTRIC nonlinear LASER CRYSTALS

PLASMONS SUPPORTED BY METALIC NANOSTRUCTURES

Optical properties – Plasmon Resonances

- Fluorescence
- Laser emission
- SHG

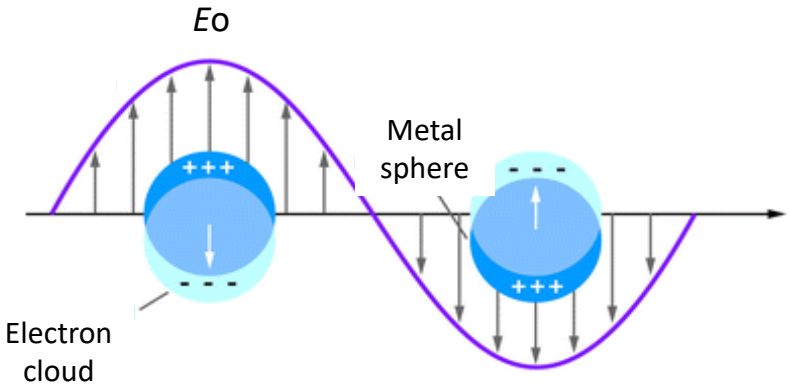
Metallic nanostructures on nonlinear solid state lasers materials for the development of novel photonic devices

Why Noble metal nanoparticles ?

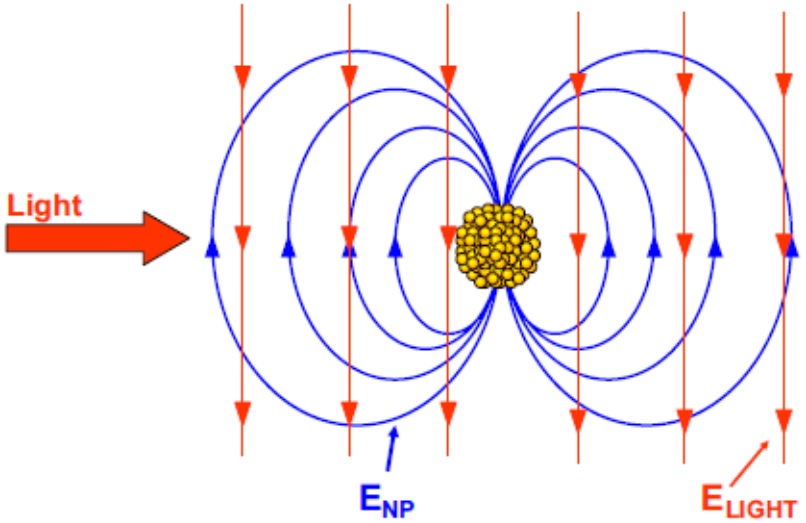
Noble metal nanoparticles
Localized Surface Plasmons (LSP)

Possibility of coupling light with **collective oscillations of their conduction band electrons**

→ **localized surface plasmon (LSP) resonances**



K. A.Willets et al. Annu. Rev. Phys. Chem.**58**, 266 (2007)



M A Garcia *J. Phys. D: Appl. Phys.* **44** 283001 2011

- **Antenna effect: Bridge the gap between near field and far field**
- **Highly confined Electromagnetic waves in the metal-dielectric interface**

Why Ferroelectrics?

❖ **Spontaneous polarization** → Fabrication of alternate domain patterns

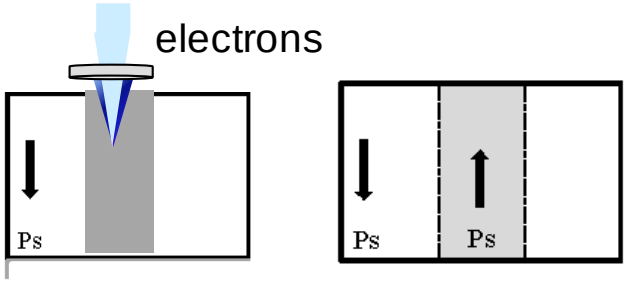
Efficient NONLINEAR Devices

❖ **Incorporation of optically active RE^{3+}** → Multifunctional Solid State Lasers

❖ **Presence of surface charge** → **Templates to assembly nanostructures**: organics, biological or **metals**

Fabrication of Alternate domain patterns

Ferroelectric crystals
$LiNbO_3$, RTP, BMF,…

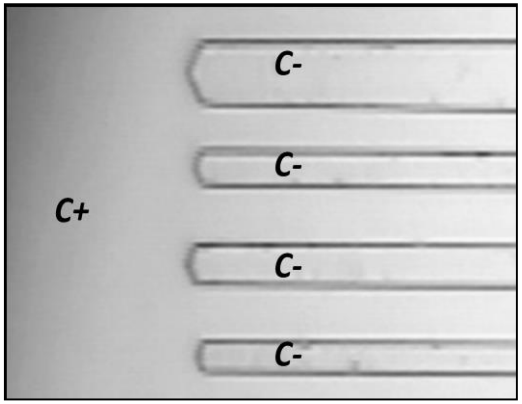


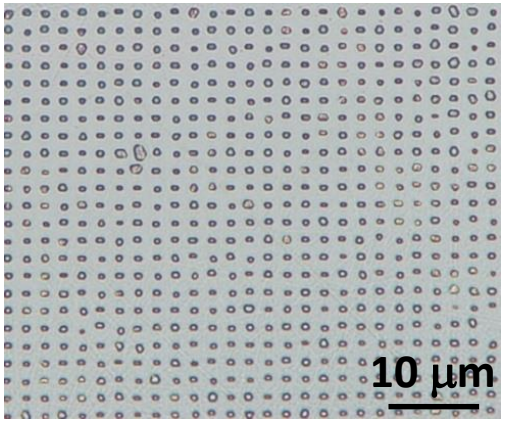
Direct electron beam writing

(DEBW)

1D

(*PPLN*)





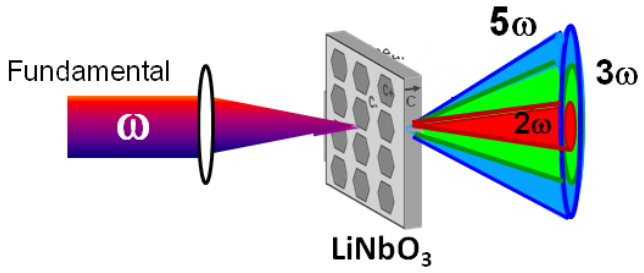
2D

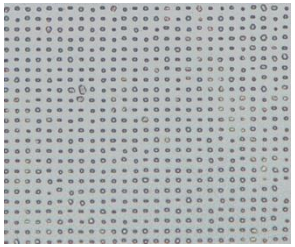
Λ~ 2μm

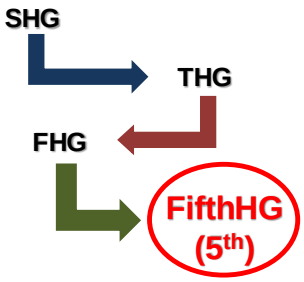
Applied Physics Letters **102,** 042910 (2013)

Ferroelectric domain engineering

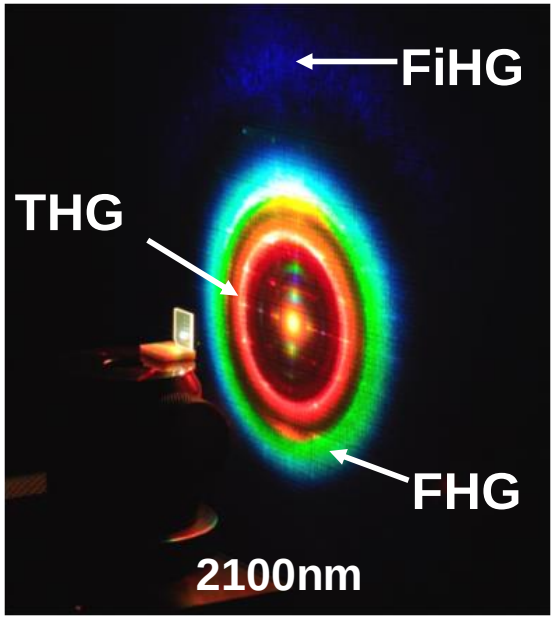
Nonlinear frequency conversion processes







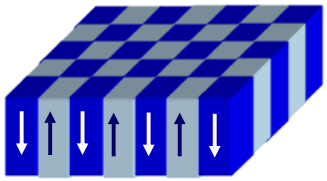
Cascade χ^2



Optics Express **20**, 29940 (2012)

Simultaneous generation of multiple harmonics (from 2nd to 5th)

Ferroelectric patterns as artificial templates



❑ **Assembling of metallic Ag nanoparticles**

❑ **Ferroelectric lithography**

Photo-reduction of Ag^{+} cations at the polar surface of ferroelectrics

Photo-deposition of Ag nanoparticles

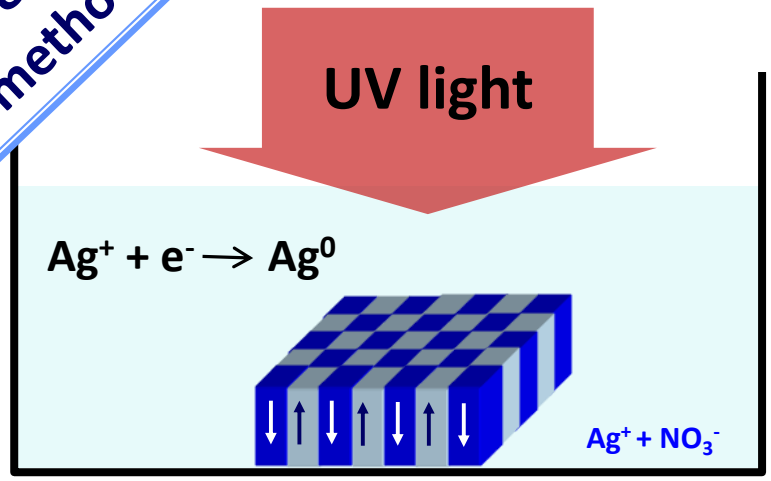
❑ Ferroelectric lithography

Ferroelectrics → Polar surface → surface charges, electric fields

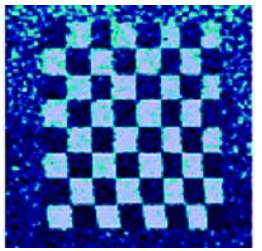
Above Bandgap UV irradiation ⟶ **selective Ag photo-reduction** on the surface

$$Ag^{+} + e^{-} \rightarrow Ag^{0}$$

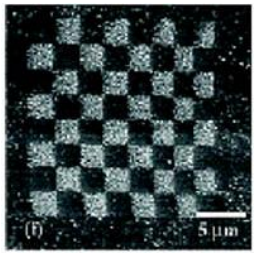
Scalable & Low cost method



Ferroelectric Pattern



Ag deposition

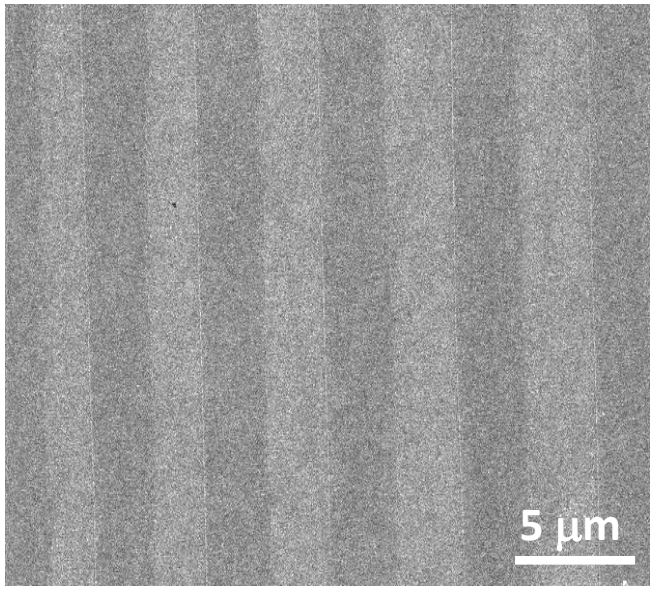


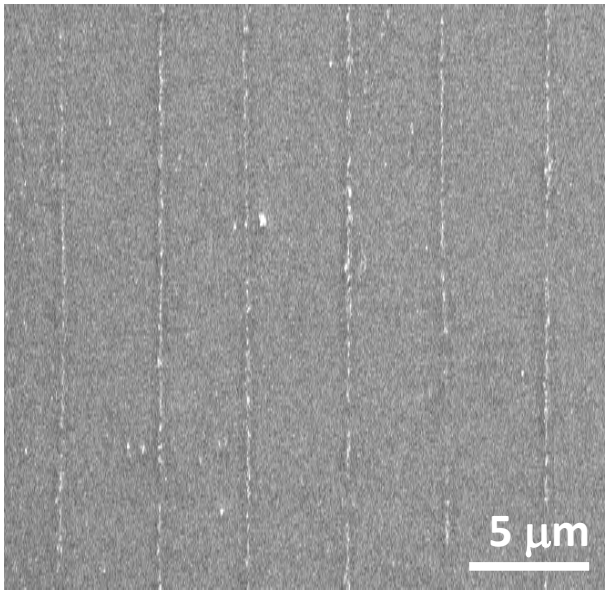
Kalinin et al. *Adv Mat* **16** 9 (2004)

Photo-deposition of Ag nanoparticles

❑ Ferroelectric lithography

1D PPLN





Positive domains (+c)

Domain walls

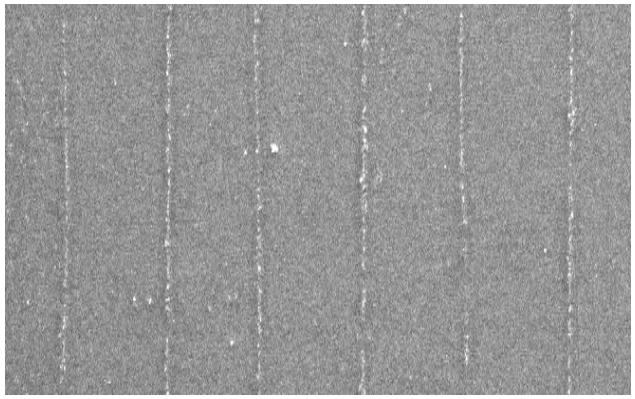
Selective Spatial deposition

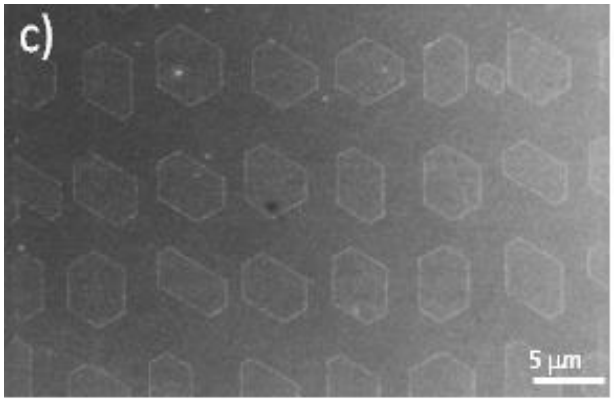
❑ Ferroelectric lithography

PPLN

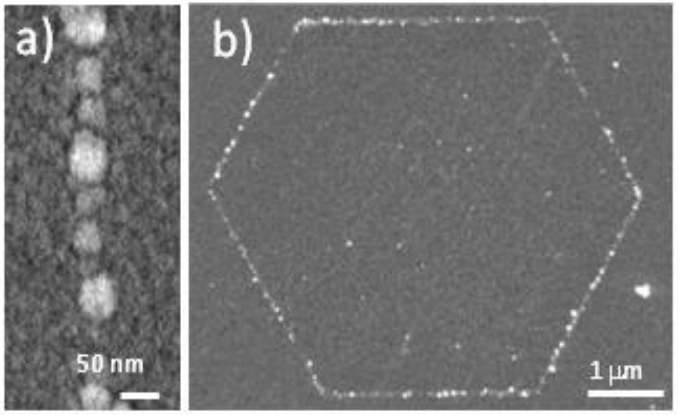
Arrays of hexagonal domains

1D





2D

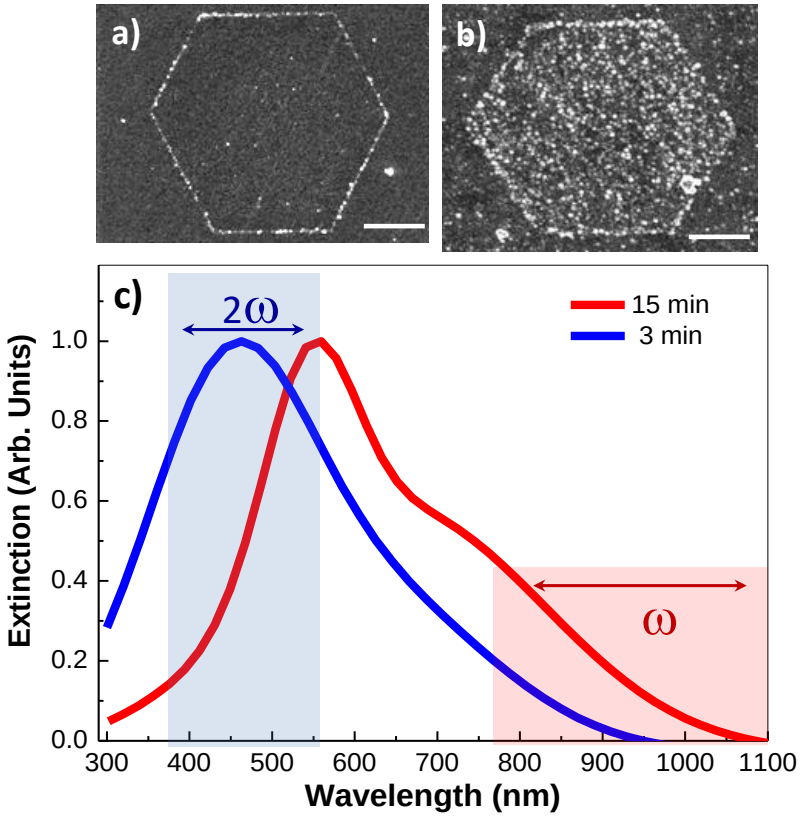


Linear Chains

Plasmonic Necklaces

Large areas of ordered metallic arrays

Plasmonic resonances: Dependence on the size & geometry



❖ **Spectral tuning of LSP resonances**

A)

Solid state laser
Optically active ferroelectric crystal

Plasmonic nanostructures

SOLID STATE LASER IN THE NANOSCALE

B)

Frequency converter crystal
Ferroelectric crystal

Plasmonic nanostructures

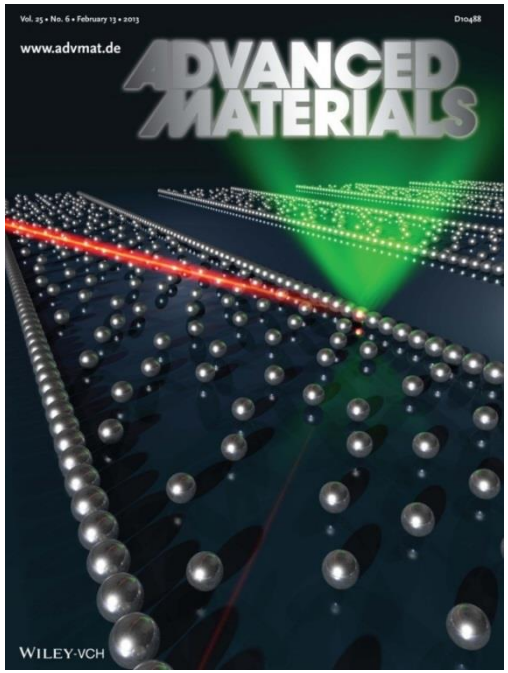
SHG enhancement in the NANOSCALE

Plasmon assisted Nd^{3+} based nanolaser

Ferroelectric domain pattern in $LiNbO_3:Nd^{3+}$

as template for Ag Nps deposition

Nd^{3+} doped PPLN



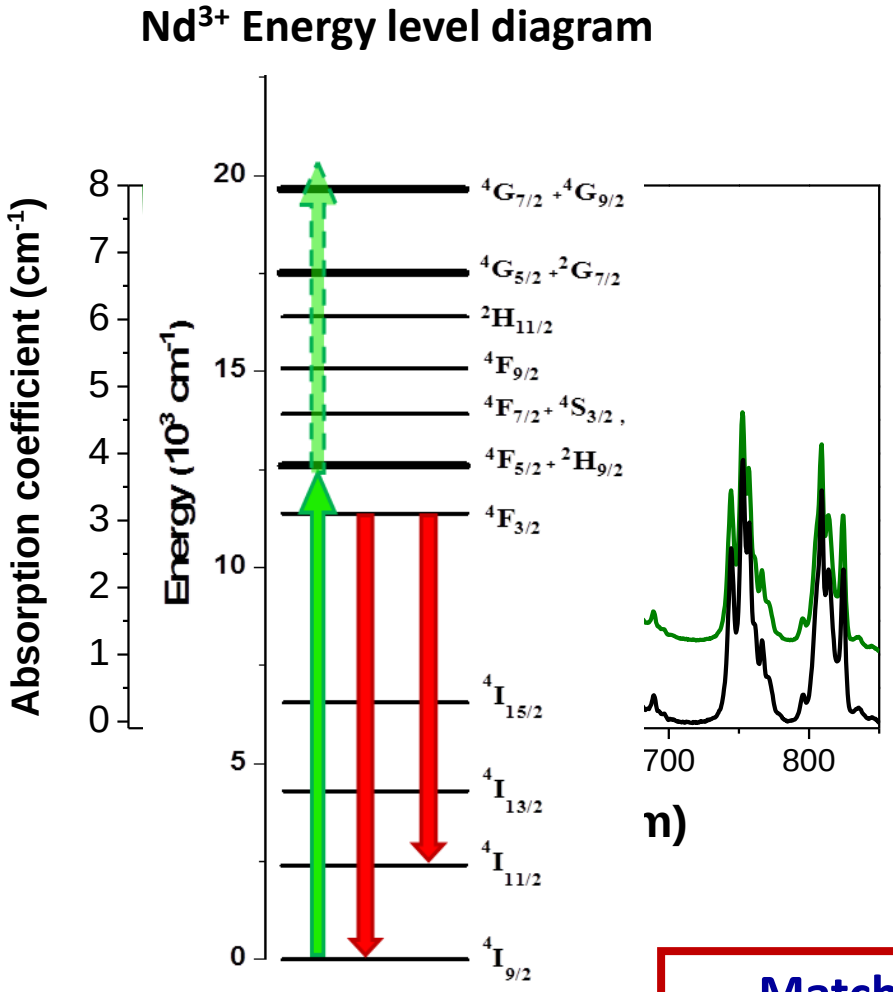
- Effect of Ag Nps on the luminescence
- Effect of the Ag NPs on the laser action of $Nd^{3+}:LiNbO_3$

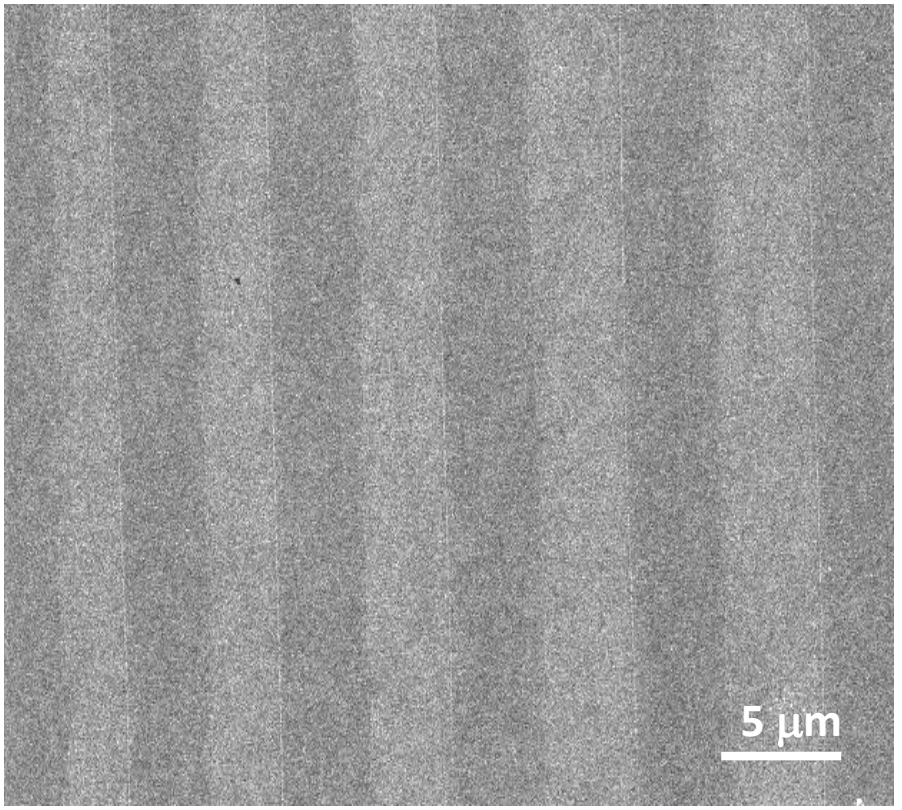
Materials Views
www.MaterialsViews.com
ADVANCED MATERIALS
www.advmat.de

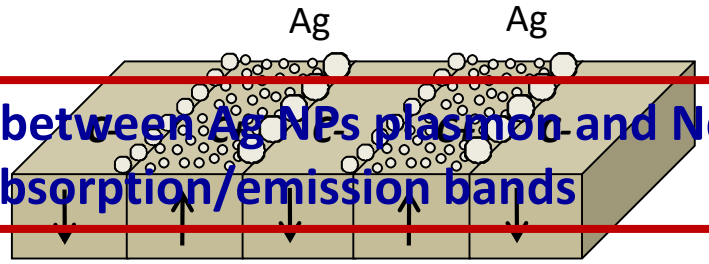
Spontaneous Emission and Nonlinear Response Enhancement by Silver Nanoparticles in a Nd^{3+}-Doped Periodically Poled $LiNbO_3$ Laser Crystal

Eduardo Yraola, Pablo Molina, José L. Plaza, Mariola O. Ramírez, and Luisa E. Bausá*

Optical characterization: Nd^{3+} and Ag NPs absorption

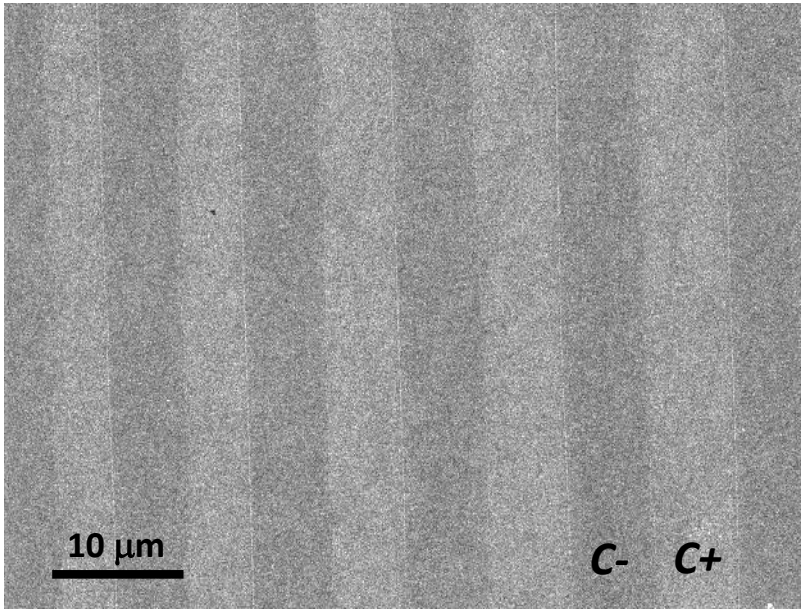




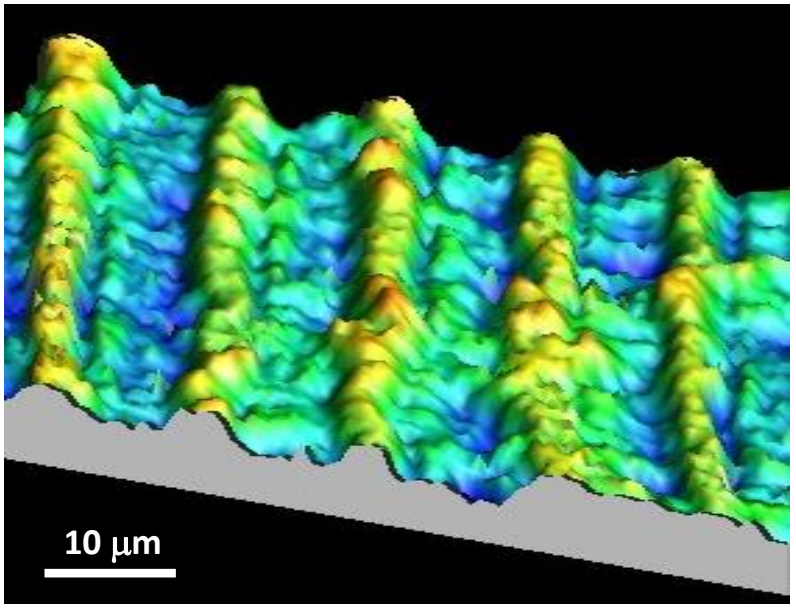


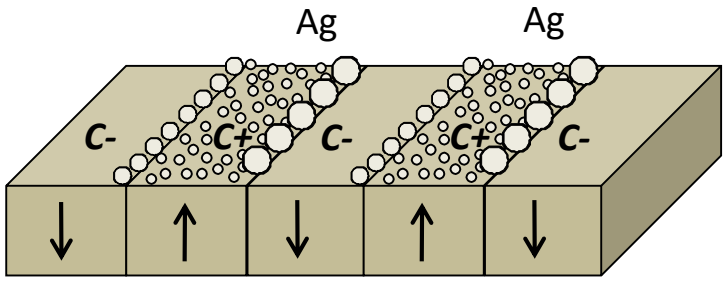
Matching between Ag NPs plasmon and Nd^{3+} absorption/emission bands

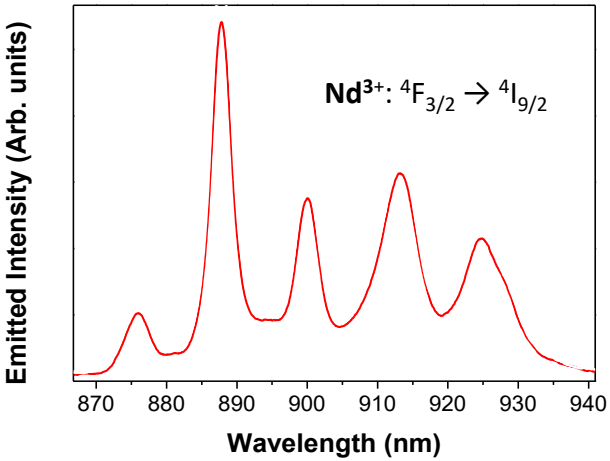
Optical characterization: Nd^{3+} Luminescence



Unpolarized Confocal μ-luminescence



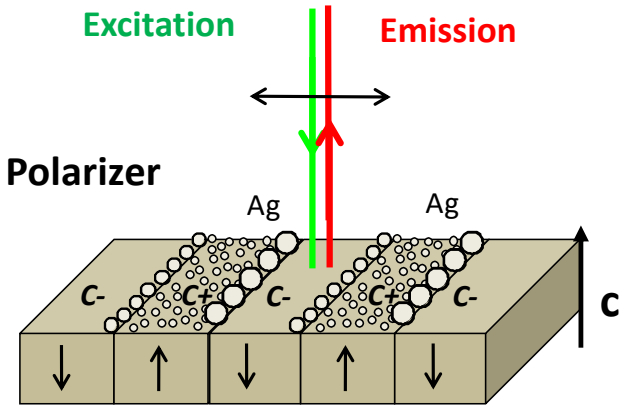


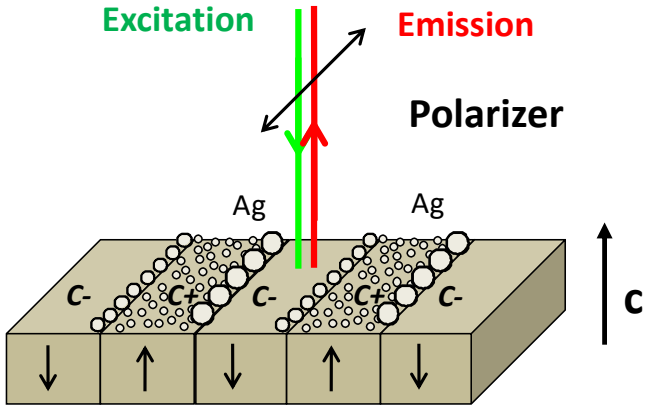


PL enhancement attributed to the coupling of transitions of Nd^{3+} ions to the localized surface plasmons

Polarization dependence

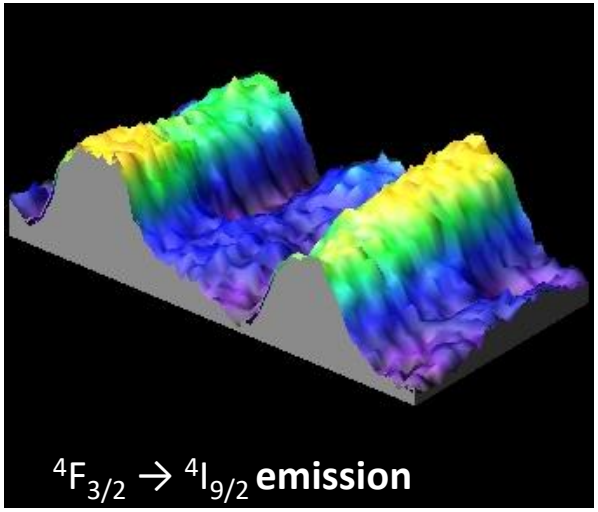
Ag NP ramdomly distributed / Ag Nps chains on the domain walls

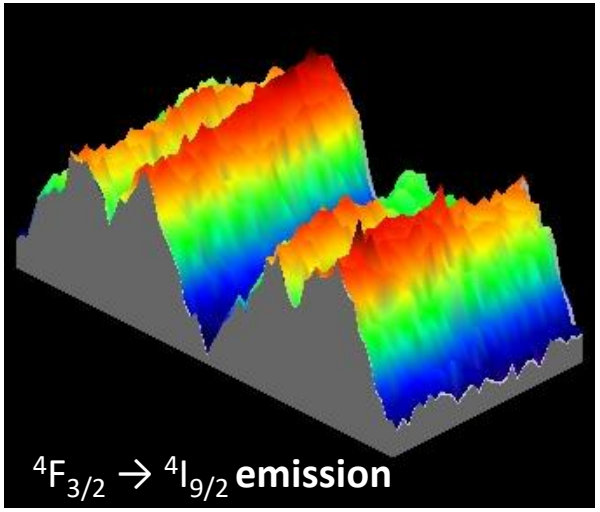




I max

I min





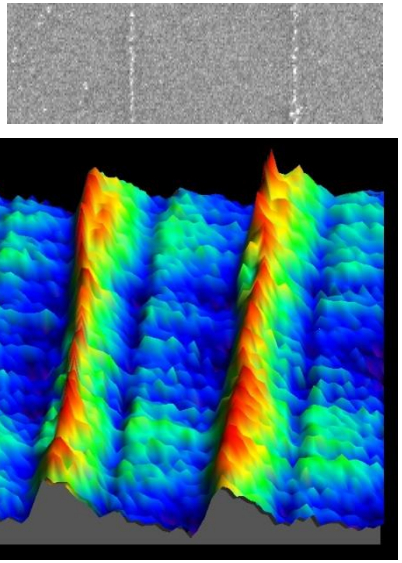
E. Yraola et al Adv. Mat. 25, 910 (2013)

Strong polarization dependent behaviour → Directional enhancement of the PL

Resonant directional coupling between Nd^{3+} transitions and the LSP of the Ag NP chains

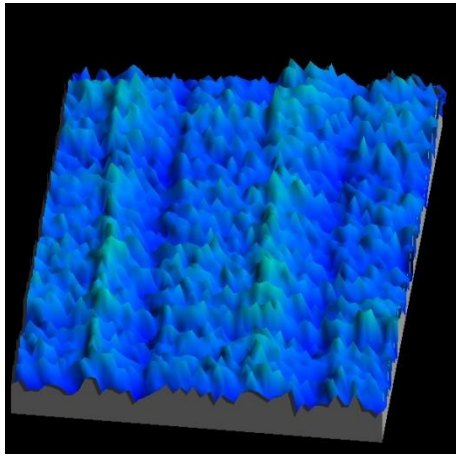
E II
Chain axis

E ↕

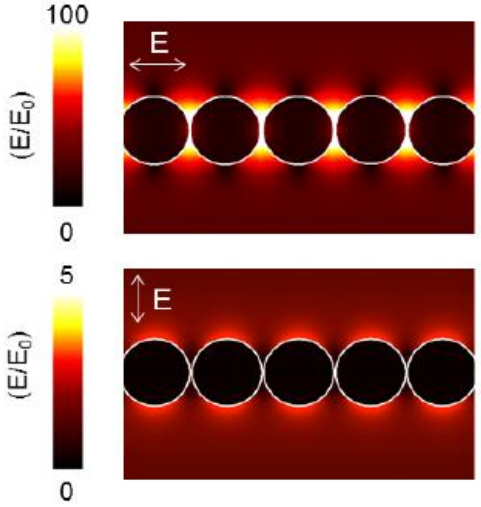


E ⊥
Chain axis

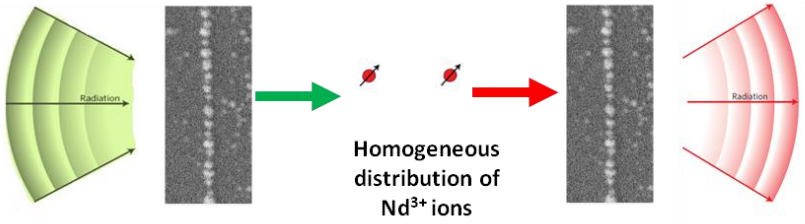
E ⟷



Strong near-field dipolar interaction between adjacent Ag nanoparticles



Coupling between antennas;



Excitation enhancement

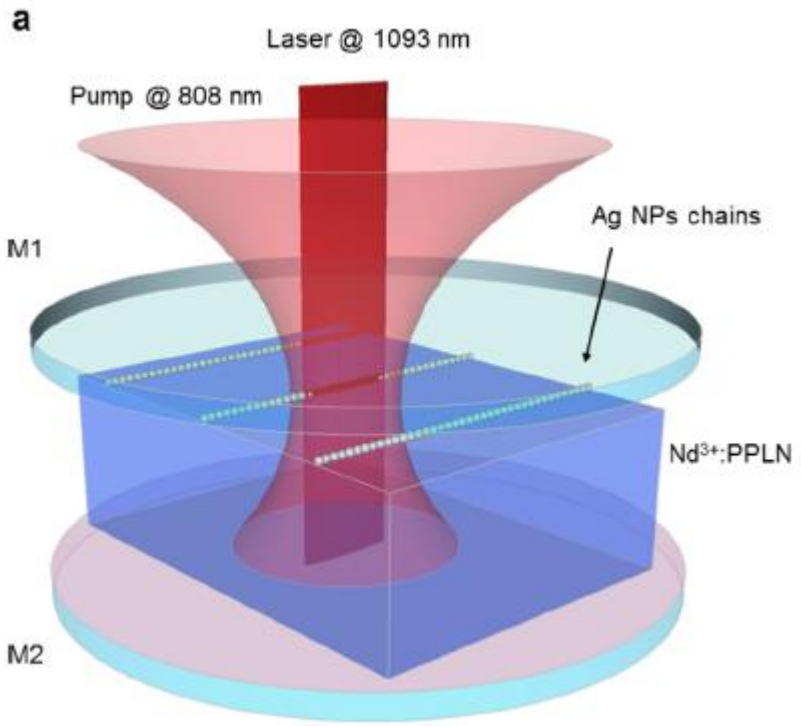
$^4I_{9/2} \longrightarrow {}^2G_{11/2} + {}^2G_{9/2}$

Emission enhancement

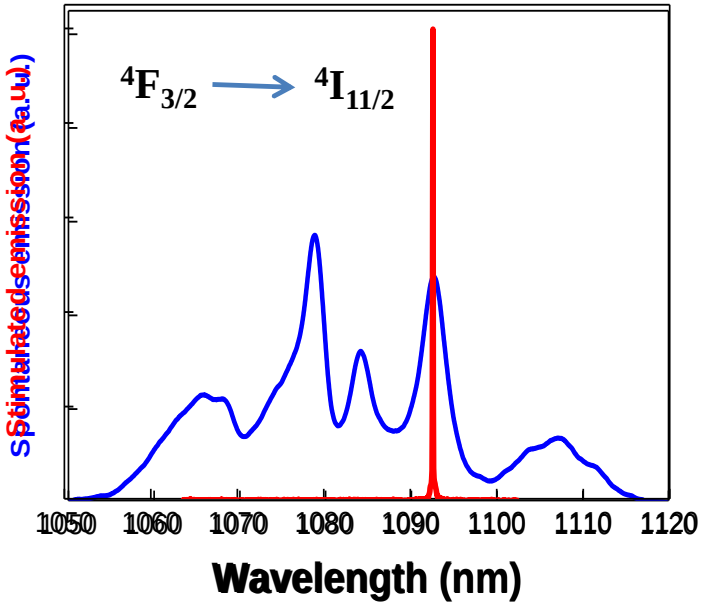
$^4F_{3/2} \searrow {}^4I_{9/2}$, $^4I_{11/2}$

Nd^{3+} based Solid-state nanolaser

Effects of the Ag NPs on the laser action of Nd^{3+} :LNB



Plane-parallel Optical Resonator



Nano Letters 16, 895, (2016)

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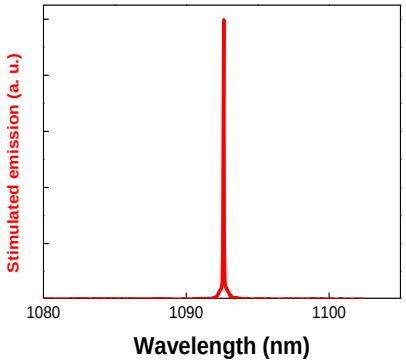
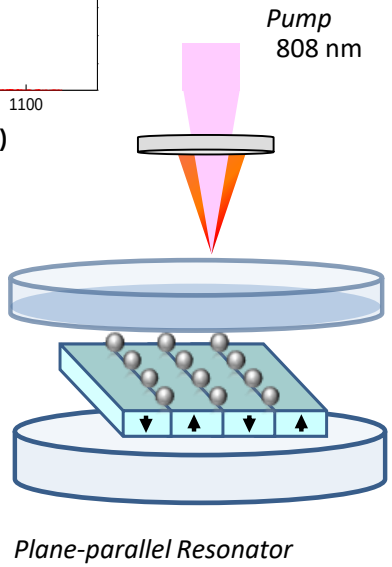
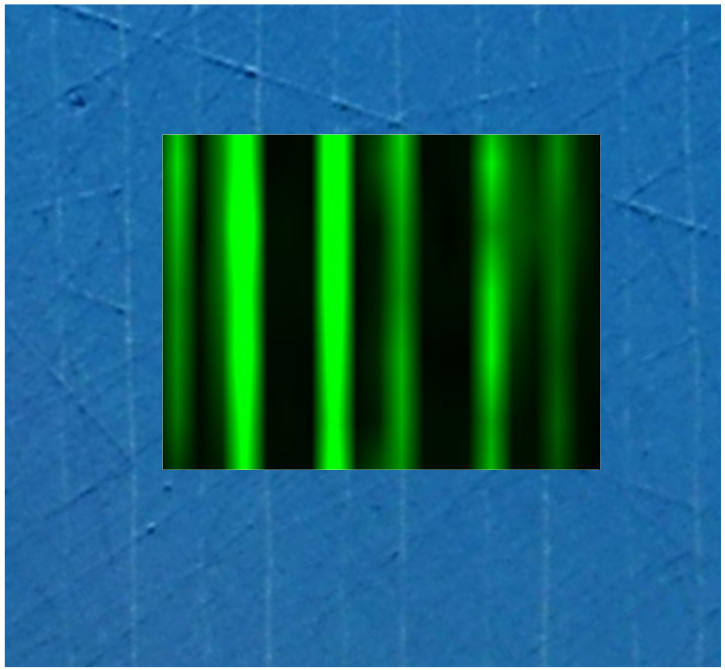
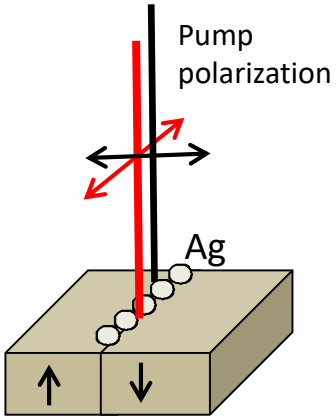


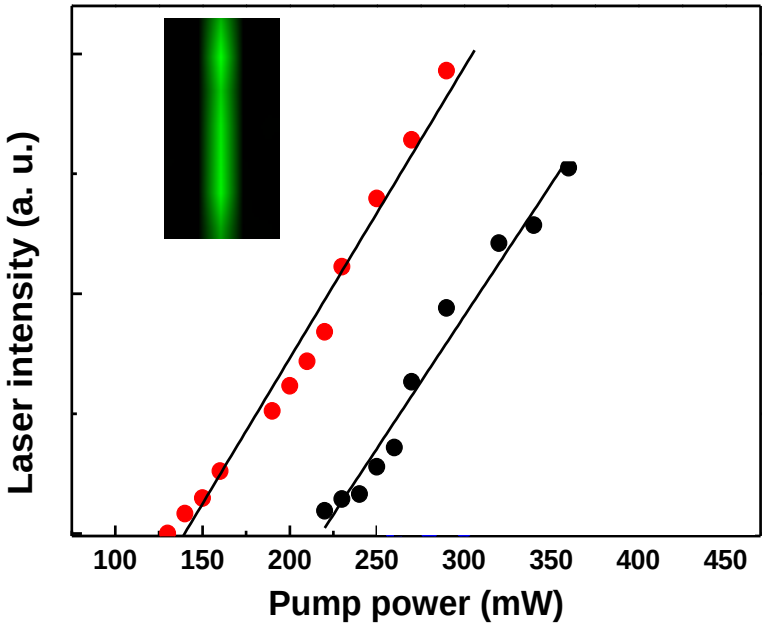




Room temperature CW laser action at the nanoscale

Laser performance at room temperature



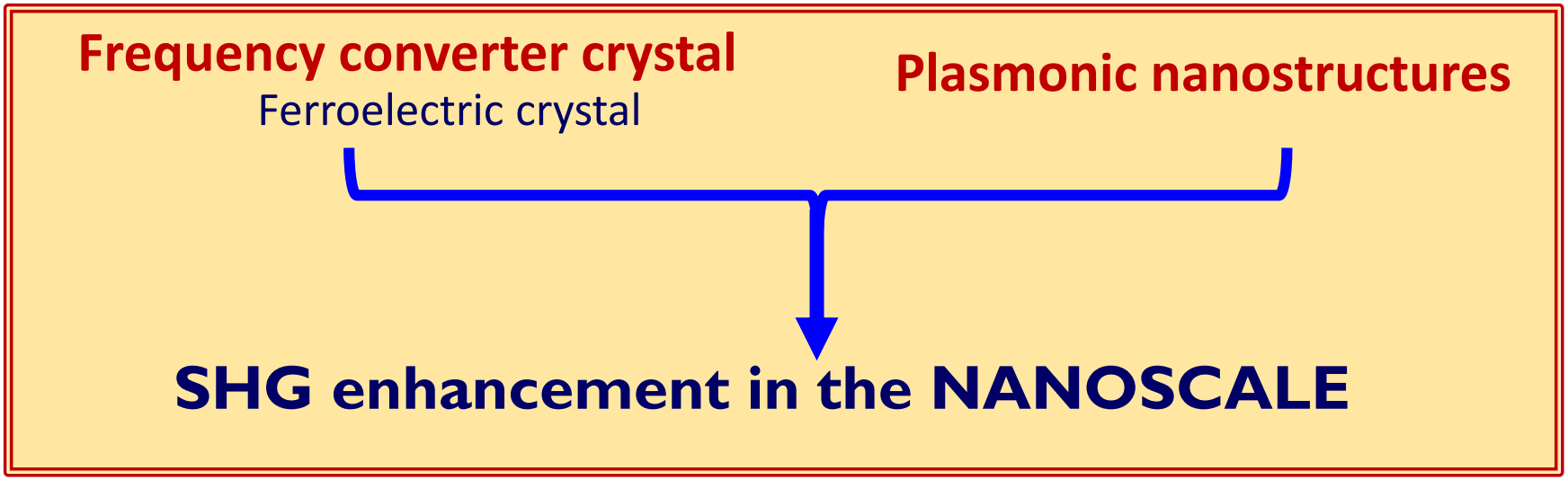


Nano Letters 16, 895, (2016)

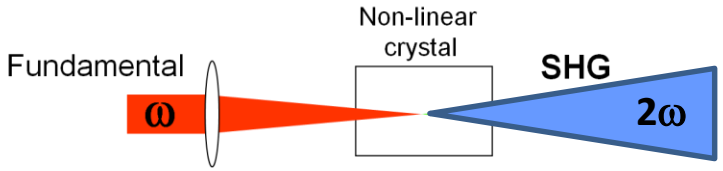
RT & CW Laser performance along the *c* axis at 1093 nm, pumping at 808 nm

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

The laser slope efficiency increases in a factor of 15

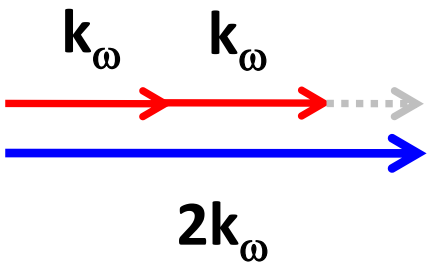

Frequency converter crystal
Ferroelectric crystal
Plasmonic nanostructures
SHG enhancement in the NANOSCALE

Second Harmonic Generation (SHG)

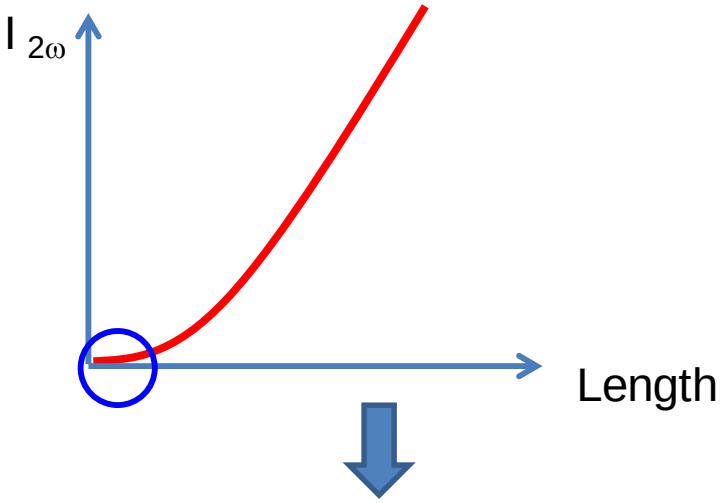


$$P_i^{2\omega} = \chi_{ijk}^{(2)} E_j^{\omega} E_k^{\omega}$$

❖ Efficient Frequency Conversion Processes: **PHASE MATCHING**

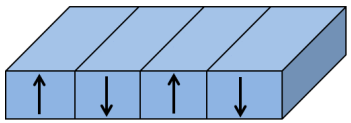


$$2k^{\omega} \neq k^{2\omega}$$



❑ Birrefringent Phase Matching (BPM)

❑ Quasi-Phase Matching (QPM)

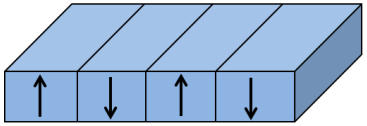


❖ **SHG inherently weak in nanoscale dimensions**

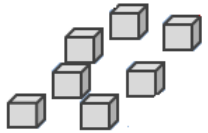
Second Harmonic Generation (SHG)

Ferroelectric system

(Periodically poled)

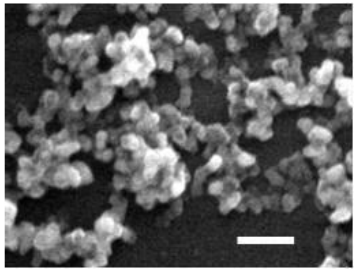


Plasmonic resonances provided by metallic nanostructures

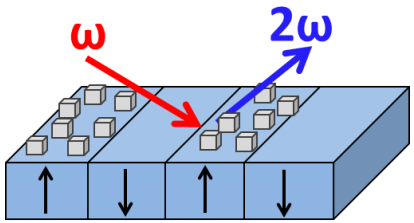


Enhanced SHG process at the nanoscale

❑ *Silver Aggregates*



(RTP)



❑ *Plasmonic Necklaces*

(LNB)

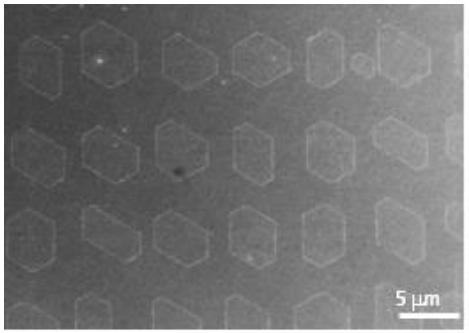


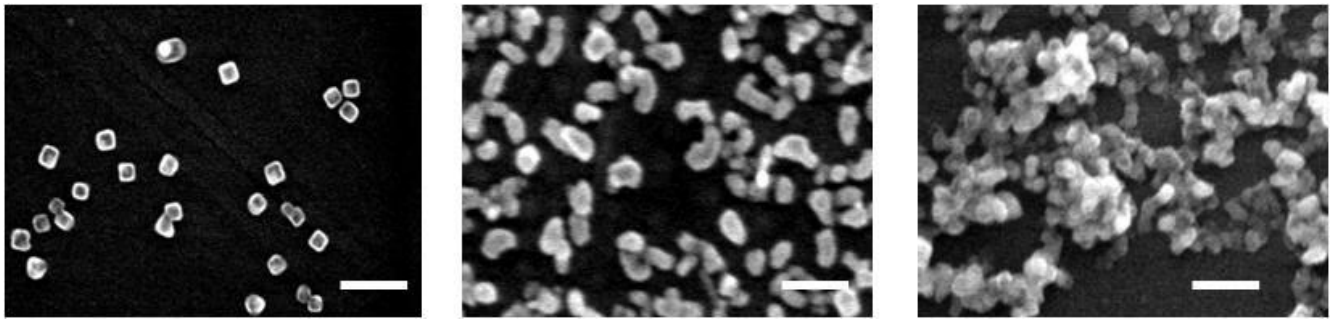
Photo-deposition of Ag nanoparticles in RTP

❖ **Increasing the illumination time (UV irradiation)**

(2 min) (4 min) (8 min)

Photo-deposition time

Nanocubes *Elongated NPs* *Complex nanostructures*

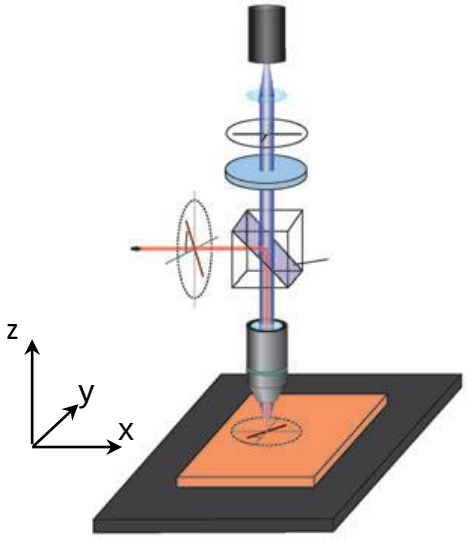


Isolated Nanocubes

Aggregates of complex nanostructures

Ag aggregates when increasing the time of the photochemical process

Second Harmonic Generation (RTP)



RTP Polar Surface

Pump: 840 nm
SHG: 420 nm

RTP second order nonlinear response

$$\begin{pmatrix} P_x^{2\omega} \\ P_y^{2\omega} \\ P_z^{2\omega} \end{pmatrix} = \begin{pmatrix} 0 & 0 & 0 & 0 & d_{15} & 0 \\ 0 & 0 & 0 & d_{24} & 0 & 0 \\ d_{31} & d_{32} & d_{33} & 0 & 0 & 0 \end{pmatrix} \begin{pmatrix} E_x^2 \\ E_y^2 \\ E_z^2 \\ E_y E_z \\ E_x E_z \\ E_x E_y \end{pmatrix}$$

$$P_x^{2\omega} = d_{15} E_x \cancel{E_z}$$

$$P_y^{2\omega} = d_{24} E_y \cancel{E_z}$$

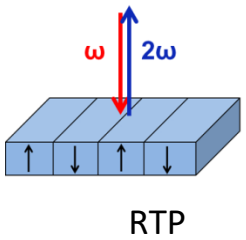
Fundamental beam propagates parallel to *c*
(z axis of the nonlinear tensor)

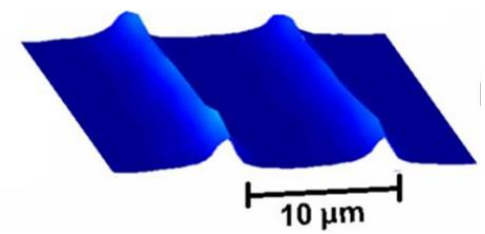
Bulk SHG is forbidden by symmetry rules

SHG @ Domain Walls

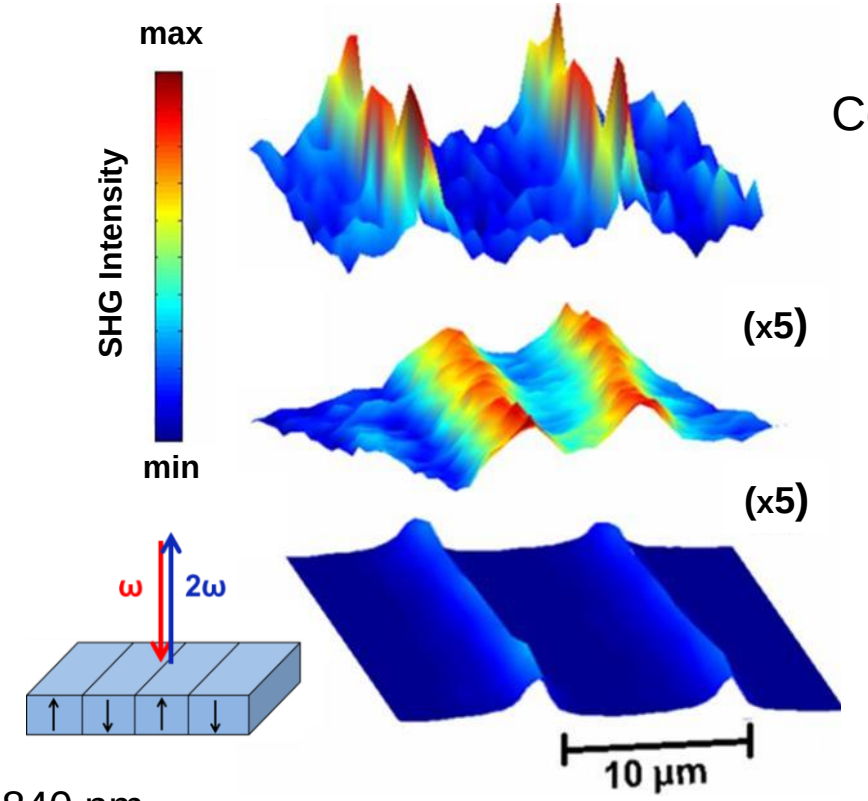
Symmetry Relaxation at interfaces

Domain walls: nanometric SHG sources



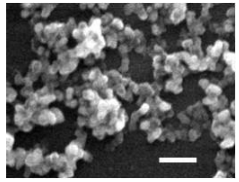


SHG in hybrid metal-RTP systems



Type B
Complex Aggregates

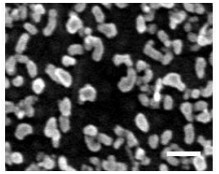
60-fold



(NIR)

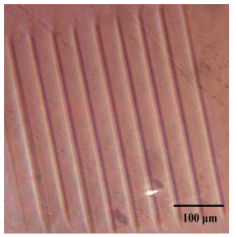
Type A
Elongated NPs

5-fold



(VIS)

Bare RTP

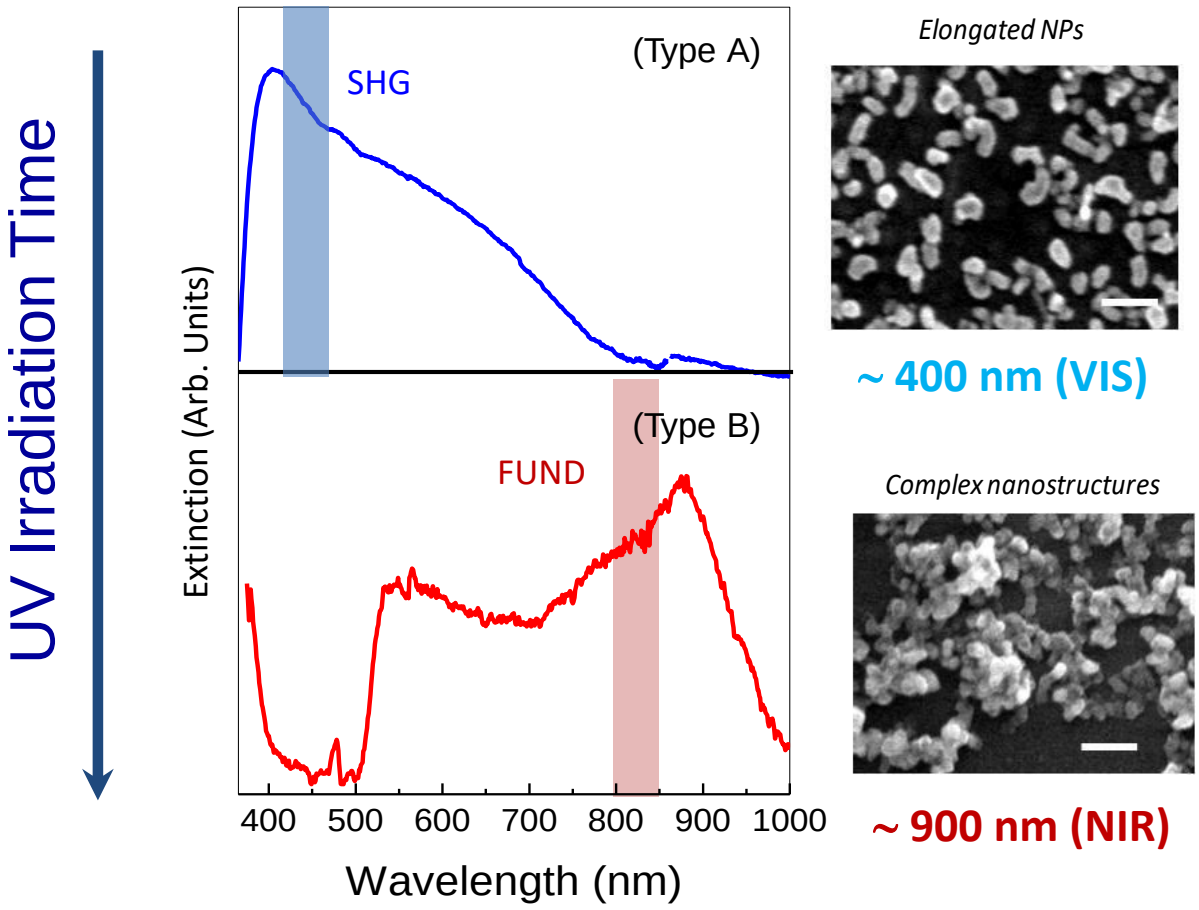


Pump: 840 nm
SHG: 420 nm

After metallization, the SHG signal is enhanced and remains localized at domain walls

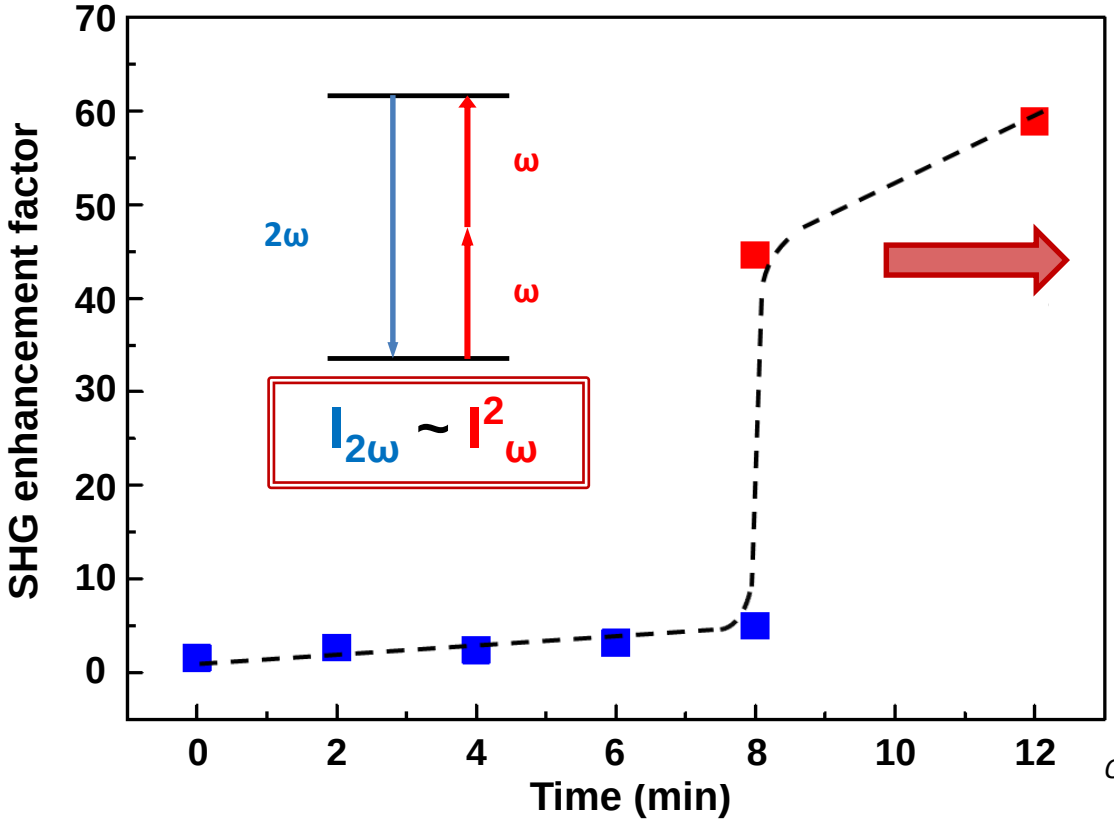
Plasmonic resonances in hybrid metal-RTP system

- **Spectral tuning of LSP resonances: from VIS to NIR**



SHG in hybrid metal-RTP systems

❖ **Enhancement factors**



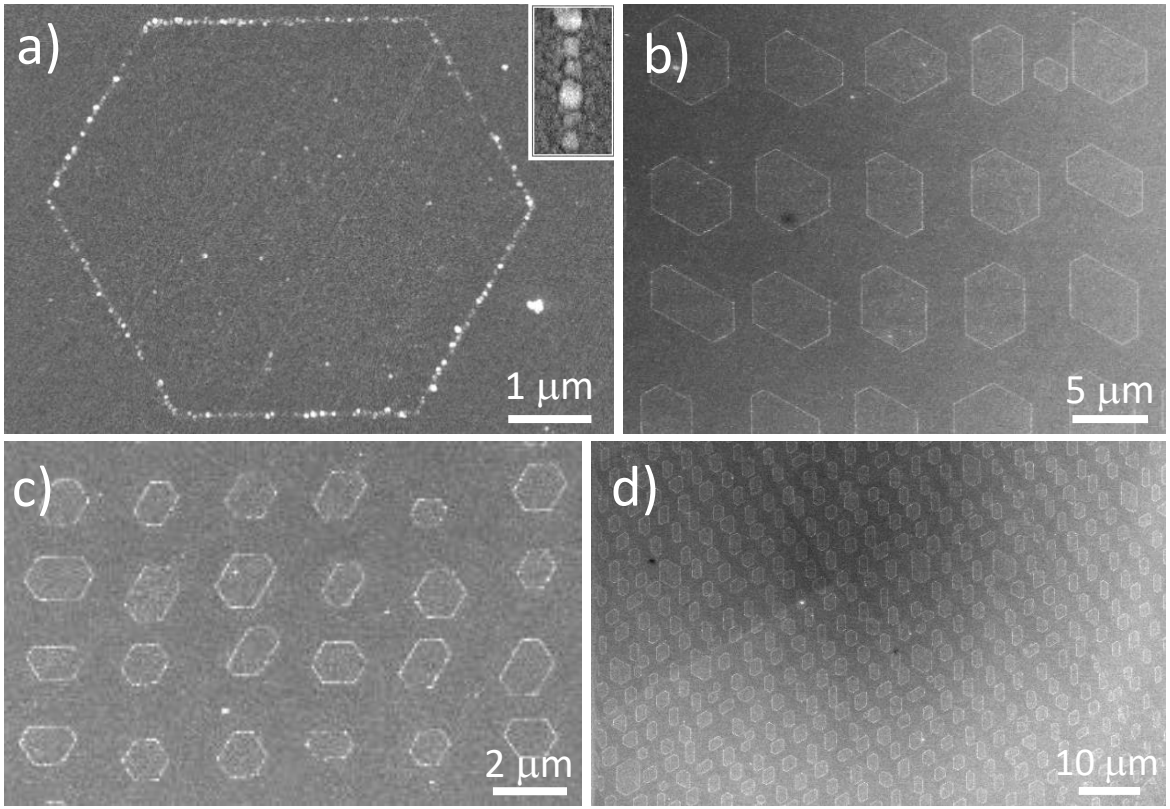
The local field is quadratically boosted

Resonant Pump

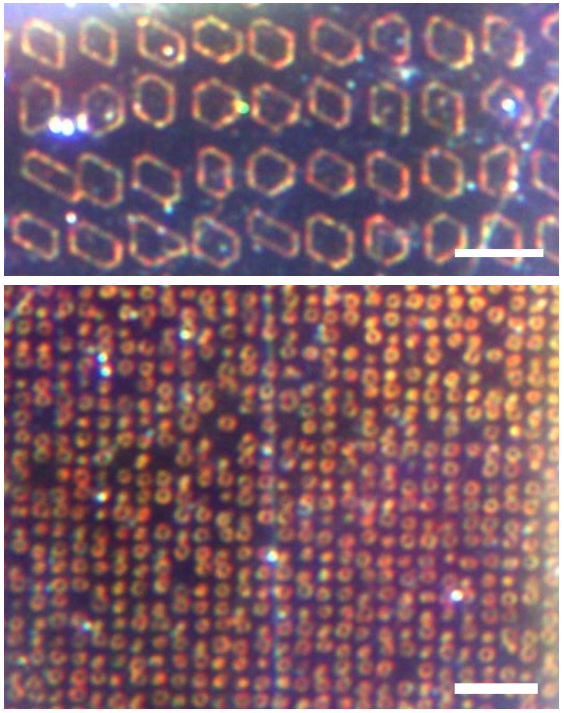
Optics Express **24,** 8491 (2016)

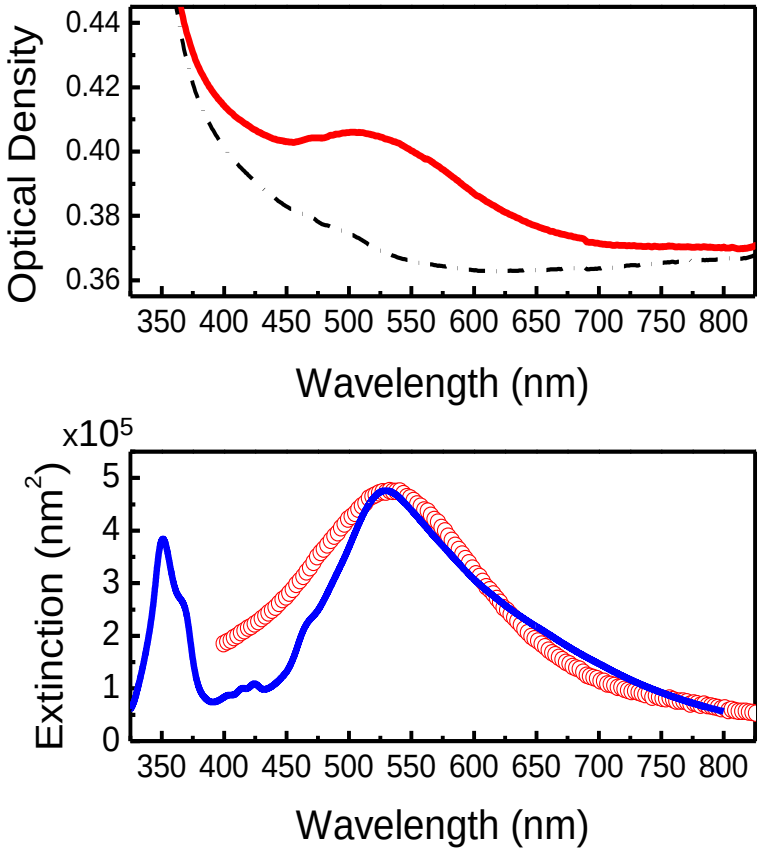
❑ Two well differentiated SHG enhancement regimes

Plasmonic necklaces for enhanced SHG



Plasmonic response of hexagonal necklaces



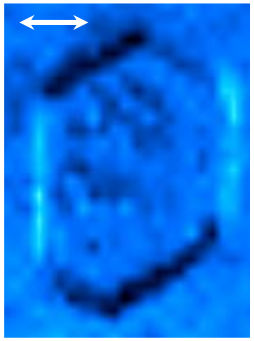


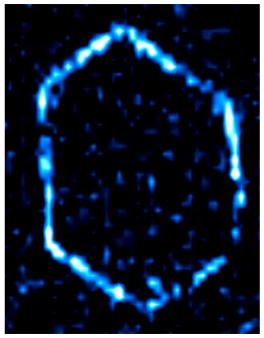
Broad radiative collective mode

Enhancement of SHG in hexagonal plasmonic necklaces

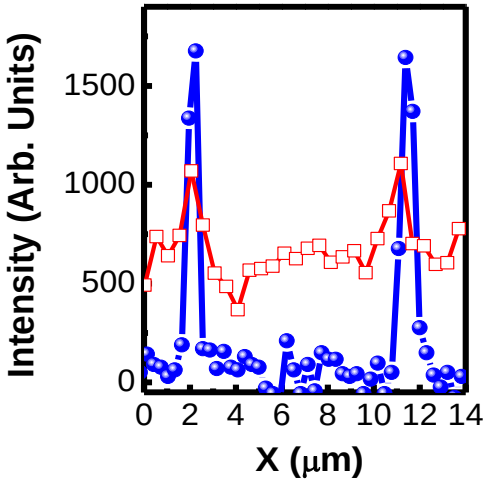
❑ Confocal SHG

Bare $LiNbO_3$

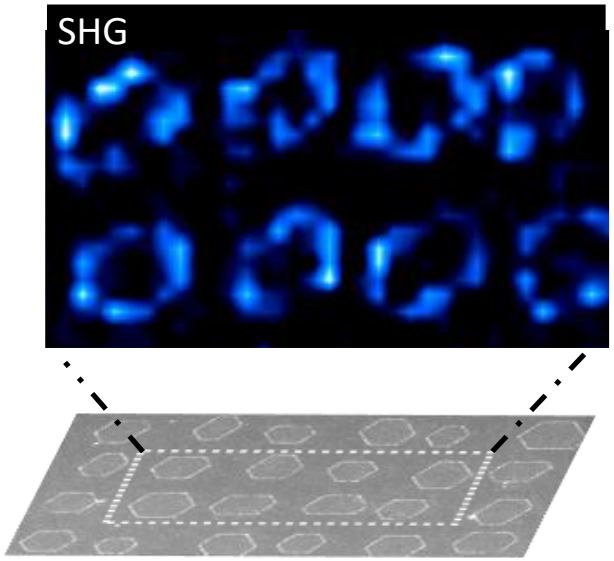


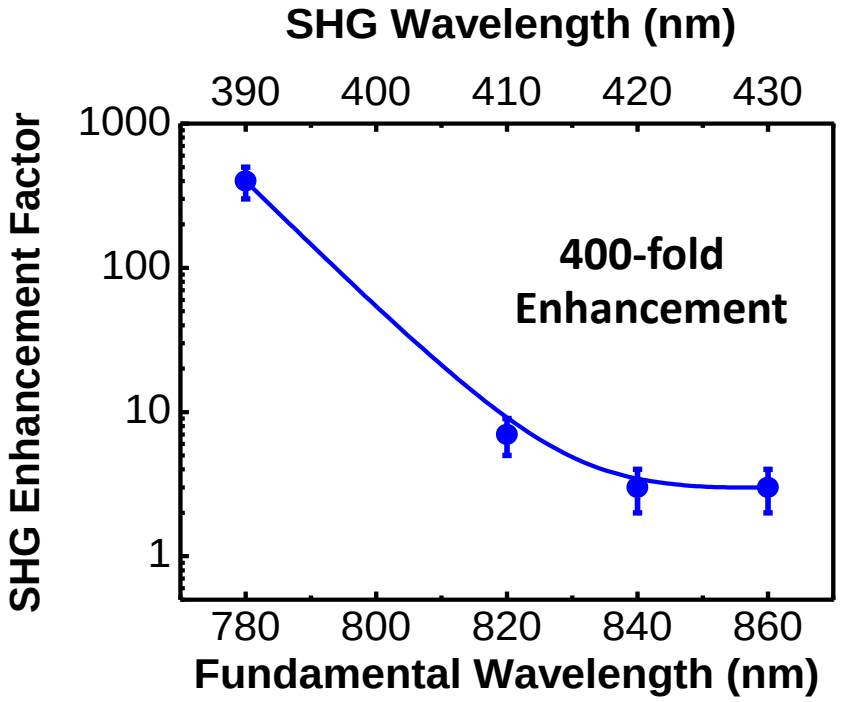


Hybrid Metal/ LNB

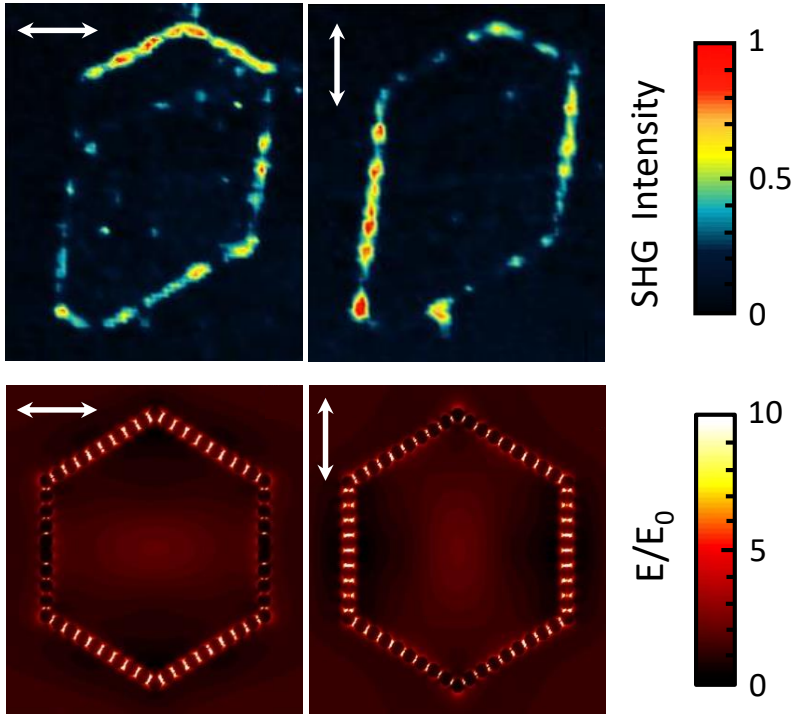


Enhancement of SHG in 2D arrays of hexagonal plasmonic necklaces





Polarization dependence of the enhanced SHG



Summary & Conclusions

1. Possibility of assembling of Ag NPs on ferroelectric domains and domain walls in 1D and 2D geometries

2. Nanolasing around the Ag NPs chains on Nd^{3+} :LNB

→ Strong reduction of the pump power at threshold (50 %)

→ The laser efficiency increases in a factor of 15

3. Plasmon enhanced SHG in hybrid metal-ferroelectrics

→ Up to 400-fold enhancement in the near UV spectral region

Development of novel coherent light sources operating at the nanoscale

Thank you!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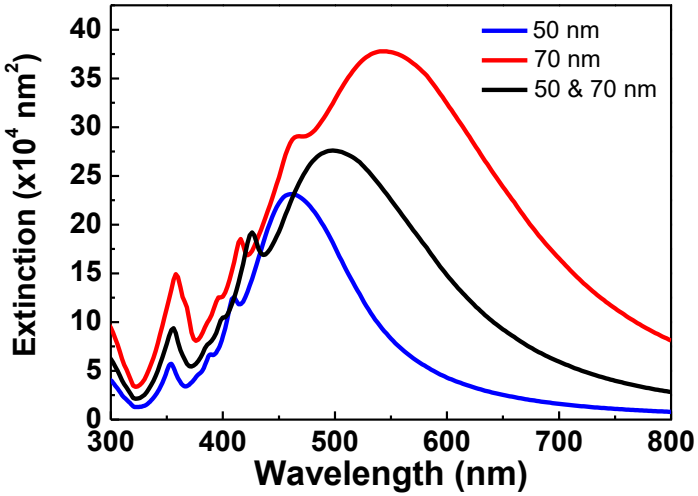
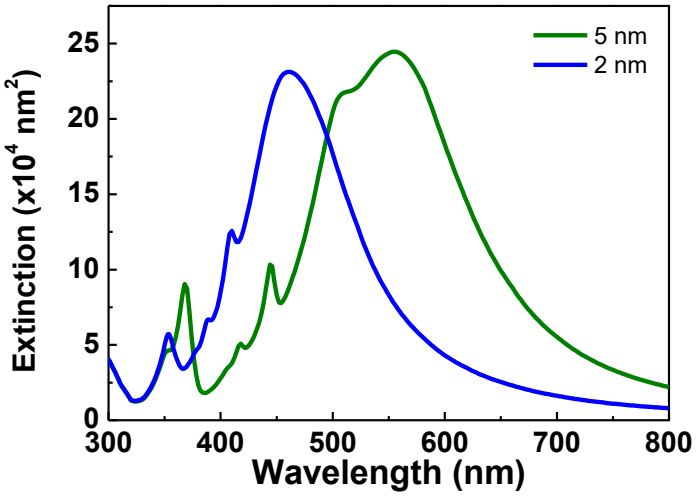
Linear Ag NPs chains

Absorption and extinction cross section

Effect of the NPs size



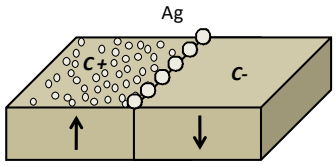
Effect of the inter-particle distance



- A red shift of the plasmonic resonance when the size of the silver NPs is increased
- Mixing NPs of different sizes results into modes located at intermediate wavelengths

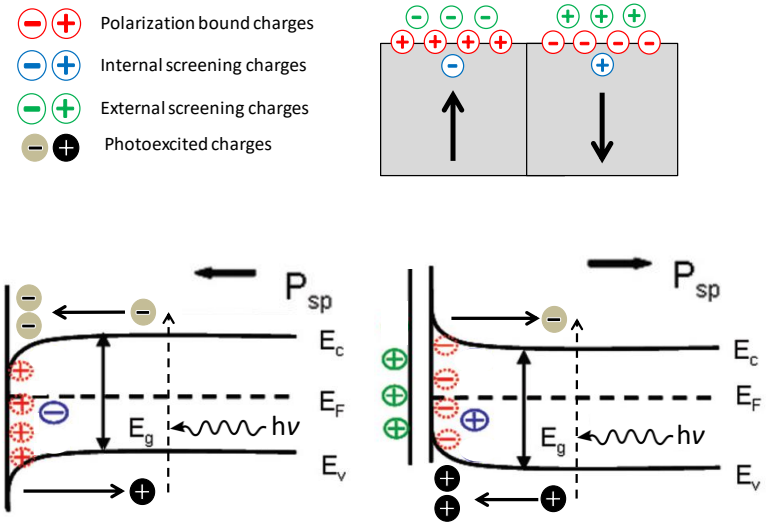
Decreasing the distance between metallic NPs blue-shifts the resonance

Fabrication process: Photoreduction of metallic nanostructures



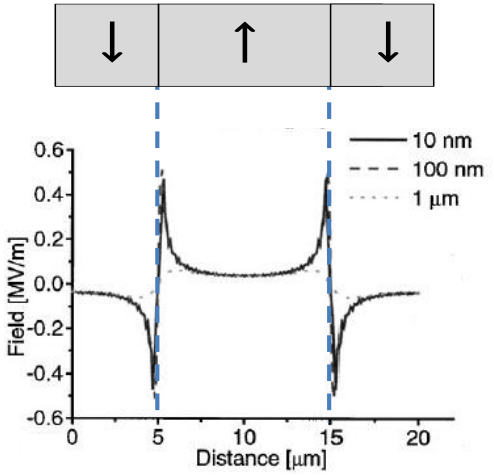
$LiNbO_3:Nd^{3+}$

Domain surface decoration mechanism



Band bending near the surface

Domain walls decoration mechanism



S.V. Kalinin and D.A. Bonnell *Phys. Rev. B,* **63** 125411 (2001)

Strong E_z component in the vicinity of the domain walls.