(Non-)reciprocity and nanotechnology: demonstration of a nonlinear optical diode

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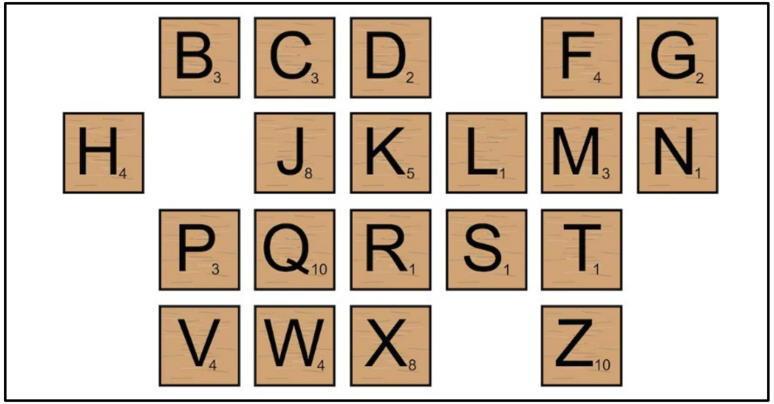
EPFL

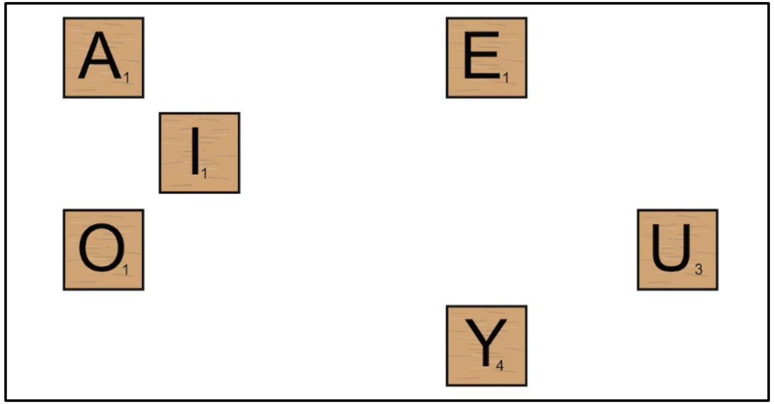

DOUBLE WORD
TRIPLE LETTER
DOUBLE LETTER
DOUBLE LETTER
DOUBLE LETTER
DOUBLE LETTER
DOUBLE LETTER
TRIPLE LETTER
S 1
C 3
R 1
A 1
B 3
B 3
L 1
E 1

Using the entire alphabet

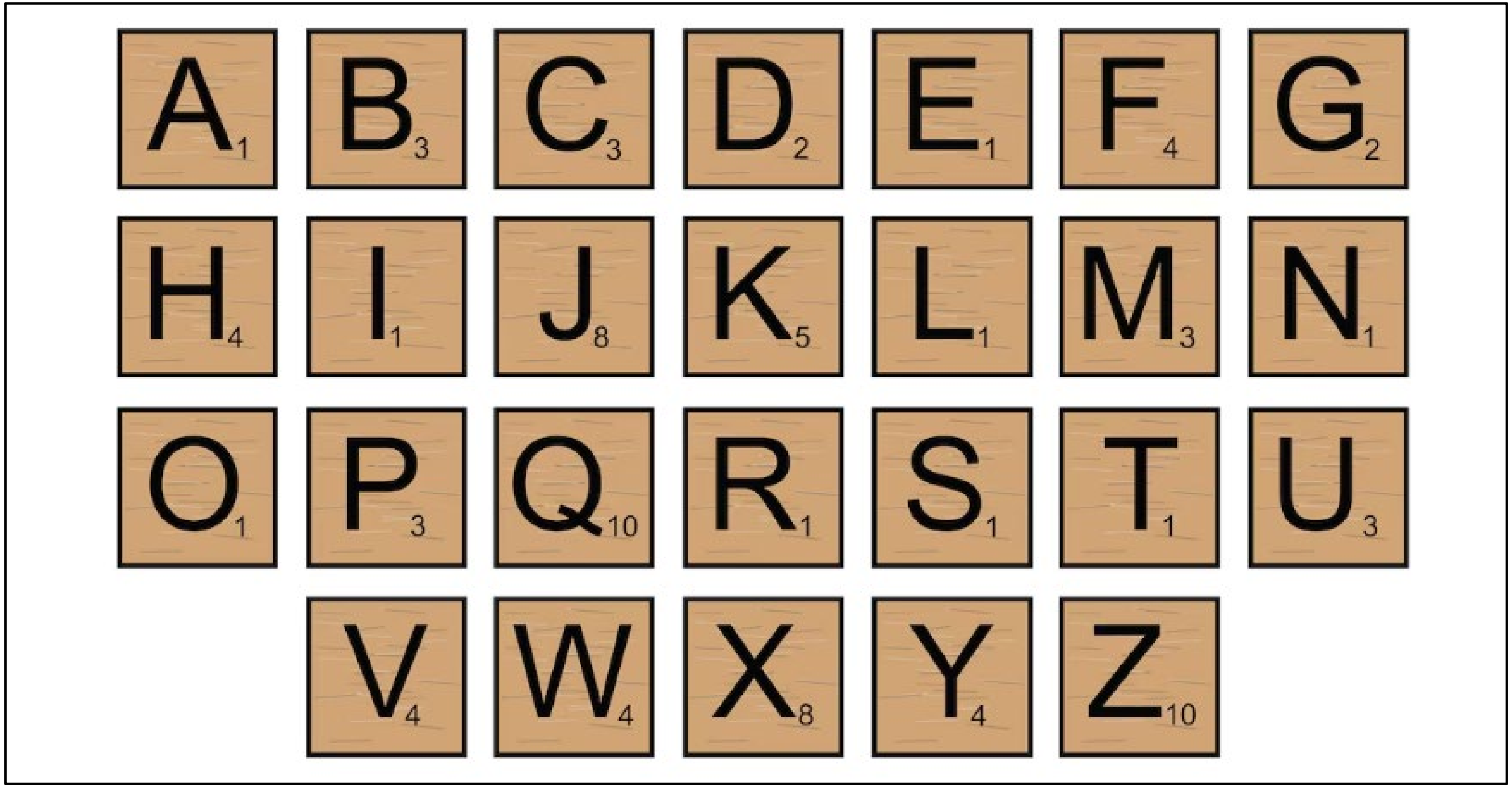
Set 1



Set 2



Using the entire alphabet



Using the entire electromagnetic alphabet

$$\nabla \times \mathbf{E}(\mathbf{r},t) = -\frac{\partial \mathbf{B}(\mathbf{r},t)}{\partial t}$$

$$\nabla \times \mathbf{H}(\mathbf{r},t) = \frac{\partial \mathbf{D}(\mathbf{r},t)}{\partial t} + \mathbf{J}(\mathbf{r},t)$$

$$\nabla \cdot \mathbf{D}(\mathbf{r},t) = \rho(\mathbf{r},t)$$

$$\nabla \cdot \mathbf{B}(\mathbf{r},t) = 0$$

$$\begin{pmatrix} \mathbf{p} \\ \mathbf{m} \end{pmatrix} = \begin{pmatrix} \underline{\underline{\alpha}}_{ee} & \underline{\underline{\alpha}}_{em} \\ \underline{\underline{\alpha}}_{me} & \underline{\underline{\alpha}}_{mm} \end{pmatrix} \cdot \begin{pmatrix} \mathbf{E} \\ \mathbf{H} \end{pmatrix}$$

magneto-electric coupling terms (bianisotropy)

K. Achouri, Advanced Photonics vol. 5, p. 046001 (2023)

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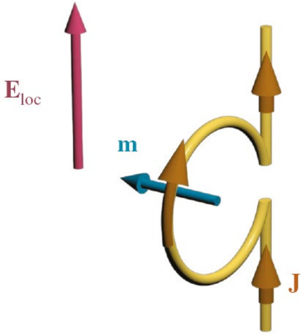
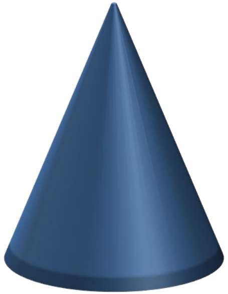
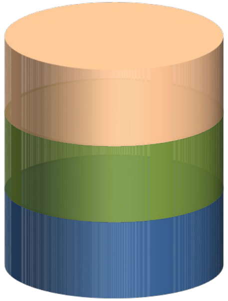






- Strong bianisotropy requires breaking the symmetry of the system…
- … including in the out-of-plane direction…
- … which is very challenging for nanotechnology !
- It can also be achieved by using simple shapes built from different materials!

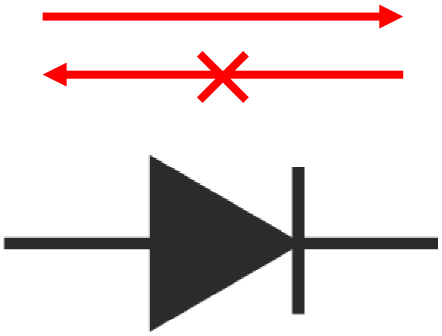
K. Achouri, Advanced Photonics vol. 5, p. 046001 (2023)

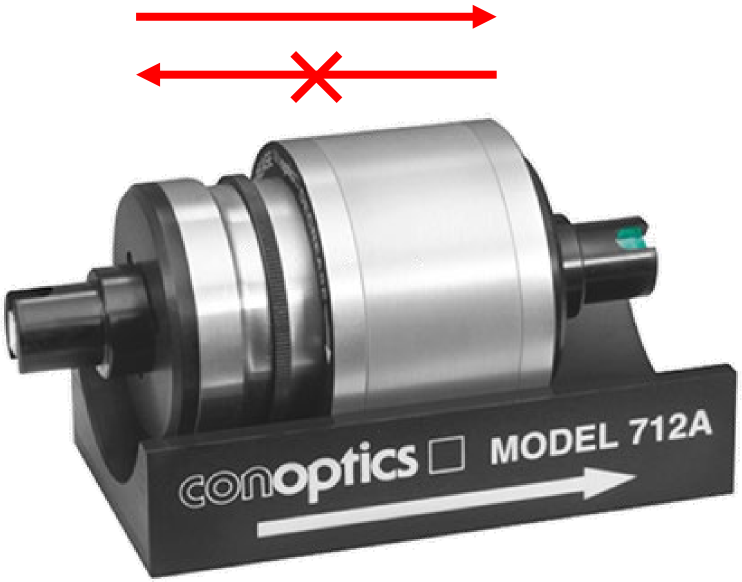
Outline

- Introduction
- Nonreciprocal devices
- Second harmonic generation in plasmonic systems
- Putting numerical techniques to good use
- Nanotechnologies
- Nonlinear optical diode
- Is it non-reciprocal?
- Summary

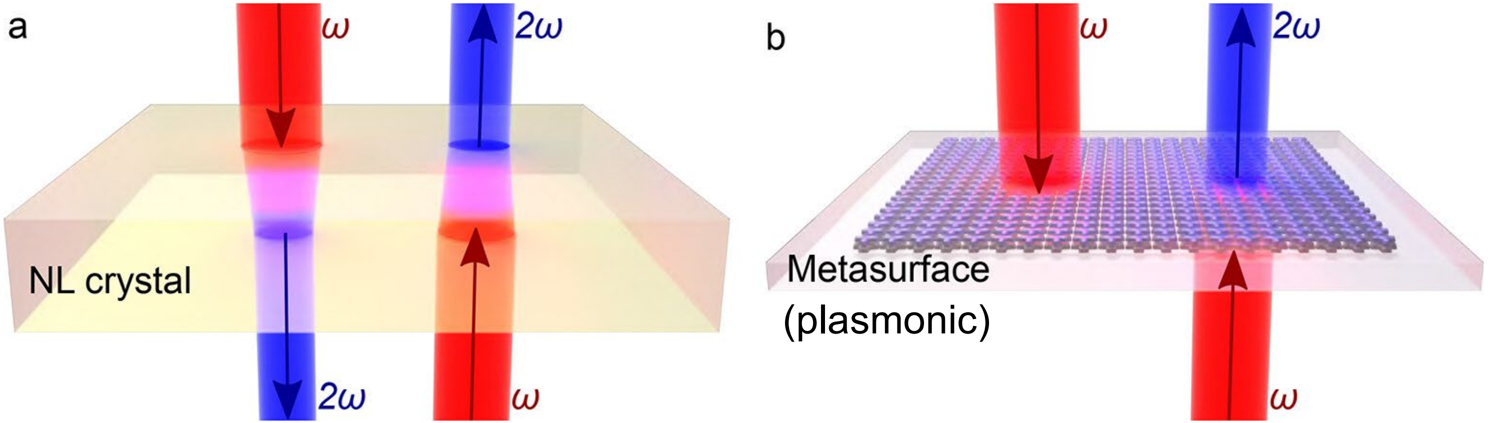
Nonreciprocal devices

- Allow the signal to pass only in one direction
- Are key for signal processing
- In optics, require an external time-odd bias, such as a magnetic field



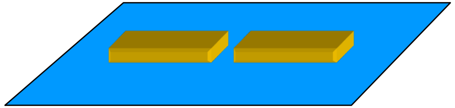


Asymmetric second harmonic generation (SHG)

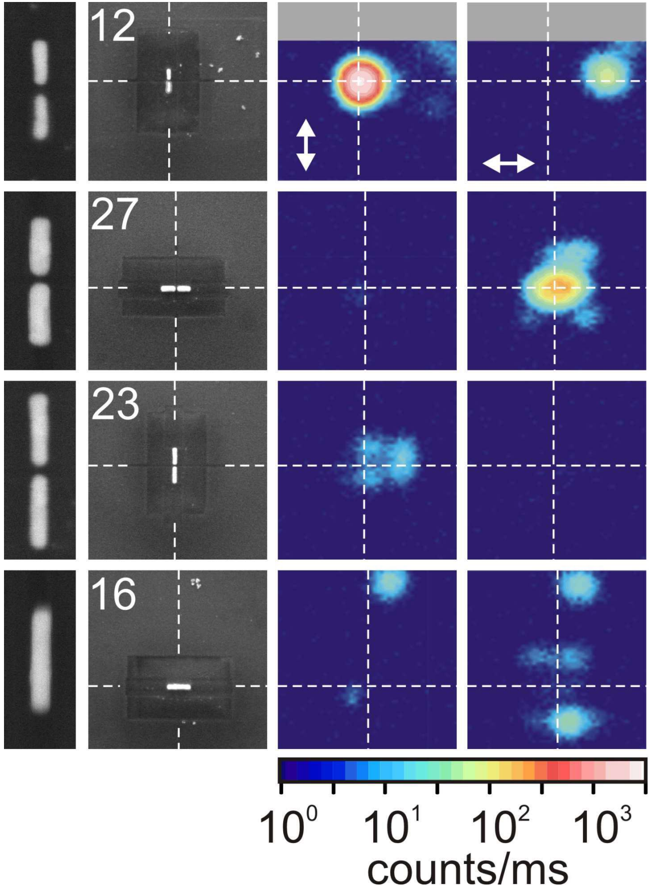


S. Boroviks, Nano Letters, vol. 23, p. 3362 (2023)

Plasmonic dipole antenna

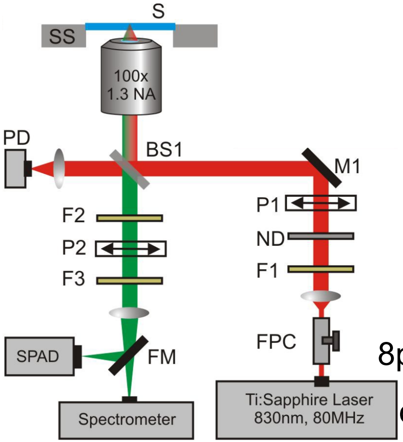


Focused ion beam fabrication:
- 40nm thick gold
- Length: 190…400nm
- Width: 45nm
- Gap: 20nm
- Glass substrate

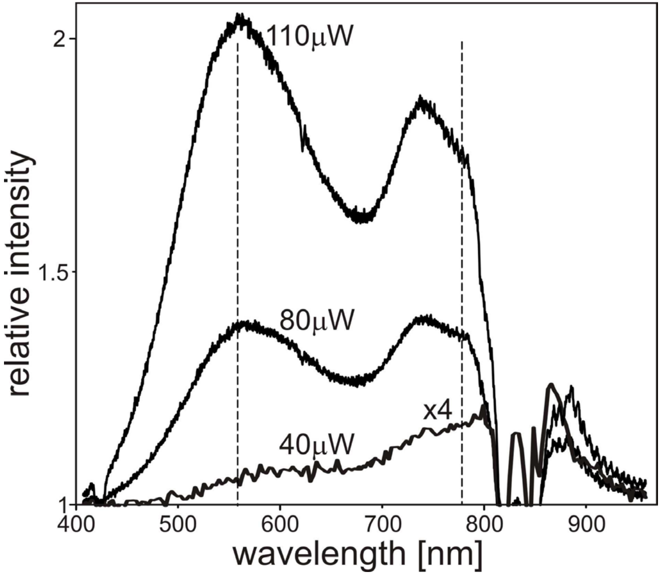


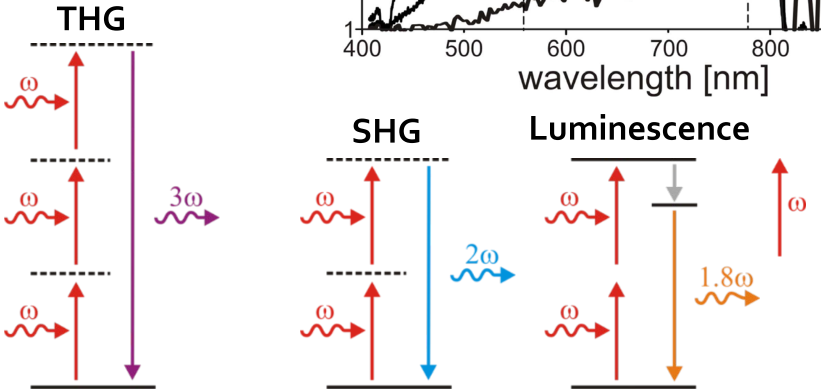
F. Mühlschlegel et al. Science vol. 308, p. 1607 (2005)

Plasmonic dipole antenna – Nonlinear effects



8ps pulse duration





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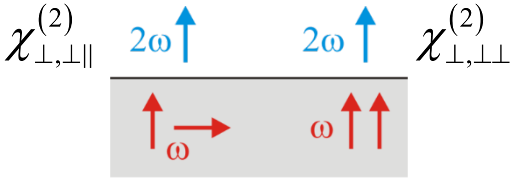
F. Mühlschlegel et al. Science vol. 308, p. 1607 (2005)

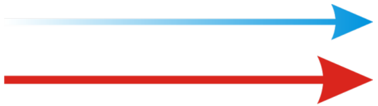
Second order susceptibility tensor

$$P_\alpha(2\omega) = \varepsilon_0 \chi^{(2)}_{\alpha,\beta\gamma} E_\beta(\omega) E_\gamma(\omega)$$

- **In the bulk:** $\alpha, \beta, \gamma = x, y$ or z
- **At an interface:** $\alpha, \beta, \gamma = \|$ or $\perp$



- **For plasmonic metals, the $\chi^{(2)}_{\perp,\perp\perp}$ component dominates**
- **Classical SHG requires phase matching between the waves at ω and 2ω that travel together, which is complicated and requires an anisotropic crystal and control of polarization:**



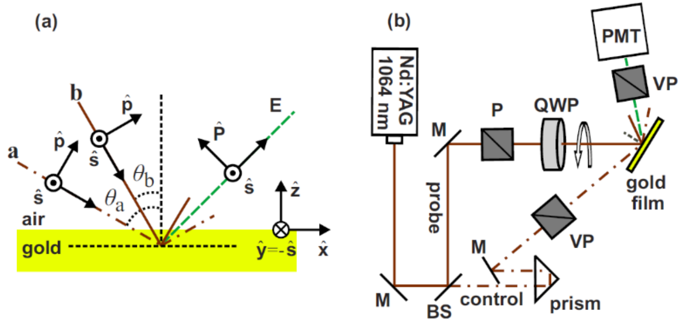
$$P_\alpha(2\omega) = \chi^{(2)}_{\alpha,\beta\beta} E_\beta(\omega) E_\beta(\omega)$$

- **This is (generally) not the case for SHG in plasmonic nanostructures ("zero phase matching")**

J. Butet, ACS Nano vol. 9, p. 10545 (2015)

SHG: Surface vs bulk contributions

- **Measurement of bulk and surface contributions:**



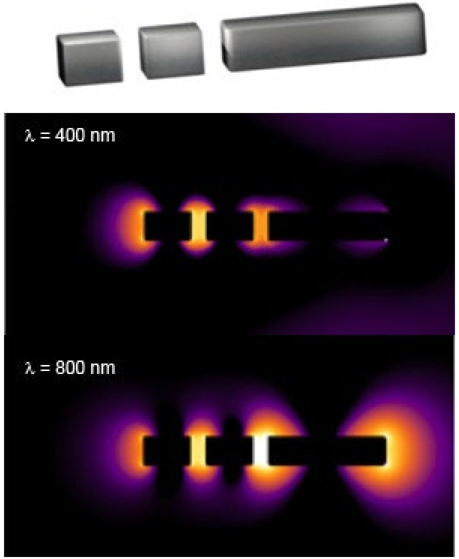
Contribution to the measured SH signals				Character
f_{ppp}	f_{pss}	f_{sps}	f_{sps}	
1.3				Surface and bulk
1.5	1.0			Surface and bulk
3.5		2.2	0.6	Surface only
0.004		0.2	0.1	Bulk only

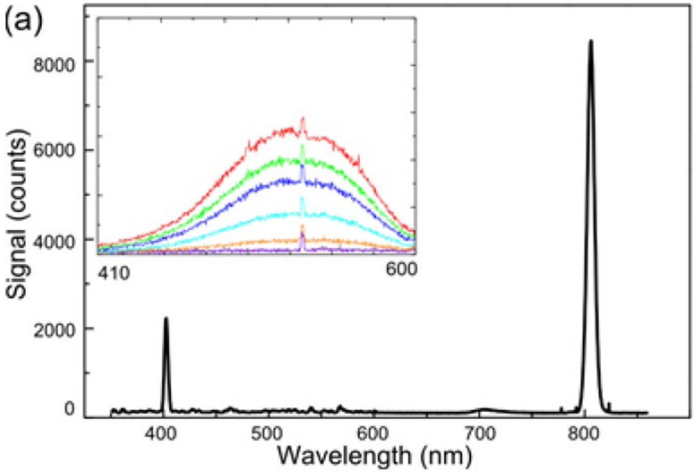
- **For plasmonic metals the surface contributions dominate over the bulk; there are however specific experimental conditions, where $\chi^{(2)}_{\text{bulk}}$ cannot be neglected**

F. X. Wang, Physical Review B vol. 80, p. 233402 (2009)

Double resonant antenna and second harmonic generation

- **A plasmonic nanostructure with two resonances: at the fundamental and at the second harmonic frequency**

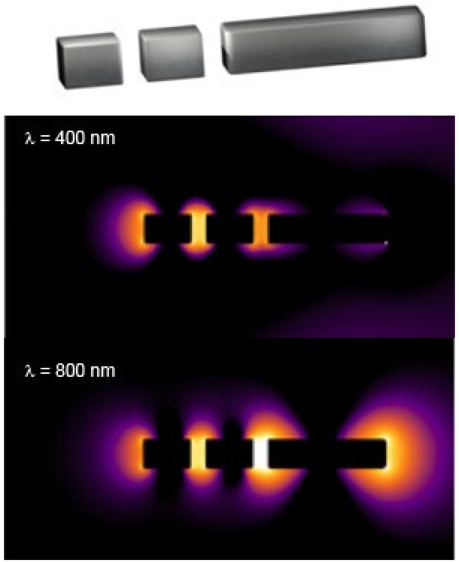


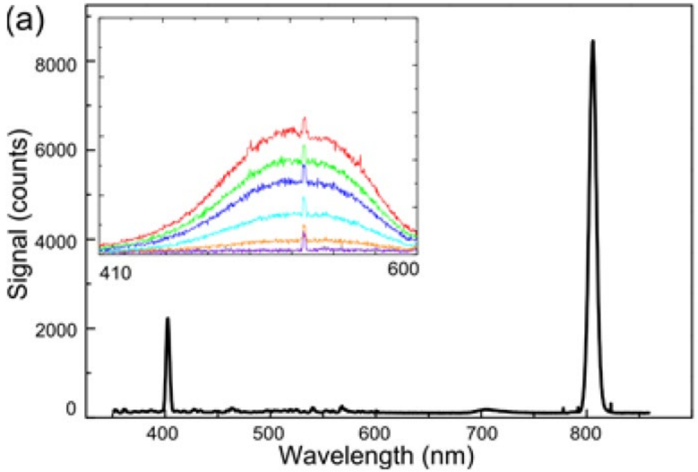


K. Thyagarajan, Optics Express, vol. 20, p. 12860 (2012)

Double resonant antenna and second harmonic generation

- **How does SHG work in such a structure?**
- **What is the role of the different parts in the antenna?**





K. Thyagarajan, Optics Express, vol. 20, p. 12860 (2012)

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- Nonreciprocal devices
- Second harmonic generation in plasmonic systems
- **Putting numerical techniques to good use**
- Nanotechnologies
- Nonlinear optical diode
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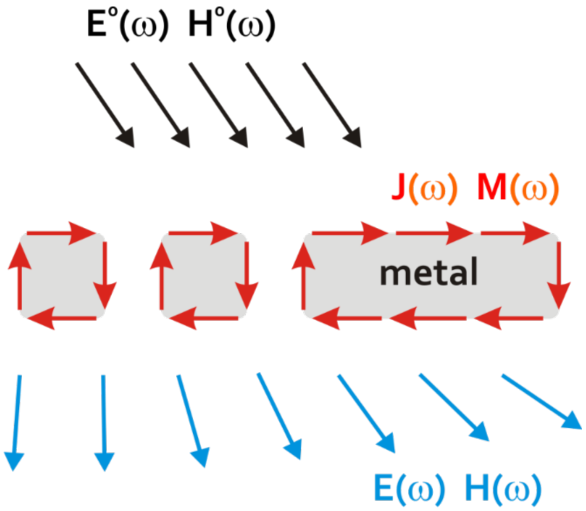
Review on numerical methods for nanophotonics/plasmonics

B. Gallinet, J. Butet, and O.J.F. Martin
Laser & Photonics Reviews vol. 9,
p. 577 (2015)

Surface integral equation (SIE) method – Linear regime

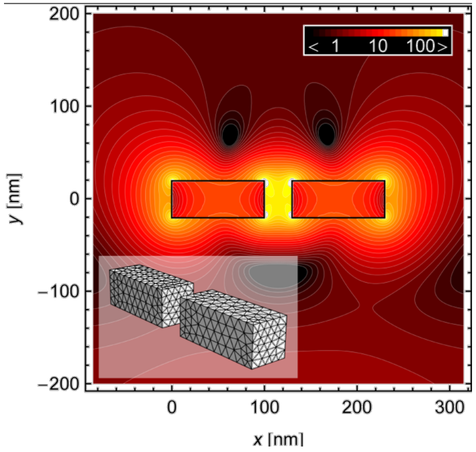


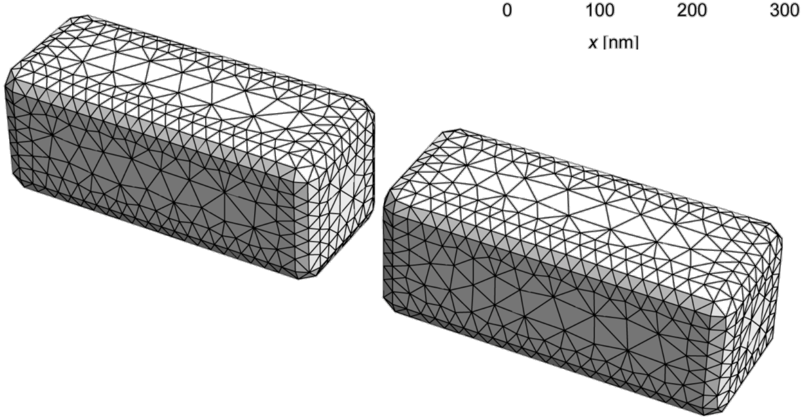
- Light scattered is computed by means of surface currents on the scatterers

A.M. Kern, J. Optical Soc. America A vol. 26, p. 732 (2009)

Modelling plasmonic structures accurately

- Finite elements
  surface integral formulation:
  - Fast
  - Versatile
  - Very accurate
  - Useful for comparison
    with experiments

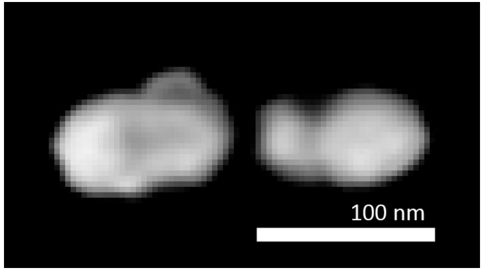


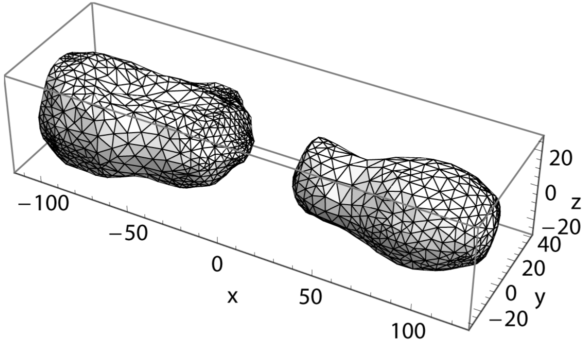


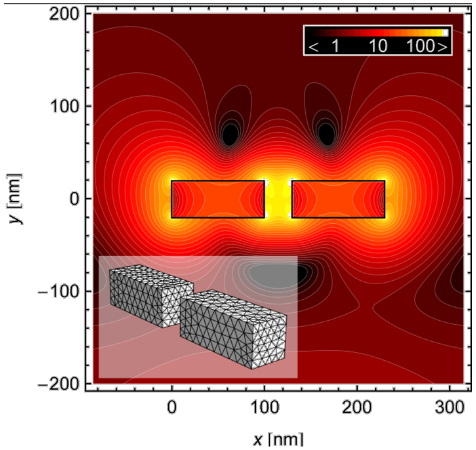
A.M. Kern, J. Optical Soc. America A vol. 26, p. 732 (2009)

Modelling realistic nanostructures

- Sometimes the nanofabrication is not so perfect…

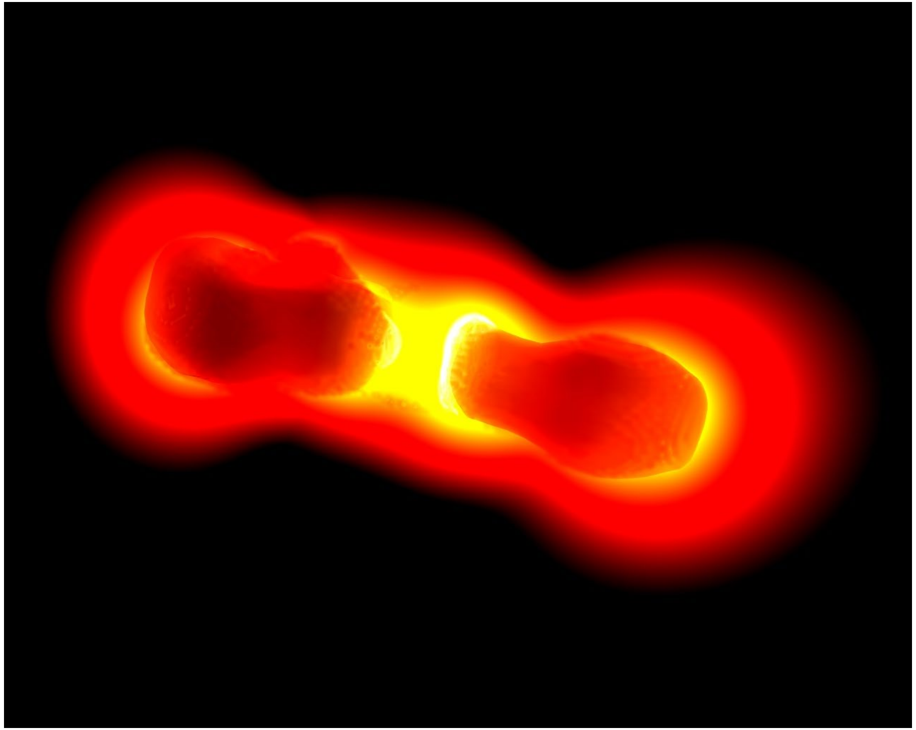






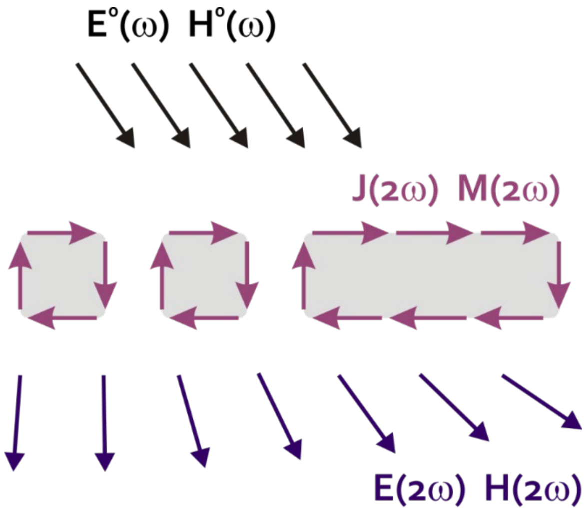
A.M. Kern, Nano Letters vol. 11, p. 482 (2011)

Modelling realistic nanostructures



A.M. Kern, Nano Letters vol. 11, p. 482 (2011)

Surface integral equation (SIE) method – Computing SHG

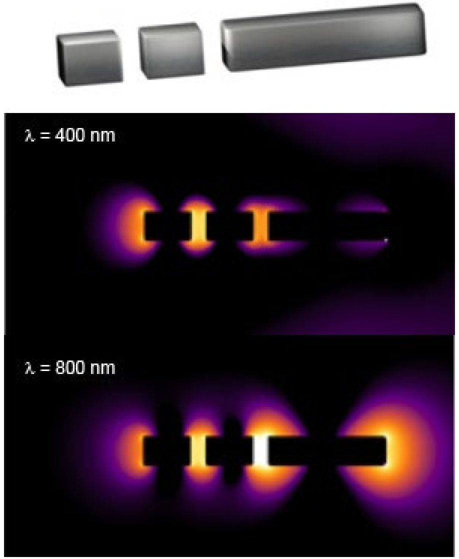


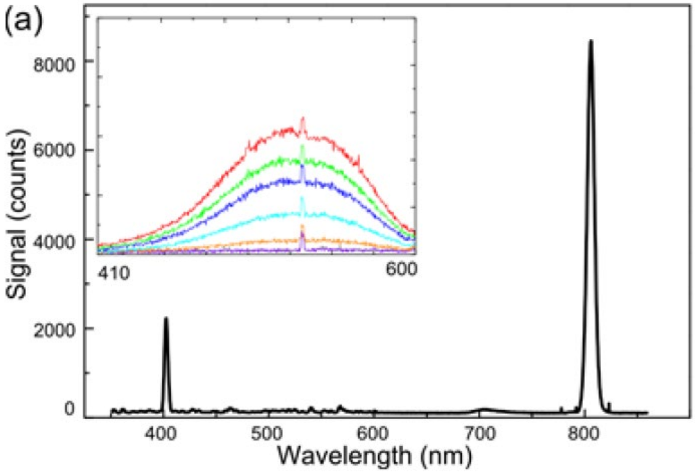
- Compute electric and magnetic surface currents at ω
- For each mesh, compute the normal field component at ω inside the metal
- Use $P_n(2\omega) = \chi^{(2)}_{n,nn} E_n(\omega) E_n(\omega)$ to obtain the surface currents at 2 ω
- Solve again the scattering problem with those currents to get the fields at 2 ω

J. Butet, Nano Letters vol. 13, p. 1787 (2013)

Double resonant antenna and second harmonic generation

- **How does SHG work in such a structure?**
- **What is the role of the different parts in the antenna?**

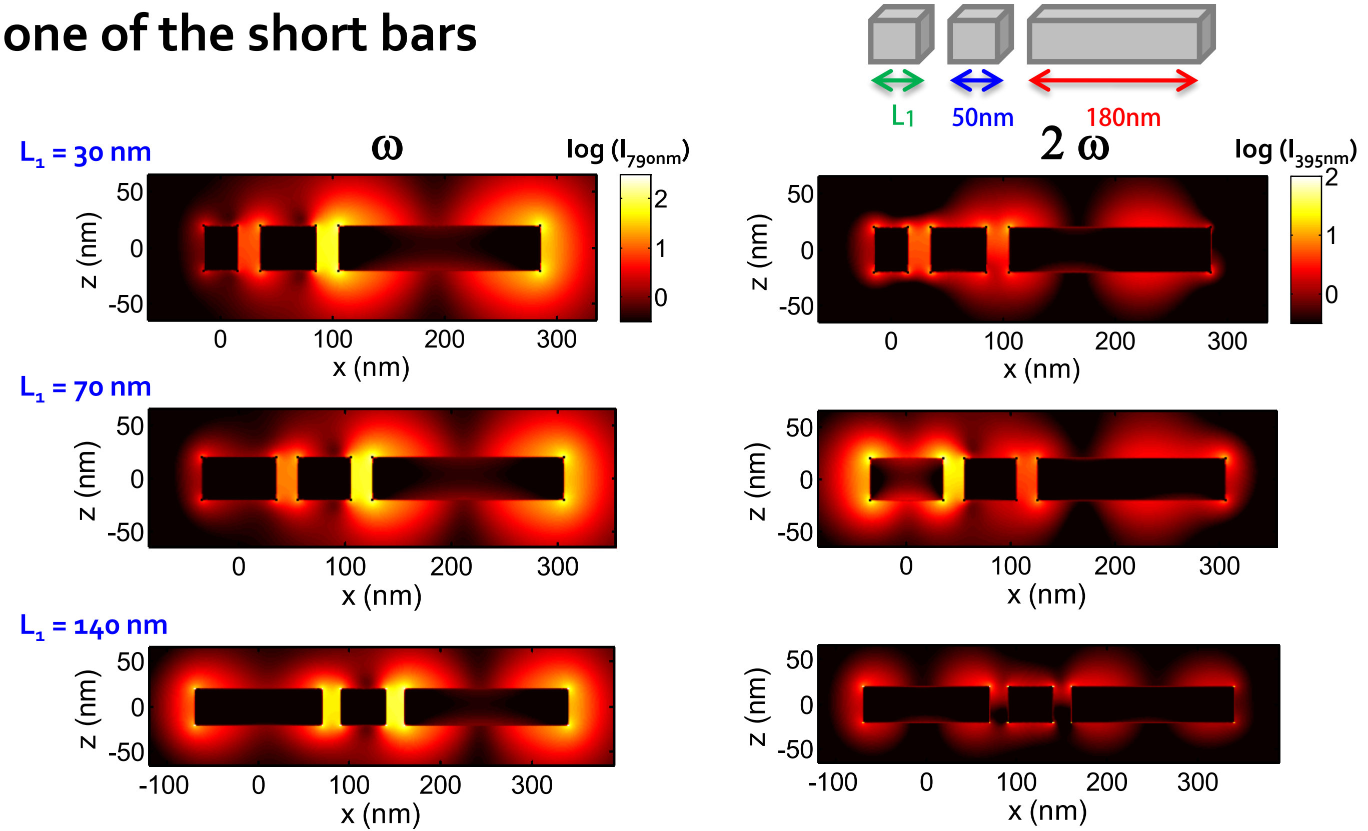




K. Thyagarajan, Optics Express, vol. 20, p. 12860 (2012)

Elucidating the role of the different parts of the antenna

- Tuning one of the short bars



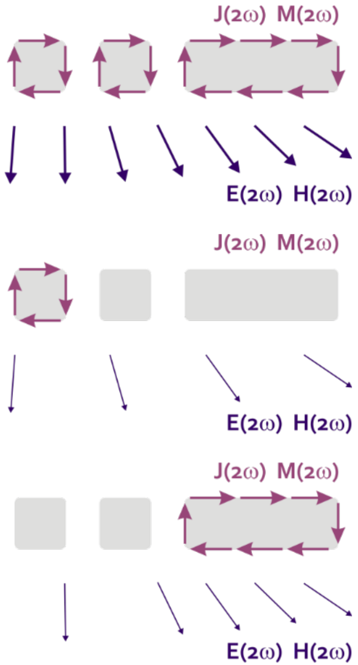
- It looks like the short bar determines the SHG emission

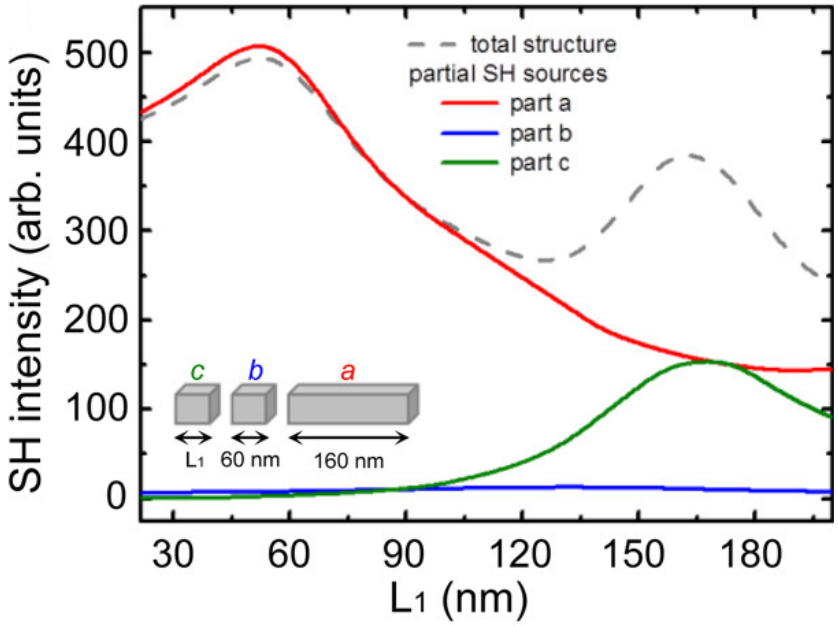
www.nanophotonics.ch

K.Y. Yang, ACS Photonics vol. 4, p. 1522 (2017)

Elucidating the role of the different parts of the antenna

- It looks like the short bar determines the SHG emission…

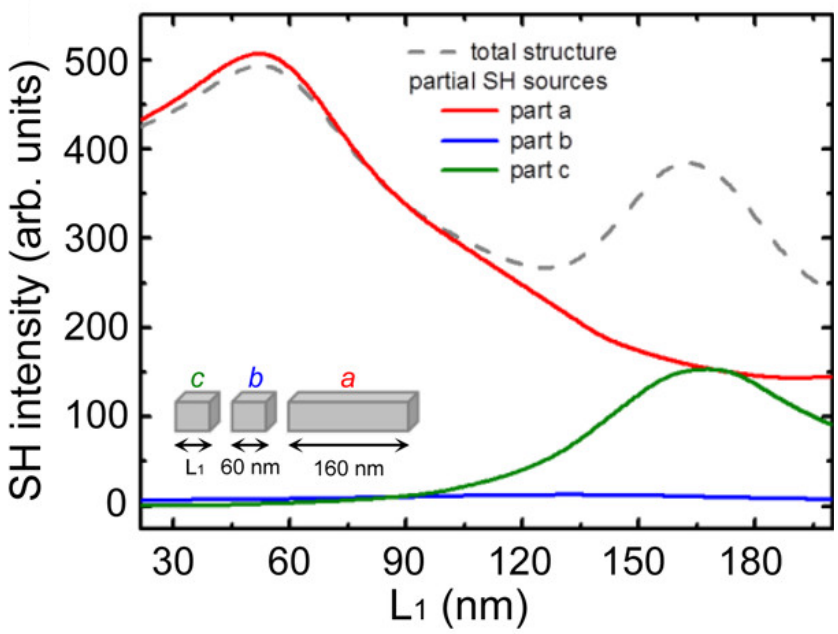




K.Y. Yang, ACS Photonics vol. 4, p. 1522 (2017)

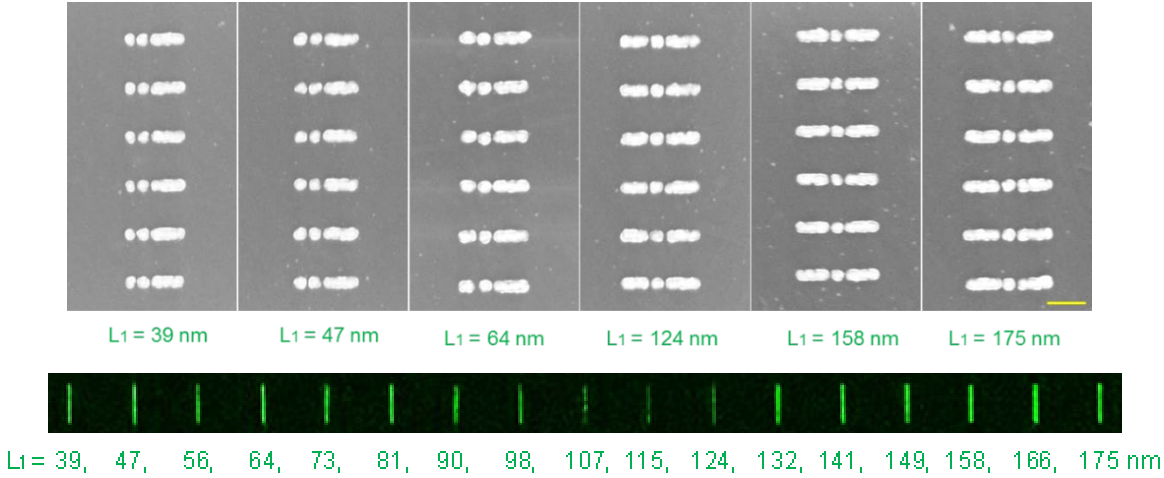
Elucidating the role of the different parts of the antenna

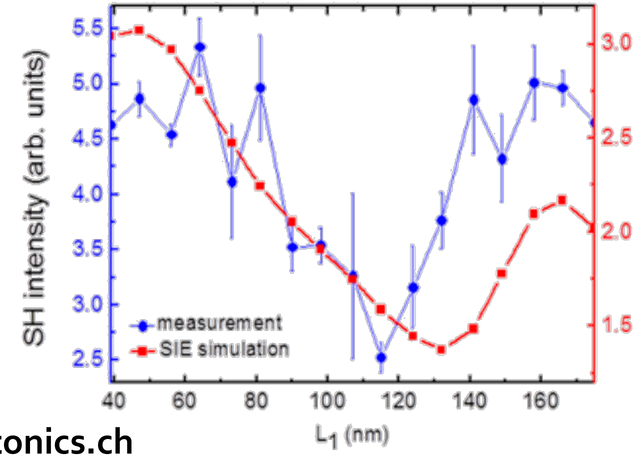
- **It looks like the short bar determines the SHG emission…**
- **… but things are more complicated!**
  - **The small bar tunes the structure**
  - **SHG comes from the large bar**



K.Y. Yang, ACS Photonics vol. 4, p. 1522 (2017)

Elucidating the role of the different parts of the antenna





- **Good agreement between measurements and simulations**

K.Y. Yang, ACS Photonics vol. 4, p. 1522 (2017)

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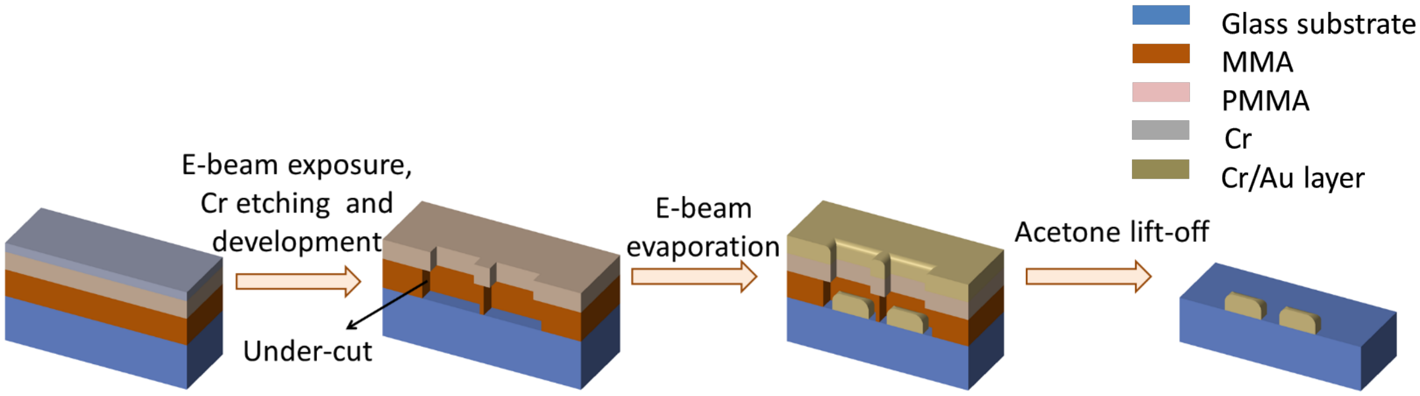
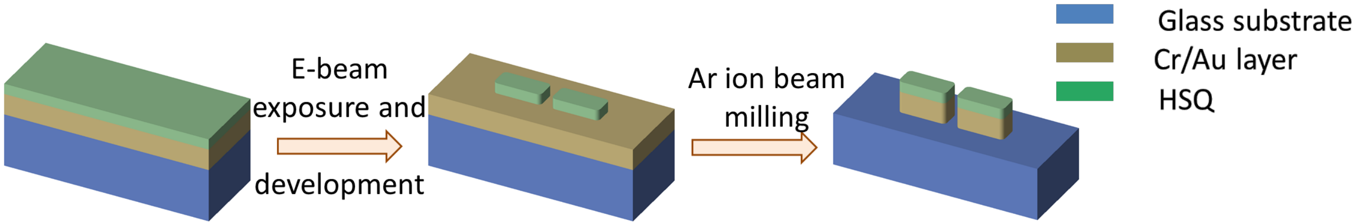
Nanofabrication of plasmonic structures

- **Lift-off (positive resist) vs. ion etching (negative resist)**

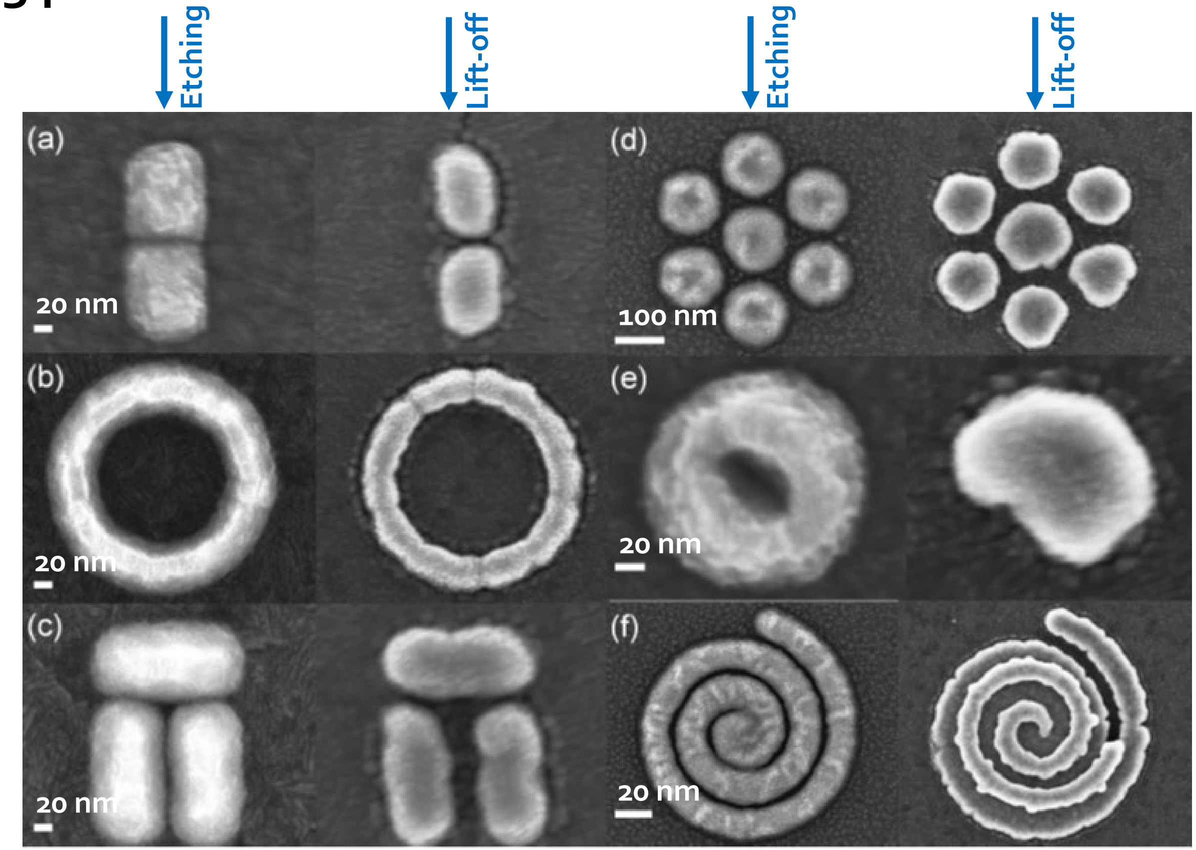




B. Abasahl, Nanotechnology vol. 32, p. 475202 (2021)

Lift-off vs. ion etching (gold nanostructures)

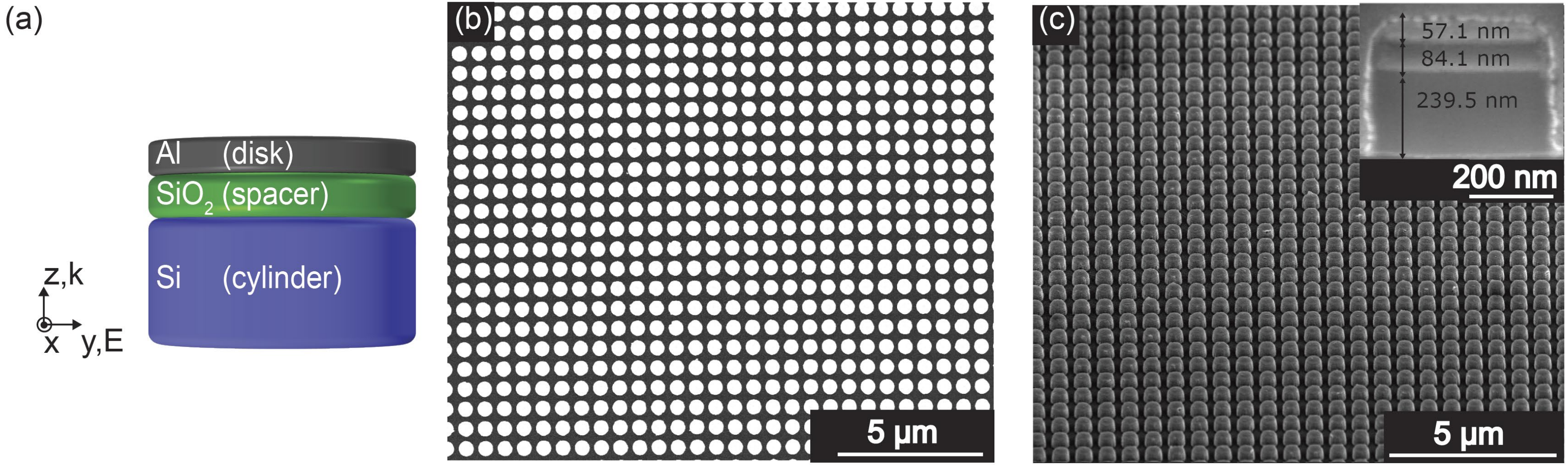
- Ion etching provides smaller features and better control



B. Abasahl, Nanotechnology vol. 32, p. 475202 (2021)

Hybrid metasurface for sensing

- **Etching can fabricate quite thick nanostructures**

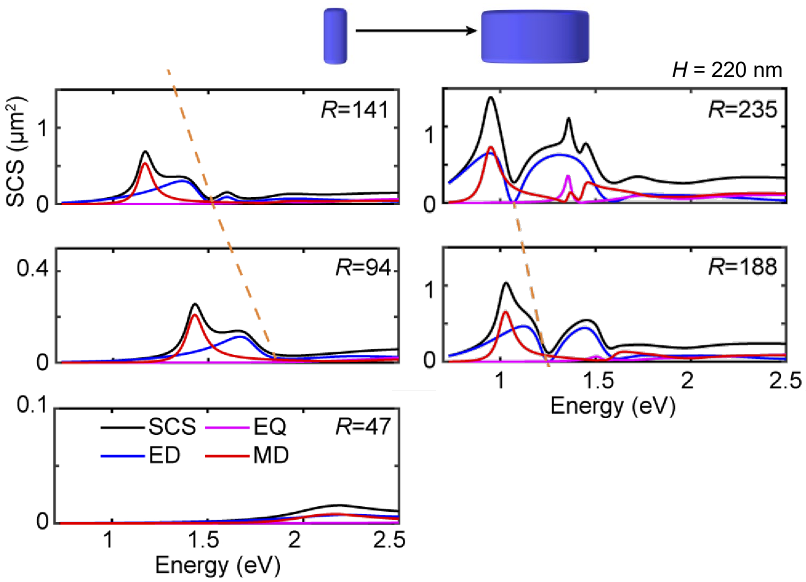


D. Ray, Nano Letters vol. 20, p. 8752 (2020)

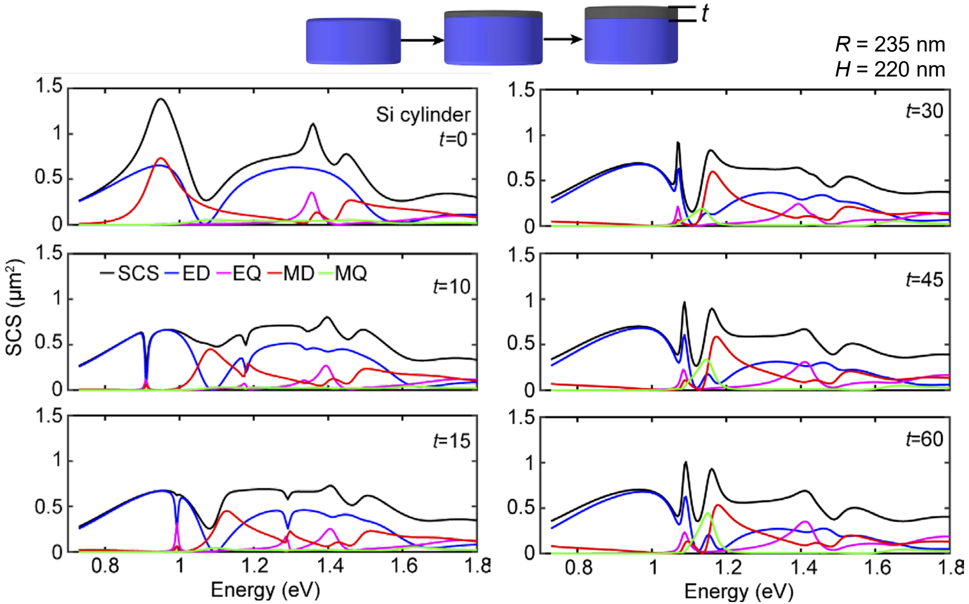
Hybrid nanostructures – Many degrees of freedom to tune the response

- **Dielectric structures support a magnetic dipole that can hybridize with the electric dipole of a plasmonic structure to produce a very rich response**

Si cylinder



Si cylinder + Ag disk



D. Ray, Optics Express vol. 29, p. 24056 (2021)

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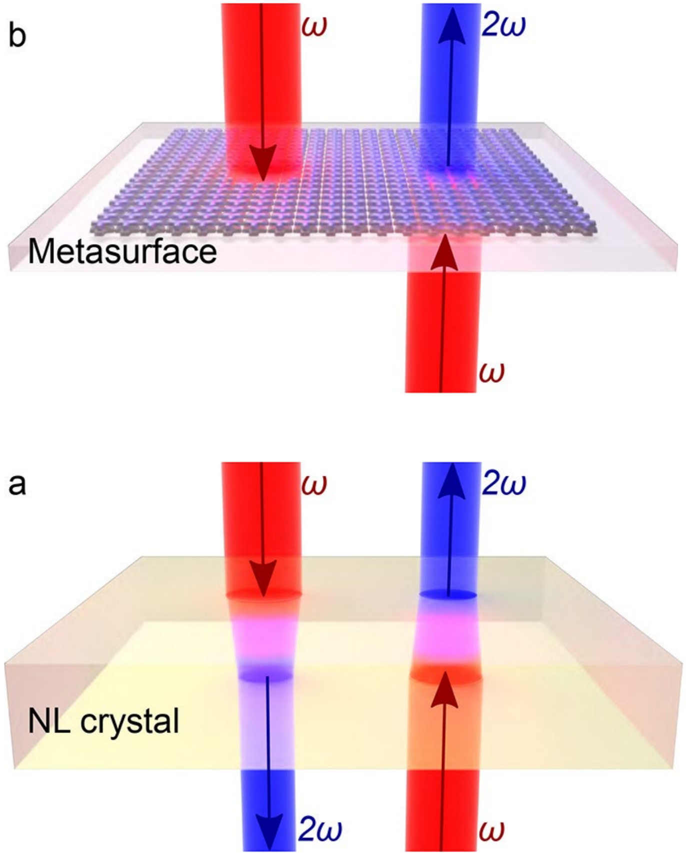
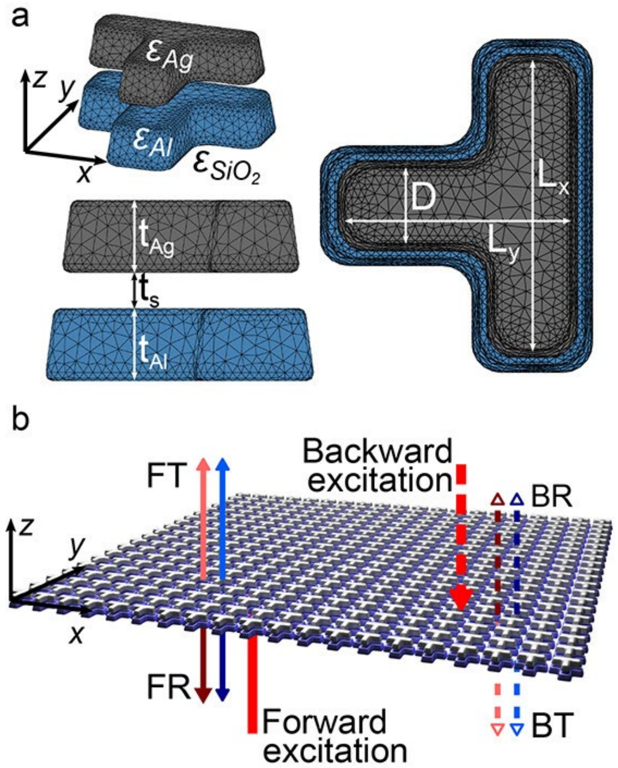
Asymmetric nonlinear metasurface – Meta-atom design



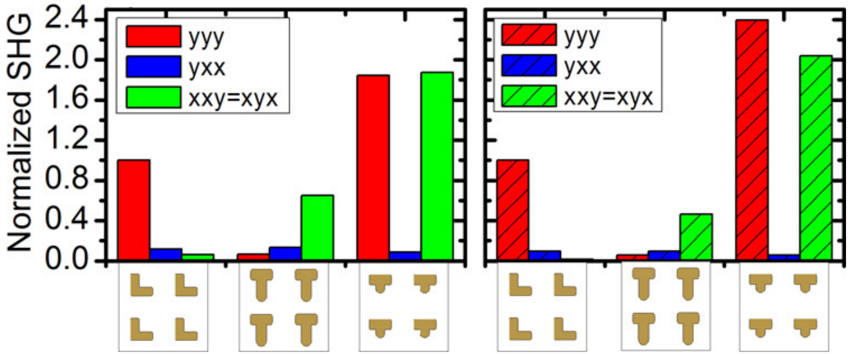


S. Boroviks, Nano Letters, vol. 23, p. 3362 (2023)

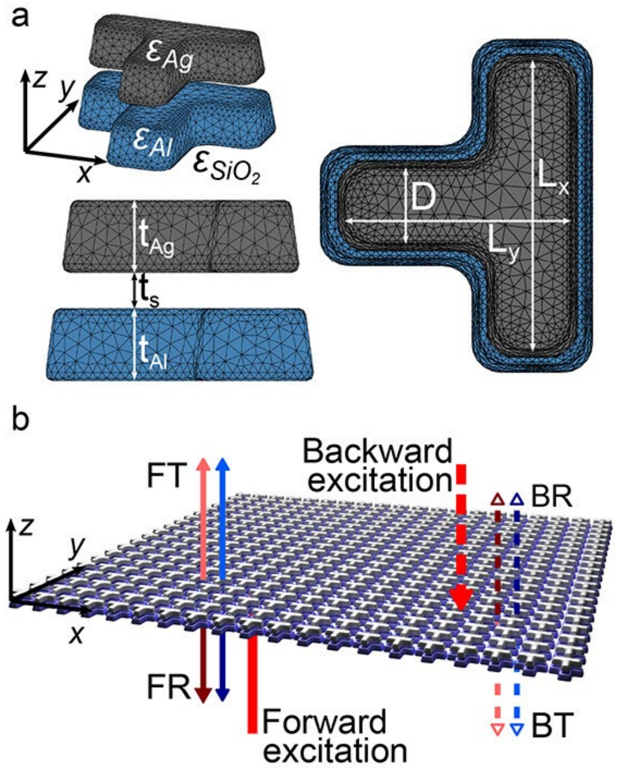
Asymmetric nonlinear metasurface – Meta-atom design

- T-shape plasmonic nanostructure
- Excitation polarization along the x-direction
- SHG produced along the y-direction

SHG intensity



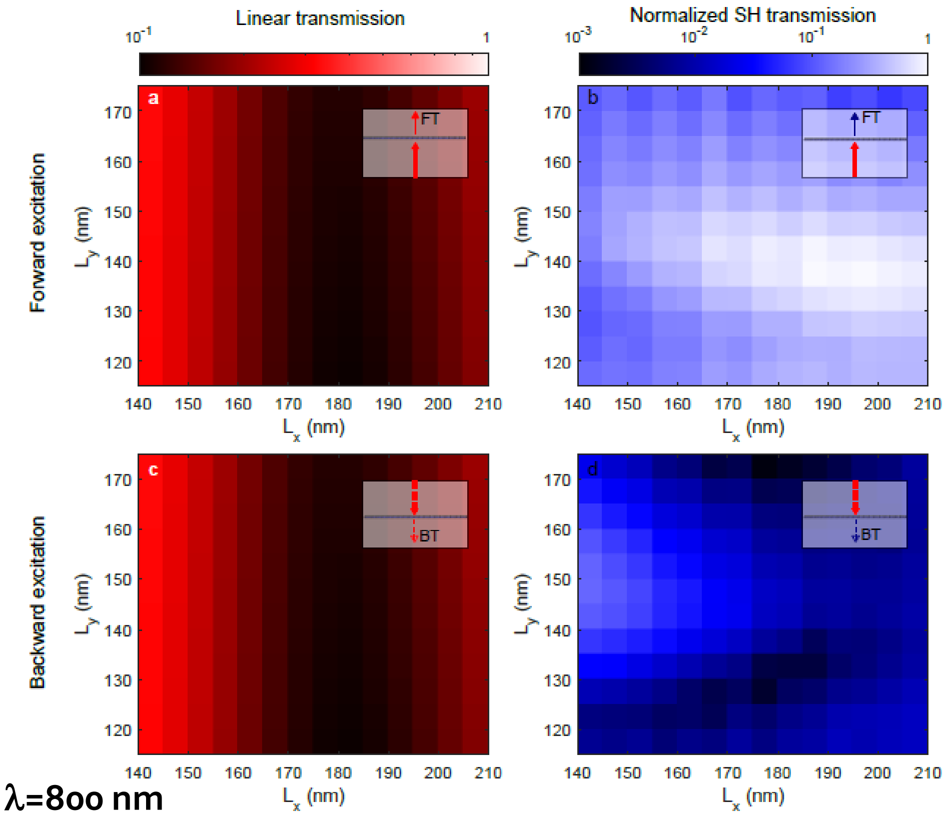
R. Czaplicki, Nano Letters vol. 15, p. 530 (2015)

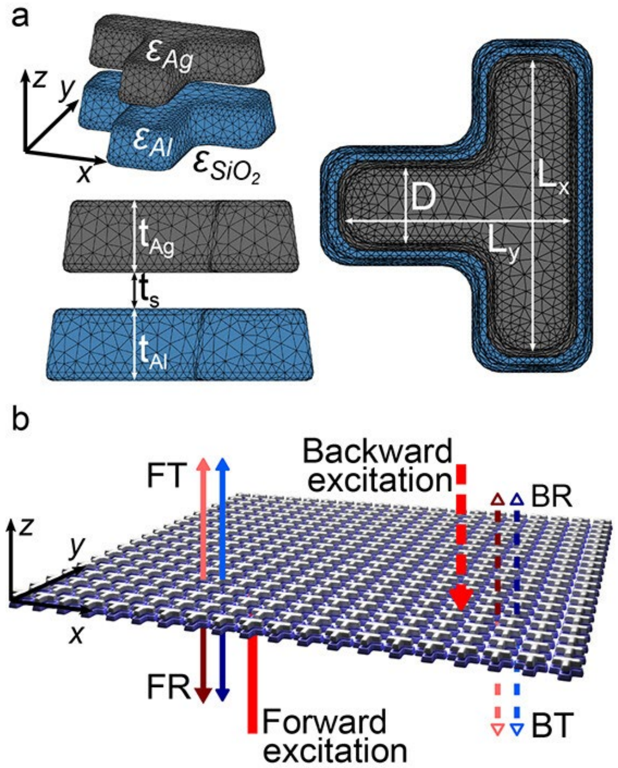


S. Boroviks, Nano Letters, vol. 23, p. 3362 (2023)

Asymmetric nonlinear metasurface – Meta-atom design

- **A T-shape provides independent control for the resonances at ω and 2ω**





λ=800 nm

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S. Boroviks, Nano Letters, vol. 23, p. 3362 (2023)

Asymmetric nonlinear metasurface – Meta-atom design

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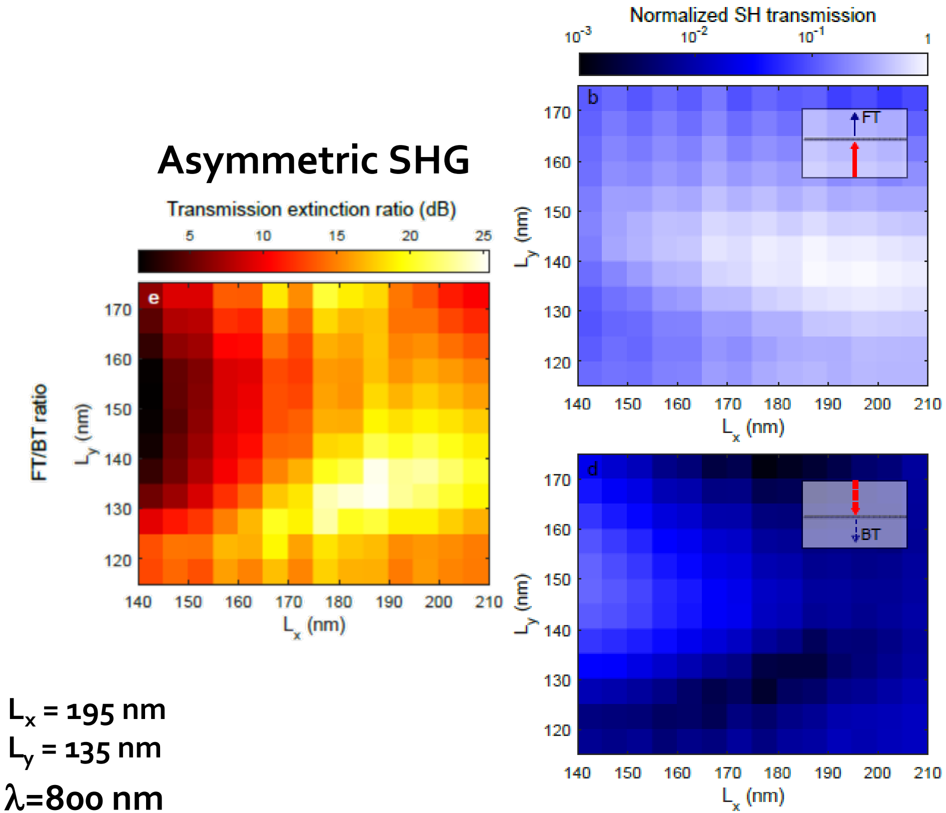
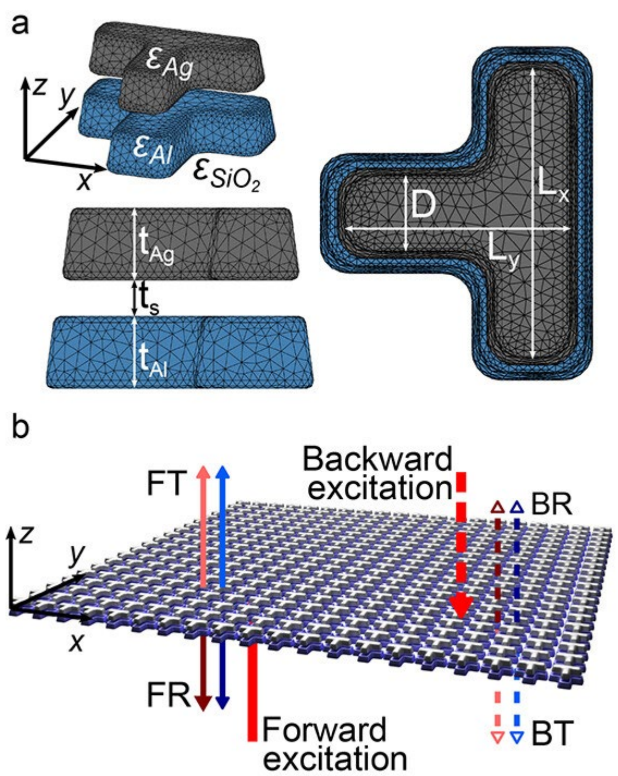
Asymmetric SHG





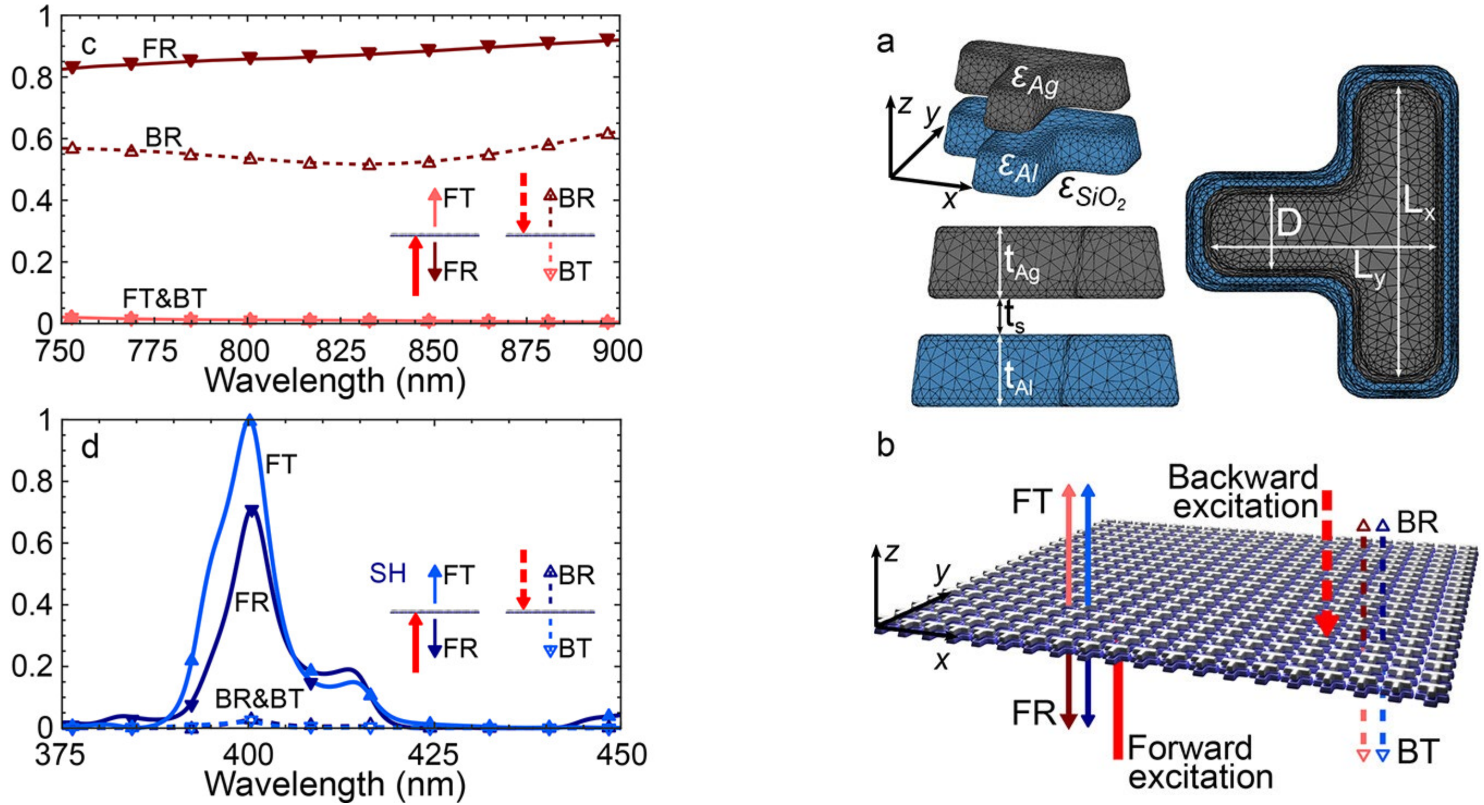
L_x = 195 nm
L_y = 135 nm
λ=800 nm

www.nanophotonics.ch

S. Boroviks, Nano Letters, vol. 23, p. 3362 (2023)

Asymmetric nonlinear metasurface – Meta-atom design

- **Numerical simulations in the linear and nonlinear regimes**



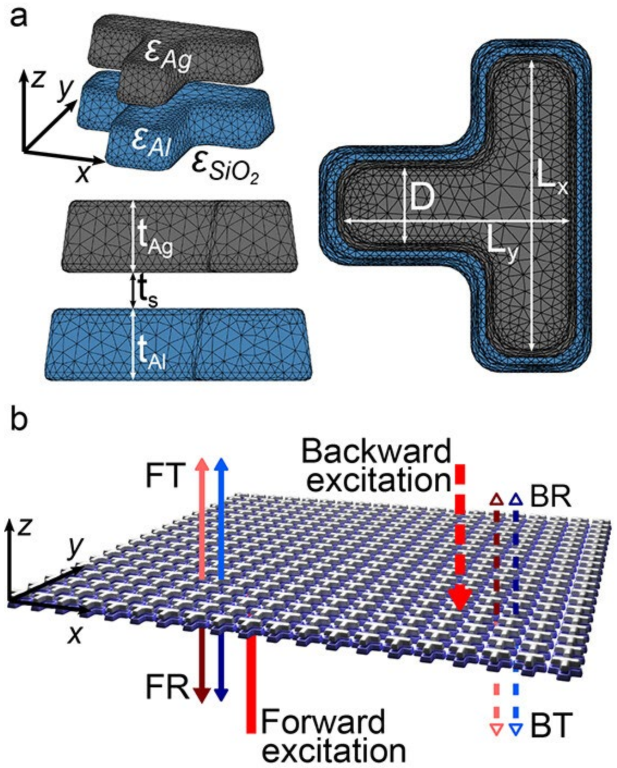
L_x = 195 nm
L_y = 135 nm

www.nanophotonics.ch

S. Boroviks, Nano Letters, vol. 23, p. 3362 (2023)

Nanofabrication

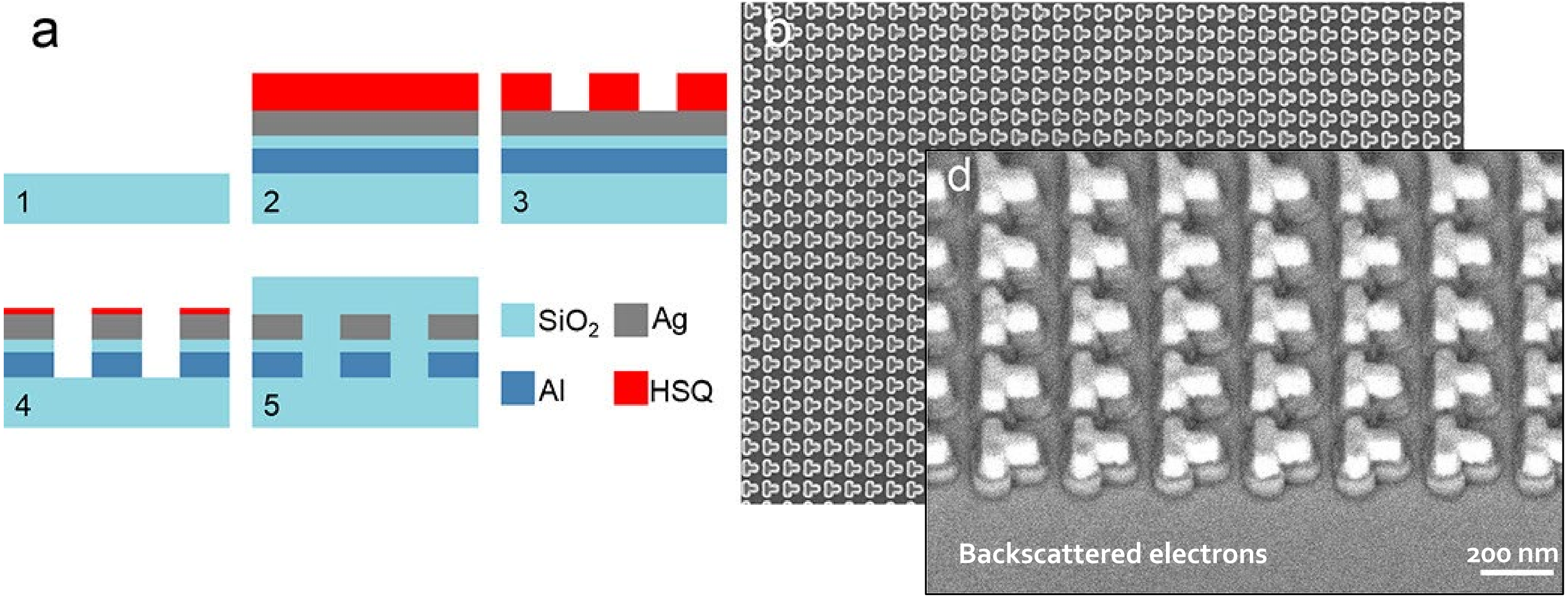
- Array of Al – Ag meta-atoms (each 50 nm thick)
- L_x = 195 nm, L_y = 135 nm
- Square lattice with period Λ = 250 nm (avoid diffraction orders at ω and 2ω)
- 25 nm SiO_2 spacer (n = 1.49)
- Embedded in SiO_2 (symmetric environment)



S. Boroviks, Nano Letters, vol. 23, p. 3362 (2023)

Strongly asymmetric SHG – Nanofabrication using ion etching (Ar)

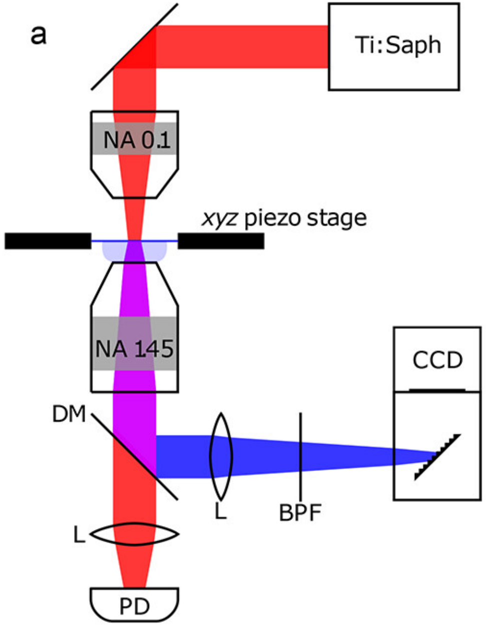
- **HSQ mask used for ion etching (Ar) the nanostructures**

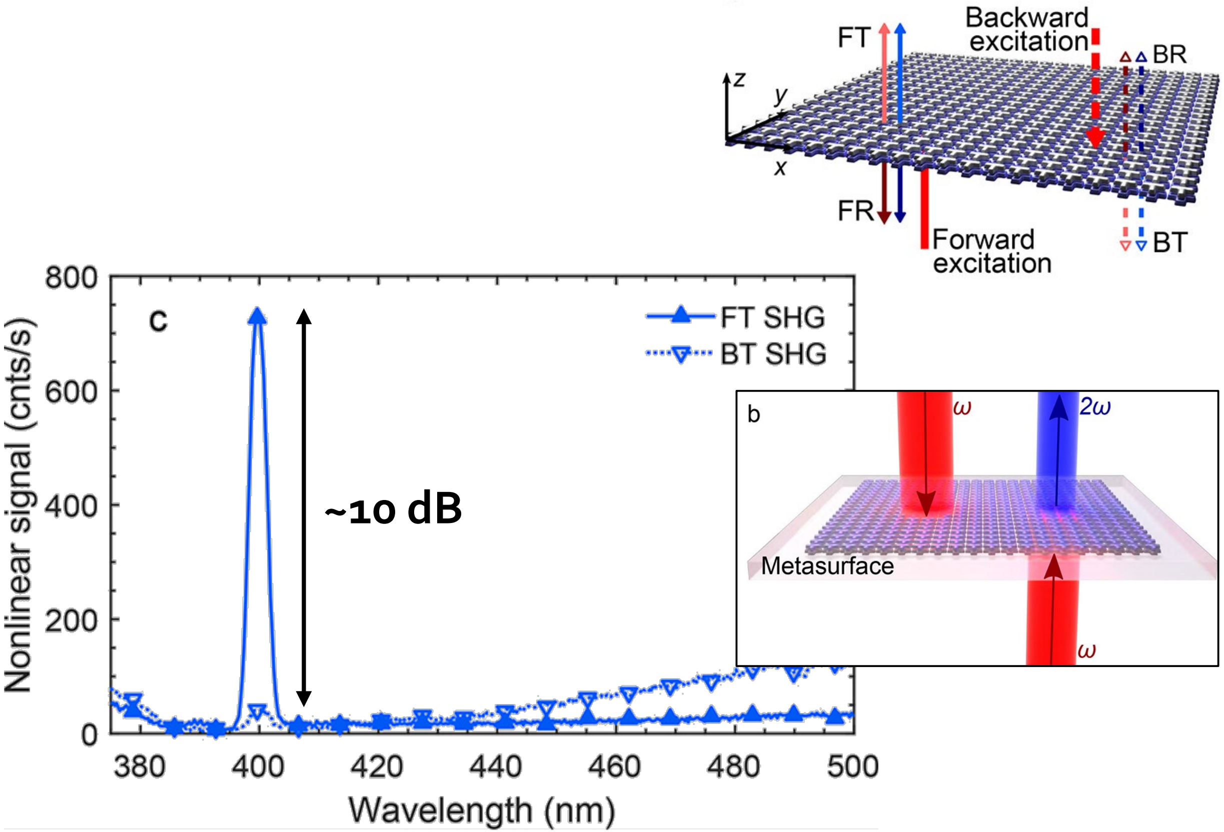


S. Boroviks, Nano Letters, vol. 23, p. 3362 (2023)

B. Abasahl, Nanotechnology vol. 32, p. 475202 (2021)

Experimental characterization



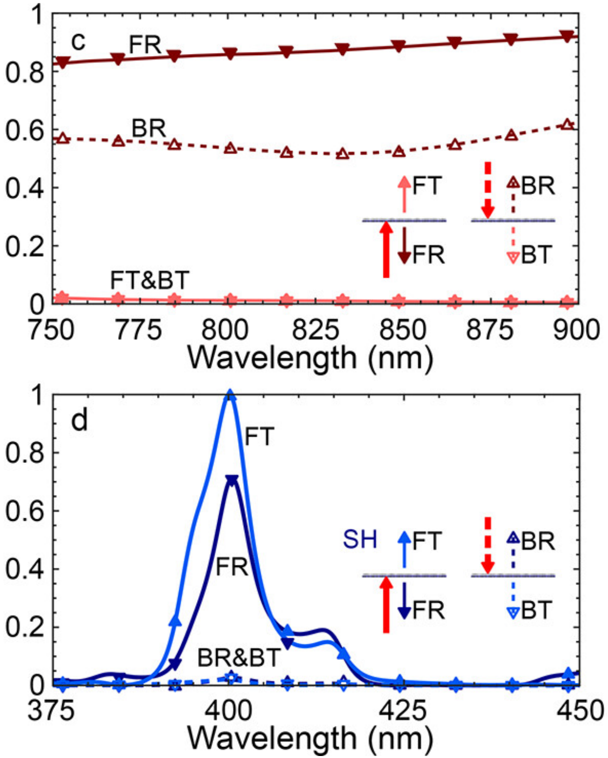


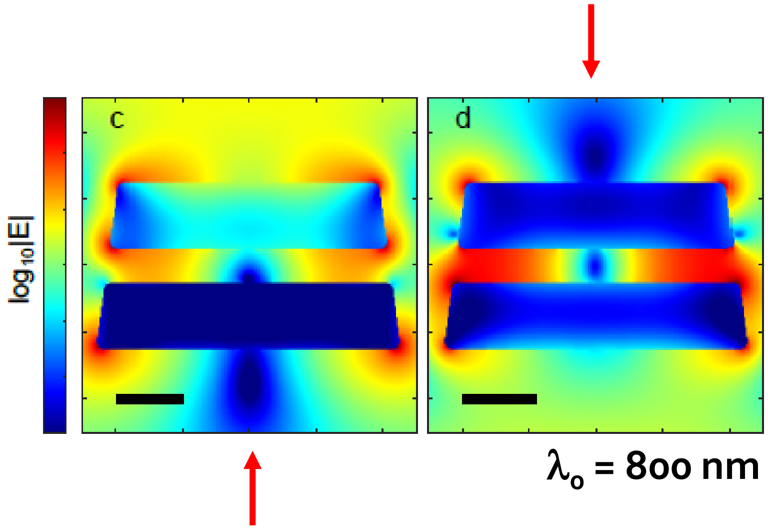
λ_o = 800 nm
120 fs pulse duration

S. Boroviks, Nano Letters, vol. 23, p. 3362 (2023)

How does it work?

- **Different field distributions depending on the illumination direction**
- **Bianisotropy (magneto-electrique coupling), asymmetric absorption**





λ_o = 800 nm

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S. Boroviks, Nano Letters, vol. 23, p. 3362 (2023)

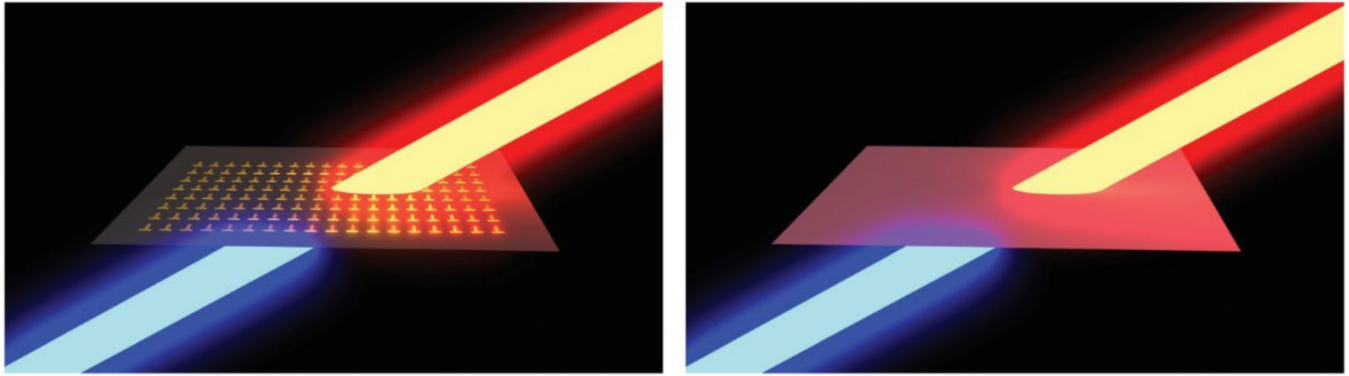
How does it work?

- **Induced linear and nonlinear polarizations**

$$\mathbf{P}^{\omega} = \overline{\overline{\chi}}_{ee}^{\omega} \cdot \mathbf{E}^{\omega} + \overline{\overline{\chi}}_{em}^{\omega} \cdot \mathbf{H}^{\omega}$$

$$\mathbf{P}^{2\omega} = \overline{\overline{\chi}}_{ee}^{2\omega} \cdot \mathbf{E}^{2\omega} + \overline{\overline{\chi}}_{em}^{2\omega} \cdot \mathbf{H}^{2\omega} + \overline{\overline{\chi}}_{eee}^{\omega} : \mathbf{E}^{\omega}\mathbf{E}^{\omega} + \overline{\overline{\chi}}_{eem}^{\omega} : \mathbf{E}^{\omega}\mathbf{H}^{\omega} + \overline{\overline{\chi}}_{emm}^{\omega} : \mathbf{H}^{\omega}\mathbf{H}^{\omega}$$

- **Retrieved numerically – Homogenization procedure based on the generalized sheet transition conditions (GSTCs)**



K. Achouri, New Journal of Physics vol. 24, p. 025006 (2022)

How does it work?

- **Induced linear and nonlinear polarizations**

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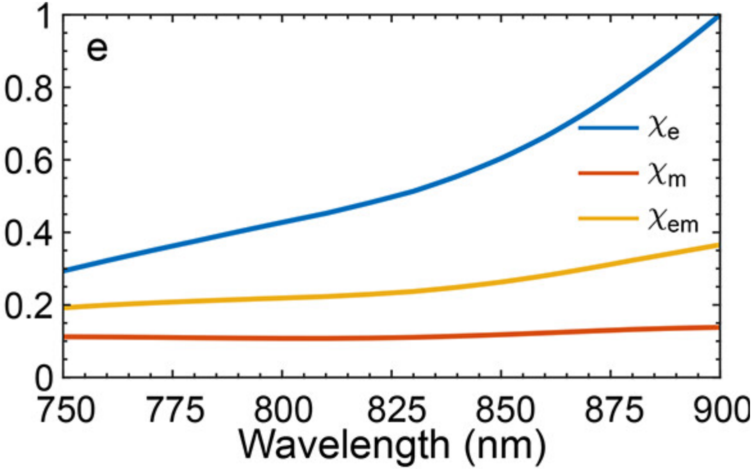
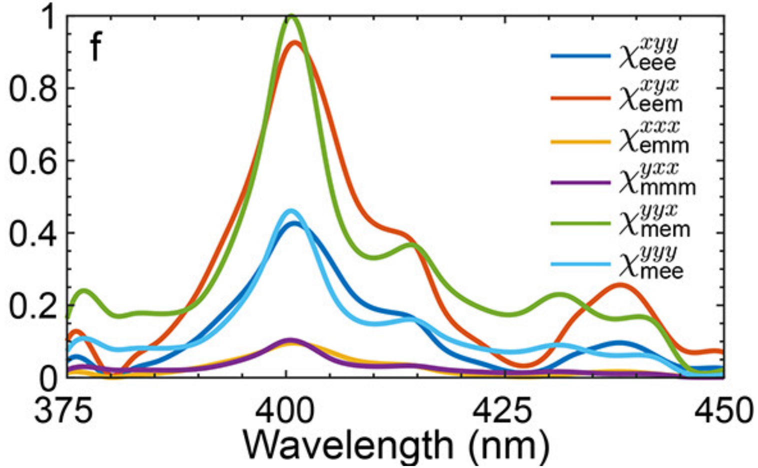
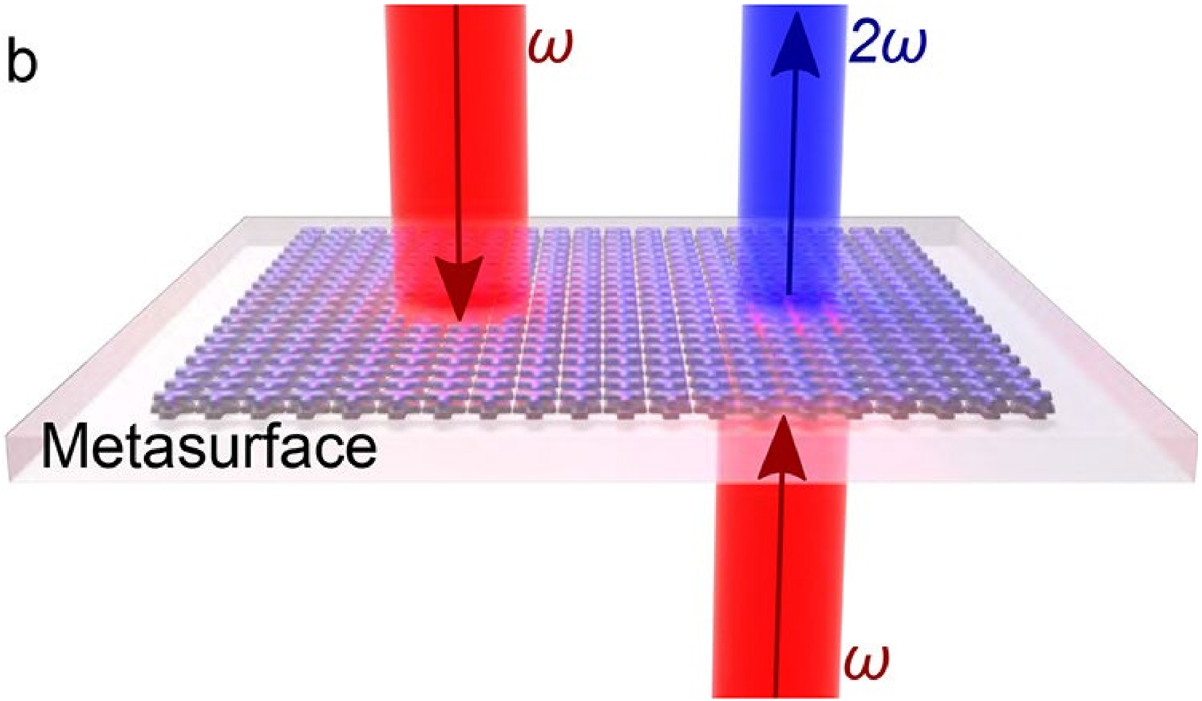
- **Linear regime: non negligible magneto-electric coupling term** χ_{em}
- **Nonlinear susceptibilities dominated by magnetic/electric excitations along orthogonal directions** $\chi_{eem}^{xyx}, \chi_{mem}^{yyx}$





S. Boroviks, Nano Letters, vol. 23, p. 3362 (2023)

Is it nonreciprocal?



S. Boroviks, Nano Letters, vol. 23, p. 3362 (2023)

Is it nonreciprocal?

PHYSICAL REVIEW APPLIED 10, 047001 (2018)

Review Article

Electromagnetic Nonreciprocity

Christophe Caloz,[1,*] Andrea Alù,[2] Sergei Tretyakov,[3] Dimitrios Sounas,[4] Karim Achouri,[5] and Zoé-Lise Deck-Léger[1]

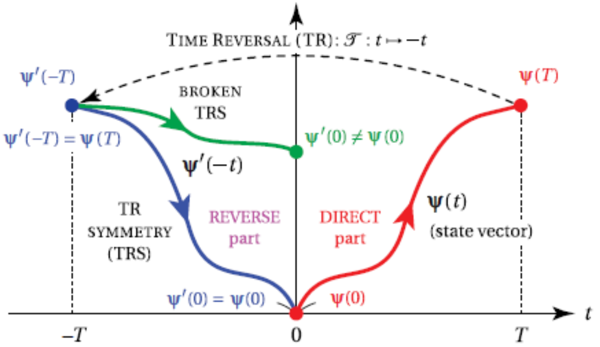
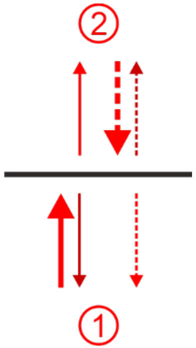


FIG. 1. Time-reversal symmetry (TRS) (red and blue curves) and broken time-reversal symmetry (red and green curves), or time-reversal asymmetry, as a general thought experiment and mathematical criterion for nonreciprocity.

K. Achouri, Physical Review B vol. 104, p. 165426 (2021)

Is it nonreciprocal?

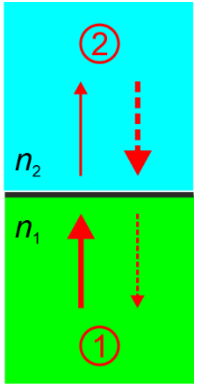
- Linear regime: 2-port system



$$\bar{\bar{\mathbf{S}}} = \begin{bmatrix} S_{11} & S_{12} \\ S_{21} & S_{22} \end{bmatrix}$$

- Reciprocity requires $\bar{\bar{\mathbf{S}}} = \bar{\bar{\mathbf{S}}}^{\mathrm{T}}$
- Equal transmission coefficients $S_{12} = S_{21}$
- No constrains on the reflection coefficients S_{11}, S_{22}

- Fresnel coefficients



$$t_{21} = \frac{2n_2}{n_2 + n_1}$$

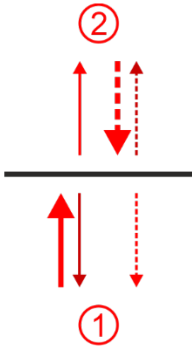
$$t_{12} = \frac{2n_1}{n_1 + n_2}$$

$$S_{12} = T_{12} = \frac{n_2}{n_1}\left|t_{12}\right|^2 = \frac{4n_1 n_2}{\left(n_1 + n_2\right)^2} = T_{21} = S_{21}$$

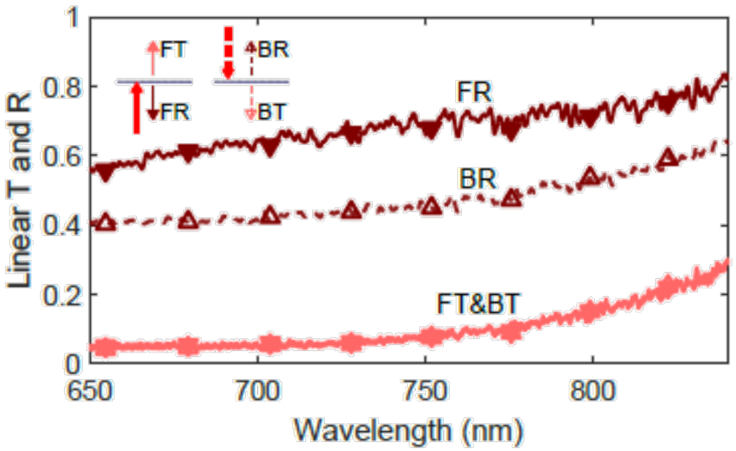
S. Boroviks, Nano Letters, vol. 23, p. 3362 (2023)

Is it nonreciprocal?

- Linear regime: 2-port system



$$\overline{\overline{\mathbf{S}}} = \begin{bmatrix} S_{11} & S_{12} \\ S_{21} & S_{22} \end{bmatrix}$$



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S. Boroviks, Nano Letters, vol. 23, p. 3362 (2023)

Is it nonreciprocal?

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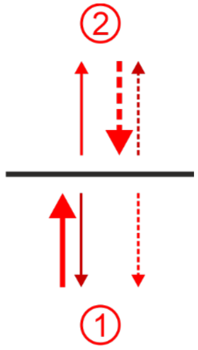

$$\bar{\bar{\mathbf{S}}} = \begin{bmatrix} S_{11} & S_{12} \\ S_{21} & S_{22} \end{bmatrix}$$

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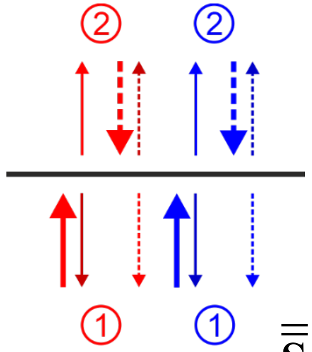
- Equal transmission coefficients $S_{12} = S_{21}$
- No constrains on the reflection coefficients S_{11}, S_{22}

- Nonlinear regime: 4-port system



$$\bar{\bar{\mathbf{S}}} = \begin{bmatrix} \bar{\bar{\mathbf{S}}}^{\omega\to\omega} & \bar{\bar{\mathbf{S}}}^{2\omega\to\omega} \\ \bar{\bar{\mathbf{S}}}^{\omega\to 2\omega} & \bar{\bar{\mathbf{S}}}^{2\omega\to 2\omega} \end{bmatrix} = \begin{bmatrix} S_{11}^{\omega\to\omega} & S_{12}^{\omega\to\omega} & S_{11}^{2\omega\to\omega} & S_{12}^{2\omega\to\omega} \\ S_{21}^{\omega\to\omega} & S_{22}^{\omega\to\omega} & S_{21}^{2\omega\to\omega} & S_{22}^{2\omega\to\omega} \\ S_{11}^{\omega\to 2\omega} & S_{12}^{\omega\to 2\omega} & S_{11}^{2\omega\to 2\omega} & S_{12}^{2\omega\to 2\omega} \\ S_{21}^{\omega\to 2\omega} & S_{22}^{\omega\to 2\omega} & S_{21}^{2\omega\to 2\omega} & S_{22}^{2\omega\to 2\omega} \end{bmatrix}$$

S. Boroviks, Nano Letters, vol. 23, p. 3362 (2023)

Is it nonreciprocal?

- Probing (non)reciprocity would require probing six (in)equalities:

$$S_{21}^{\omega\to\omega} \stackrel{?}{=} S_{12}^{\omega\to\omega} \qquad S_{12}^{\omega\to 2\omega} \stackrel{?}{=} S_{21}^{2\omega\to\omega}$$

$$S_{11}^{\omega\to 2\omega} \stackrel{?}{=} S_{11}^{2\omega\to\omega} \qquad S_{22}^{\omega\to 2\omega} \stackrel{?}{=} S_{22}^{2\omega\to\omega}$$

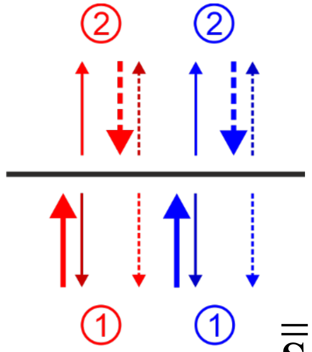
$$S_{21}^{\omega\to 2\omega} \stackrel{?}{=} S_{12}^{2\omega\to\omega} \qquad S_{21}^{2\omega\to 2\omega} \stackrel{?}{=} S_{12}^{2\omega\to 2\omega}$$

- So far, we have only shown that

$$S_{21}^{\omega\to 2\omega} \neq S_{12}^{\omega\to 2\omega}$$

(asymmetric nonlinear process)

- Nonlinear regime: 4-port system



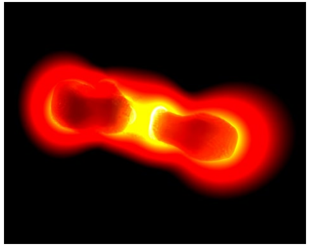
$$\overline{\overline{\mathbf{S}}} = \begin{bmatrix} \overline{\overline{\mathbf{S}}}^{\omega\to\omega} & \overline{\overline{\mathbf{S}}}^{2\omega\to\omega} \\ \overline{\overline{\mathbf{S}}}^{\omega\to 2\omega} & \overline{\overline{\mathbf{S}}}^{2\omega\to 2\omega} \end{bmatrix} =$$

$$= \begin{bmatrix} S_{11}^{\omega\to\omega} & S_{12}^{\omega\to\omega} & S_{11}^{2\omega\to\omega} & S_{12}^{2\omega\to\omega} \\ S_{21}^{\omega\to\omega} & S_{22}^{\omega\to\omega} & S_{21}^{2\omega\to\omega} & S_{22}^{2\omega\to\omega} \\ S_{11}^{\omega\to 2\omega} & \boxed{S_{12}^{\omega\to 2\omega}} & S_{11}^{2\omega\to 2\omega} & S_{12}^{2\omega\to 2\omega} \\ \boxed{S_{21}^{\omega\to 2\omega}} & S_{22}^{\omega\to 2\omega} & S_{21}^{2\omega\to 2\omega} & S_{22}^{2\omega\to 2\omega} \end{bmatrix}$$

S. Boroviks, Nano Letters, vol. 23, p. 3362 (2023)

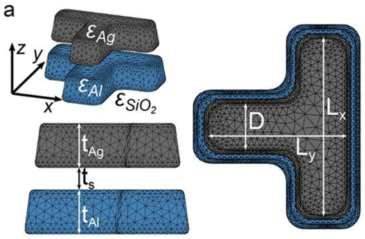
Summary

Use numerical techniques to their full extent



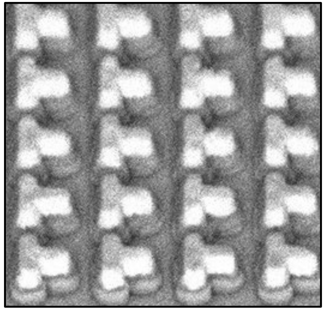
- Model realistic systems
- Perform some numerical experiments that are not feasible in practice

Simple design with strong bianisotropy



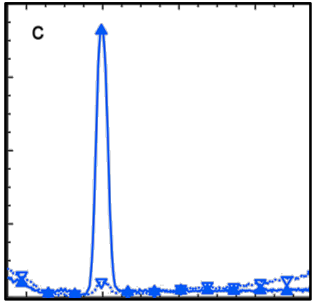
- Easily tunable
- Al and Ag give strong SHG
- Their out-of-plane combination leads to a robust bianisotropy

Simple, versatile nanotechnology



- Dry etching provides very well-defined nanostructures
- It can be used with a diversity of materials

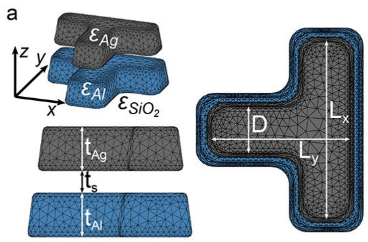
Strong, asymmetric nonlinear response



- 16 dB difference in theory, 10 dB experimentally between forward and backward SHG

Summary

Simple design with strong bianisotropy



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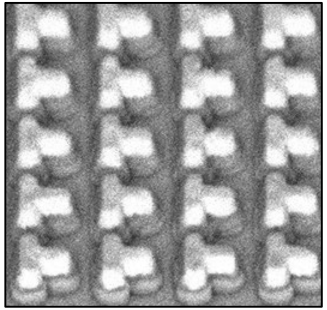
Simple, versatile nanotechnology



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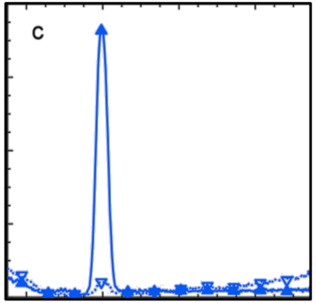
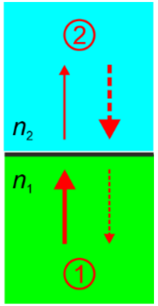


- 16 dB difference in theory, 10 dB experimentally between forward and backward SHG

No, this is not nonreciprocal!

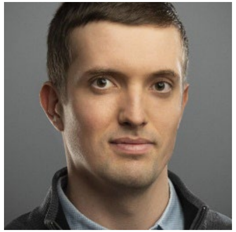


- The device is time-reversal asymmetric
- Sometimes mistaken with nonreciprocal…

Acknowledgements

Sergejs Boroviks

Karim Achouri



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Christian Santschi

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