Nonlinear and Electro-Optic Metal-Oxides for Active Photonic Devices

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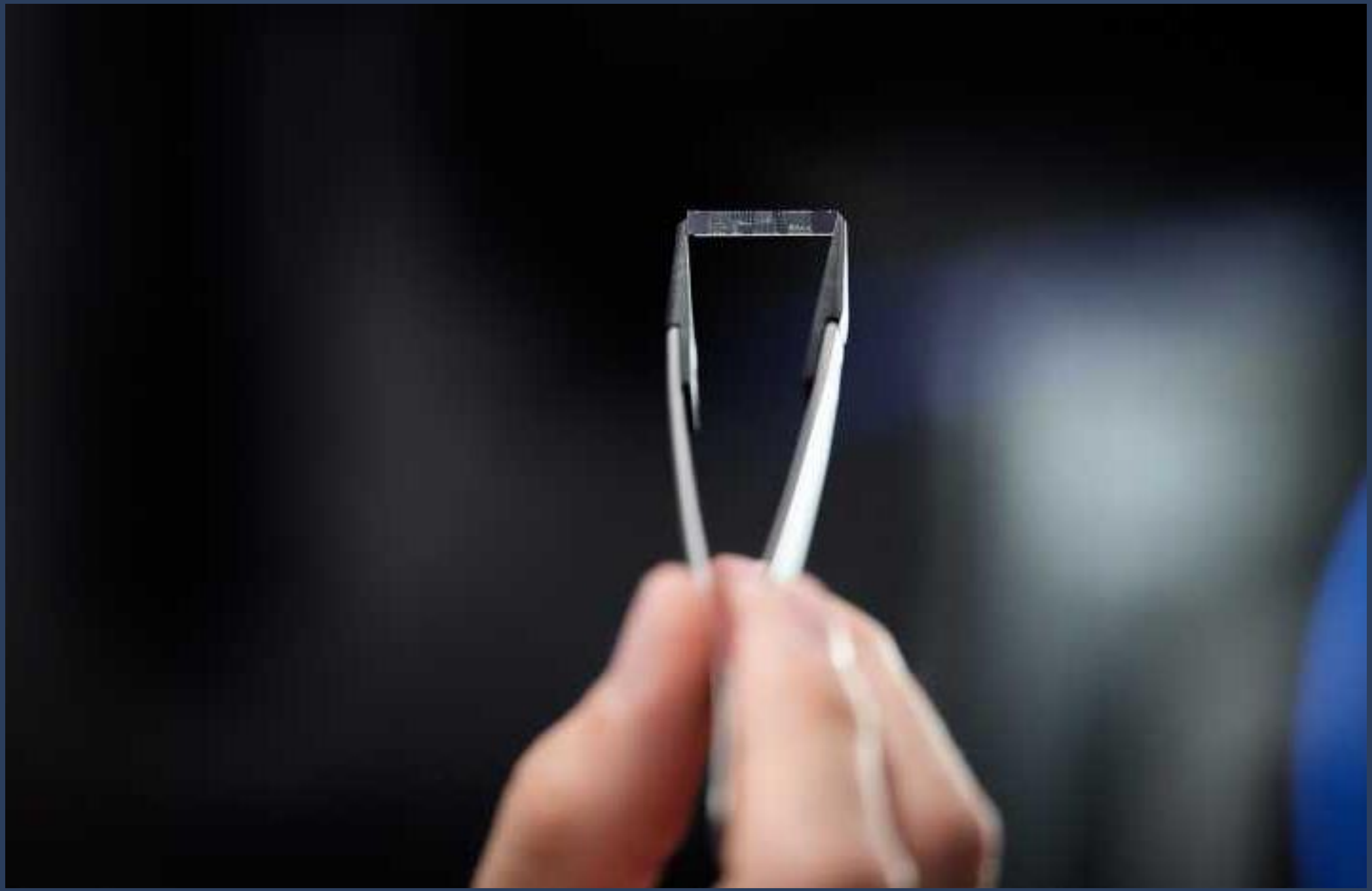

ETH Zurich / Stefan Weiss

The Optical Nanomaterial Group ONG

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FNSNF Fonds National Suisse / Schweizerischer Nationalfonds / Fondo Nazionale Svizzero / Swiss National Science Foundation

State Secretariat for Education, Research and Innovation SERI – Swiss Space Office

swiss space center

BRIDGE

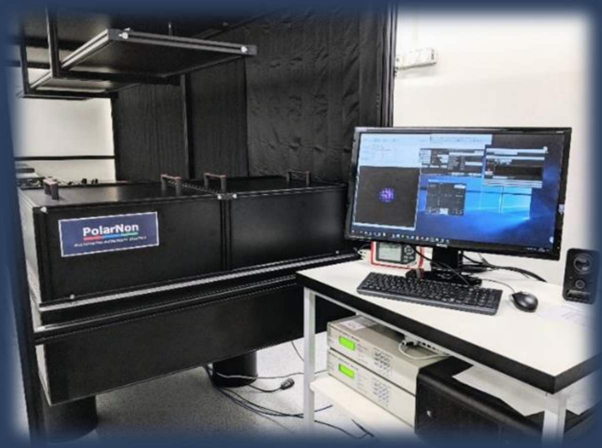
Marie Curie Actions

erc European Research Council – Established by the European Commission

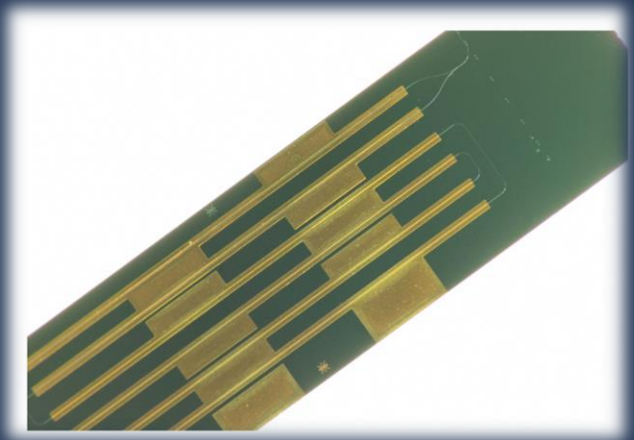
Quadratic $\chi^{(2)}$ materials as toolbox at small scale

Our focus

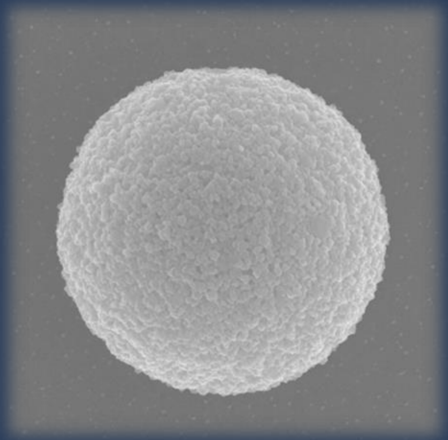
- Nonlinear and electro-optic signals at the nanoscale
- Multipolar imaging tools to study nanomaterials
- Nanofabrication with unconventional materials
- Miniaturized multifunctional photonic devices



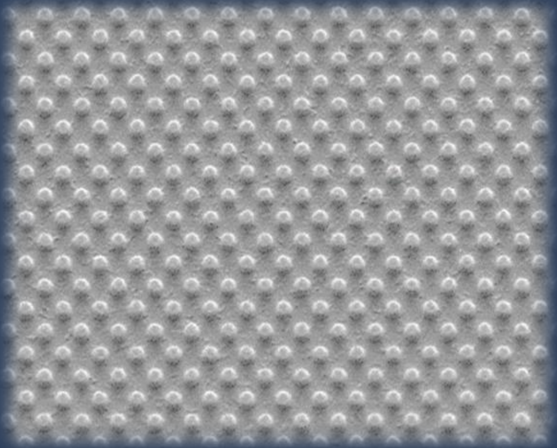
Nonlinear Imaging



Integrated Photonics



Random Media



Metastructures

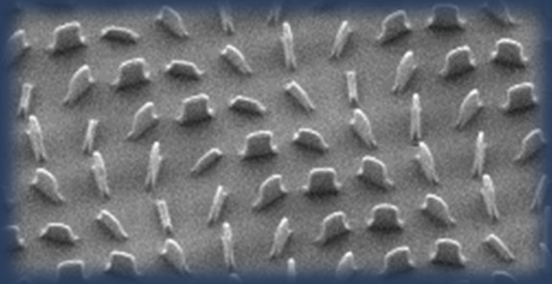
Outline

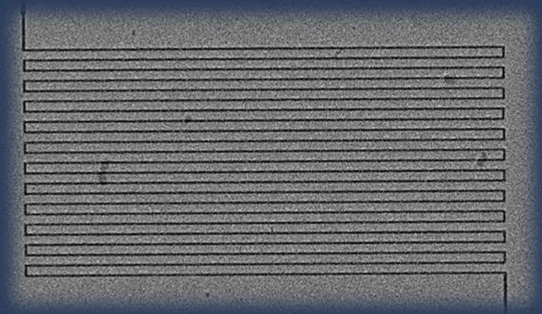
Miniaturizing $\chi^{(2)}$ materials

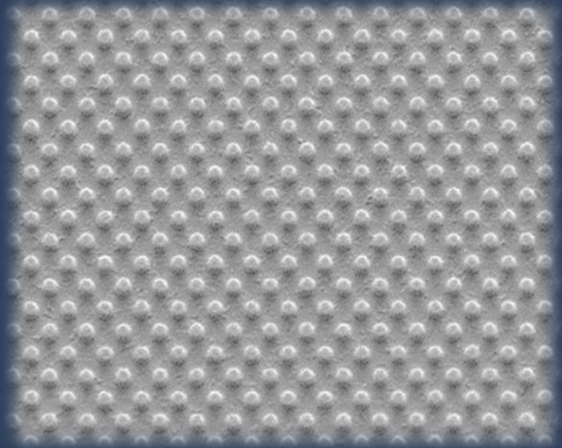
Nonlinear or electro-optic metasurfaces

- Pulsed laser deposited $BaTiO_3$
- FIB and spin coated nanoparticles
- Sol-gel nanoimprinted metalens

Miscellaneous photonic structures



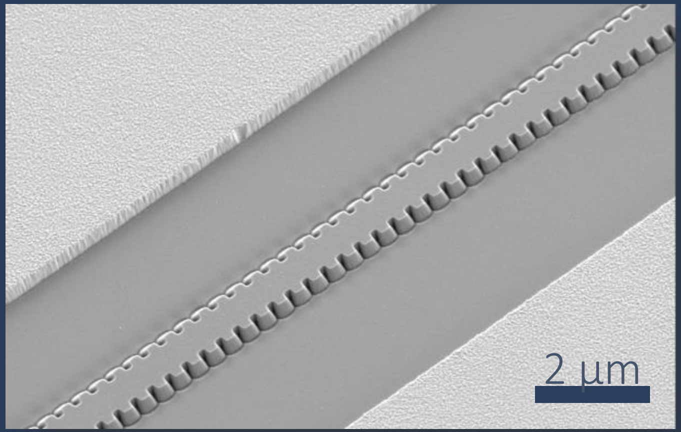




Why miniaturizing quadratic optical materials?

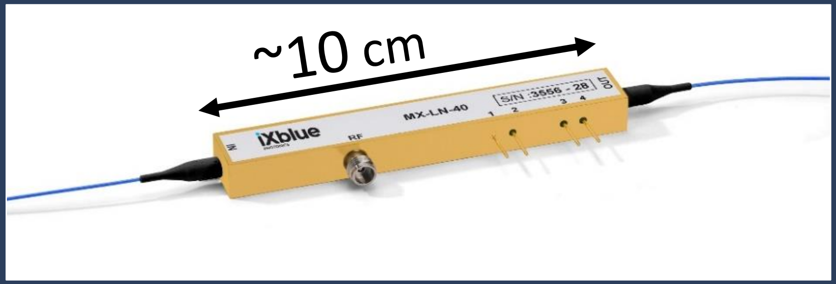
Telecommunication

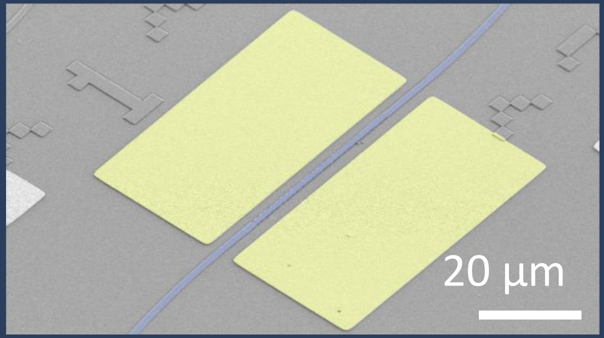
Modulators

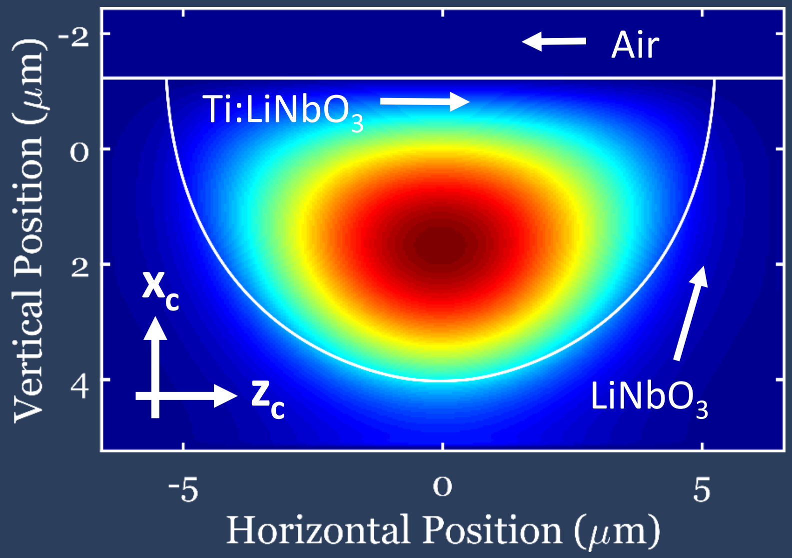


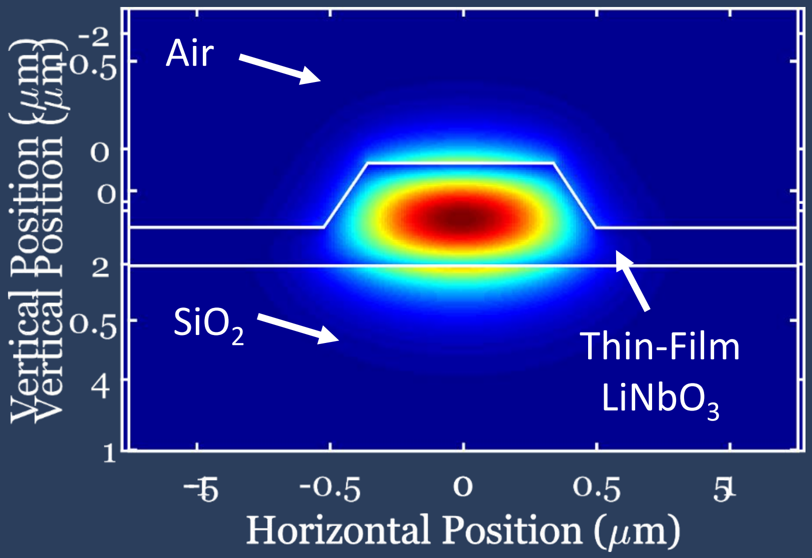
Reig Escalé, et al. OL 43(7) 2018
Pohl, et al. IEEE PTL 33 (2) 2020

Traditional vs integrated modulator design









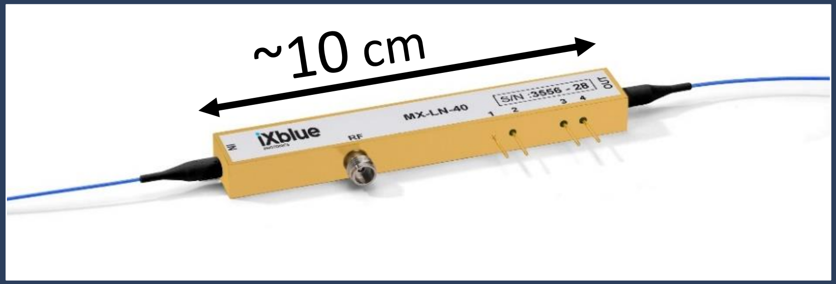
Mode Area > 30 µm²

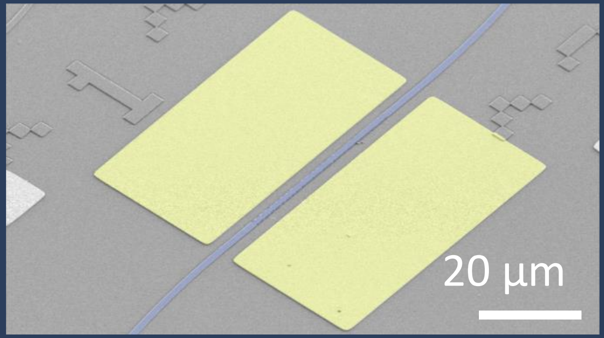
<40 Gbit/s

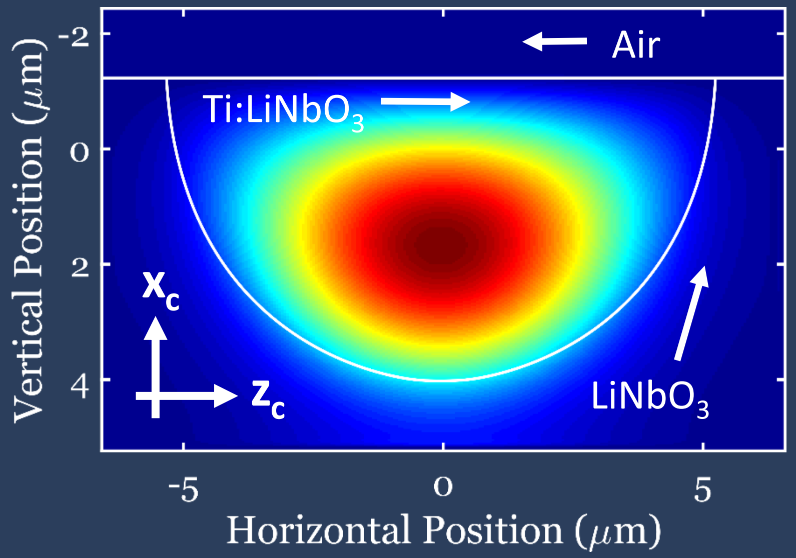
Mode Area < 1 µm²

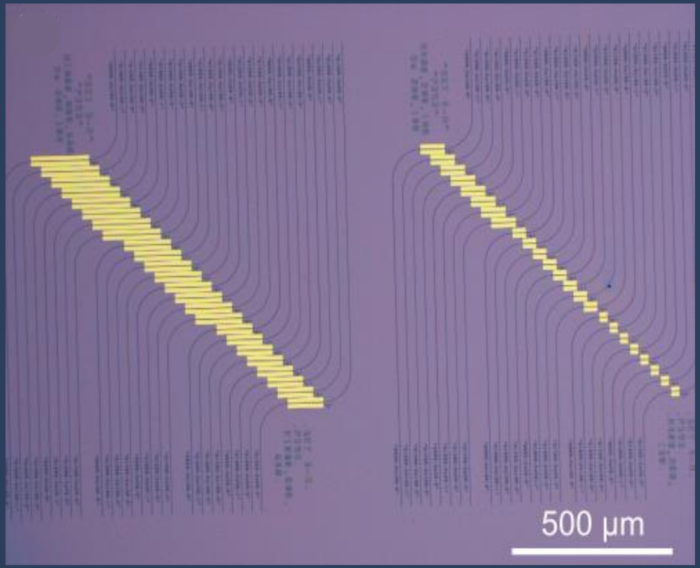
100 Gbit/s

Traditional vs integrated modulator design









Mode Area > 30 µm²

<40 Gbit/s

Mode Area < 1 µm²

100 Gbit/s

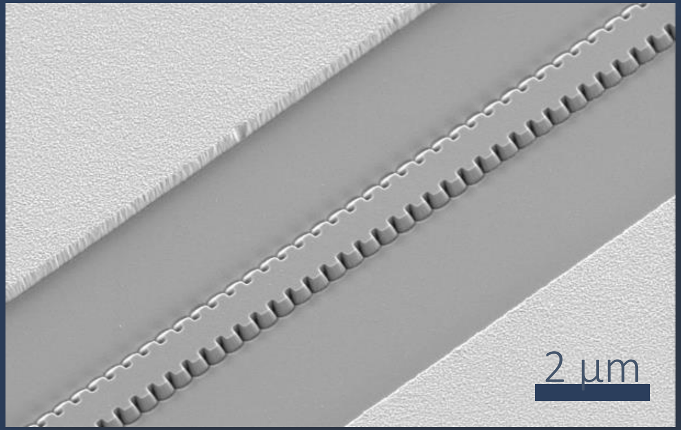
Parallelization

Why miniaturizing quadratic optical materials?

Telecommunication

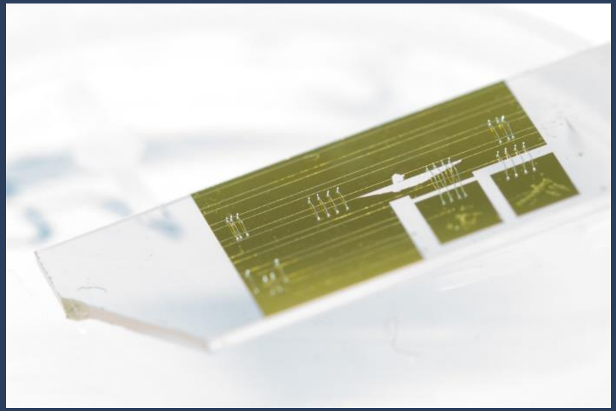
Sensor

Modulators



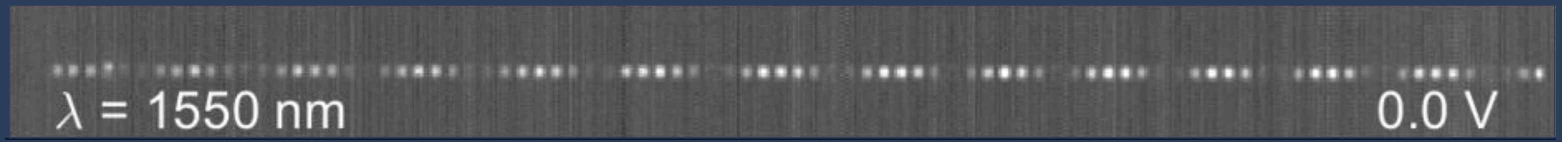
Reig Escalé, et al. OL 43(7) 2018
Pohl, et al. IEEE PTL 33 (2) 2020

Spectrometer

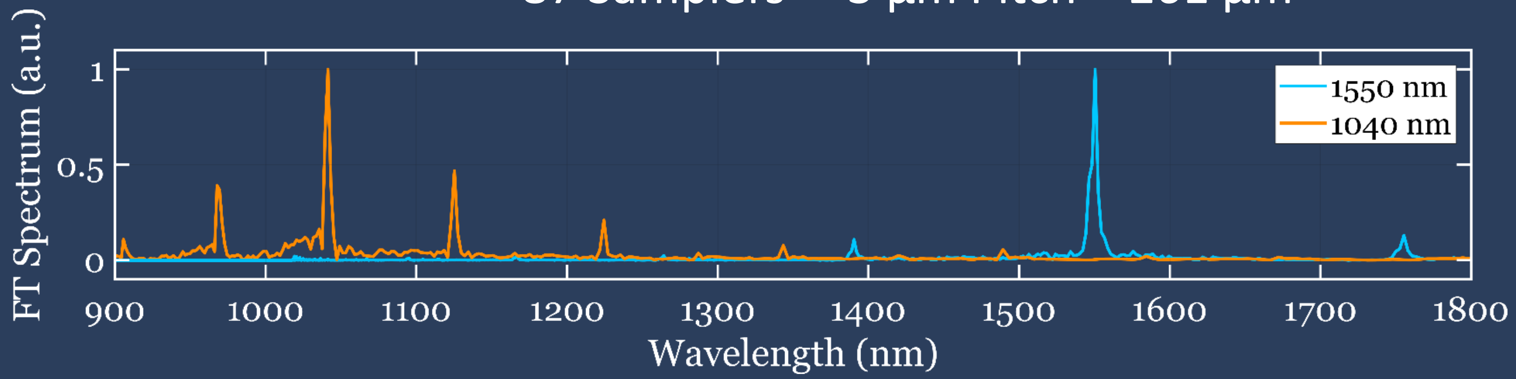


Pohl et al. Nature Photonics 14 (1) 2020

Lithium Niobate Nano Spectrometer



87 Samplers × 3 µm Pitch = 261 µm



$\Delta\lambda$ ~ 6 nm at λ = 1550 nm

>500 nm bandwidth with <20 V

λ_{SM} ~ 1000-1800 nm

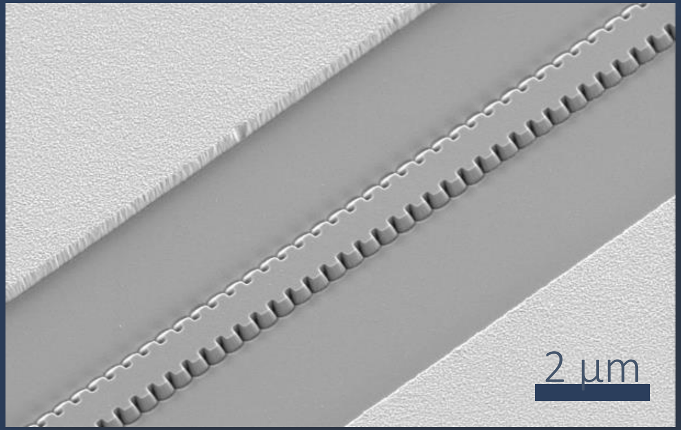
Why miniaturizing quadratic optical materials?

Telecommunication

Sensor

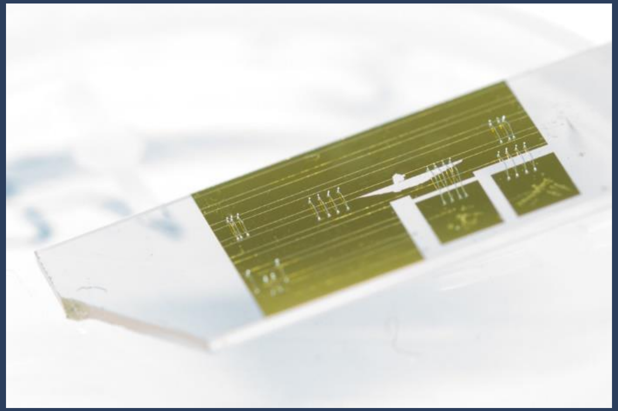
Source

Modulators



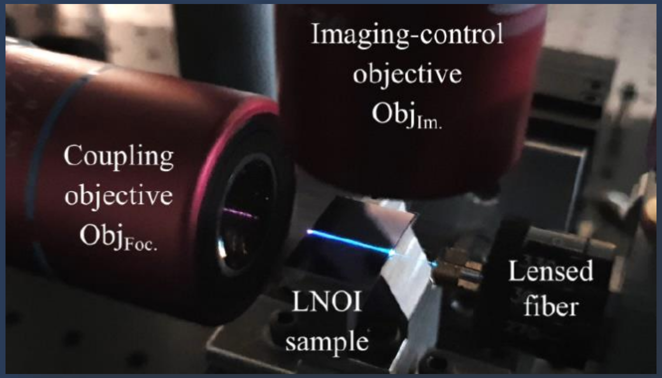
Reig Escalé, et al. OL 43(7) 2018
Pohl, et al. IEEE PTL 33 (2) 2020

Spectrometer



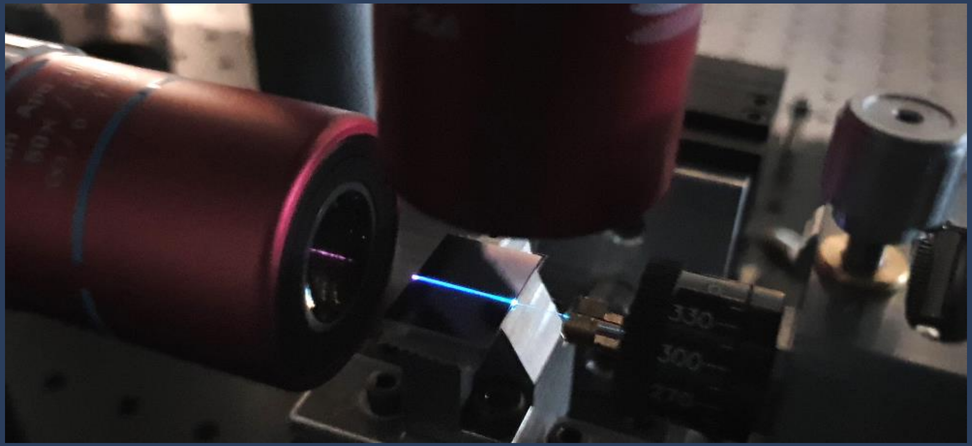
Pohl et al. Nature Photonics 14 (1) 2020

Supercontinuum

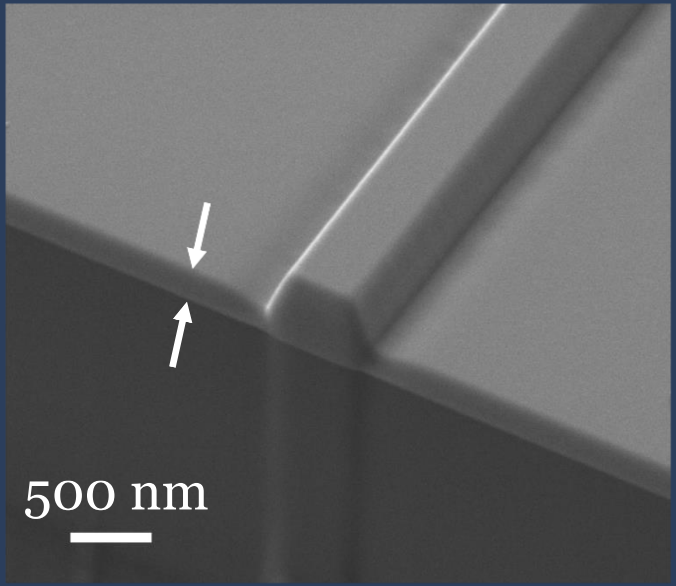


Reig Escalé, et al. APL Photonics 5 (12) 2020

Supercontinuum generation in LNOI

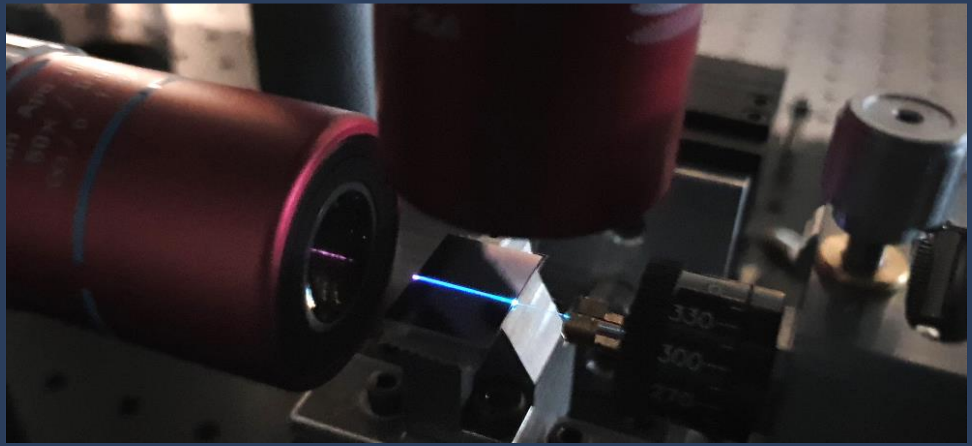


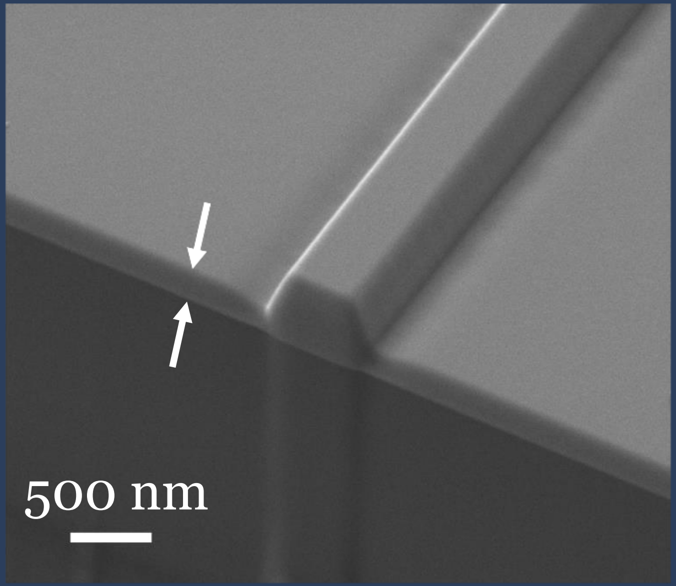
851 THz (352 nm)
in a 14-mm long rib waveguide

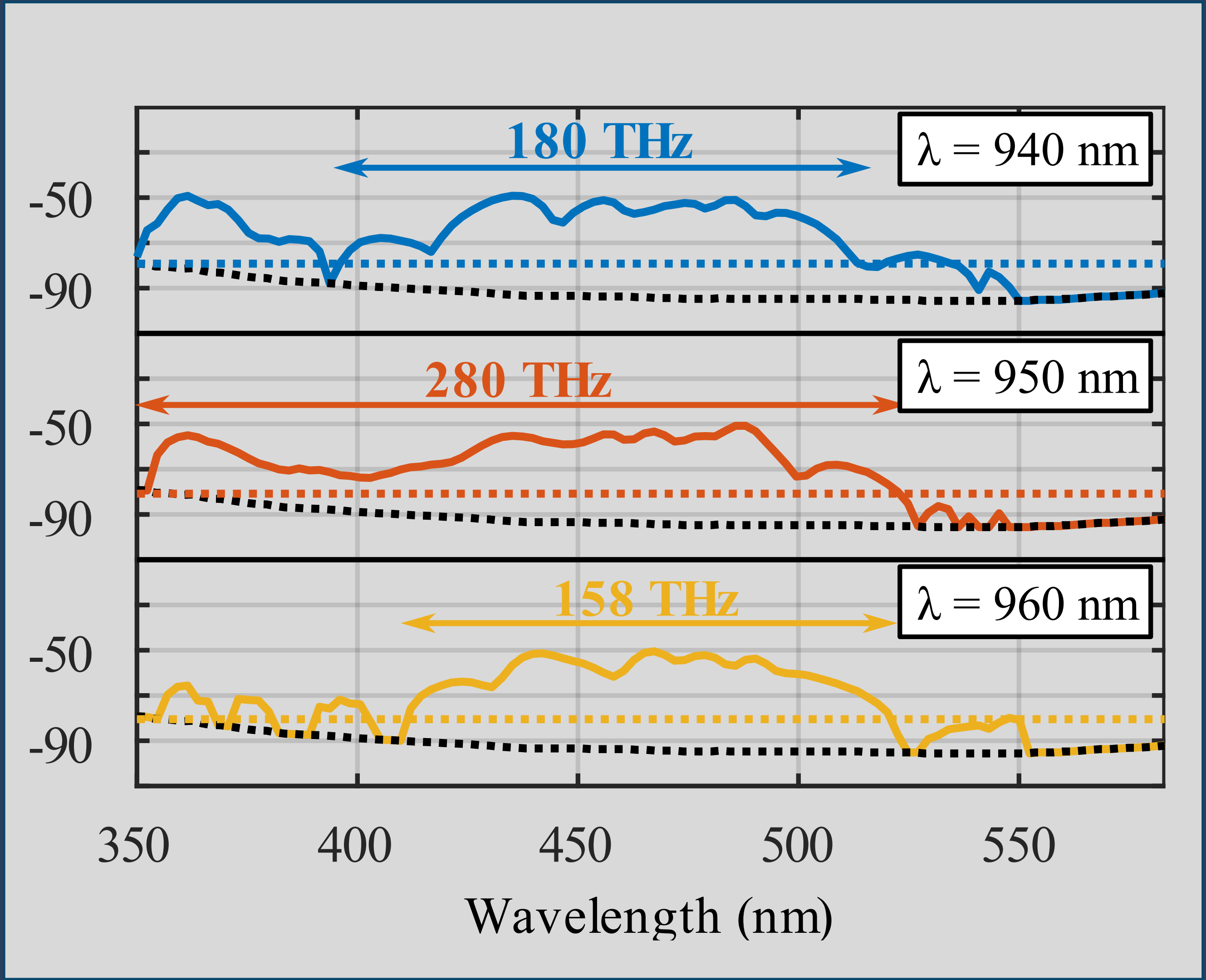


Reig Escalé et al. APL Photonics, 5, 12, 2020

Supercontinuum generation in LNOI

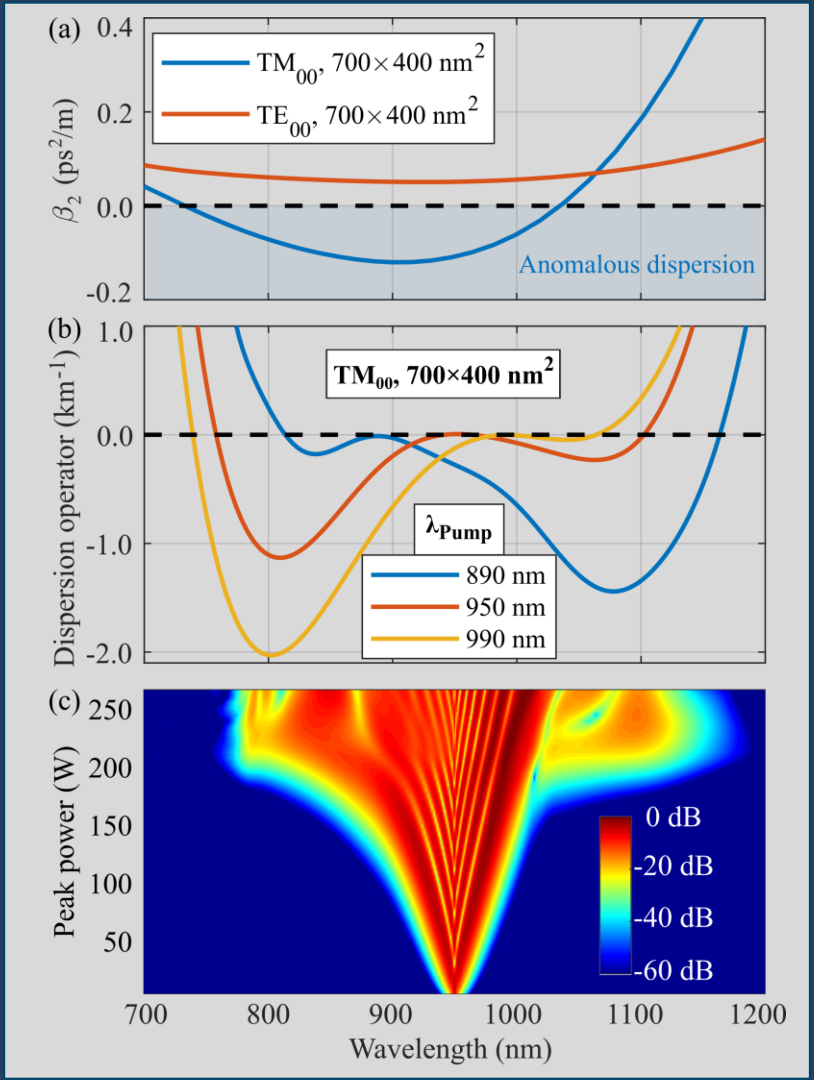


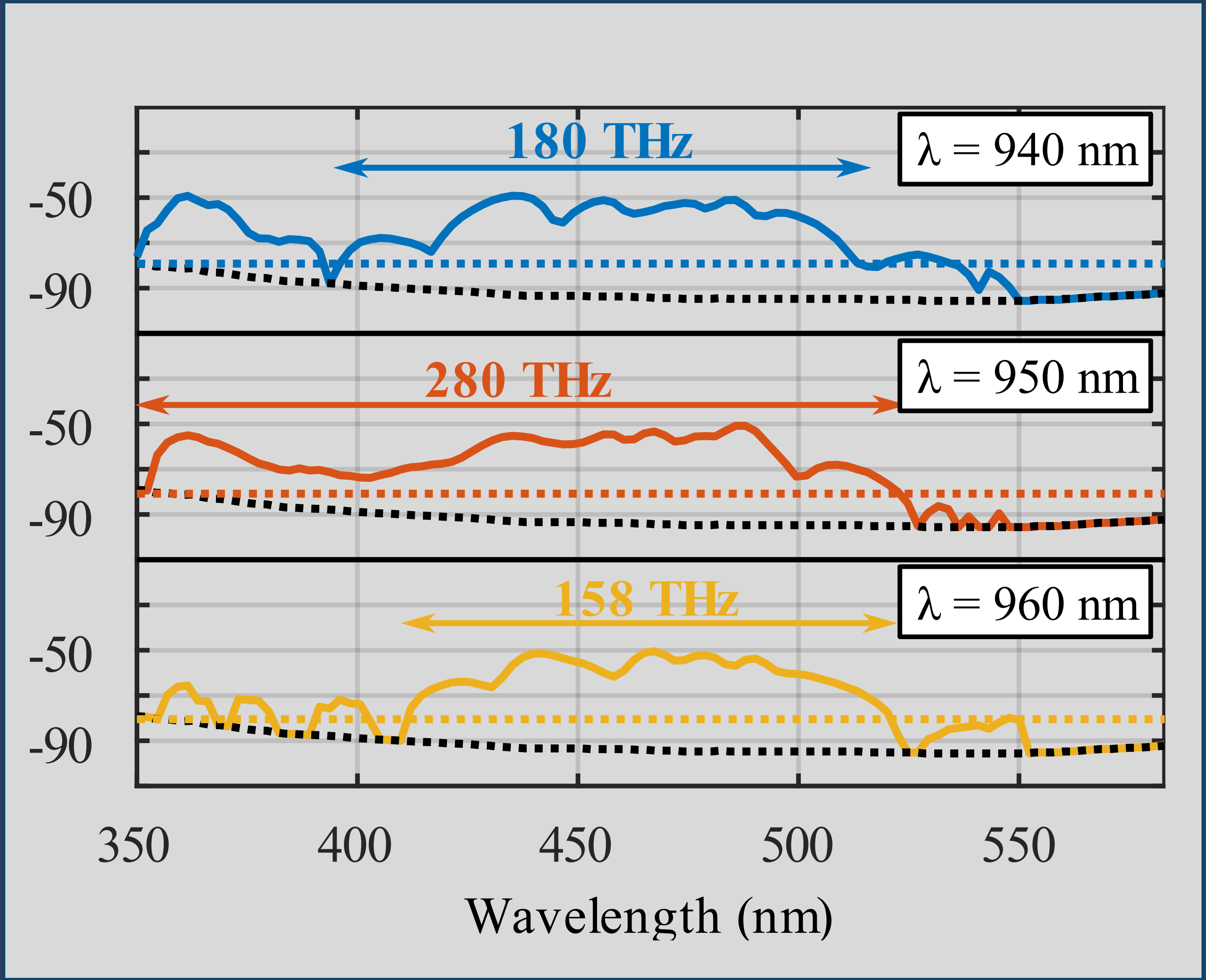




Reig Escalé et al. APL Photonics, 5, 12, 2020

Supercontinuum generation in LNOI

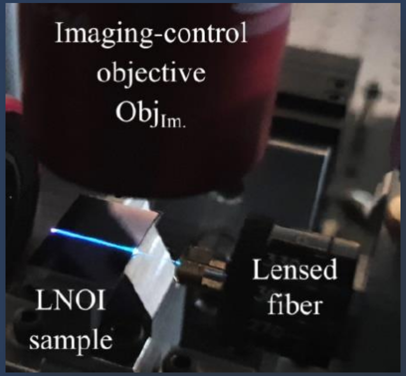




Reig Escalé et al. APL Photonics, 5, 12, 2020

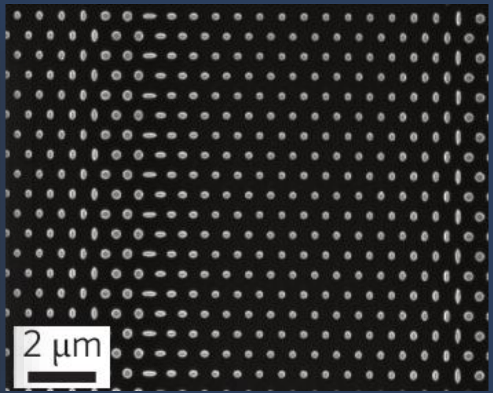
Why miniaturizing quadratic optical materials?

Sources



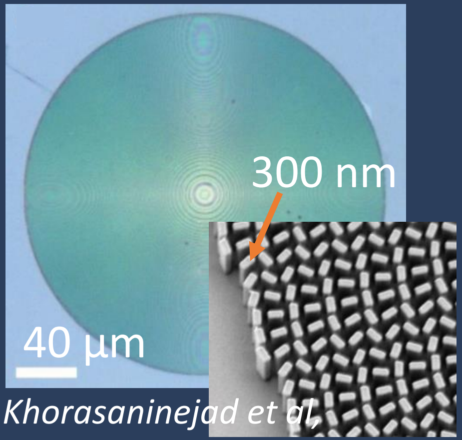
Reig Escalé, et al. APL Photonics 5 (12) 2020

Polarizers, splitters



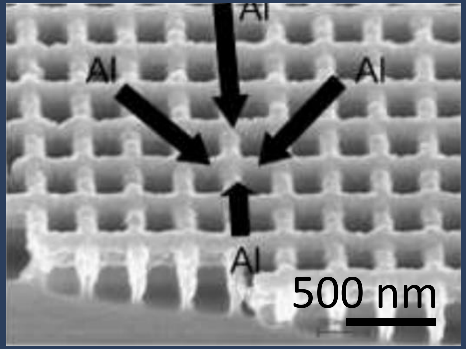
Arbabi et al, Nat. Nanotech. 10, 2015

Lenses



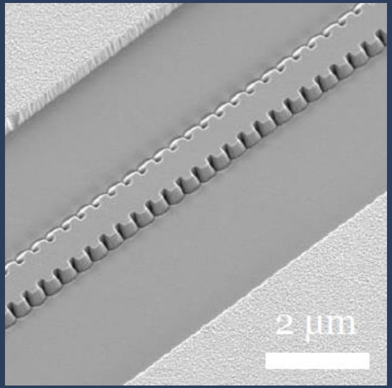
Khorasaninejad et al, Science 352, 2016

Filters



Li et al, Opt Exp 18(2), 2010

Guides

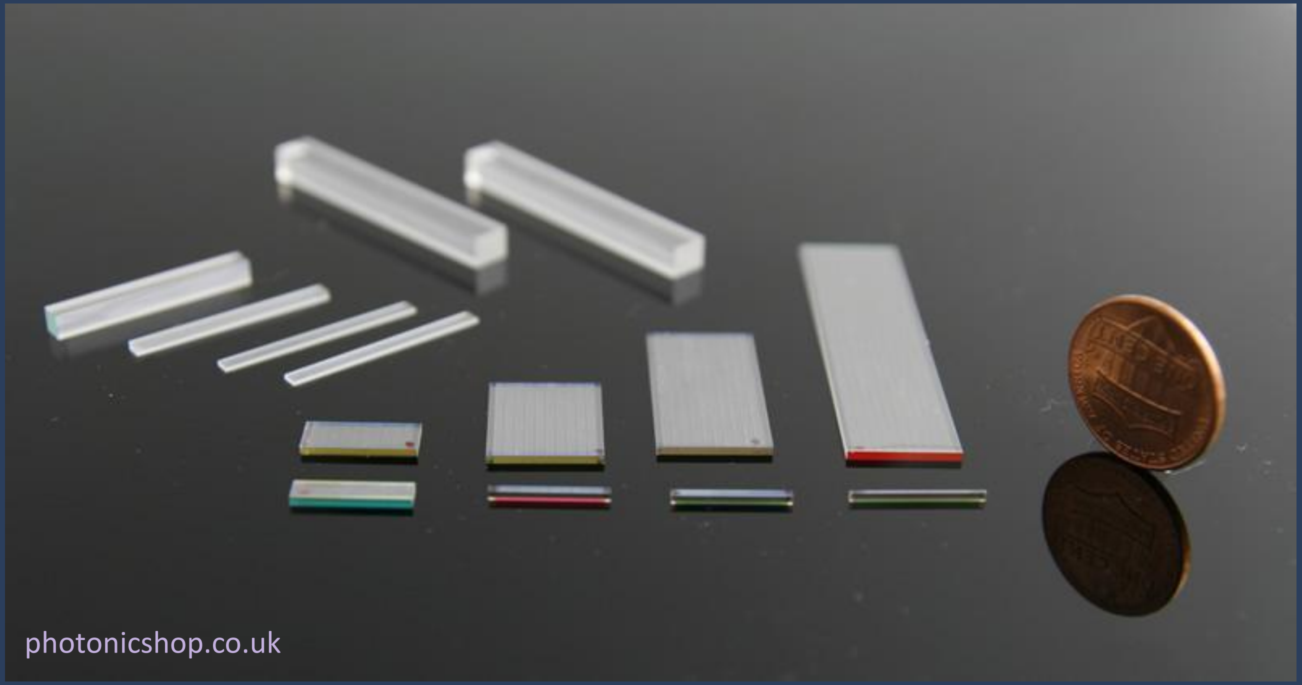


Reig Escalé et al, Opt Lett 43, 2018

www.edmundoptics.de, www.thorlabs.com

Optical material with multifunctions

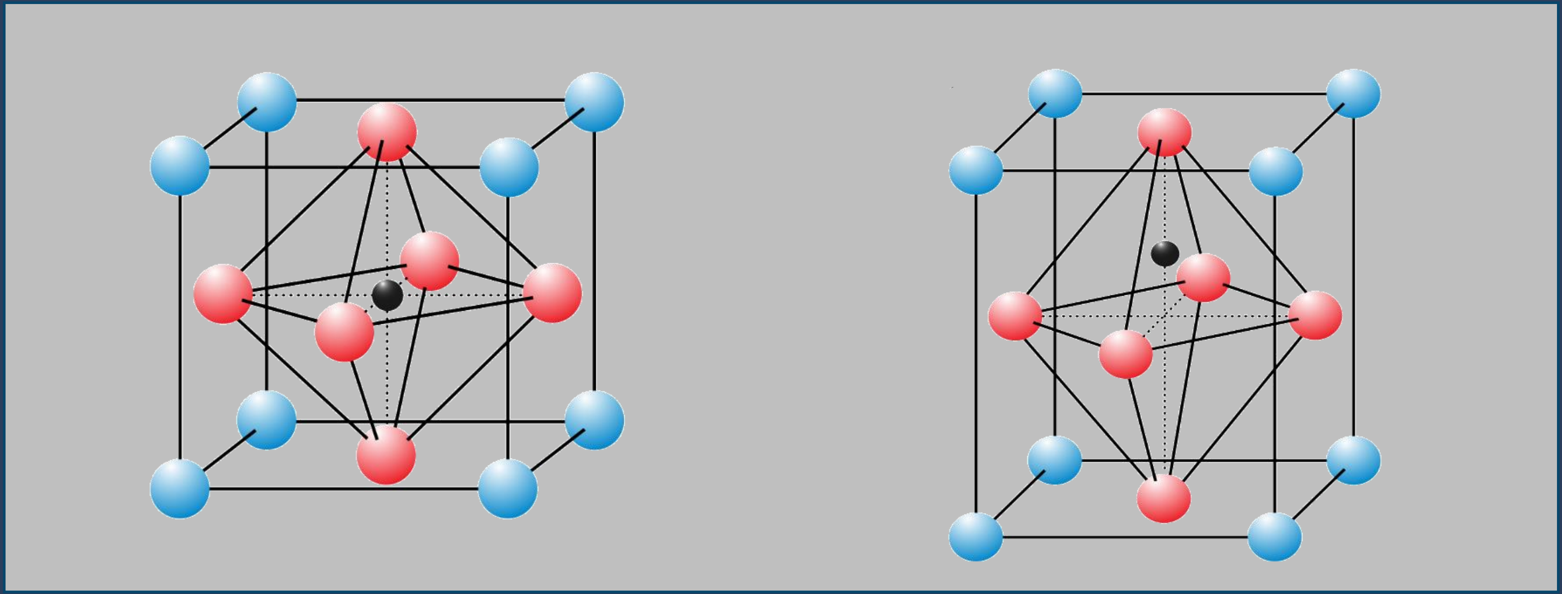
Bulk crystal



Lithium Niobate ($LiNbO_3$)
Barium Titanate ($BaTiO_3$)

Quadratic $\chi^{(2)}$ materials

Centrosymmetric | Non-Centrosymmetric

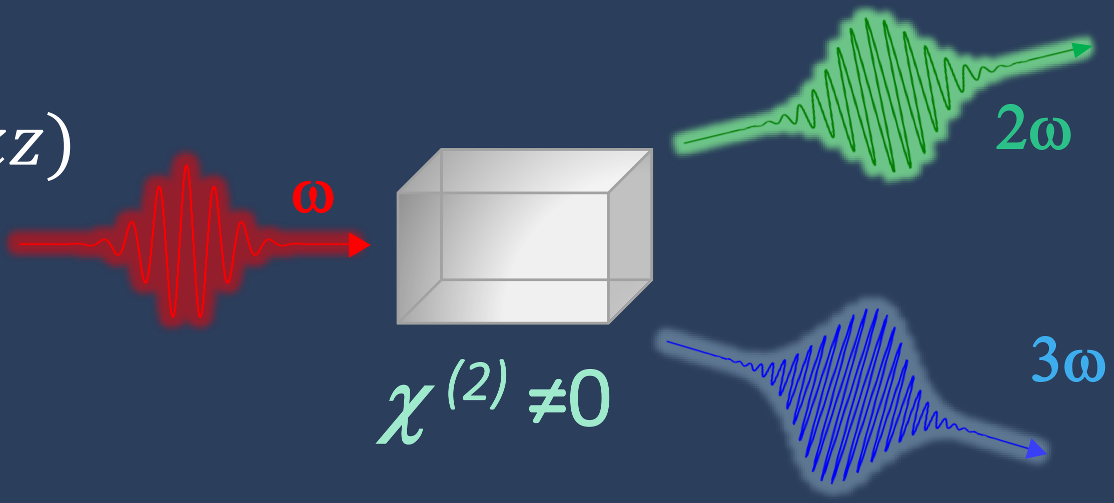


Silicon
Diamond

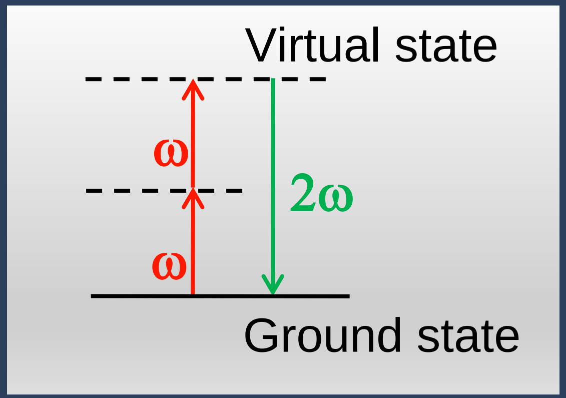
SiO_2 (crystalline quartz)
Gallium arsenide (GaAs)
Barium titanate ($BaTiO_3$)
Lithium niobate ($LiNbO_3$)

Quadratic $\chi^{(2)}$ materials

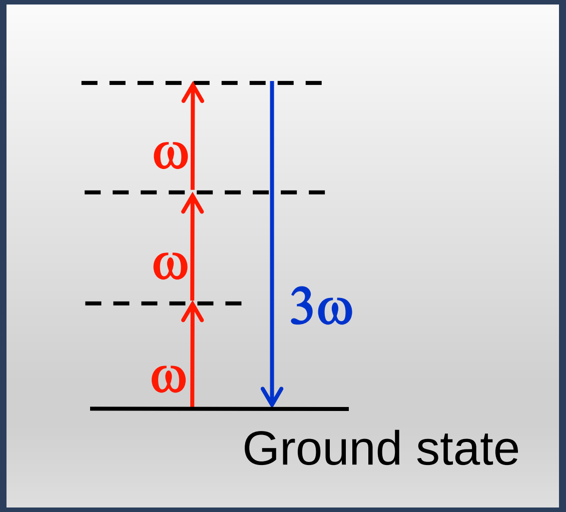
$$E(t) = A\cos(\omega t + kz)$$



Second harmonic generation



Third harmonic generation



Induced Polarization

$$\vec{P} = \varepsilon_0 \chi^{(1)} \vec{E}$$

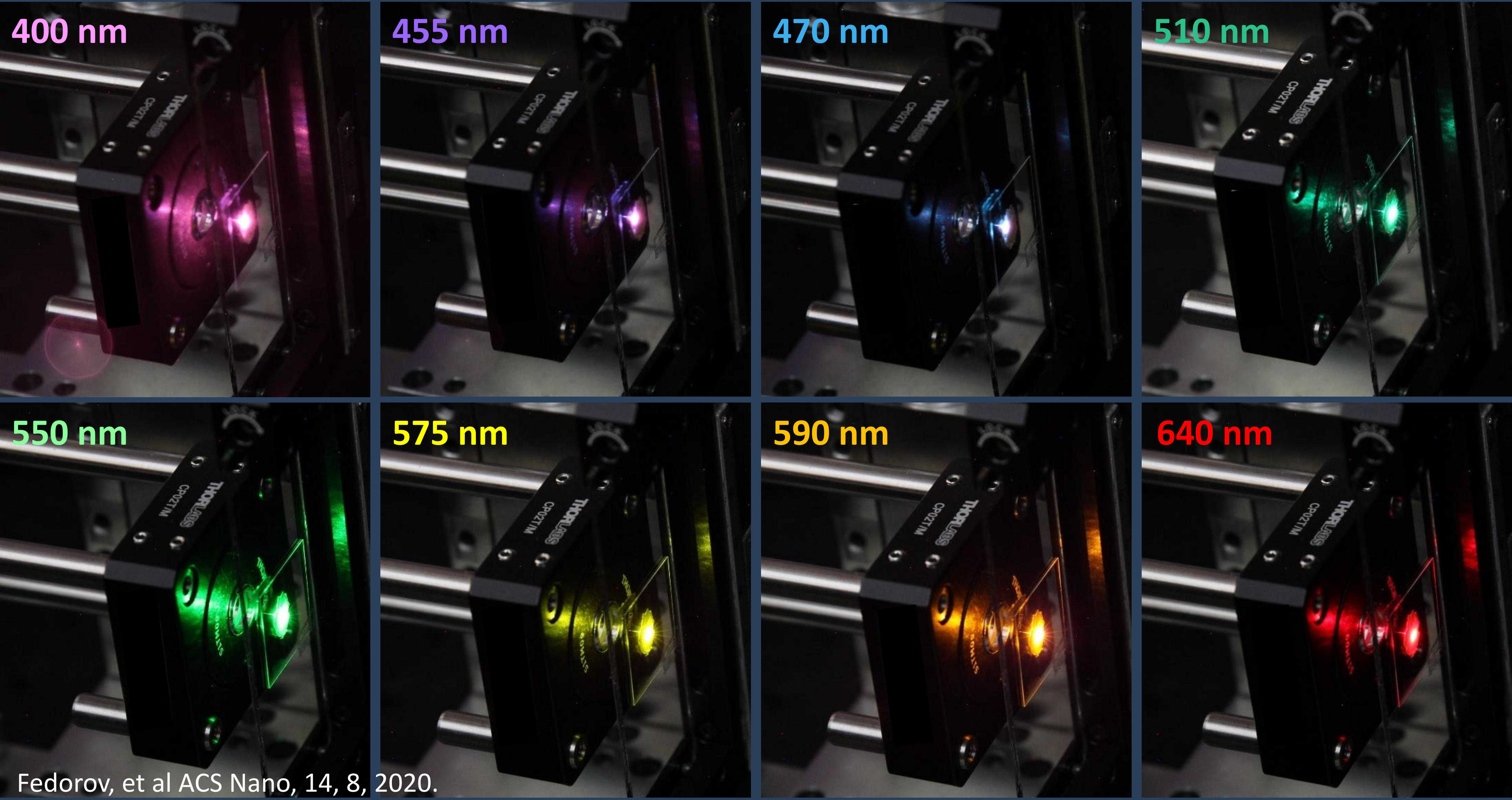
Electric susceptibility

Second order susceptibility

Third order susceptibility

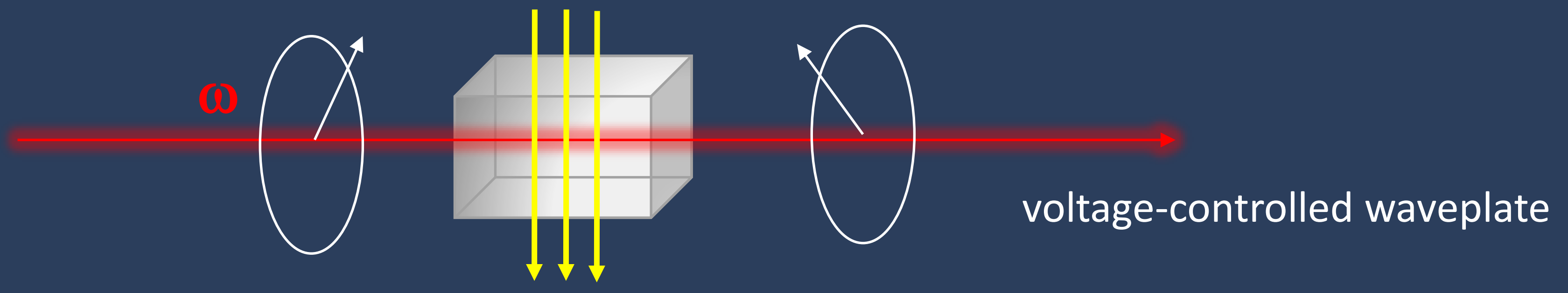
ε_0 dielectric constant of the vacuum

Franken et al., Phys Rev Lett, 7,4 1961



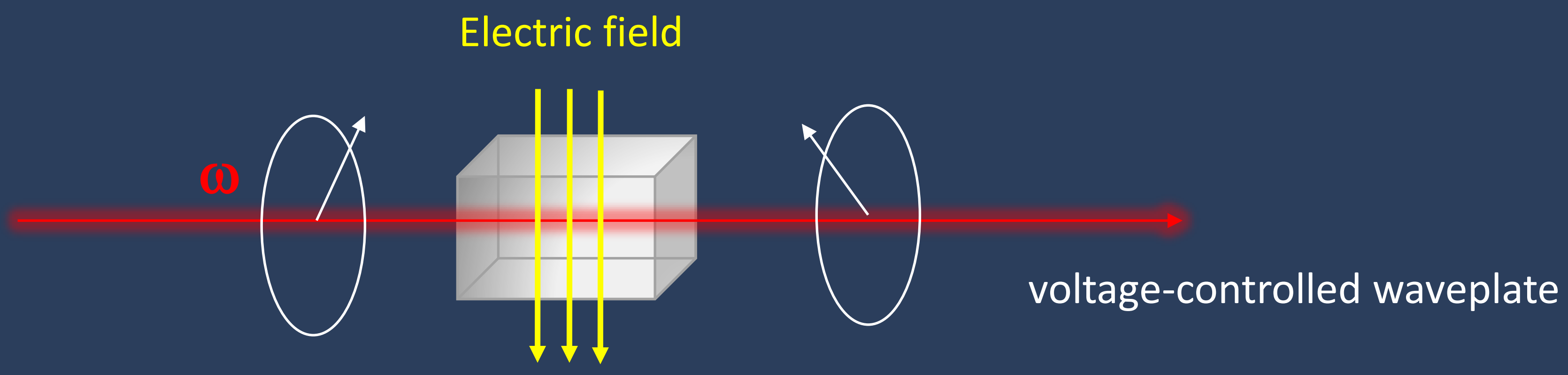
Fedorov, et al ACS Nano, 14, 8, 2020.

More properties of quadratic $\chi^{(2)}$ materials: $LiNbO_3$



- **Electro-optic**

More properties of quadratic $\chi^{(2)}$ materials: $LiNbO_3$



- **Electro-optic**

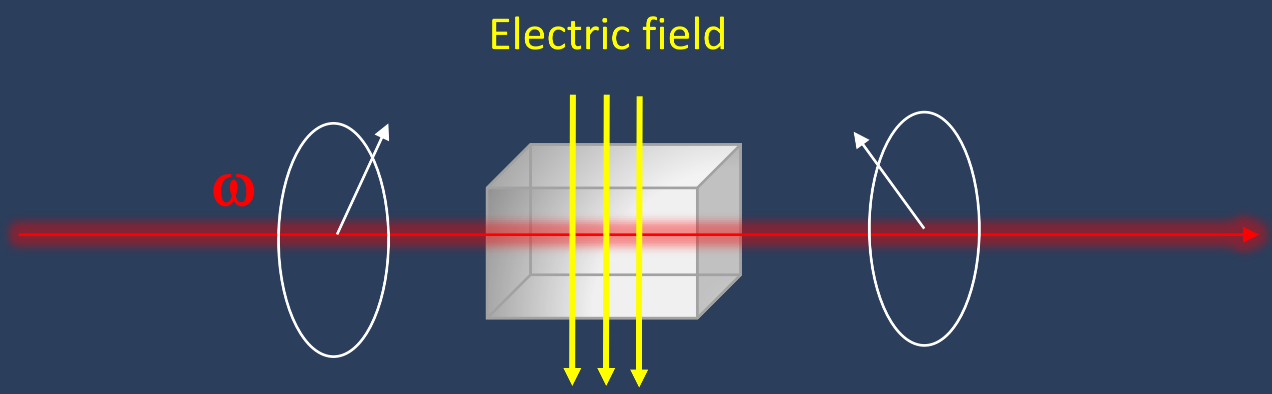
Change in the refractive index linearly proportional to the electric field

Electro-optic tensor $\chi^{(2)}$ of $LiNbO_3$

$$\begin{pmatrix} \Delta(1/n^2)_1 \\ \Delta(1/n^2)_2 \\ \Delta(1/n^2)_3 \\ \Delta(1/n^2)_4 \\ \Delta(1/n^2)_5 \\ \Delta(1/n^2)_6 \end{pmatrix} = \begin{pmatrix} 0 & -3.4 & 8.6 \\ 0 & 3.4 & 8.6 \\ 0 & 0 & 30.8 \\ 0 & 28 & 0 \\ 28 & 0 & 0 \\ -3.4 & 0 & 0 \end{pmatrix} \cdot \begin{pmatrix} E_x \\ E_y \\ E_z \end{pmatrix}$$

d_{33}= 30.8 pm/V

More properties of quadratic $\chi^{(2)}$ materials: $LiNbO_3$



- **Electro-optic**

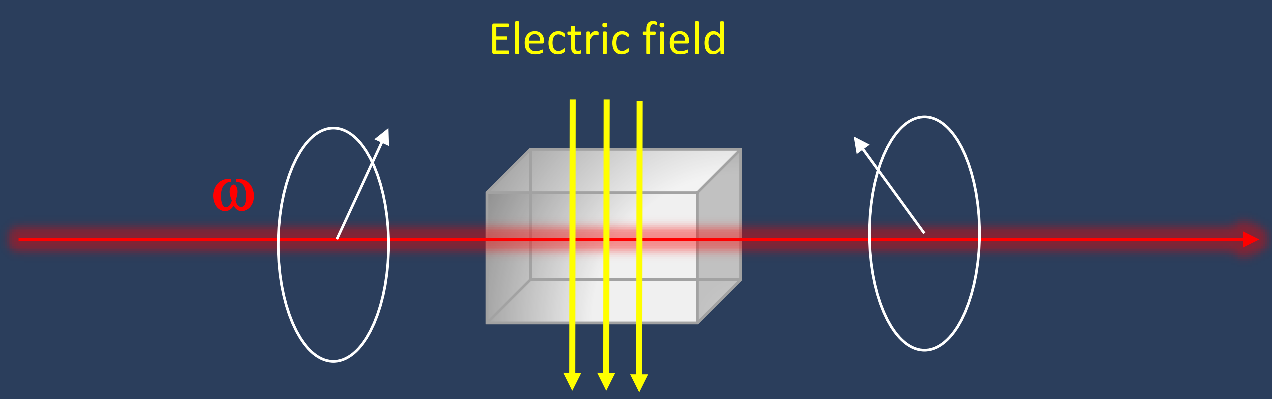
Change in the refractive index linearly proportional to the electric field

Electro-optic tensor $\chi^{(2)}$ of

$LiNbO_3$	**$BaTiO_3$**
d_{33}= 30.8 pm/V	d_{42} = 923 pm V^{-1}

Abel, S., *Nature Mater* **18,** 42–47 (2019).

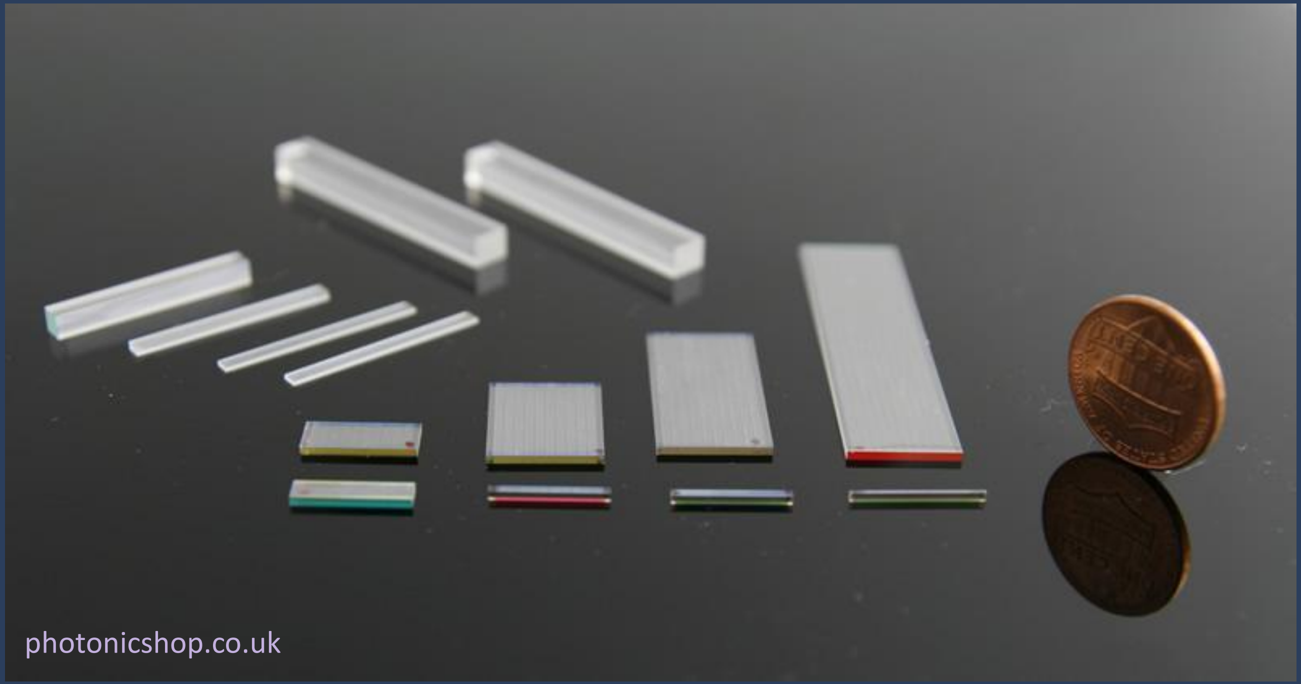
More properties of quadratic $\chi^{(2)}$ materials: $LiNbO_3$



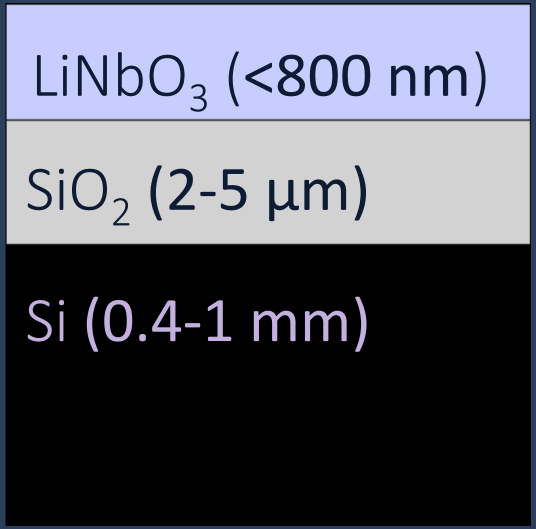
- **Electro-optic** → **Modulation**
- **Large band gap: 3-4 eV** → **Highly transparent 0.3 – 5 µm**
- **Refractive index > 2** → **Waveguiding of light**
- **Very inert** → **Robust in harsh environment**

Does this material exist at a small scale?

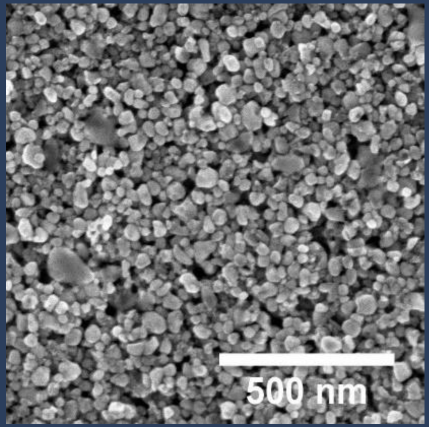
Bulk crystal



At small scale?



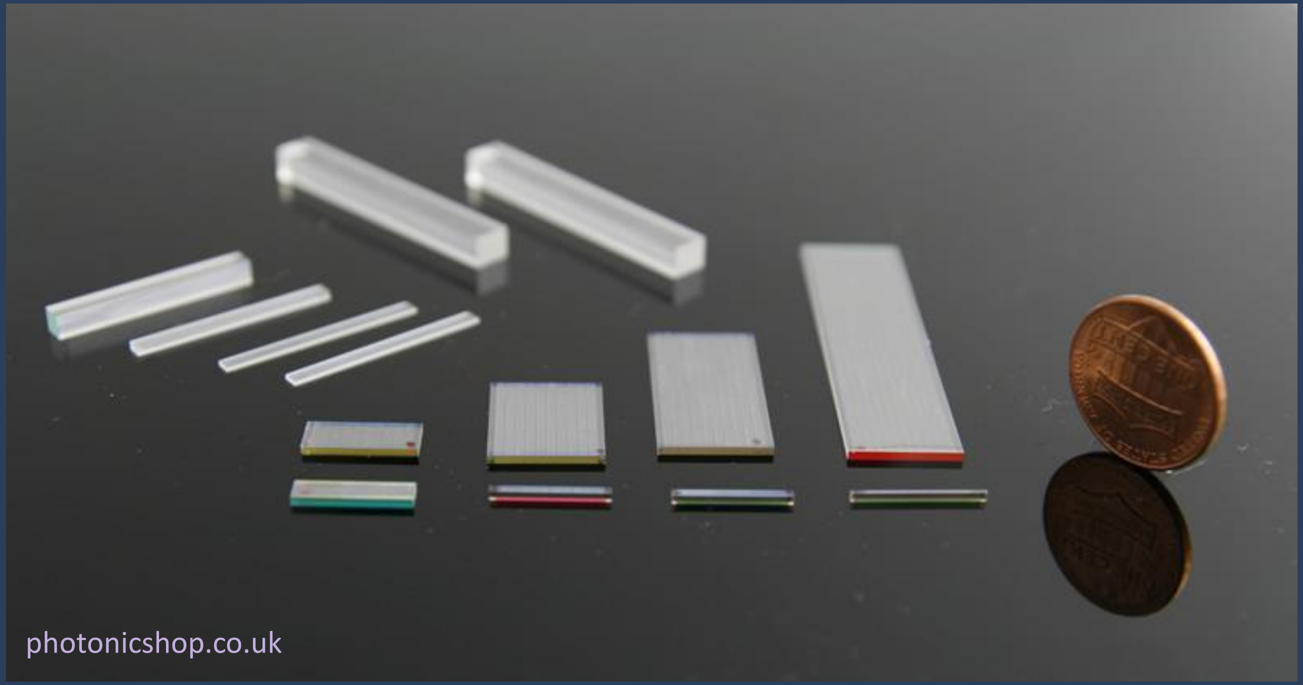
$BaTiO_3$



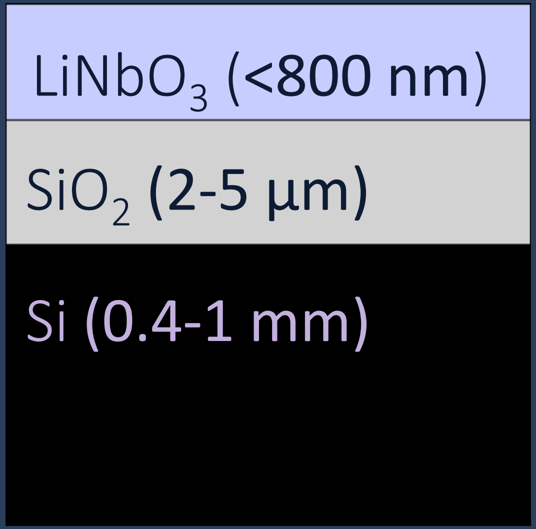
Thin films or powders?

Does this material exist at a small scale?

Bulk crystal

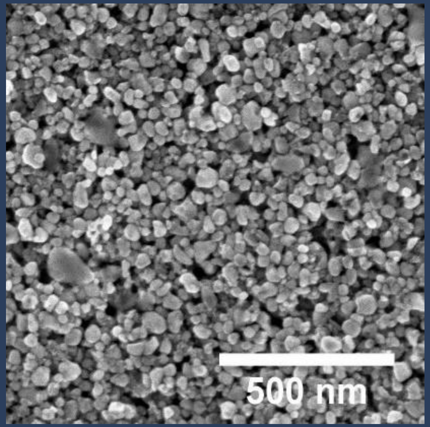


At small scale?



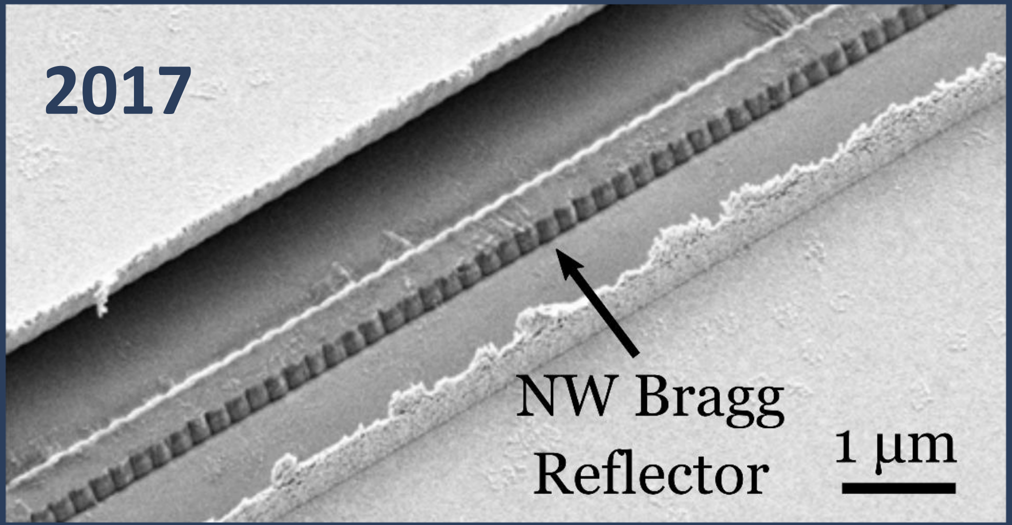
Rabiei, P.; Gunter, P. *Applied Physics Letters* **2004**, *85* (20).

$BaTiO_3$

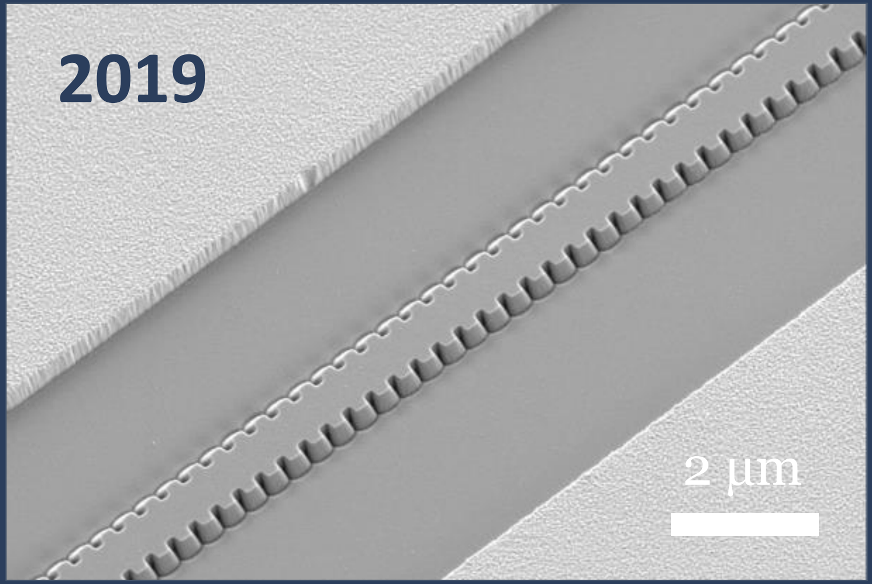


Kim, E.; ... Grange, R. ACS Nano **2013**, 7 (6).

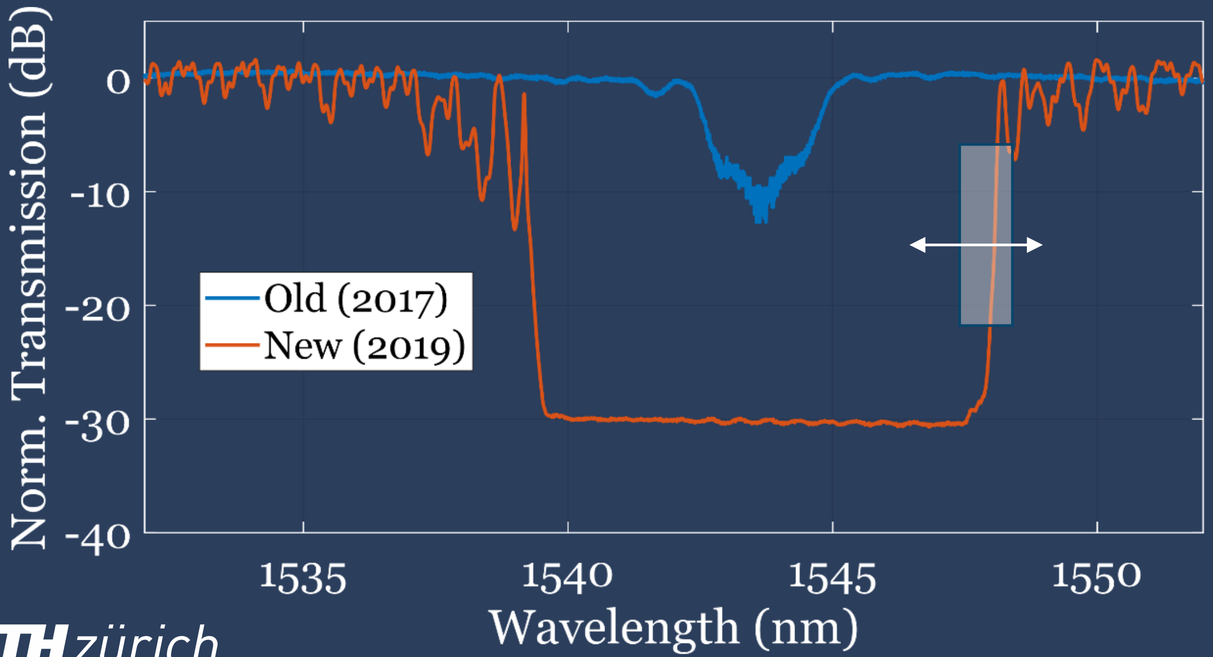
Challenges of miniaturization



Reig Escalé, et al. OL 43(7) 2018



Pohl, et al. IEEE PTL 33 (2) 2020

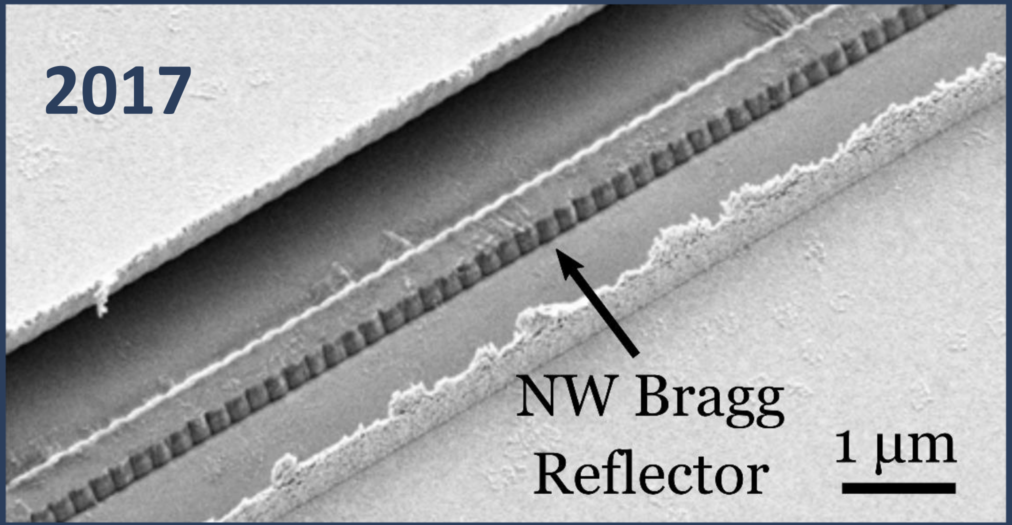


Extinction ratio from
-12 dB to more than **-30 dB**

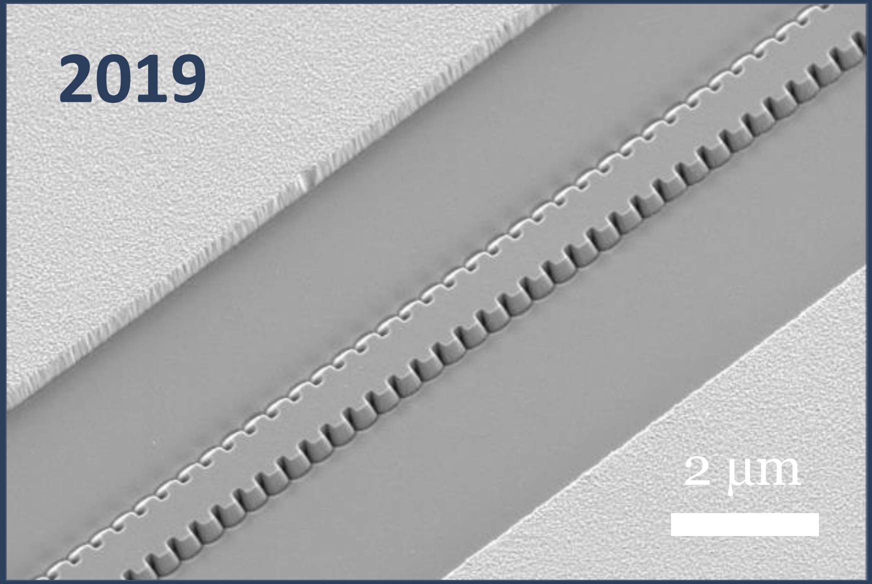
Propagation losses **< 0.1 dB/cm**

Nanofabrication at BRNC and FIRST clean rooms

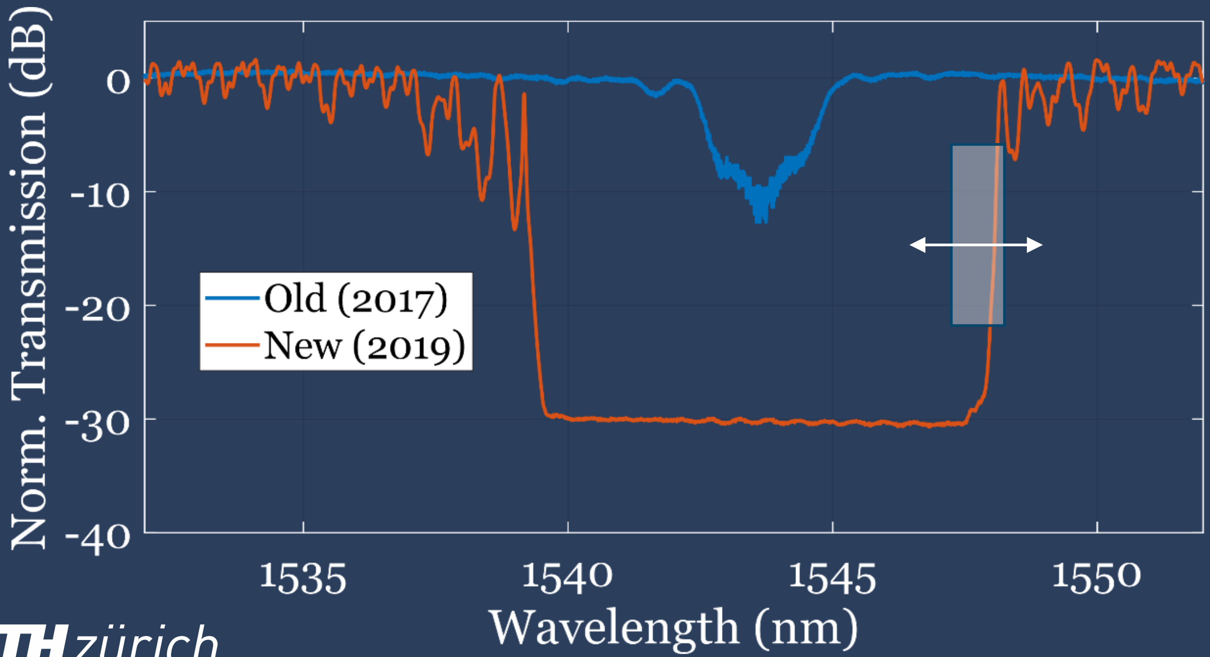
Challenges of miniaturization



Reig Escalé, et al. OL 43(7) 2018

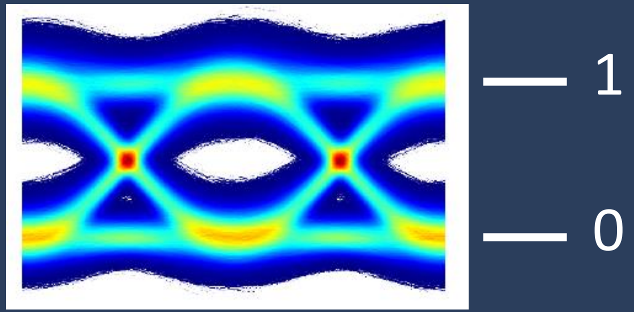


Pohl, et al. IEEE PTL 33 (2) 2020



100 Gbit/s

BER = 1.3×10^{-5}



Outline

Miniaturizing $\chi^{(2)}$ materials

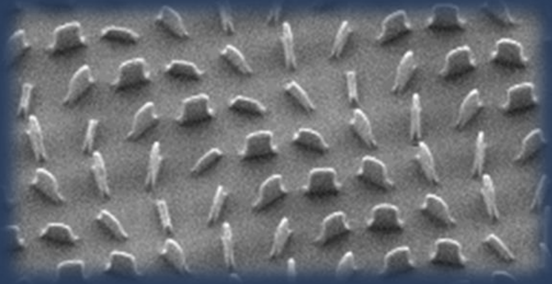
Nonlinear or electro-optic metasurfaces

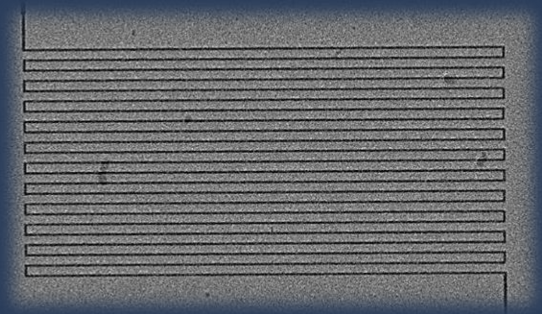
Pulsed laser deposited $BaTiO_3$

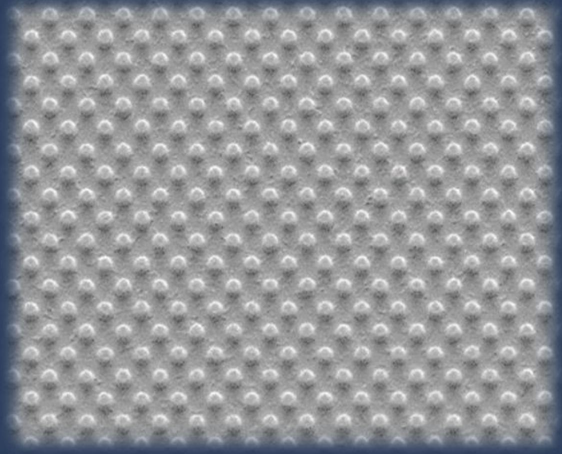
FIB and spin coated nanoparticles

Sol-gel nanoimprinted metalens

Miscellaneous photonic structures

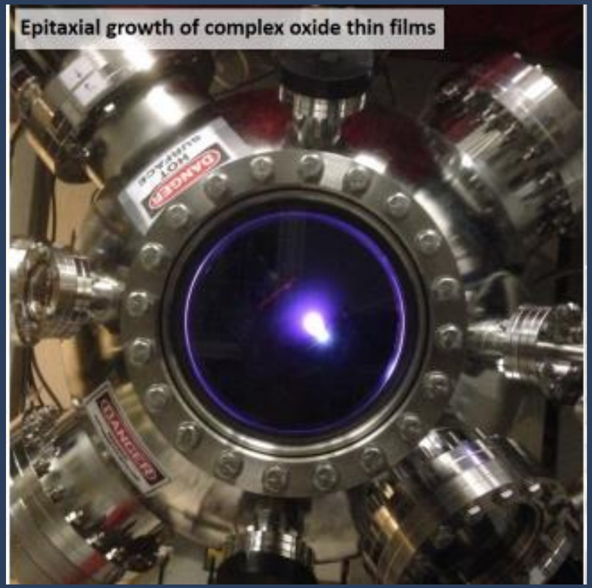


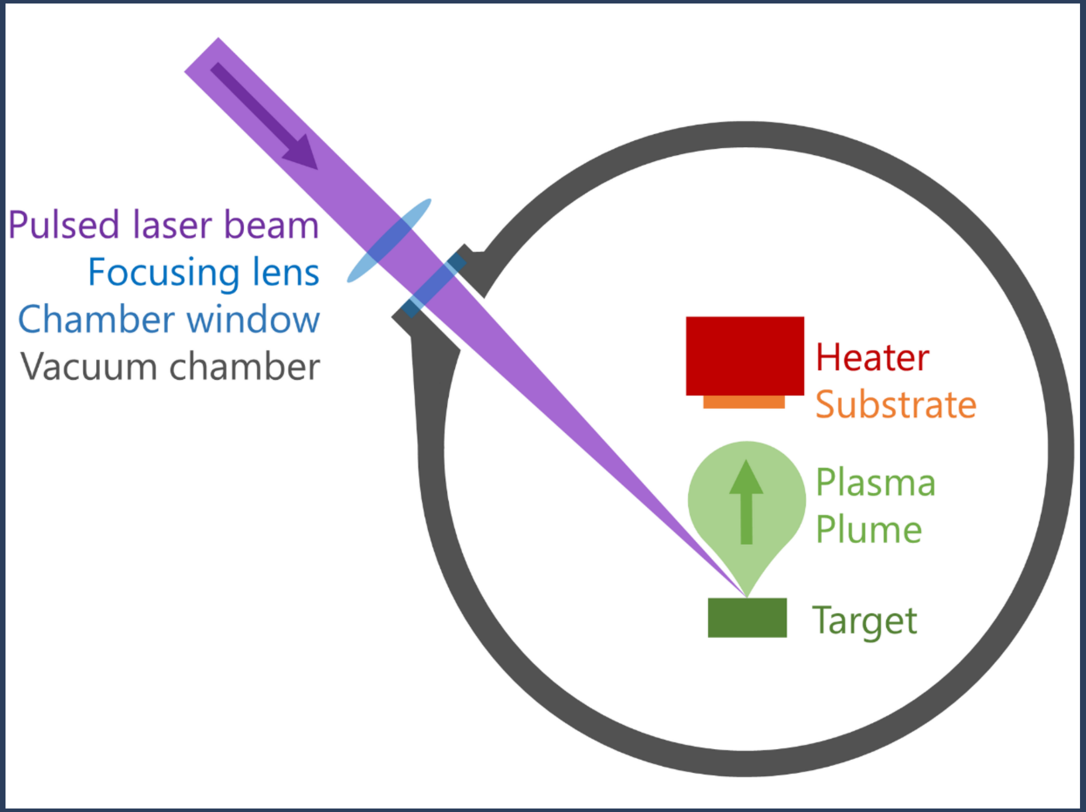




Pulsed laser deposited thin films of $BaTiO_3$

200 nm thick polycrystalline film of $BaTiO_3$





Collaboration with M. Trassin and M. Fiebig at ETH

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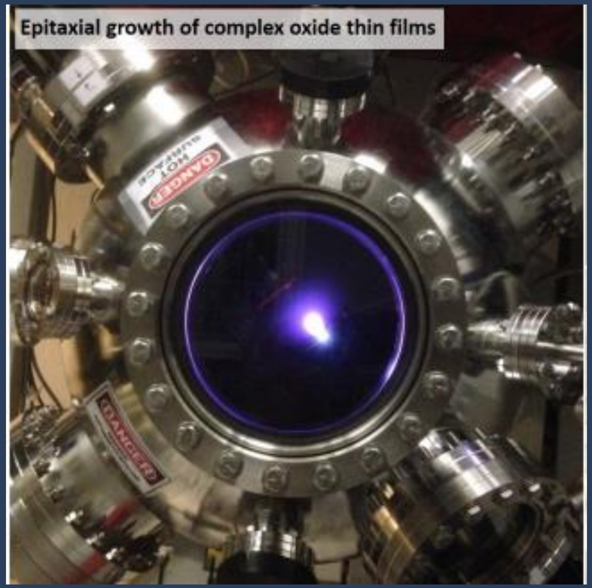
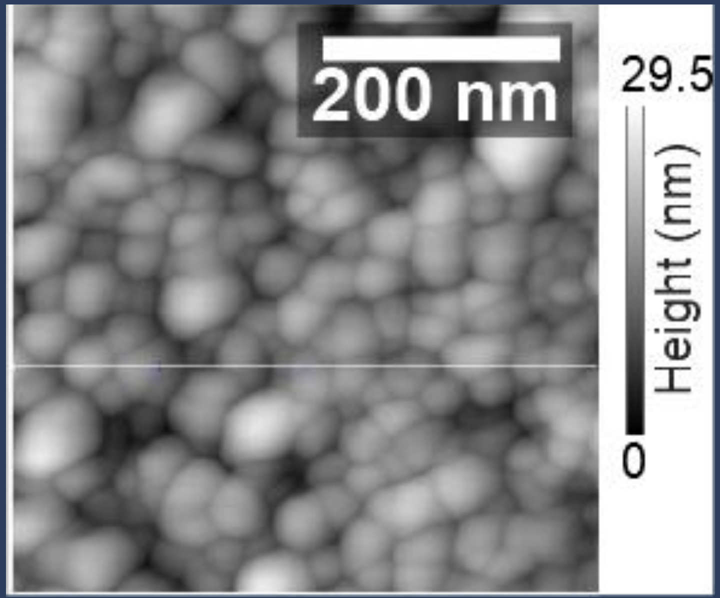
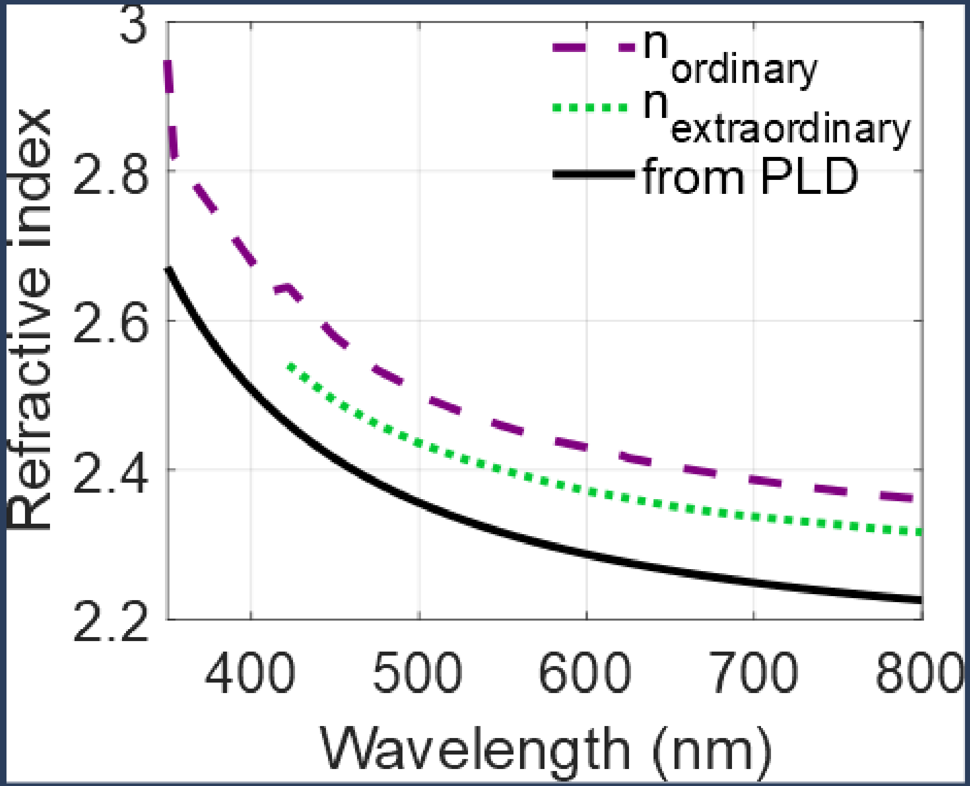


AFM



Ellipsometry



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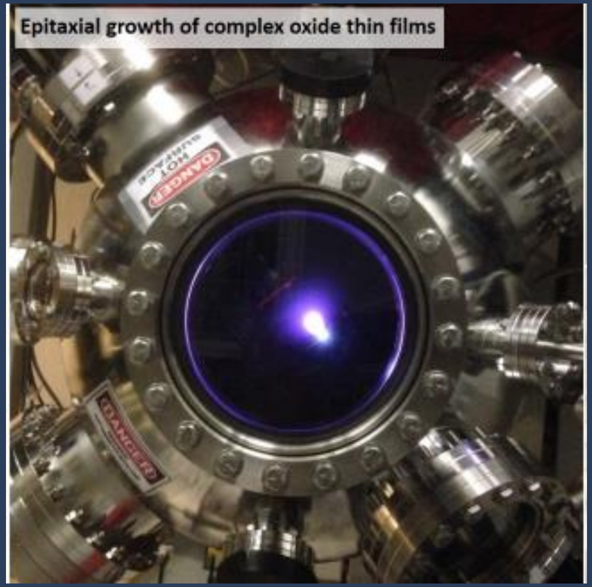
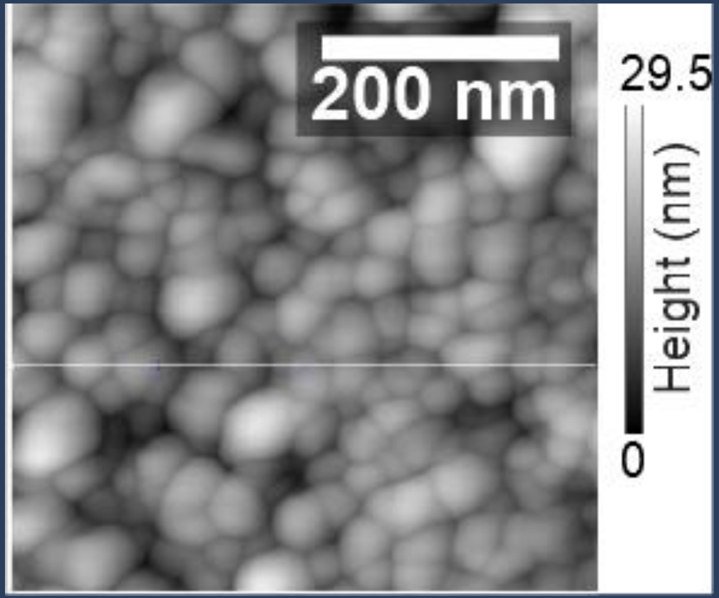
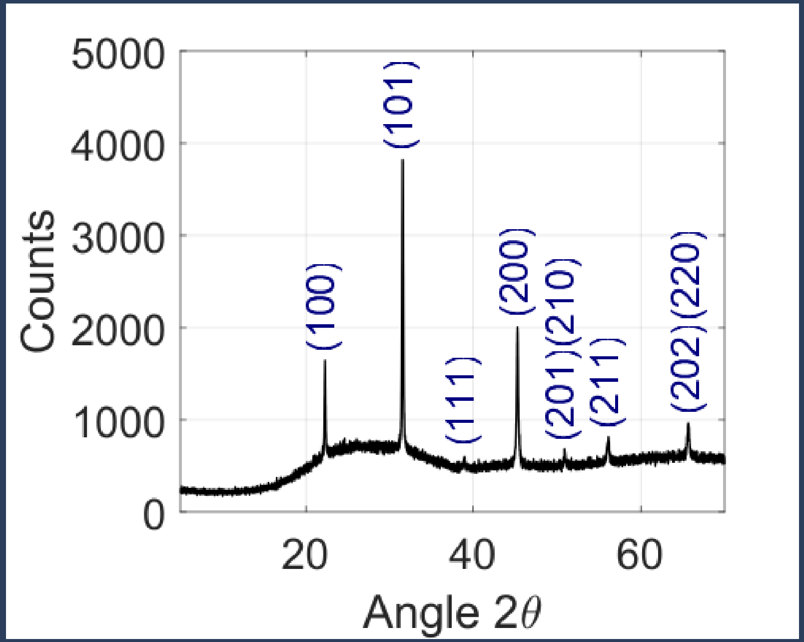


AFM



Crystalline structure:
Cubic or tetragonal

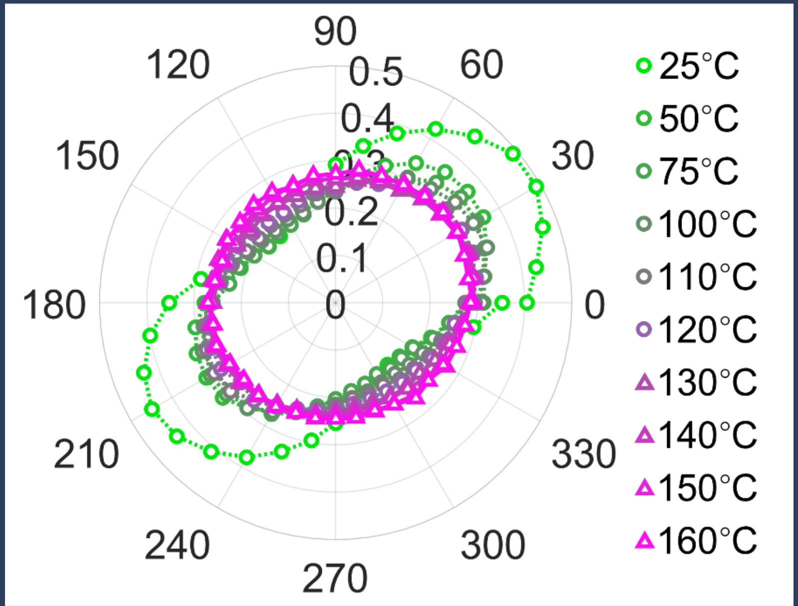


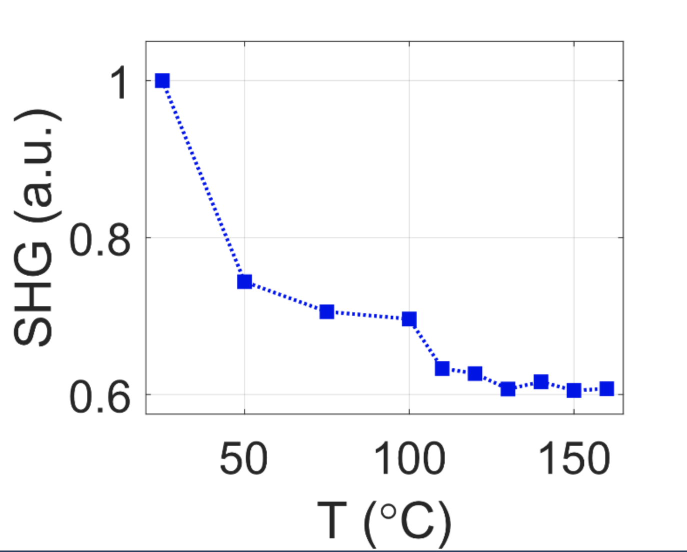
Collaboration with M. Trassin and M. Fiebig at ETH

Pulsed laser deposited thin films of $BaTiO_3$

Nonlinear optical characterization

SHG polar plots

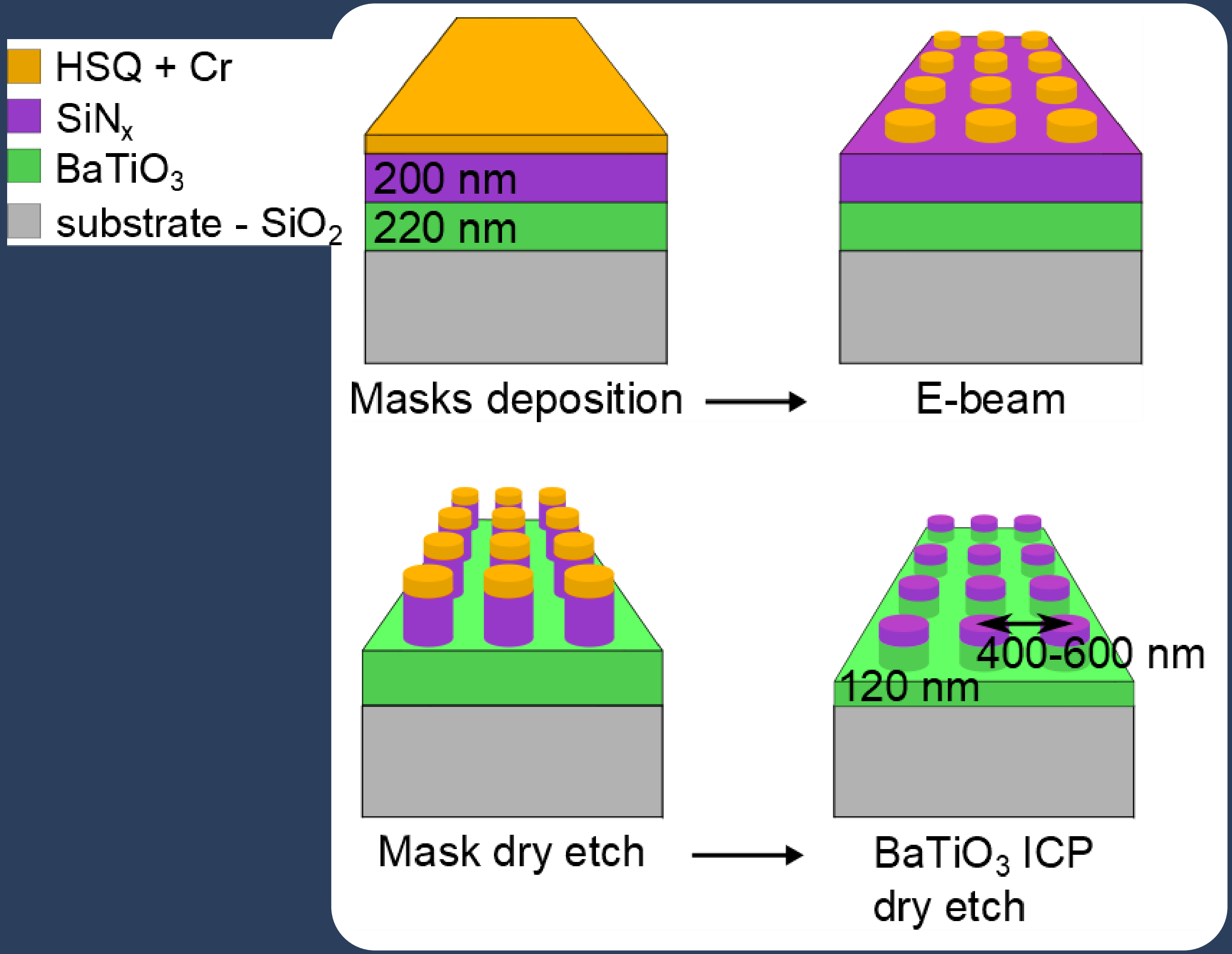




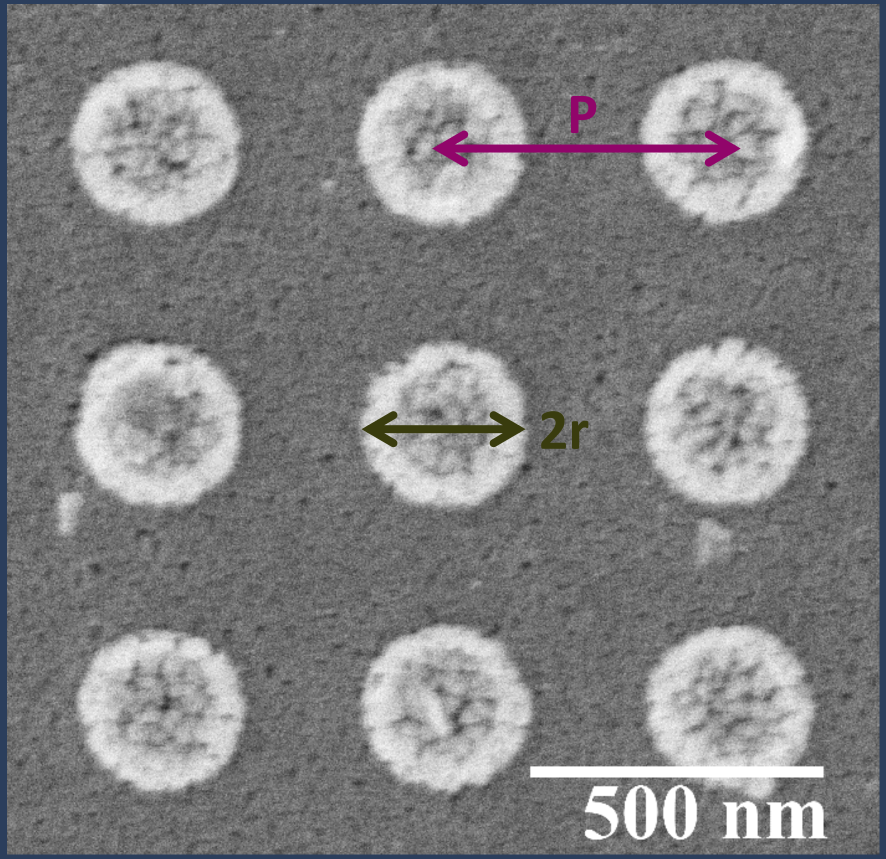
- SHG anisotropy change between 120°C and 130°C
- Phase change from tetragonal to cubic crystal structure at T_C
- Tetragonal crystal structure present at room temperature

Collaboration with M. Trassin and M. Fiebig at ETH

Top-down etching of the thin film

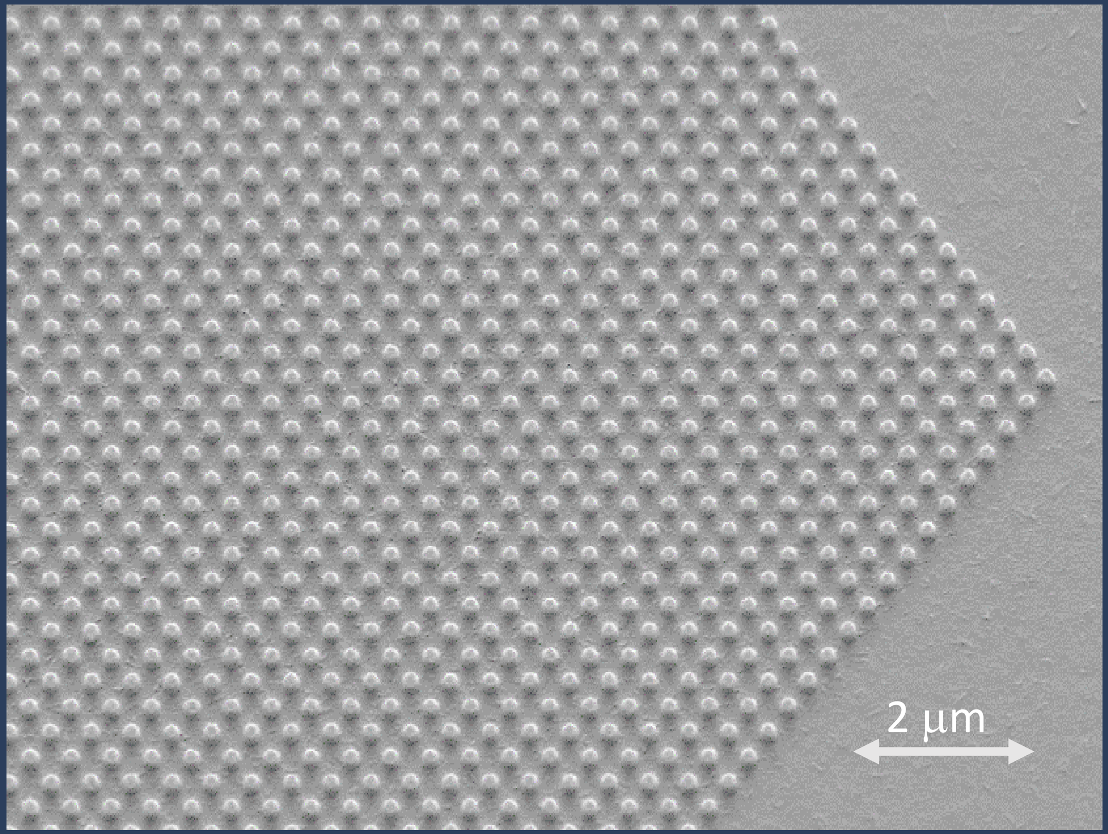


SEM image of a $BaTiO_3$ metasurface.

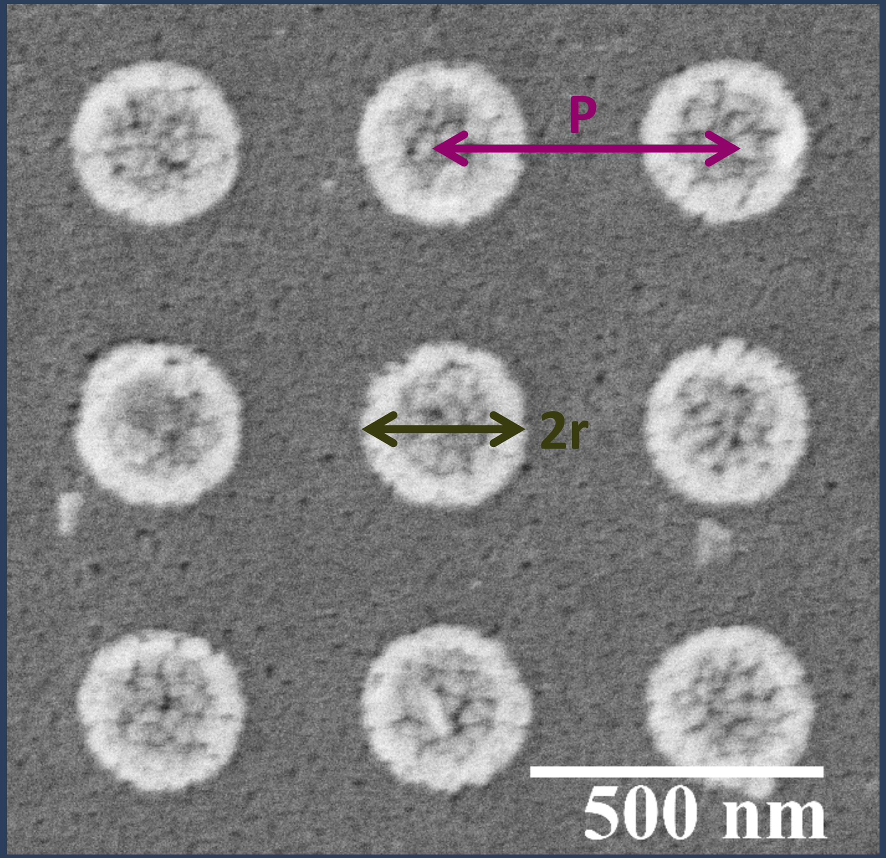


Timpu, ...Grange. Advanced Optical Materials 2019, 7 (22).

Barium titanate metasurface down to the near UV



SEM image of a $BaTiO_3$ metasurface.



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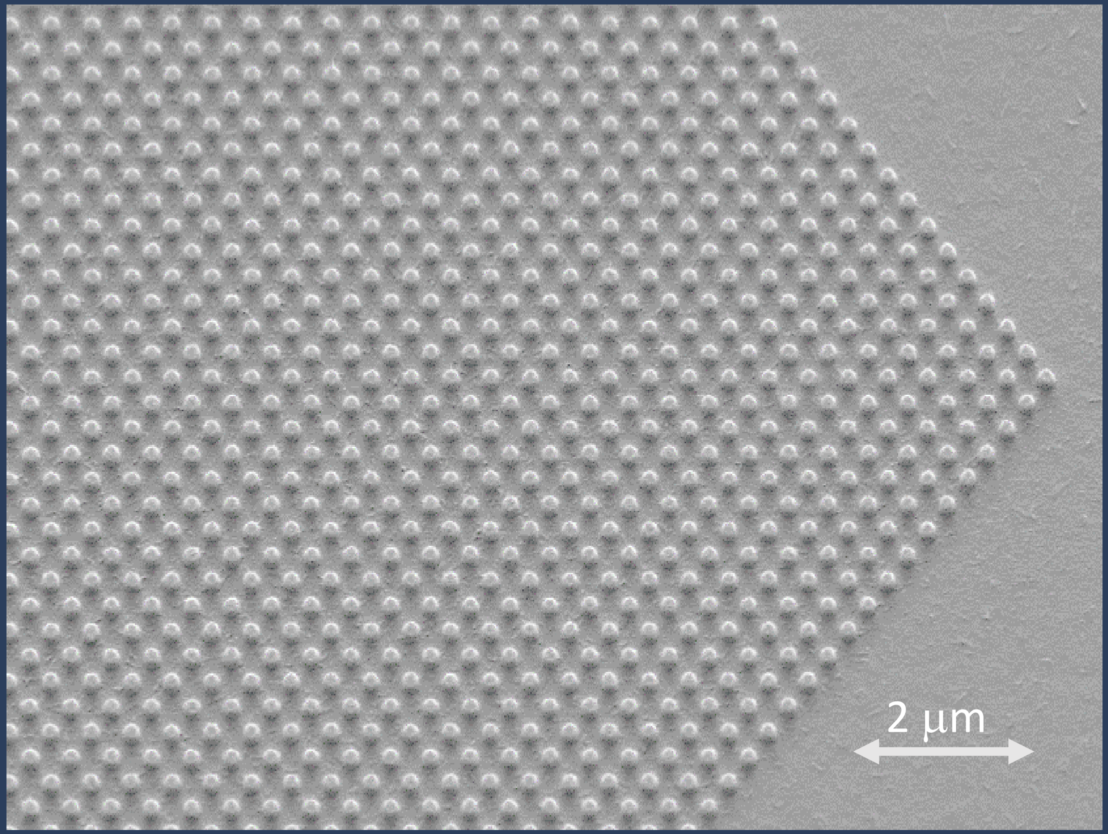
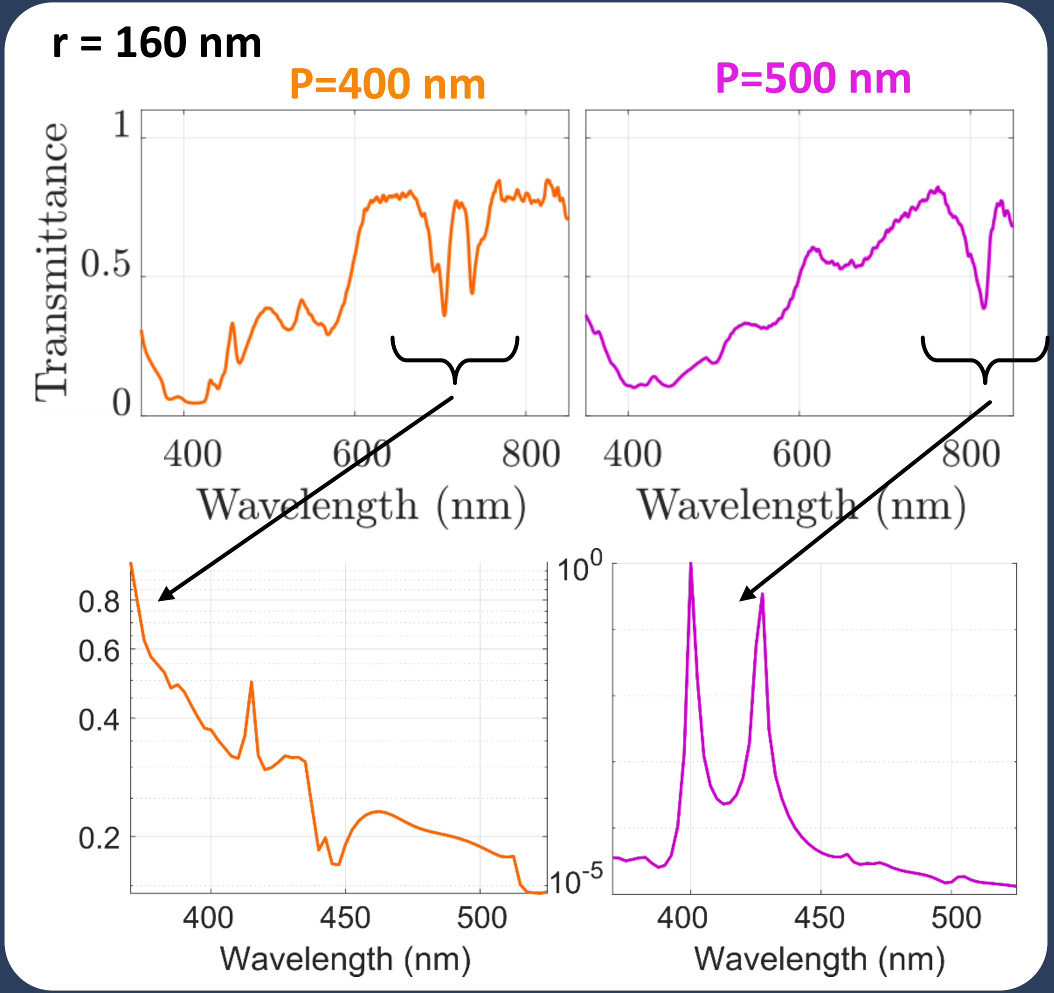
Barium titanate metasurface down to the near UV



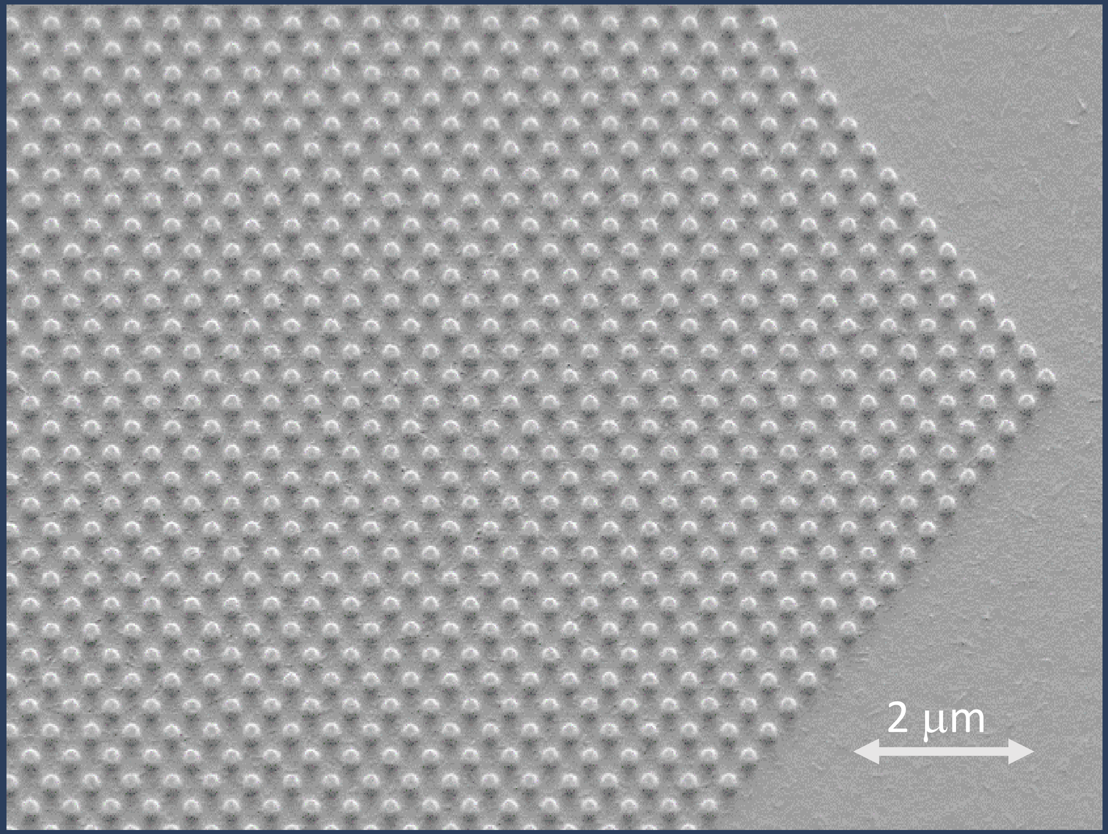
Linear optical transmittance



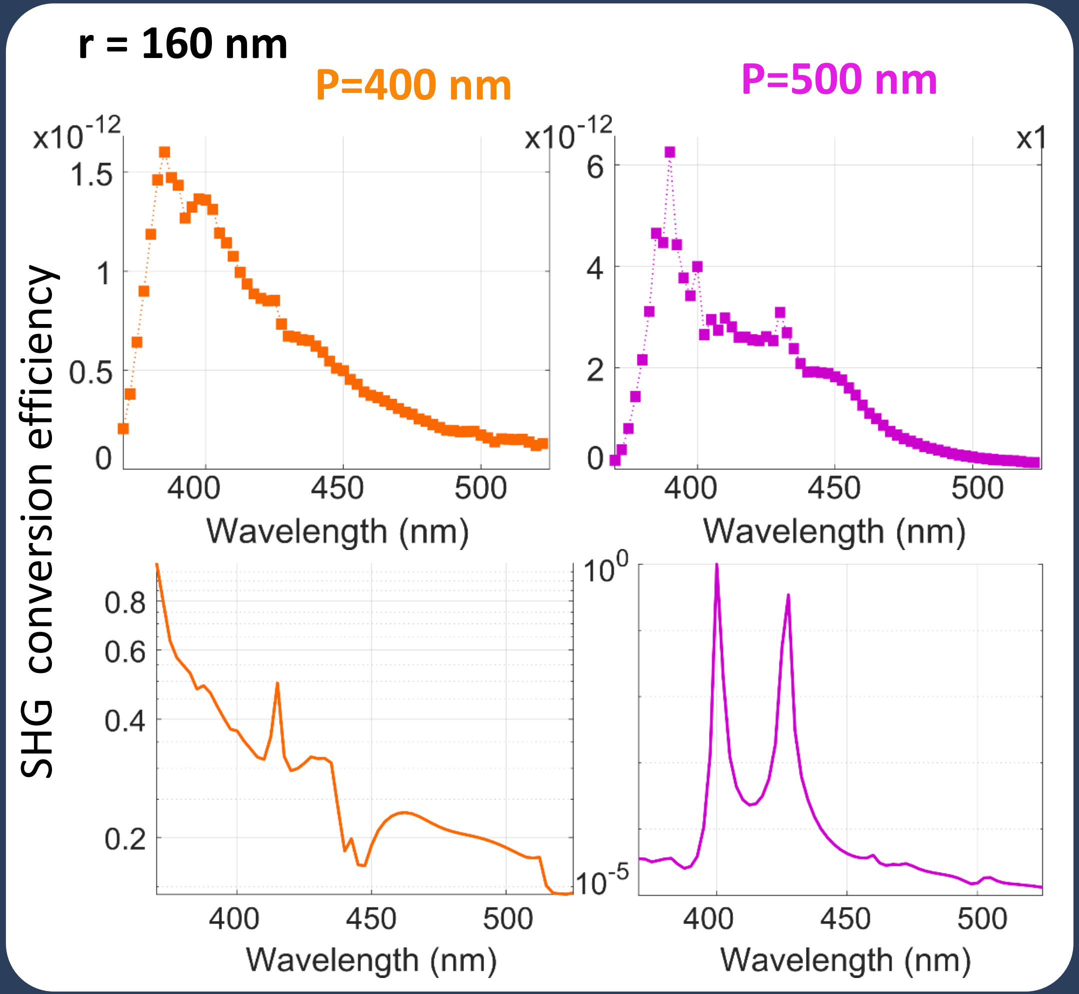
Calculation of the SHG conversion efficiency

Timpu, ...Grange. Advanced Optical Materials 2019, 7 (22).

Barium titanate metasurface down to the near UV



Measured SHG signal



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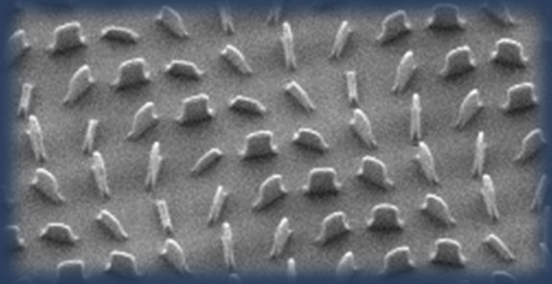
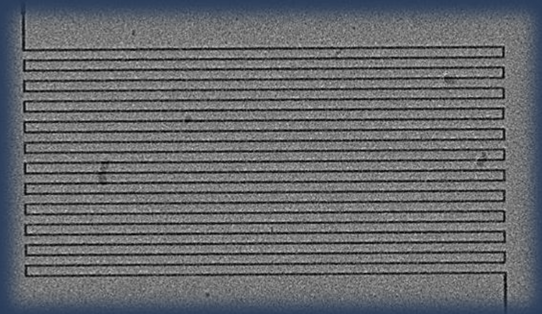
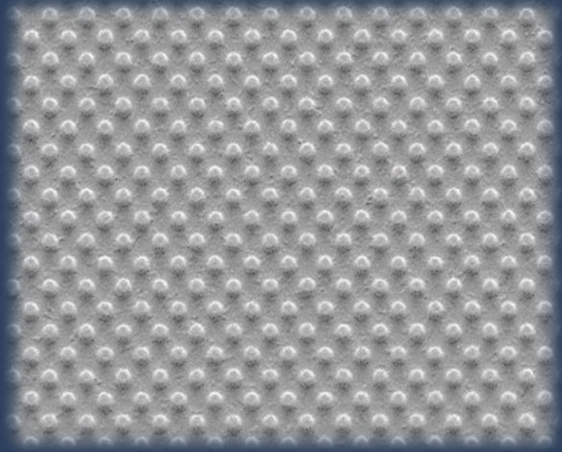
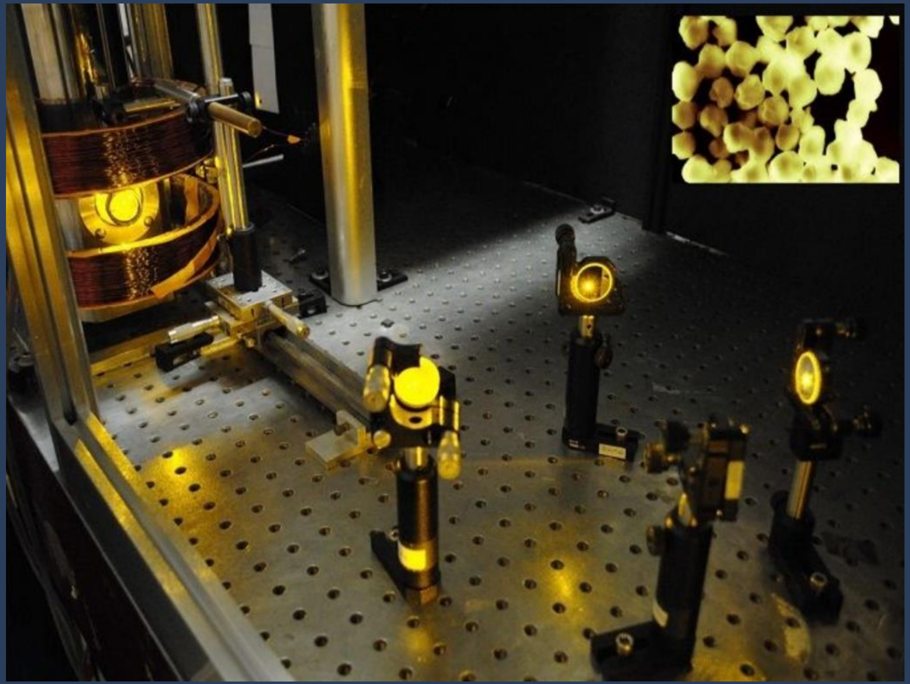






Particle-based photonic structures: advantages

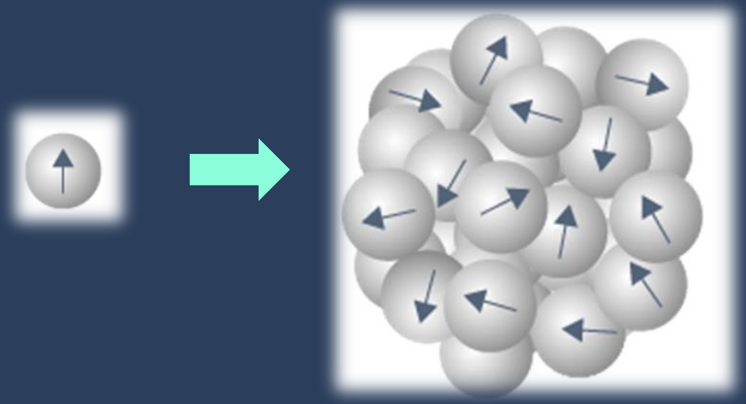
- Simplify the fabrication : avoid etching process of metal-oxides
- Use powder instead of high quality crystal : test new compounds

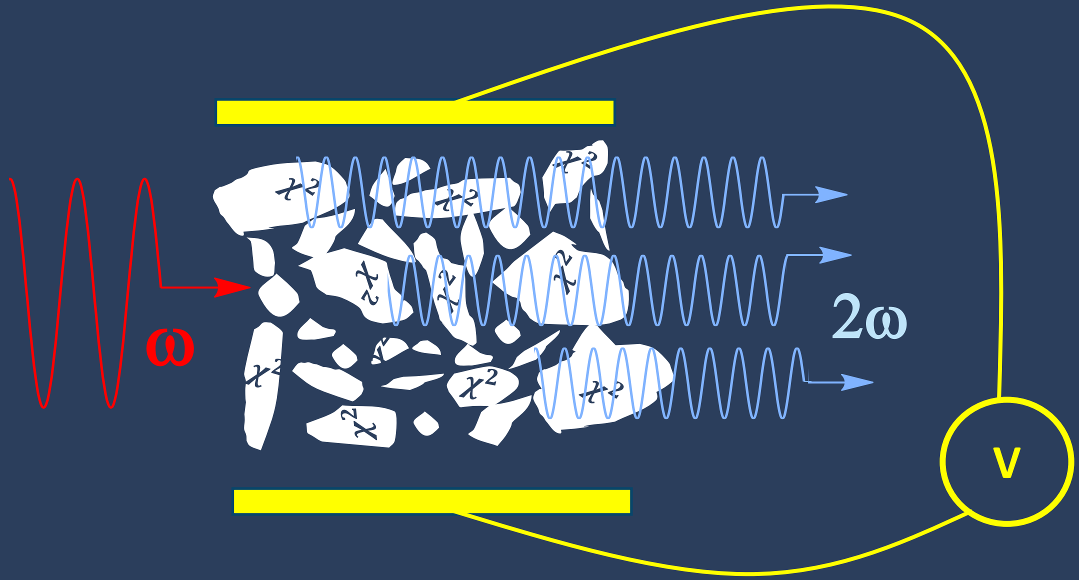


Serrano, …Goldner. All-Optical Control of Long-Lived Nuclear Spins in Rare-Earth Doped Nanoparticles. *Nat Commun* **2018**

Particle-based photonic structures: advantages

- Simplify the fabrication : avoid etching process of metal-oxides
- Use powder instead of high quality crystal : test new compounds
- Useful for nonlinear optics: relaxing the phase matching condition, broadband

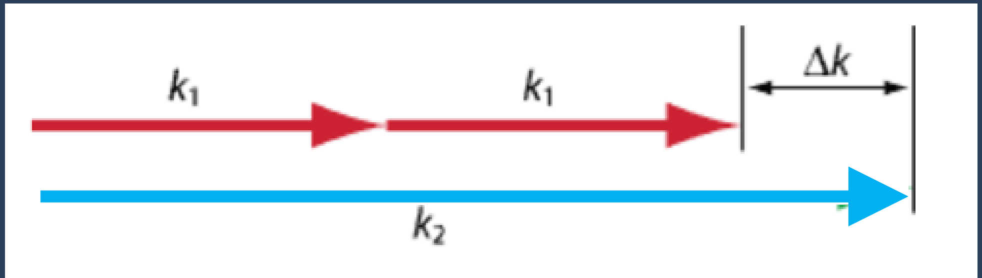




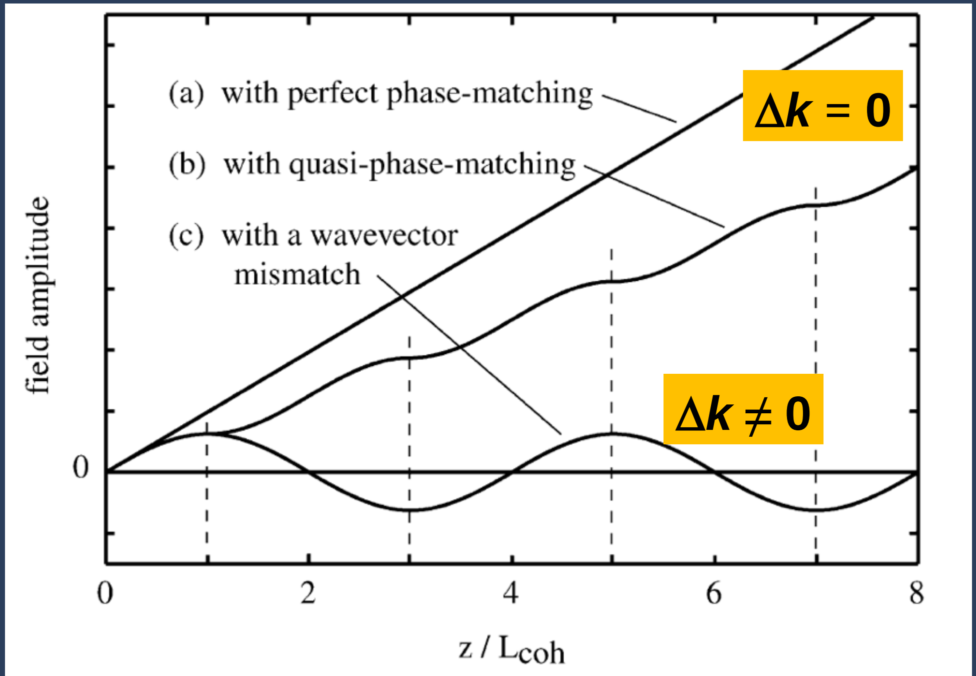
Second-order signal?

Electro-optic effect?

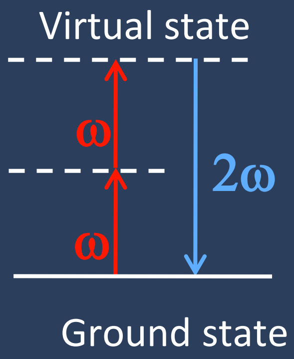
How to relax phase matching?



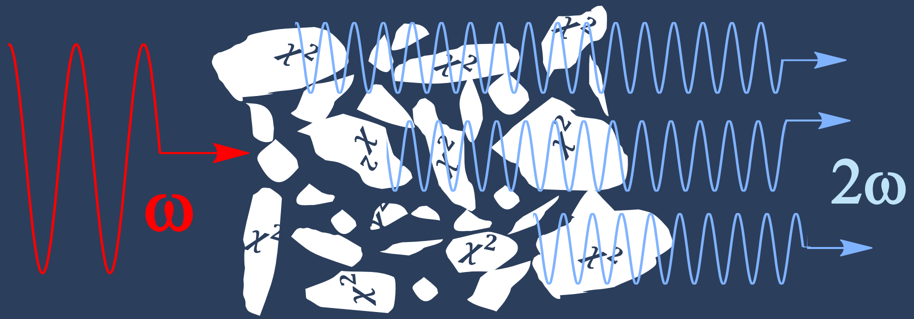
Phase mismatch due to dispersion



Boyd, Robert W. *Nonlinear optics*. 2003



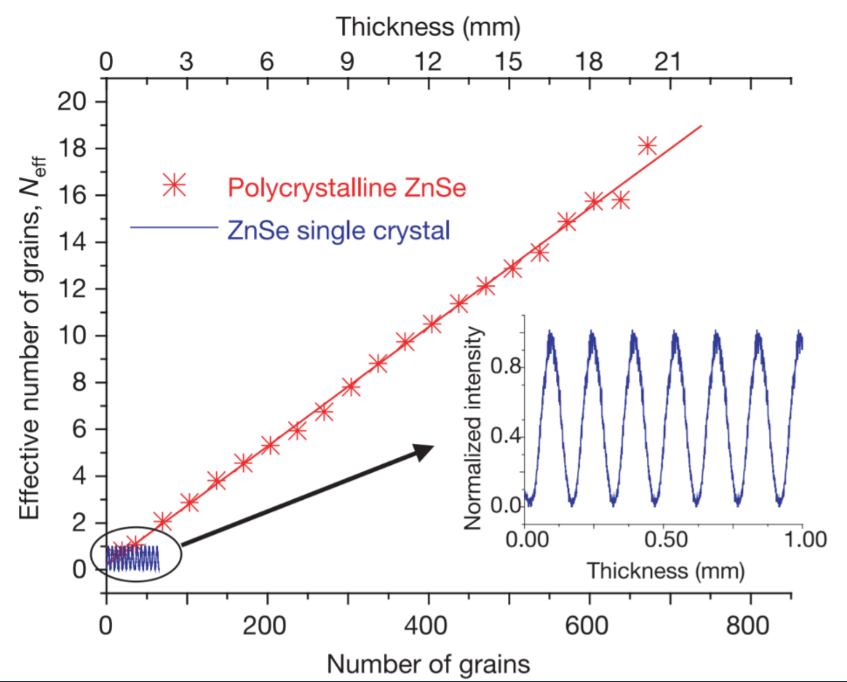
Second-Harmonic
Generation
SHG



$$\vec{P} = \varepsilon_0 \chi^{(1)} \vec{E} + \varepsilon_0 \chi^{(2)} \vec{E}^2 + \cdots$$

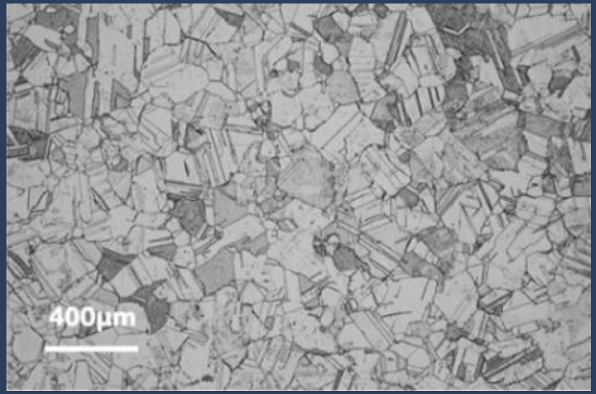
How to relax phase matching?

Random Quasi-Phase-Matching



Raybaut et al., Nature 432, 374–376 (2004)

ZnSe grains with 10s of microns in sizes

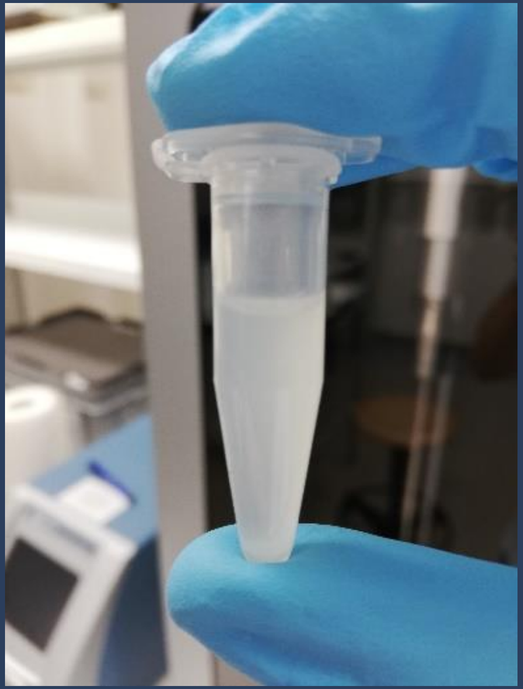


Chen and Gaume, Opt. Mater. Express 9, 400-409 (2019)

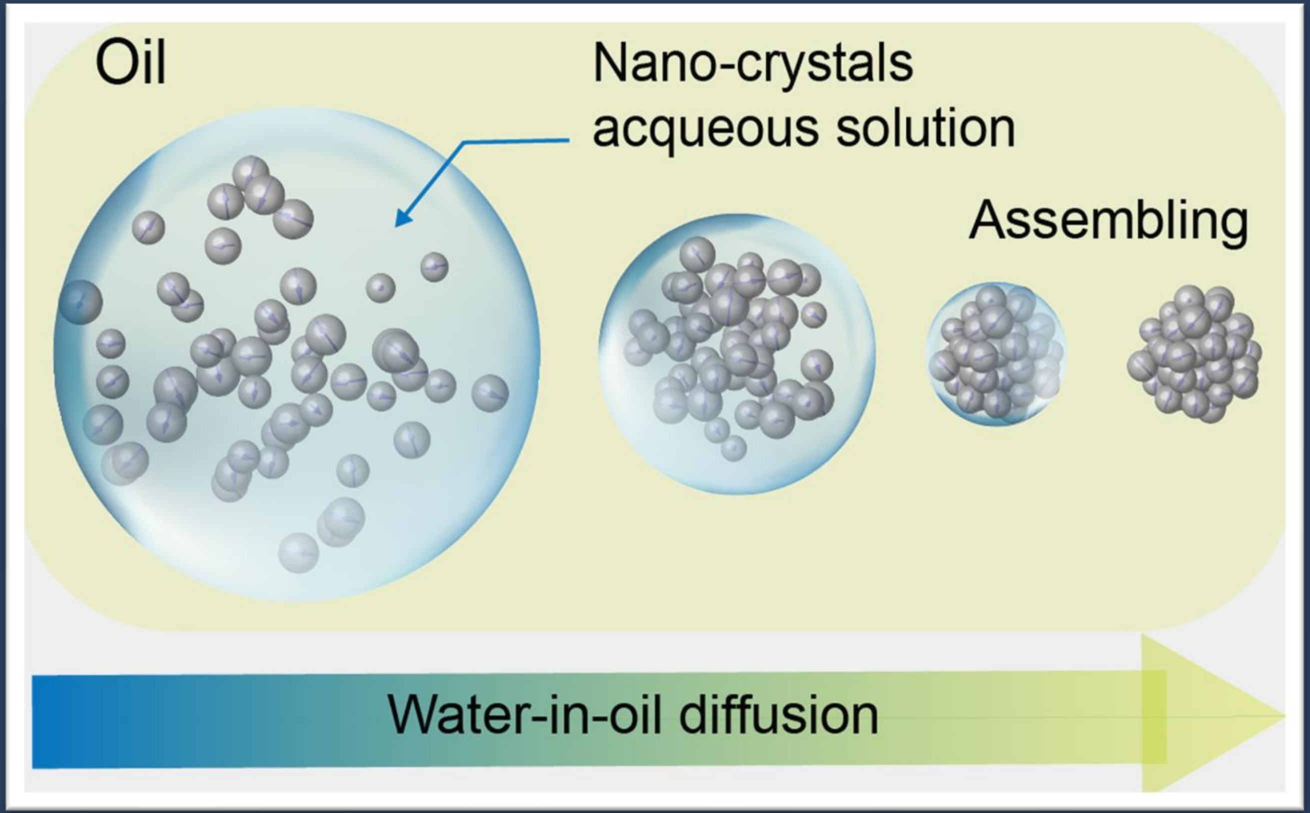
$$I_{\mathrm{SH}} \propto N^2 \propto V^2 \longrightarrow I_{\mathrm{SH}} \propto N \propto V$$

Bottom up assemblies : emulsion driven technique

$BaTiO_3$ nanoparticles in solution
Typical diameter 50 nm



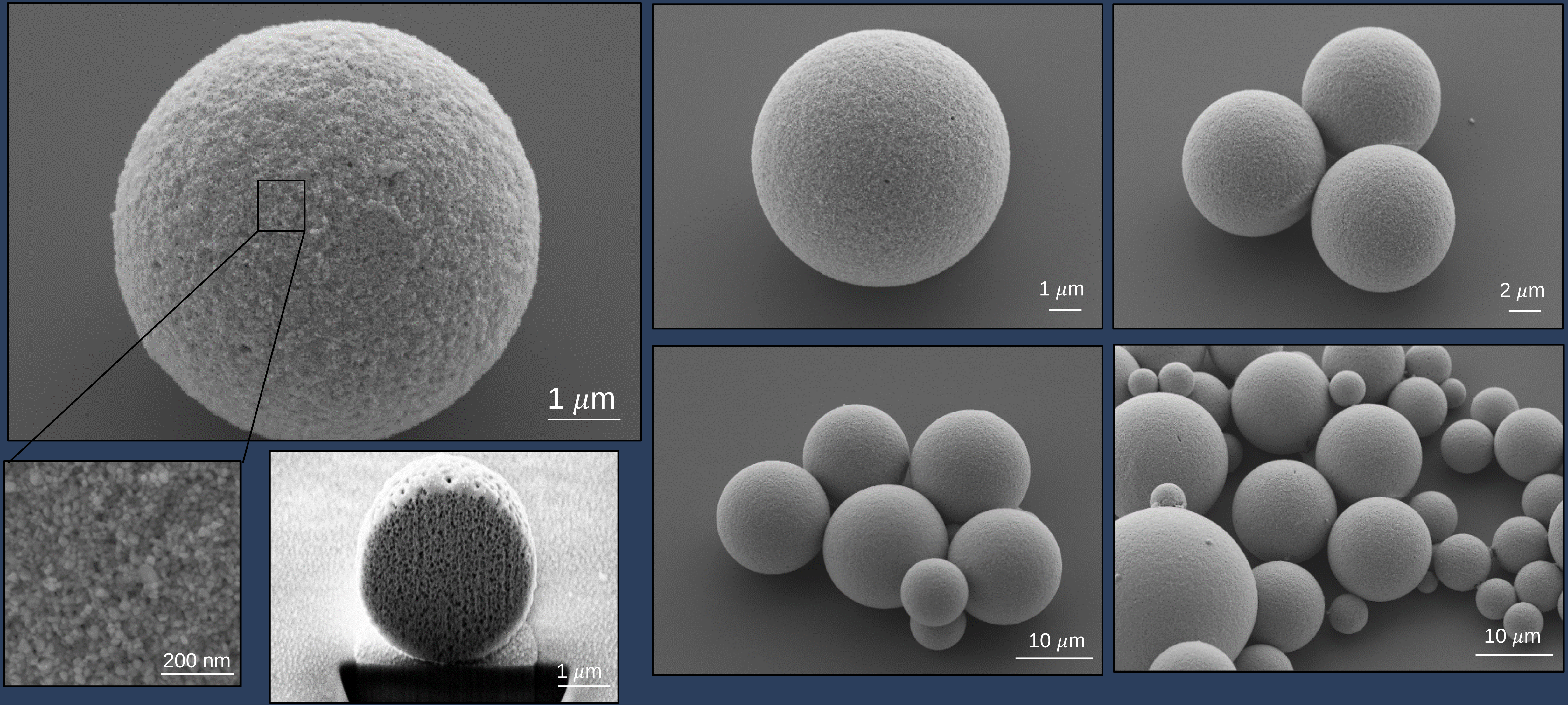
Droplets act as a template for spherical assemblies



L. Isa, D-MATL, ETHZ
M. Niederberger, D-MATL, ETHZ
S. Pratsinis, D-MAVT, ETHZ

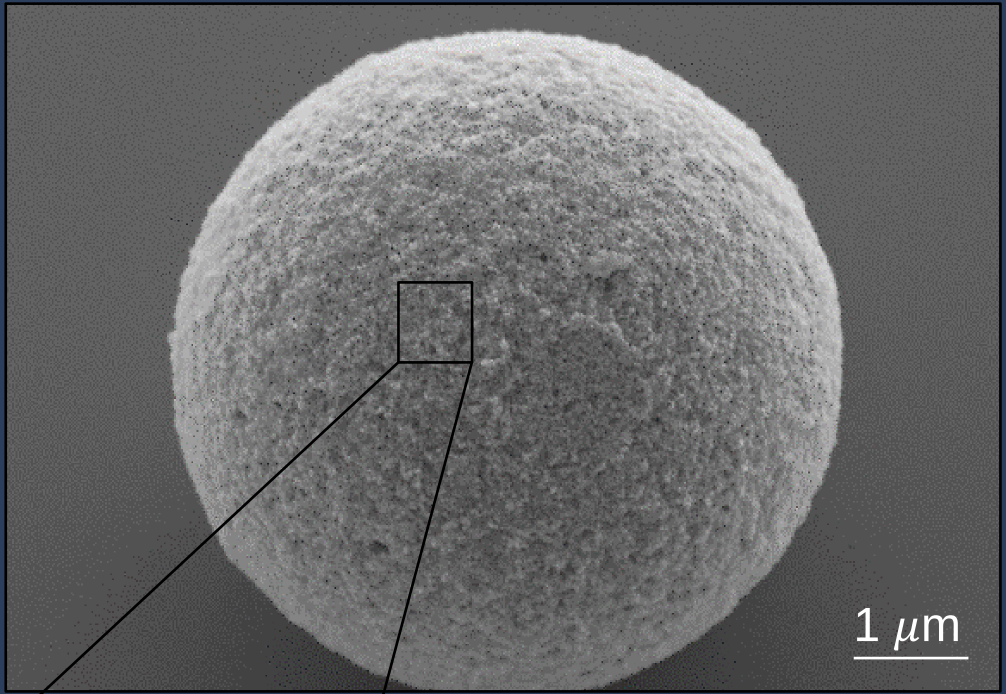
Savo *et al. Nat. Photonics* **14,** 740–747 (2020)

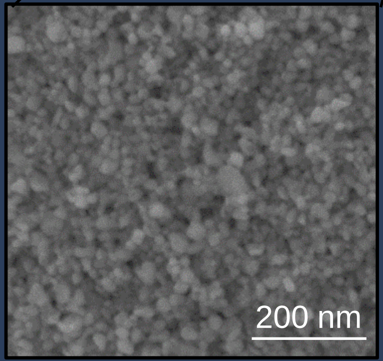
Barium titanate disordered microspheres

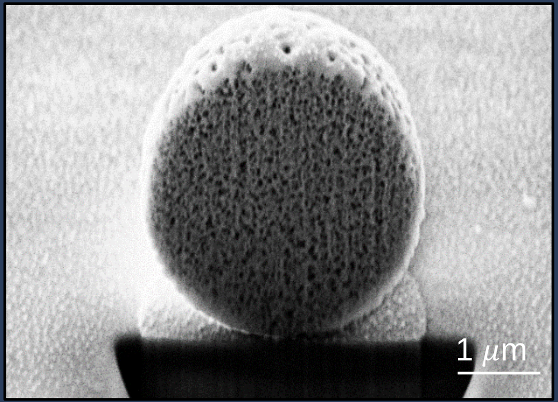


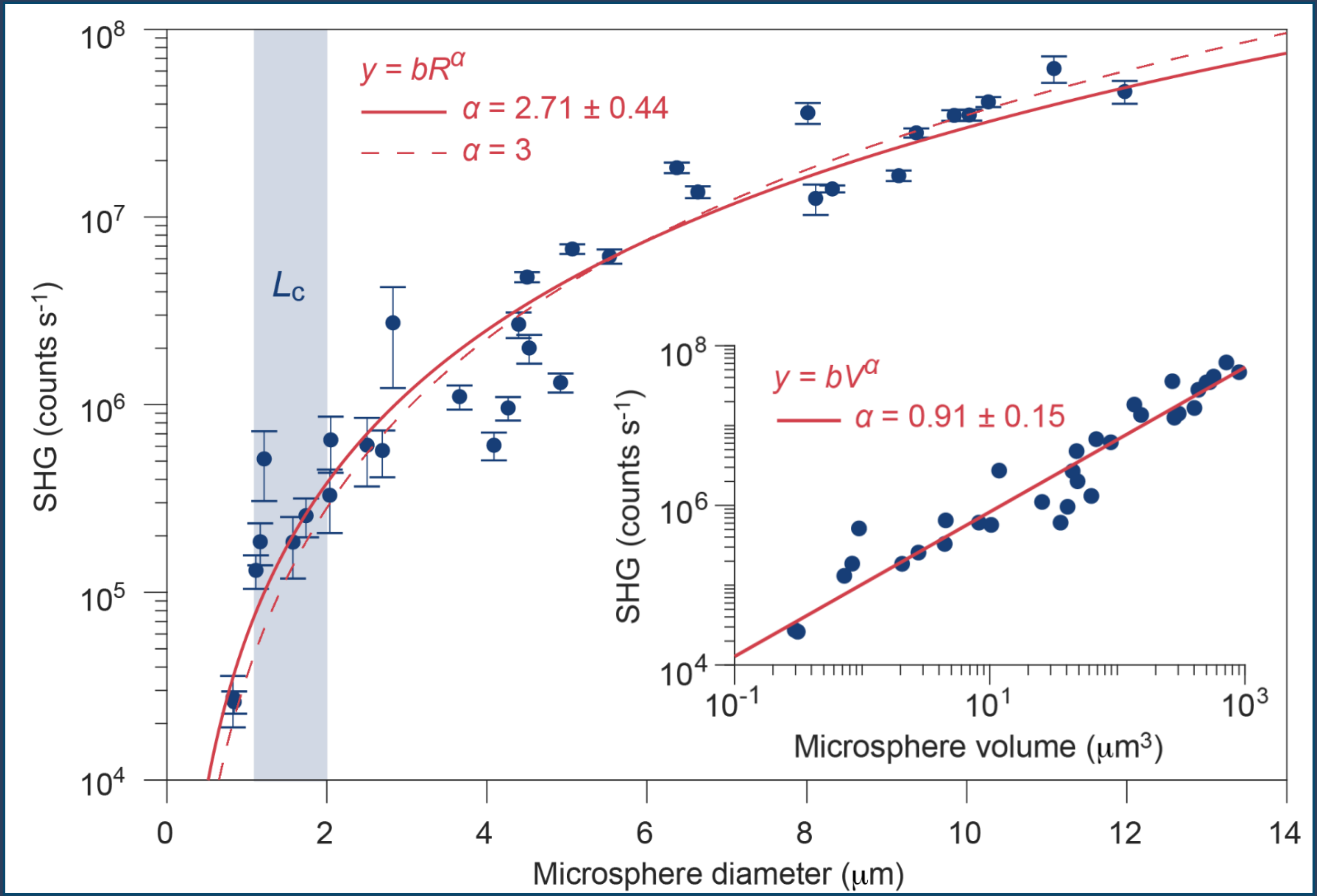
Savo *et al.* *Nat. Photonics* **14,** 740–747 (2020)

Random Quasi Phase Matching









$$I_{\mathrm{SH}} \propto N^2 \propto V^2 \longrightarrow I_{\mathrm{SH}} \propto N \propto V$$

Savo *et al. Nat. Photonics* **14,** 740–747 (2020)

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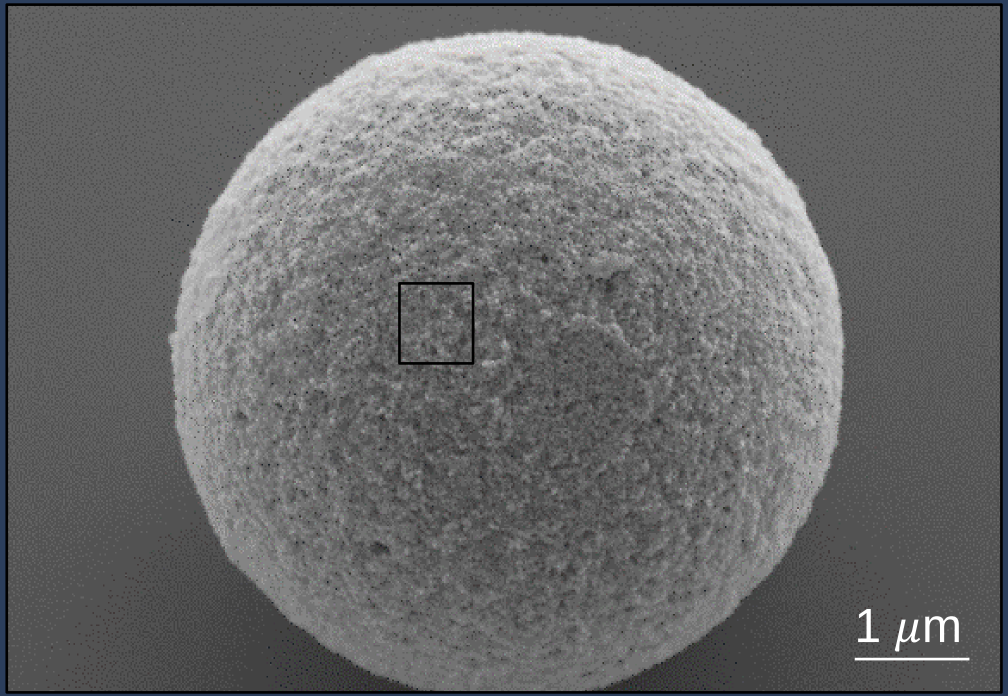
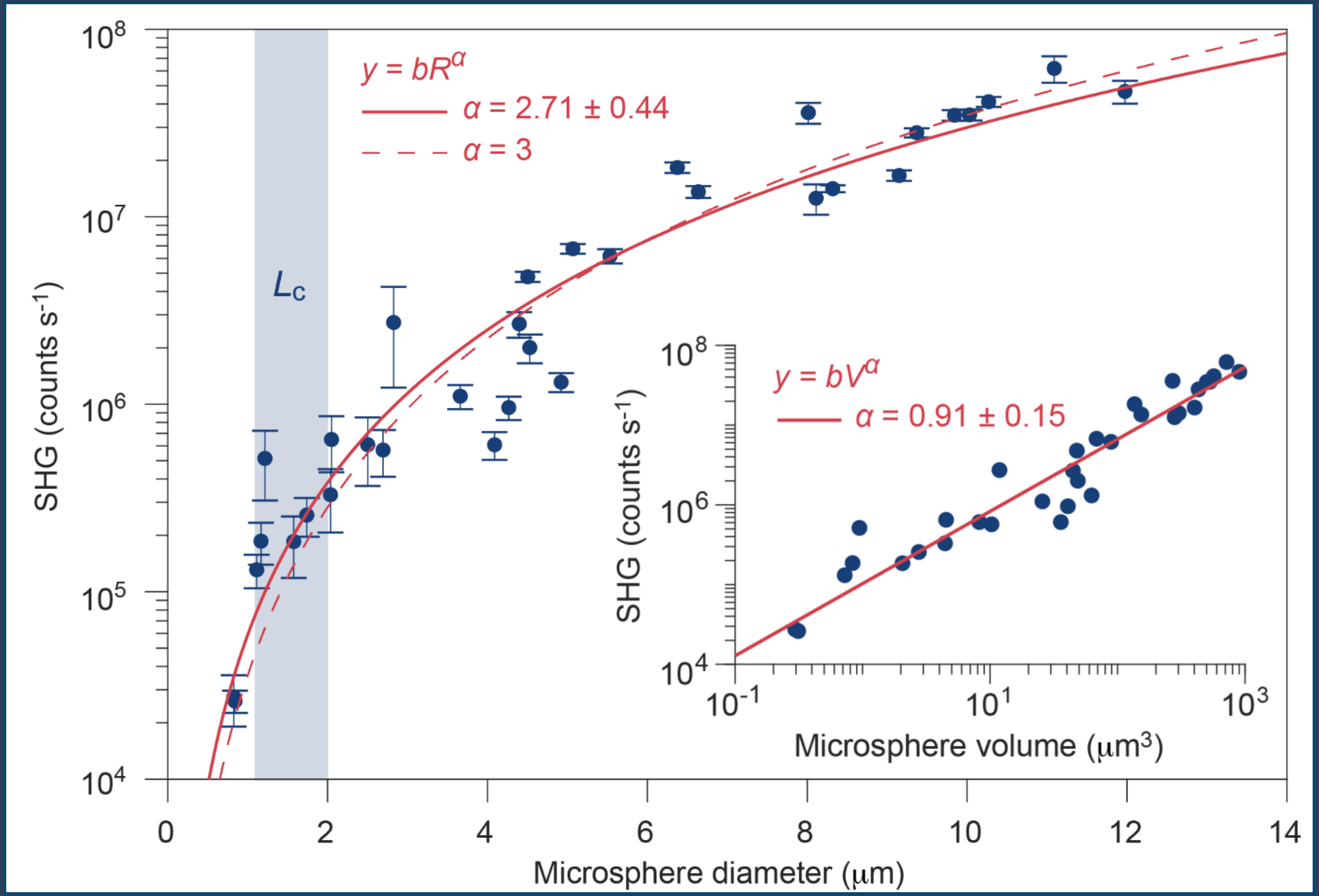
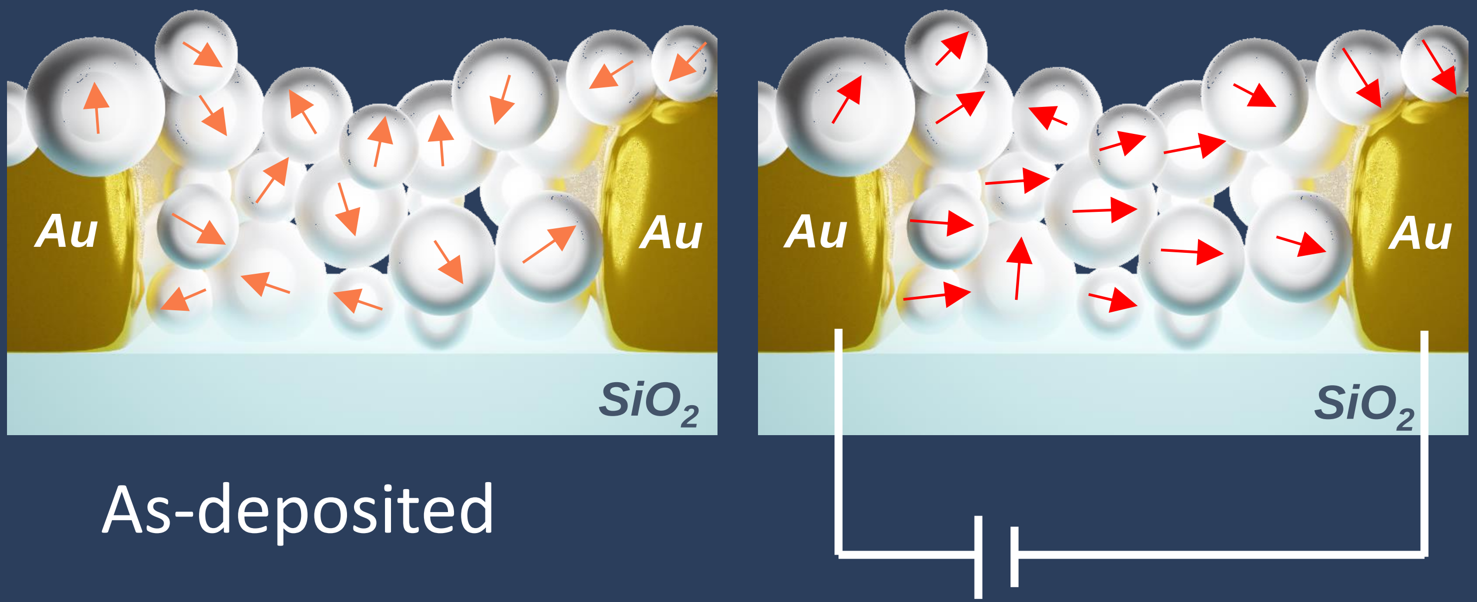




- New platform to study nonlinearities in disordered materials
- No need to match the length of a crystal with the laser source

$$I_{\mathrm{SH}} \propto N^2 \propto V^2 \longrightarrow I_{\mathrm{SH}} \propto N \propto V$$

Electro-optic Metasurface with $BaTiO_3$ Nanoparticles



As-deposited

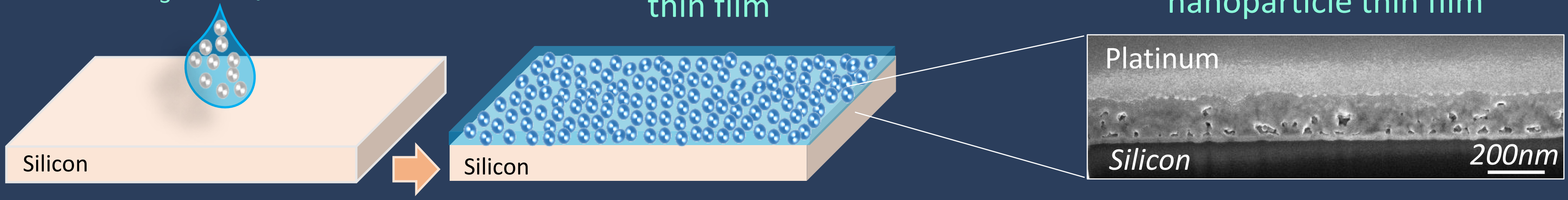
E field strength ~ 10MV/m

$BaTiO_3$ nanoparticles film and FIB nanostructuring

Drop cast & spin-coating of $BaTiO_3$ nanoparticles

$BaTiO_3$ nanoparticle thin film

SEM cross-section nanoparticle thin film



$BaTiO_3$ nps

477nm 430nm 307nm 246 nm *1 cm*

Decrease thickness

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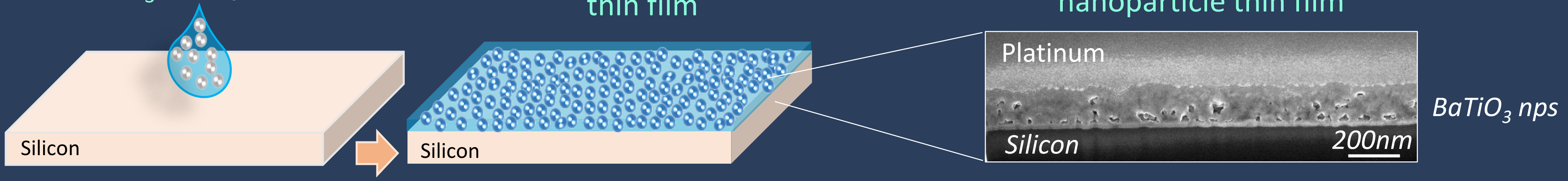
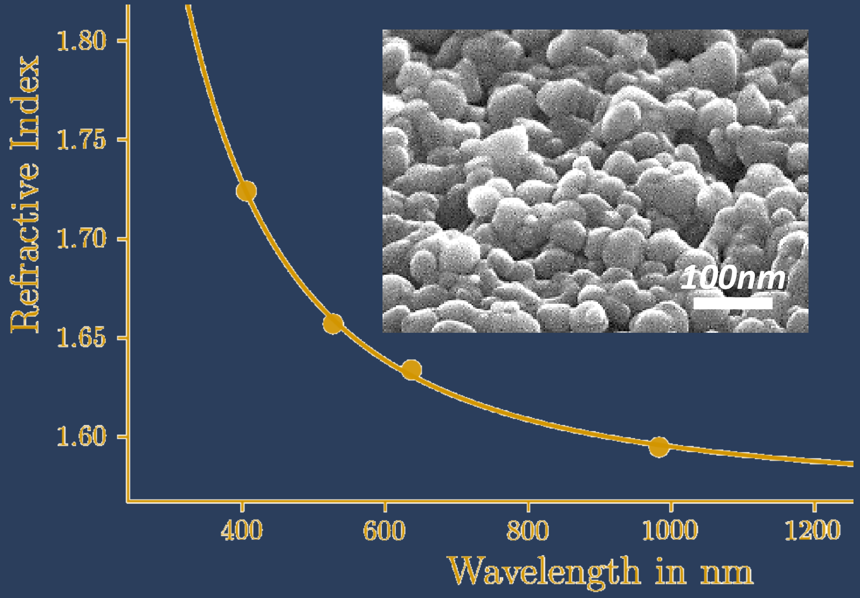
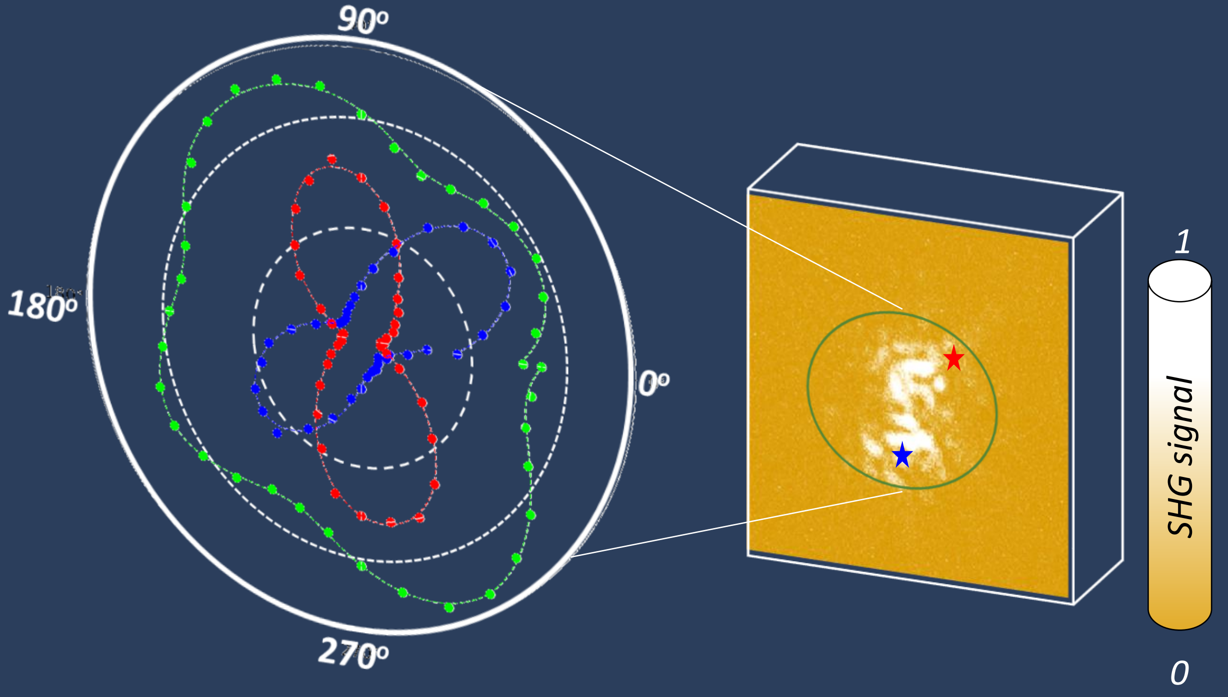


Linear optical properties



Nonlinear optical properties



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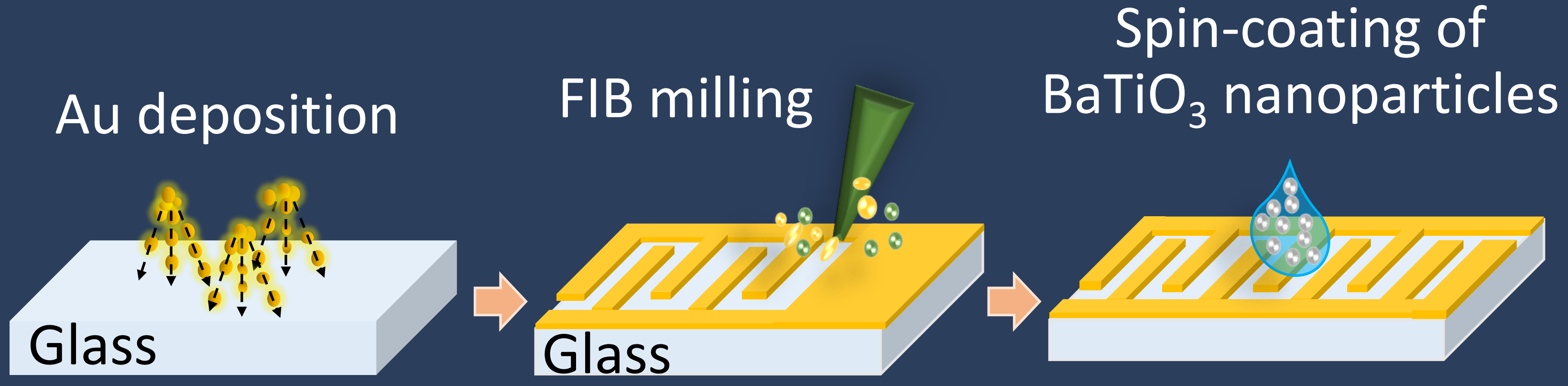
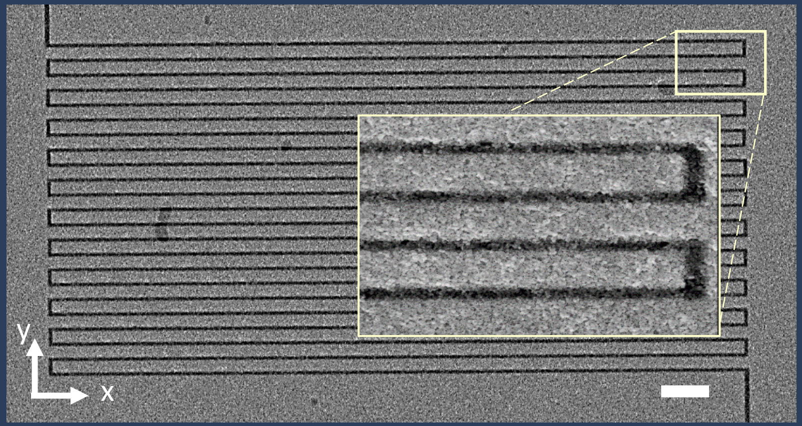
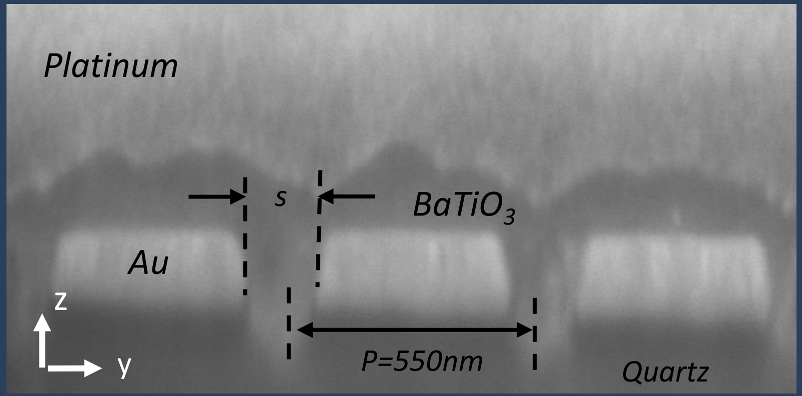
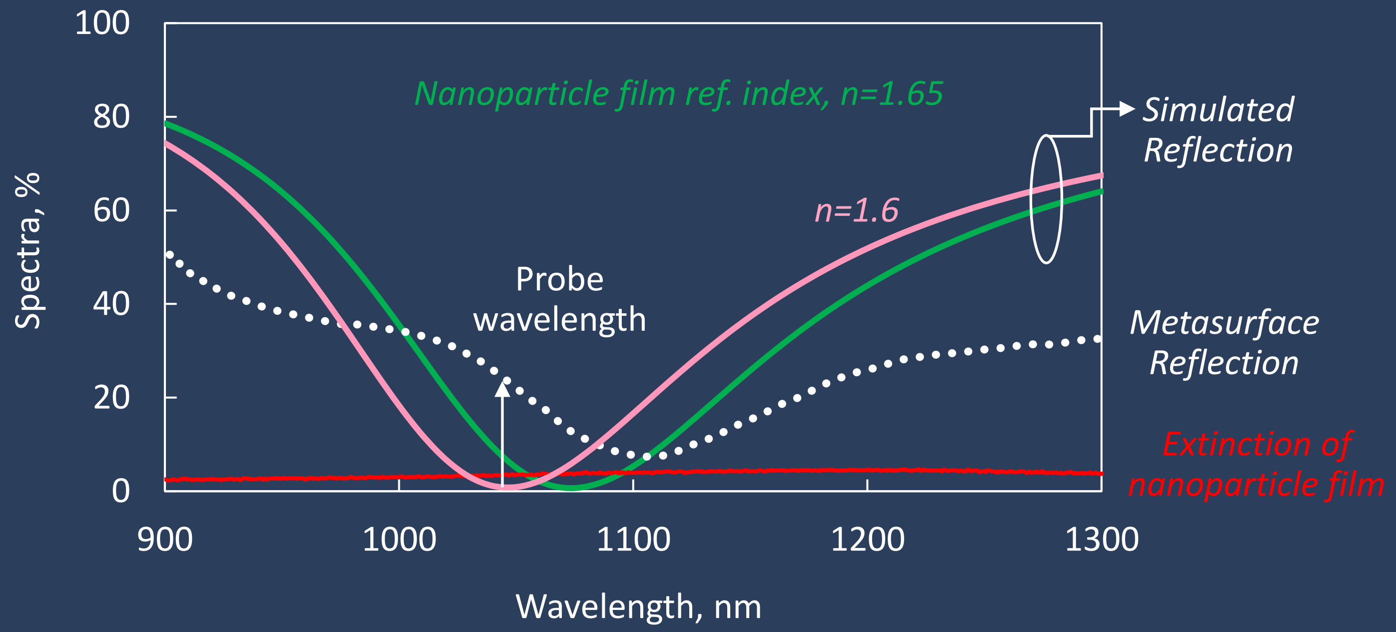




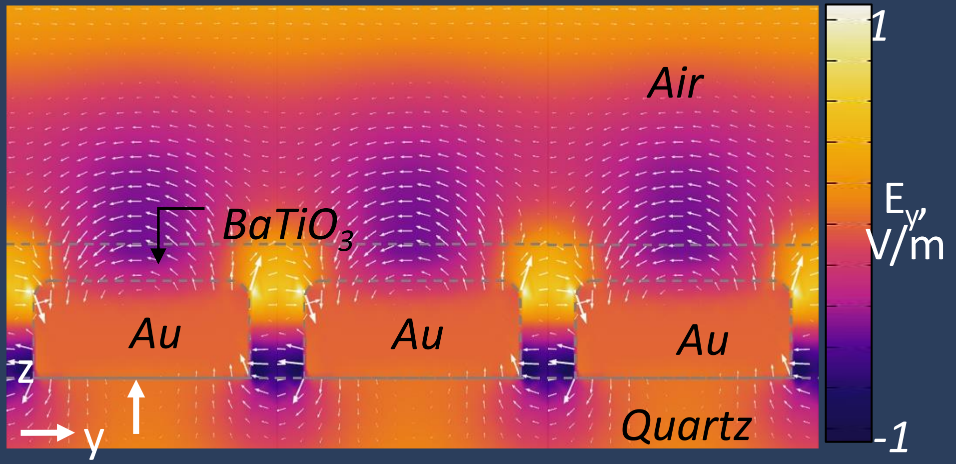




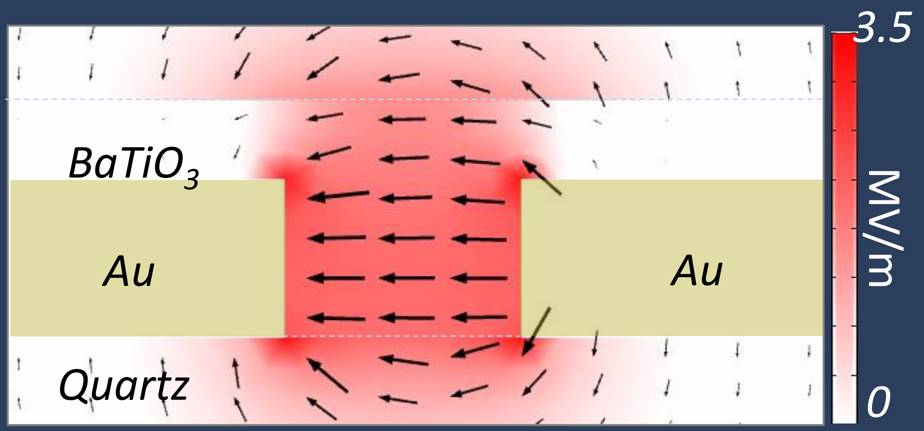
Karvounis, et al, Adv. Opt. Mat., 8, 17, 2020

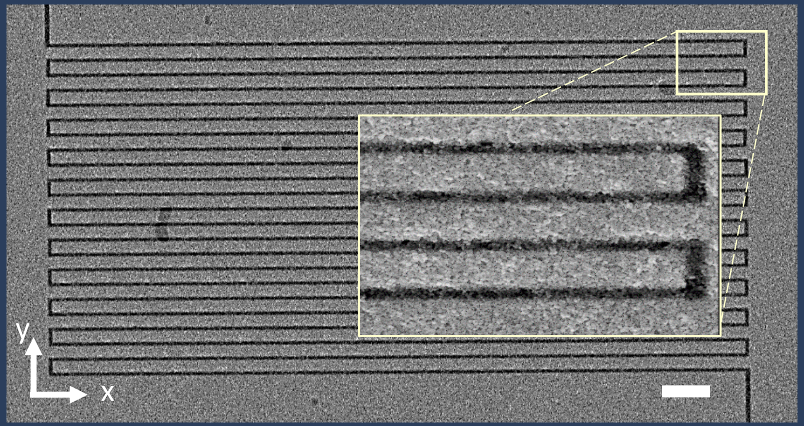
$BaTiO_3$ nanoparticles-based electro-optic metasurface

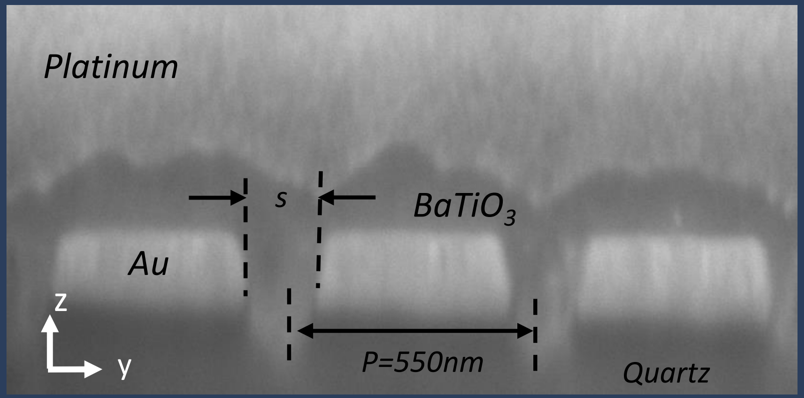
Optical field



Static electric field

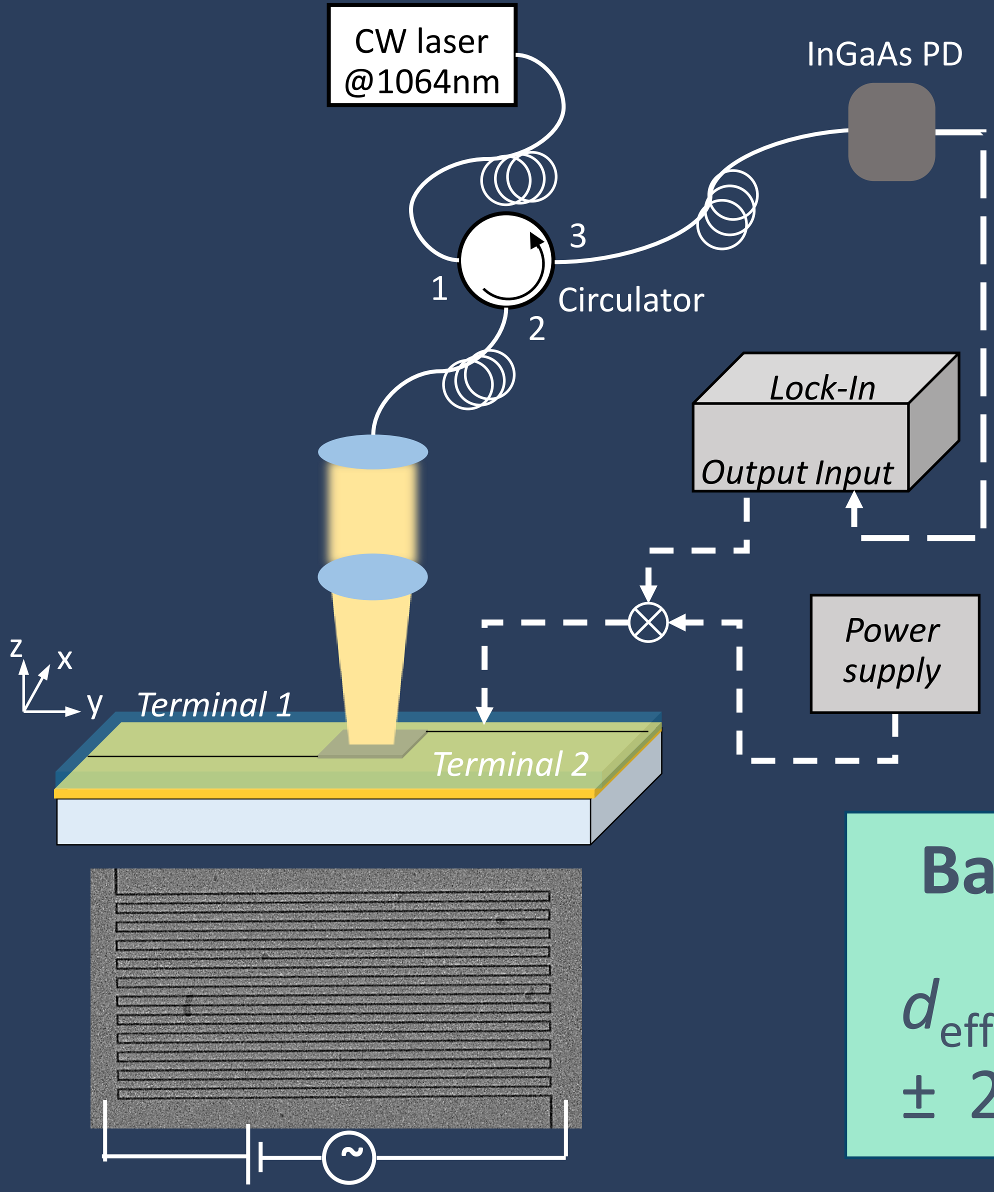


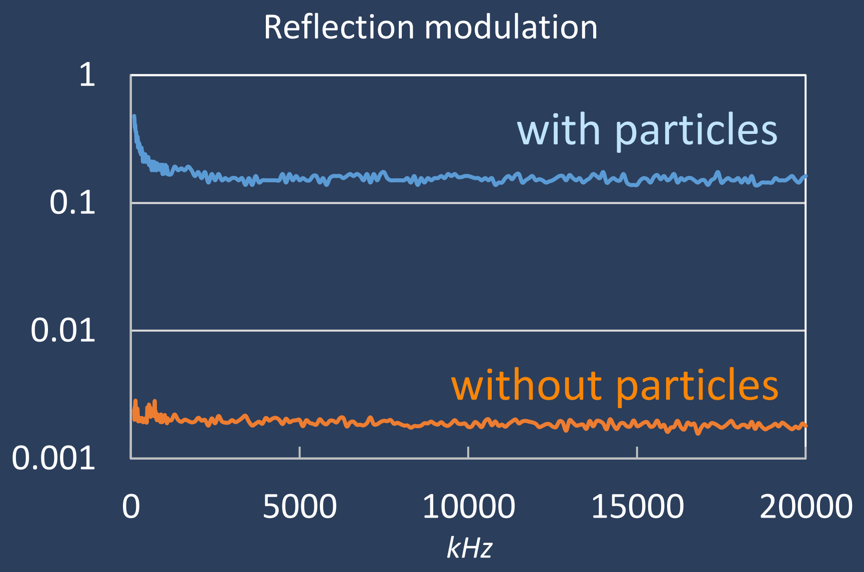


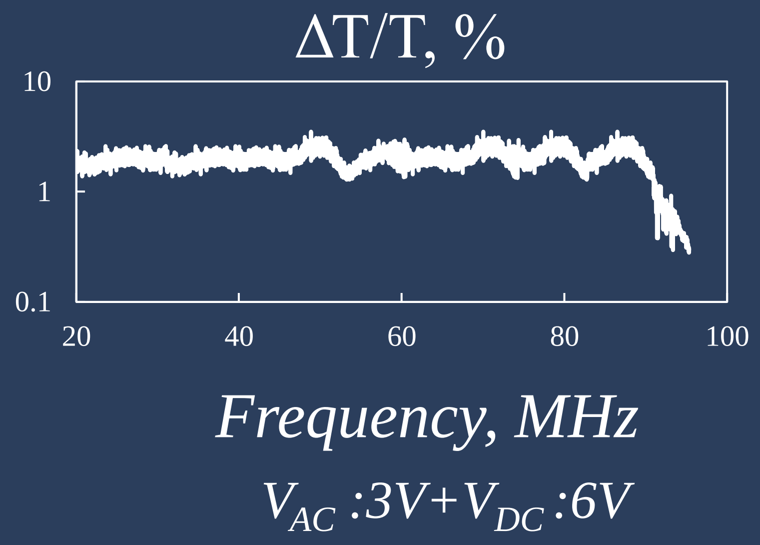


Karvounis, et al, Adv. Opt. Mat., 8, 17, 2020

$BaTiO_3$ nanoparticles-based electro-optic metasurface







$BaTiO_3$

d_{eff}= **37.04**
± 25.6 pm/V

Karvounis, et al, Adv. Opt. Mat., 8, 17, 2020

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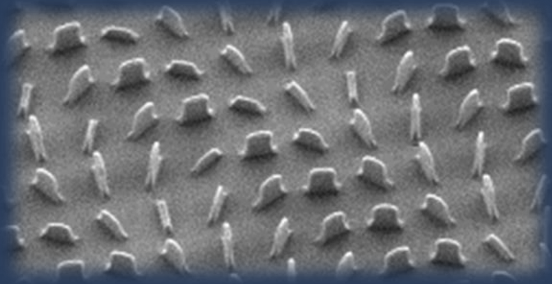
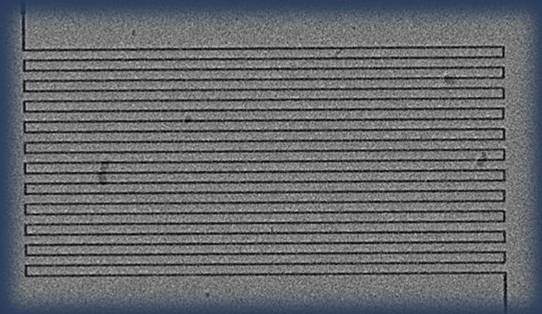
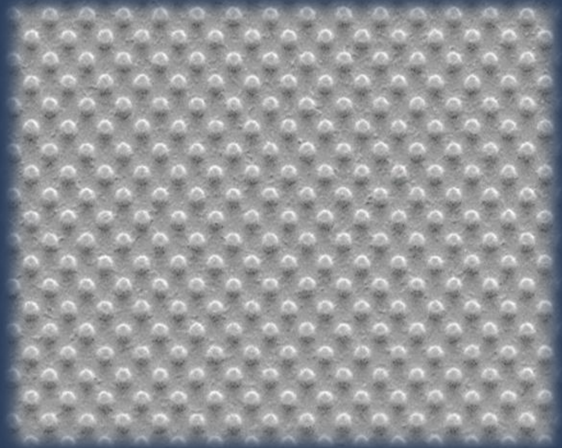
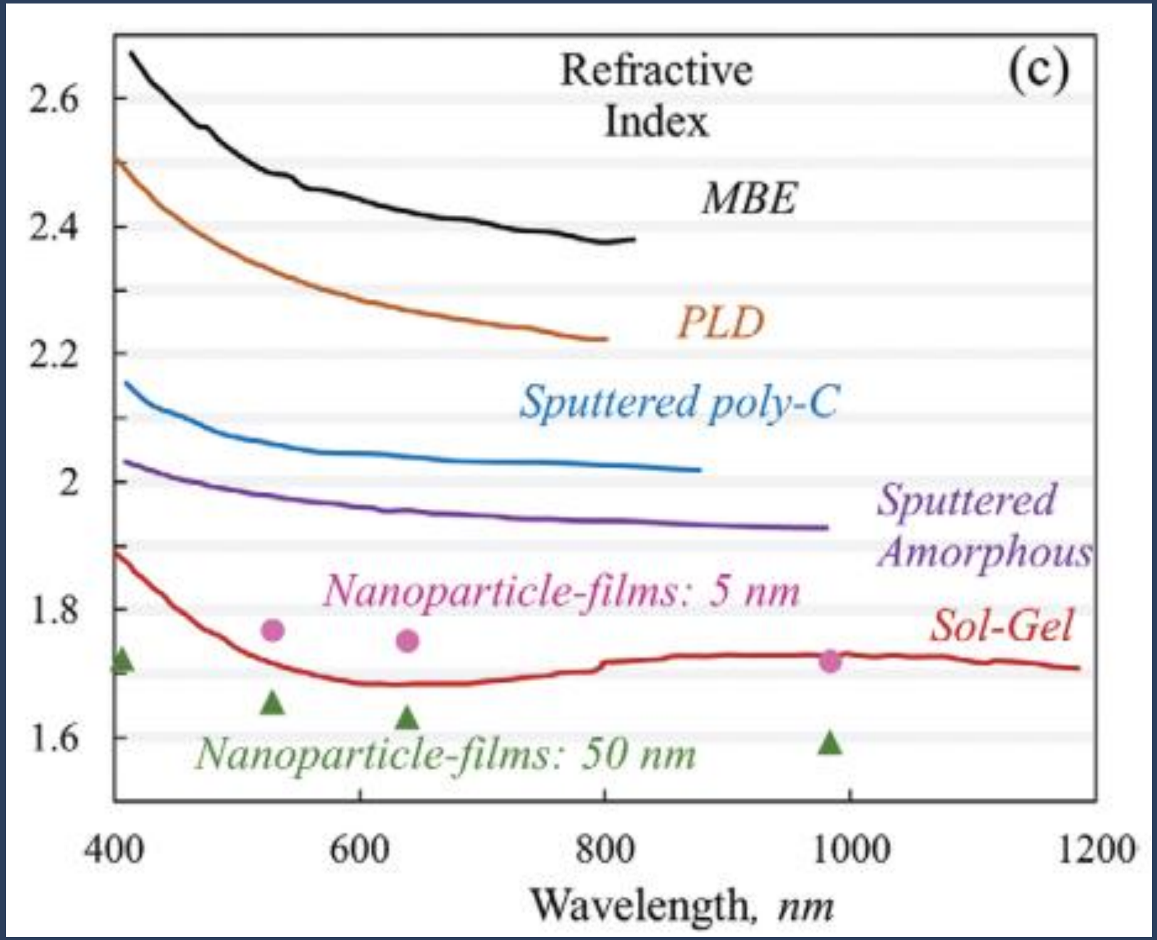


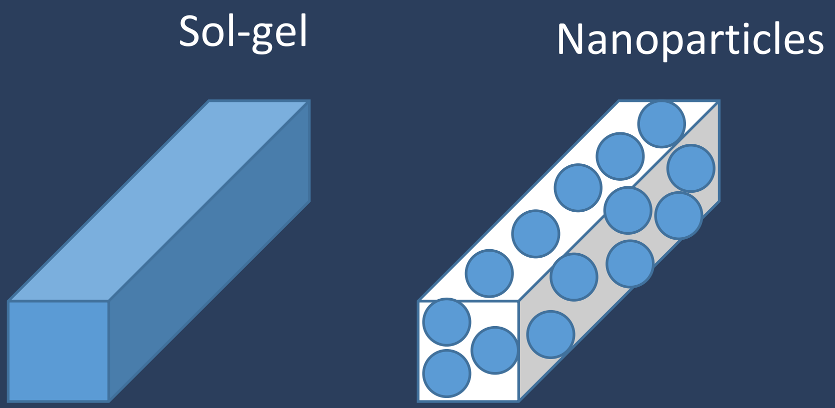




Sol-gel nanoimprint



A. Karvounis et al., Adv. Optical Mater. 2020, 8, 2001249



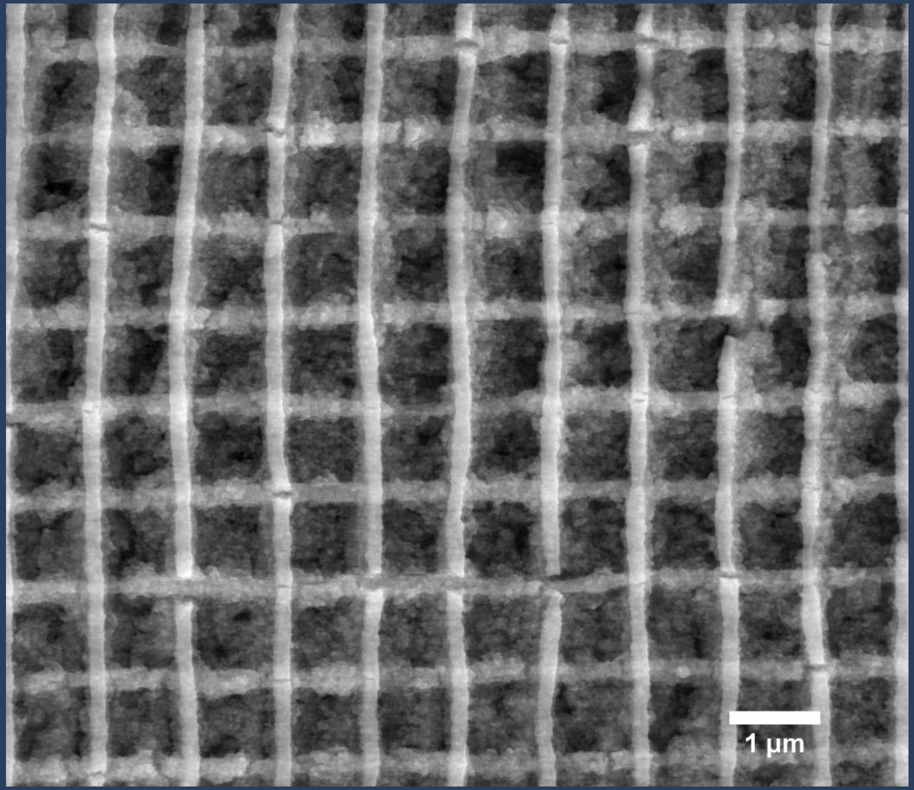
Less Scattering

Substrates have to stand 600°C

Higher refractive index

Soft Nanoimprint Lithography with nanoparticles

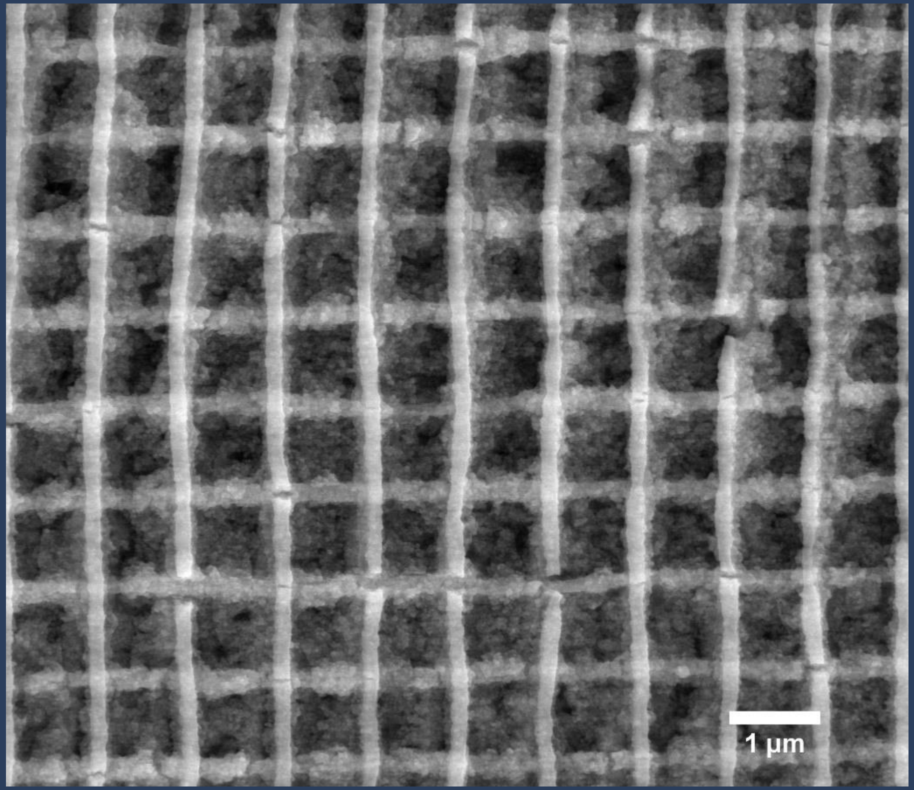
Solution-processed Barium Titanate Nonlinear
Woodpile Photonic Structures



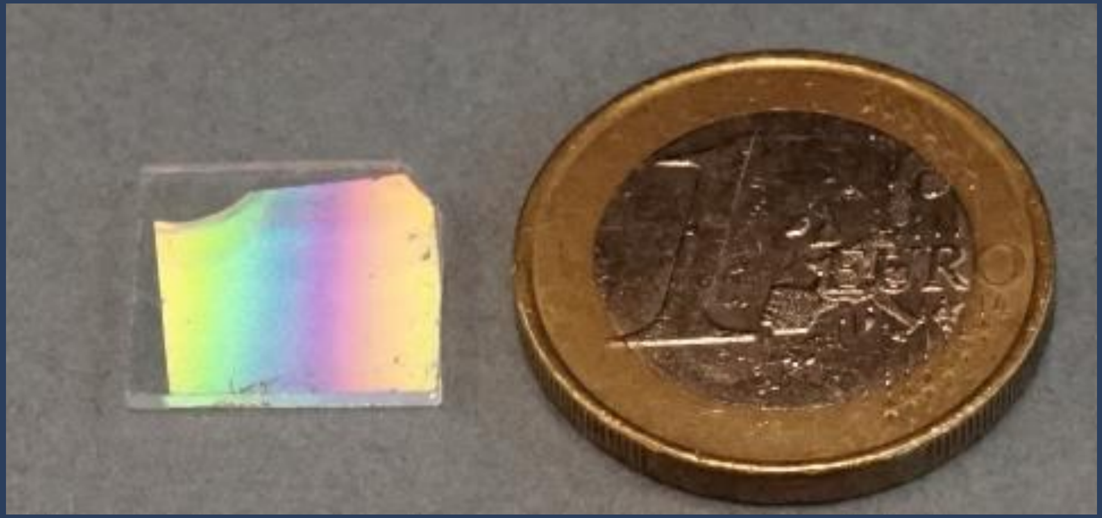
Vogler-Neuling, et al., *Phys. status solidi* **2020**, 1900755.

Soft Nanoimprint Lithography with nanoparticles

Solution-processed Barium Titanate Nonlinear Woodpile Photonic Structures



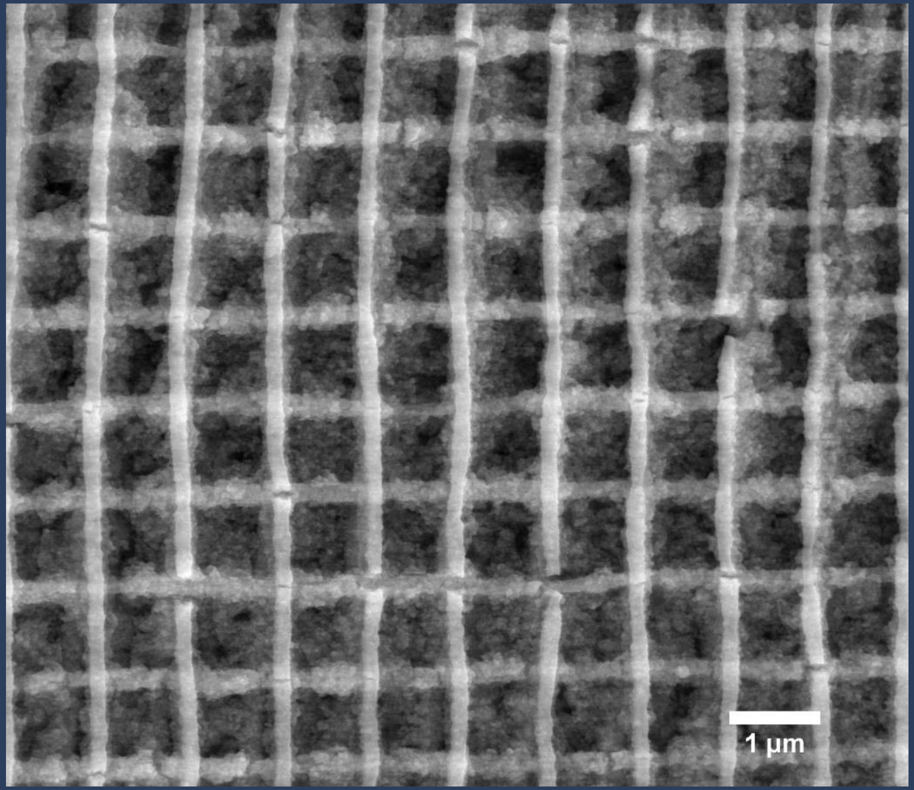
Very large surface area



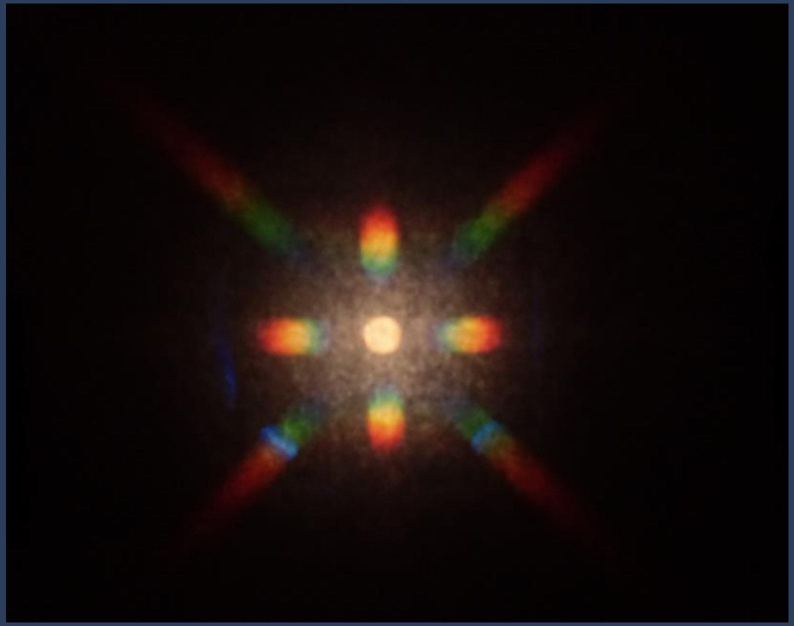
Vogler-Neuling, et al., *Phys. status solidi* **2020**, 1900755.

Soft Nanoimprint Lithography with nanoparticles

Solution-processed Barium Titanate Nonlinear Woodpile Photonic Structures



4 layers

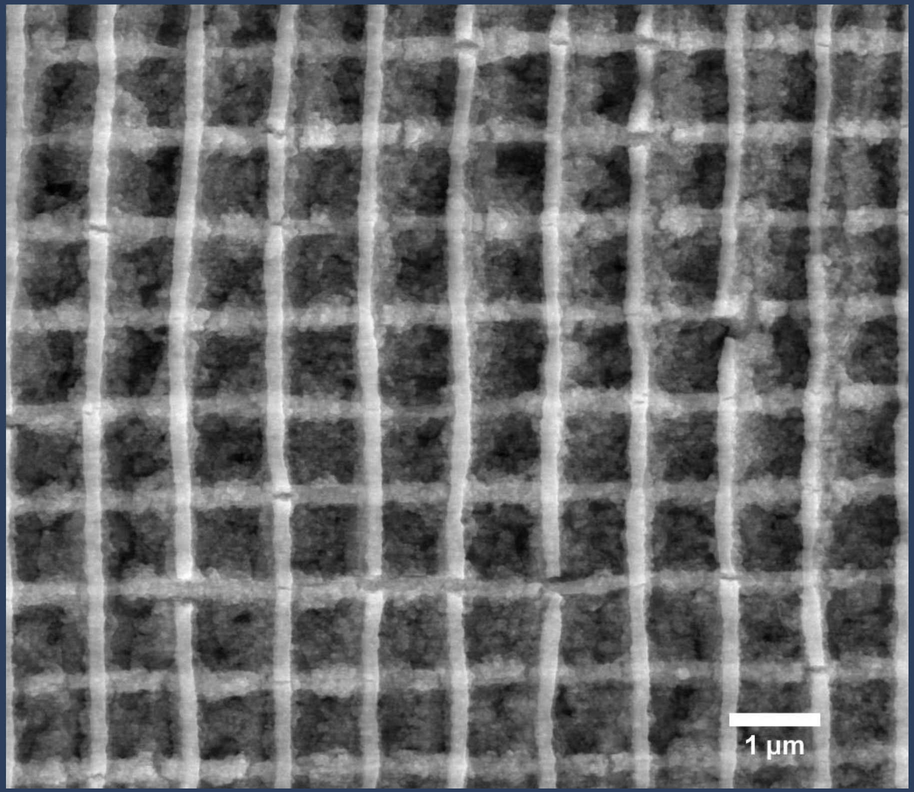


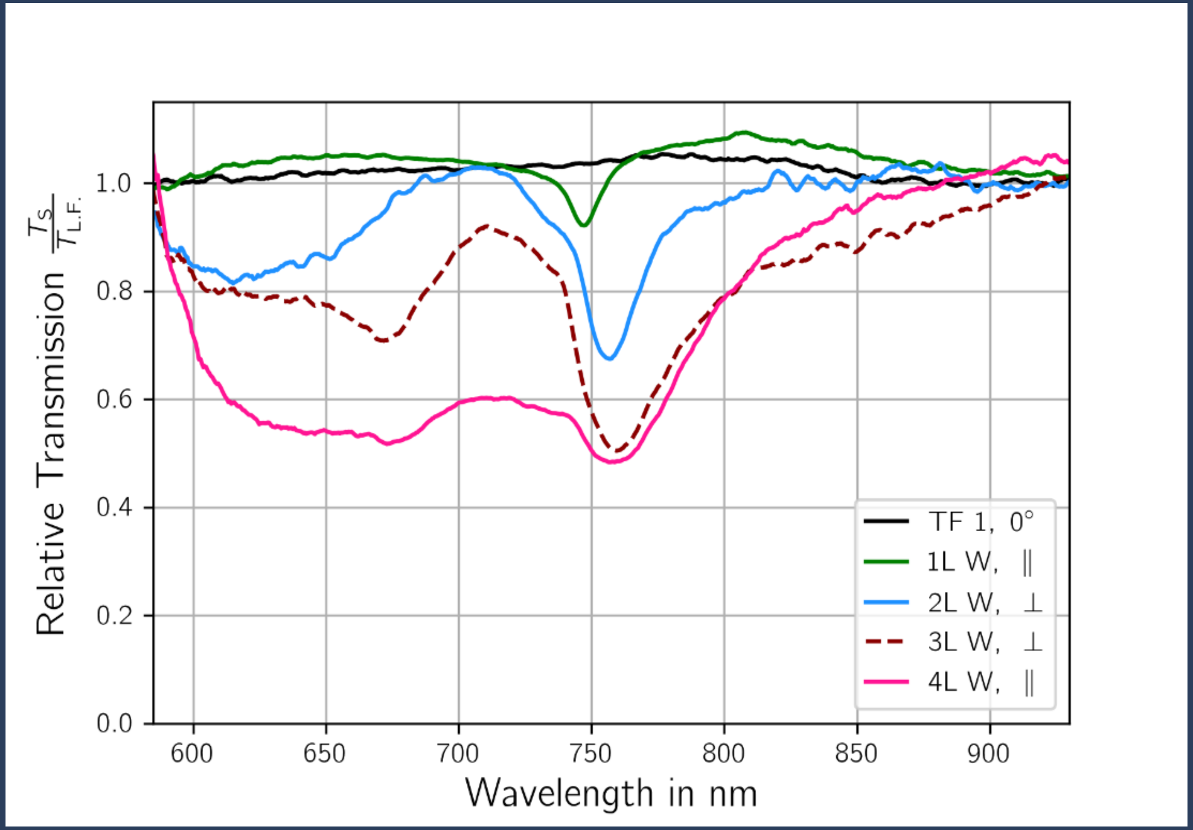
The diffraction pattern proves that there is an underlying cubic photonic crystal structure.

Vogler-Neuling, et al., *Phys. status solidi* **2020**, 1900755.

Soft Nanoimprint Lithography with nanoparticles

Solution-processed Barium Titanate Nonlinear Woodpile Photonic Structures

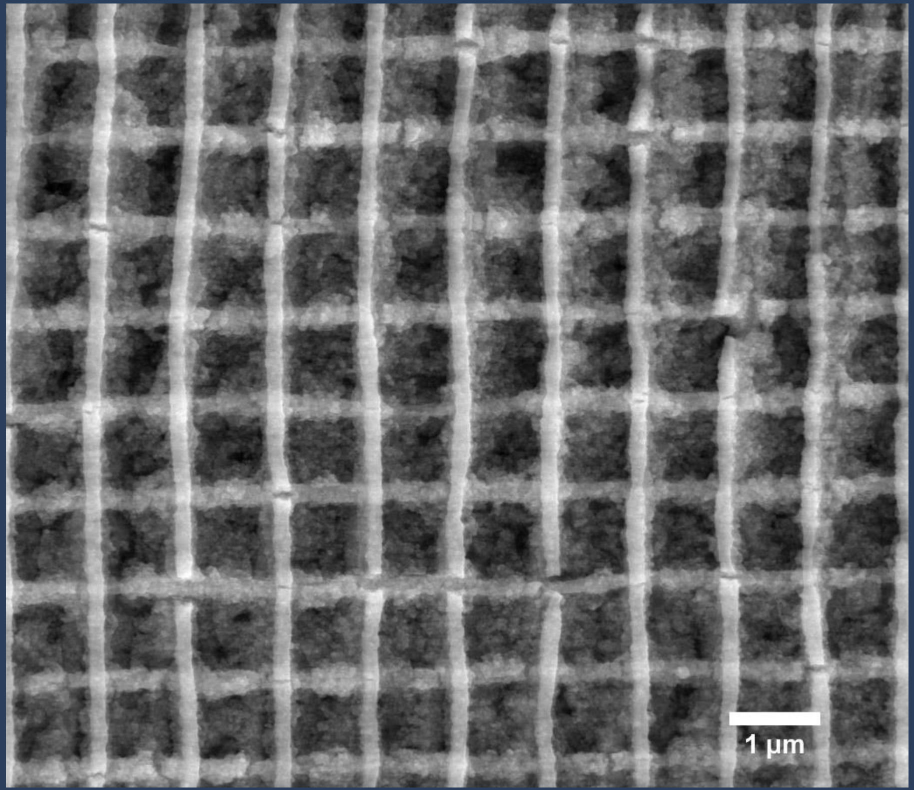


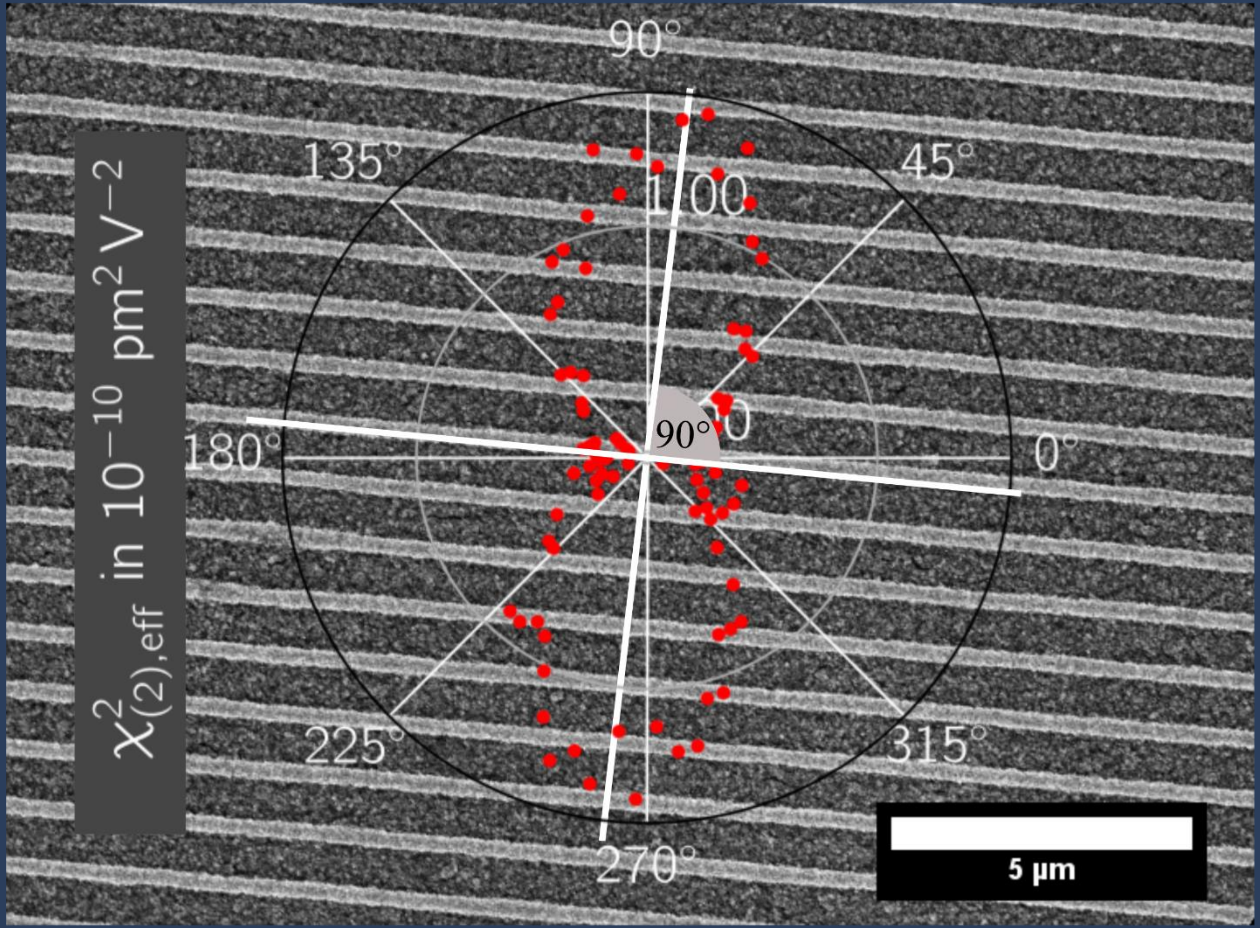


Vogler-Neuling, et al., *Phys. status solidi* **2020**, 1900755.

Soft Nanoimprint Lithography with nanoparticles

Solution-processed Barium Titanate Nonlinear Woodpile Photonic Structures





Vogler-Neuling, et al., *Phys. status solidi* **2020**, 1900755.

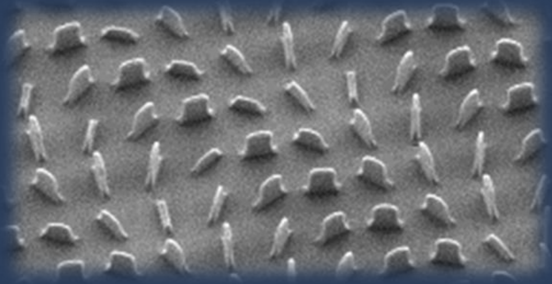
Outline

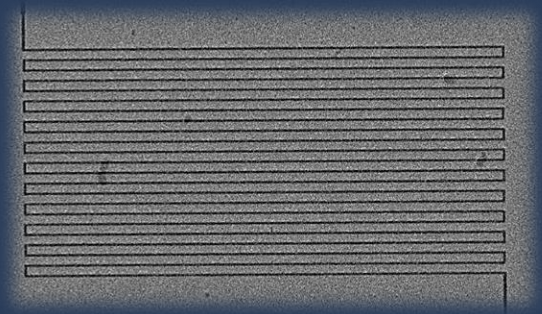
Miniaturizing $\chi^{(2)}$ materials

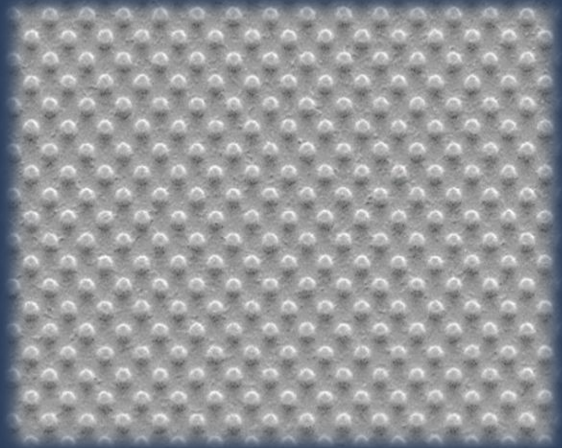
Nonlinear or electro-optic metasurfaces

- Pulsed laser deposited $BaTiO_3$
- FIB and spin coated nanoparticles
- Sol-gel nanoimprinted metalens

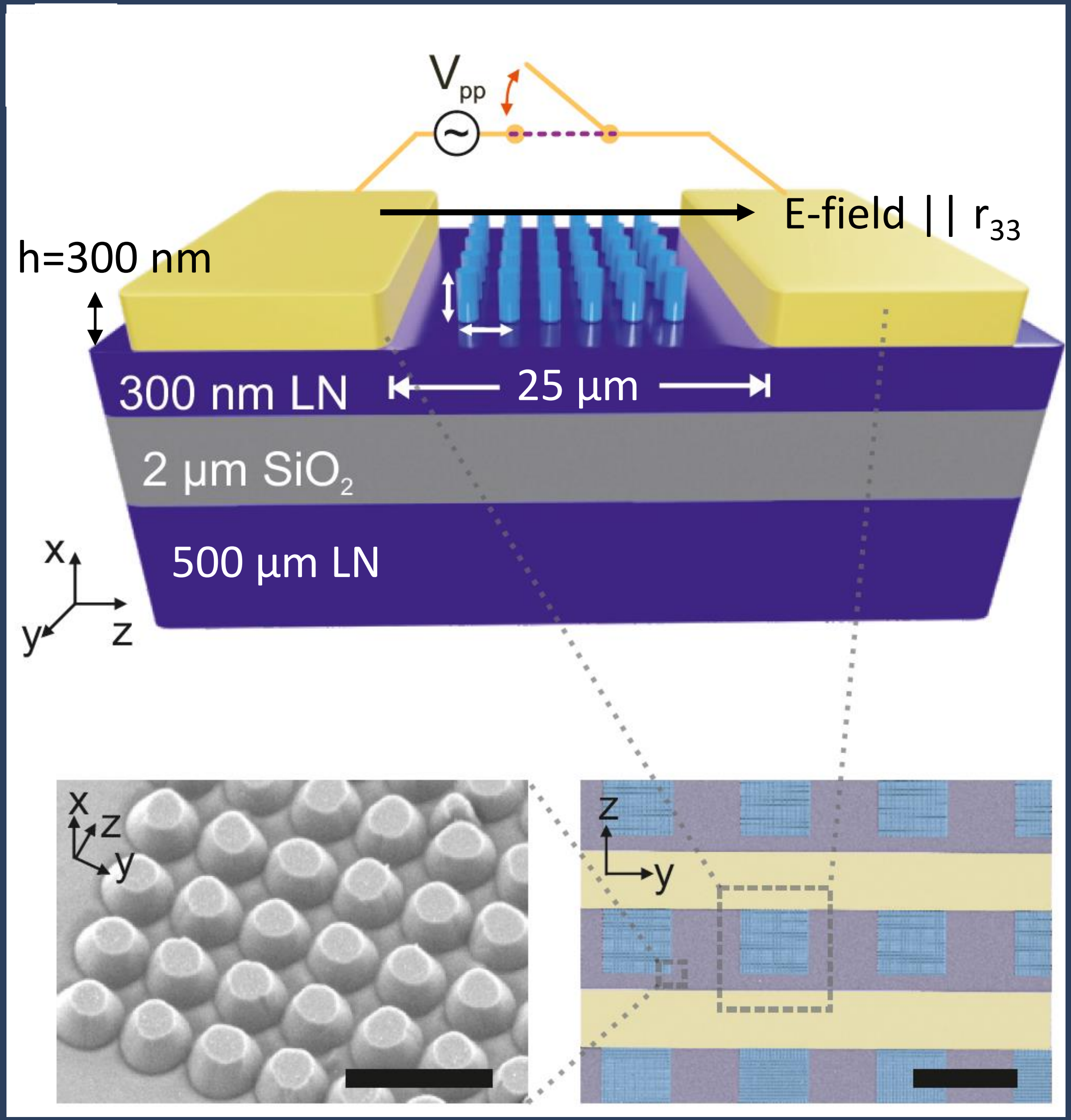
Miscellaneous photonic structures





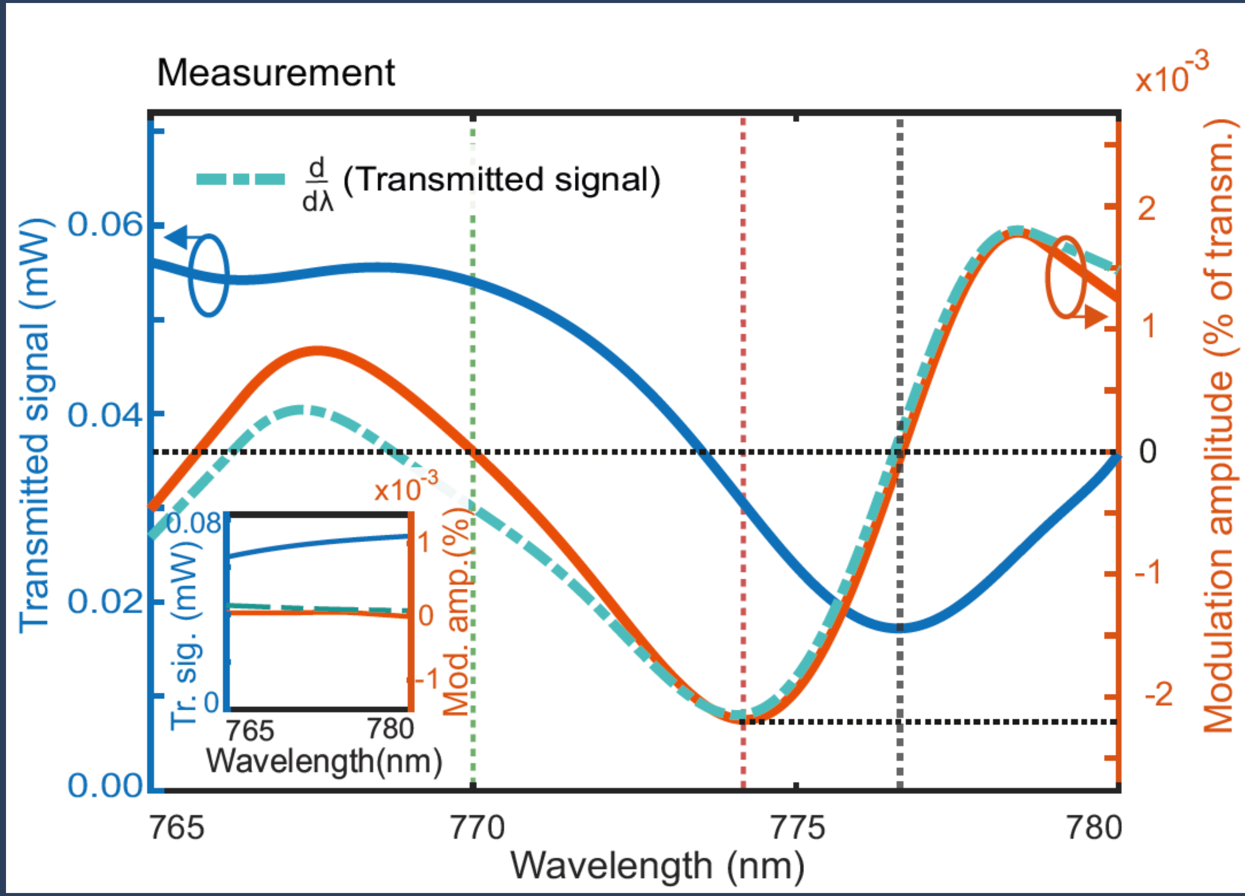


Electro-Optic Lithium Niobate Metasurfaces in the Visible



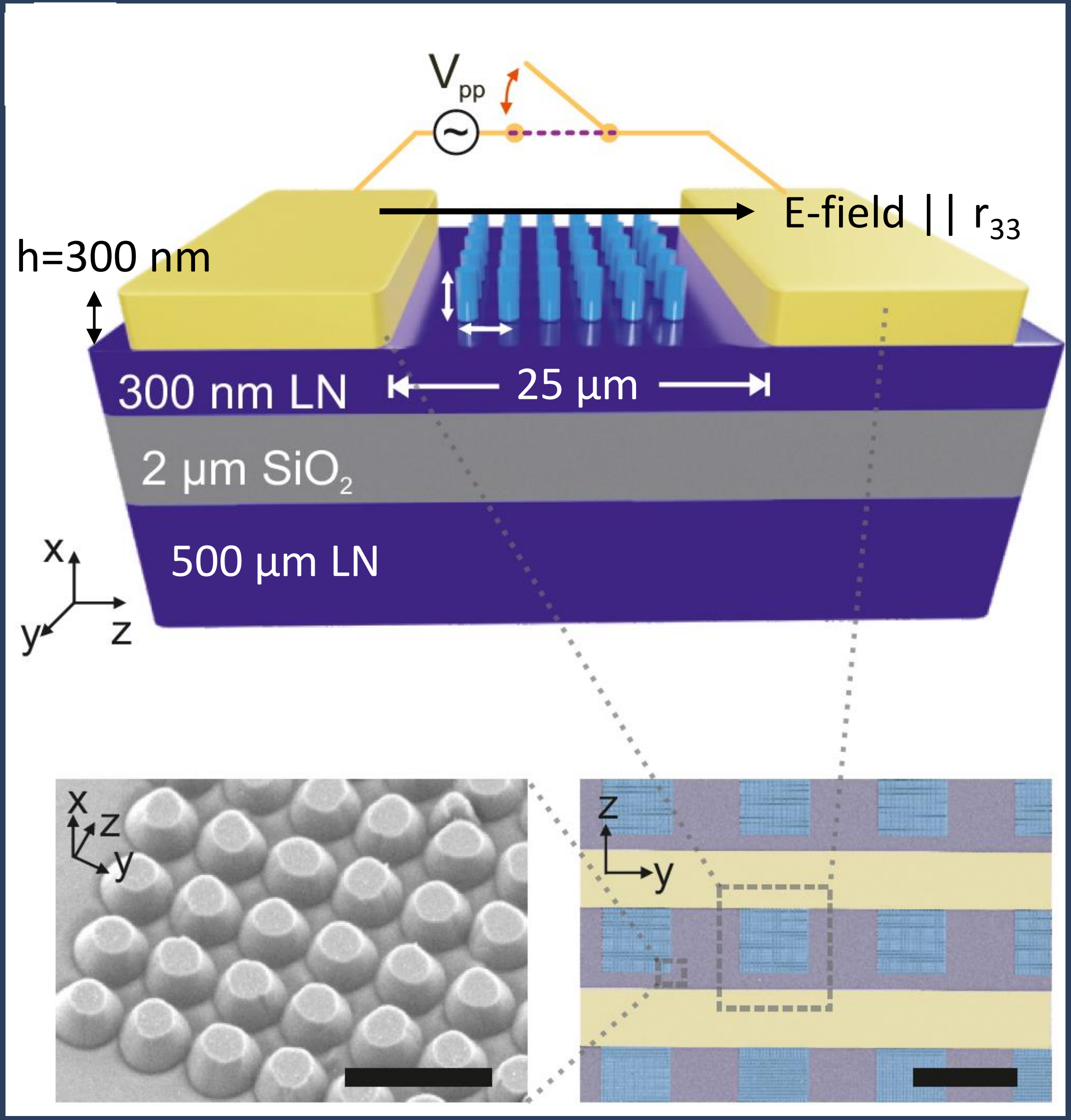
r= 89 nm, 113 nm, 135 nm, 154 nm

Enhancement of modulation amplitude **by 2 orders of magnitude** compared to the substrate



Weigand, Vogler-Neuling et al. Arxiv: http://arxiv.org/abs/2106.12232

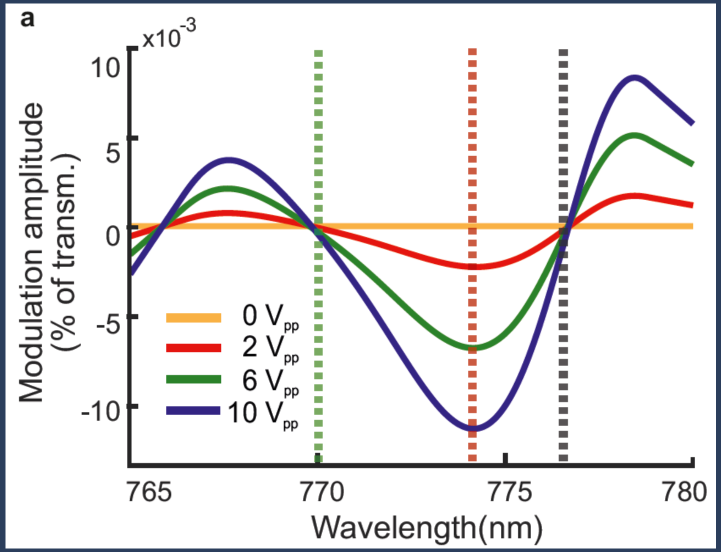
Electro-Optic Lithium Niobate Metasurfaces in the Visible

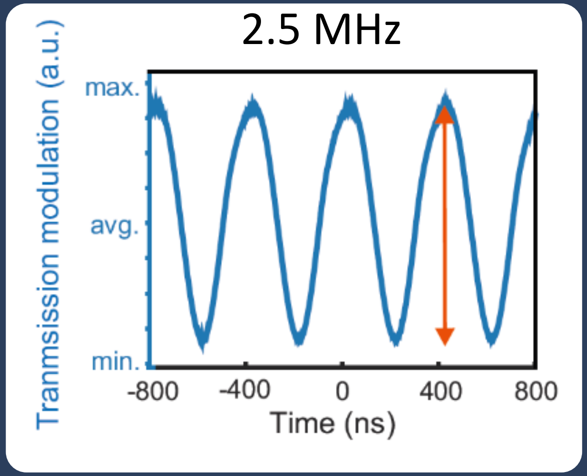


r= 89 nm, 113 nm, 135 nm, 154 nm

Modulation based on **linear electro-optic effect** enhanced at the resonance for **AC voltages below 1 V_{pp}**.

(Compatible with CMOS micro-controllers)
Modulation speeds of **2.5 MHz** could be detected





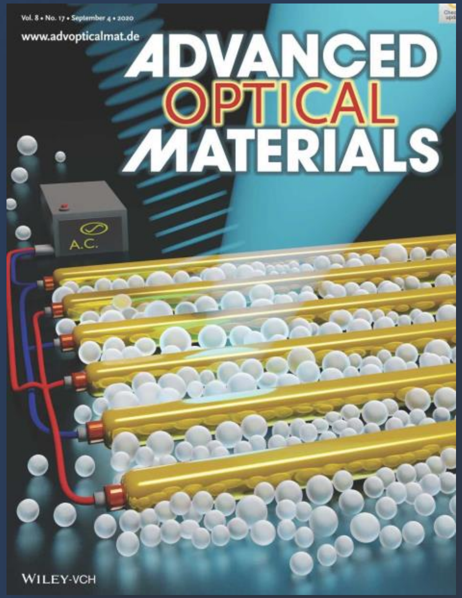
Weigand, Vogler-Neuling et al. Arxiv: http://arxiv.org/abs/2106.12232

Conclusion

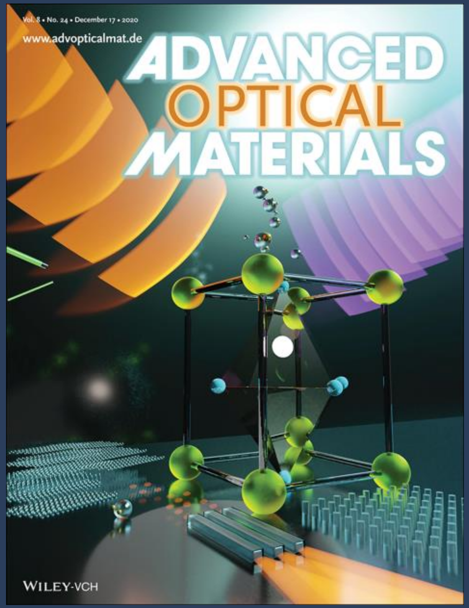
Original assemblies and the material quest is not over

→ Relaxing fabrication and new materials

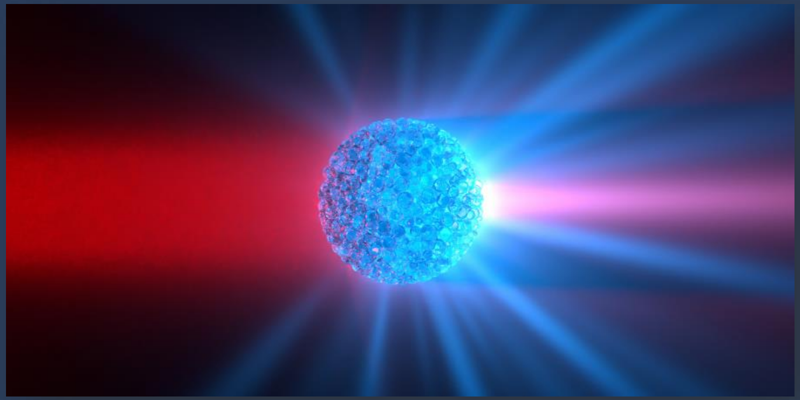
→ Not only SHG but electro-optic



Karvounis, et al, Adv. Opt. Mat., 8, 17, 2020

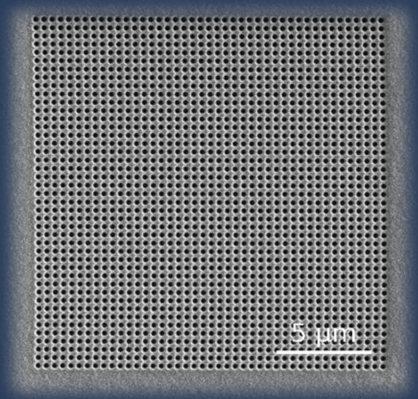


REVIEW: Karvounis et al, Adv. Opt. Mat., Nov. 20 doi.org/10.1002/adom.202001249

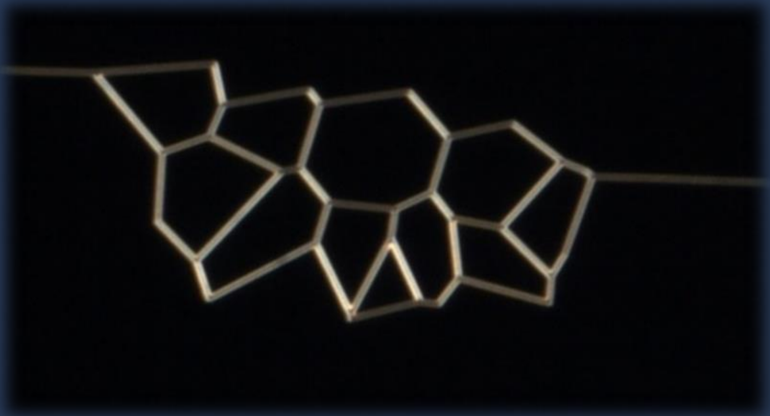


Savo *et al. Nat. Photonics* **14,** 740–747 (2020)

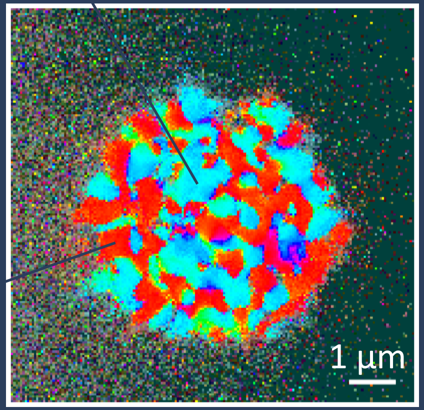
Outlook



Bottom up metasurface



Random network



Corrosion imaging

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