On The Bumpy Road Towards 3D Photonic Metamaterials

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CFN DFG

KSOP Karlsruhe School of Optics & Photonics

PHOME

Outline

- Metamaterials as effective media
- Hitherto existing realizations of 3D metamaterials
- Fabricational approaches for the NIR
- Our fabricational approach
- Recent results
- Current projects
- Conclusions and outlook

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Metamaterials as effective media

General definition:

Metamaterials are **artificially fabricated effective materials** that provide material-light interactions which are not accessible by naturally available substances.

Metamaterials as effective media

Some popular special cases:

- Magnetic photonic metamaterials ($\mu \neq 1$)
- Negative-index materials
  (μ , $\varepsilon < 0$ plus some other assumptions)
- Electric Cloaks
  (anisotropic variation of ε from 1 to 0)
- Enhanced non-linear optical response
  (high values for $\chi^{(2)}$, $\chi^{(3)}$)
- Chiral metamaterials (enhanced optical activity)

Metamaterials as effective media

Relevant equations:

1. Maxwell's equ.

$$\text{rot } \vec{E} = -\dot{\vec{B}}$$

$$\text{div } \vec{D} = \rho$$

$$\text{rot } \vec{H} = \vec{j} + \dot{\vec{D}}$$

$$\text{div } \vec{B} = 0$$

2. Material equ.

$$\vec{D} = \varepsilon_0(\vec{E} + \vec{P})$$

$$\vec{B} = \mu_0(\vec{H} + \vec{M})$$

Metamaterials as effective media

Simplify material equations:

1. $\vec{j}_{\text{av}}, \rho_{\text{av}} = 0$
2. Describe $\vec{P}(\vec{E},\vec{H})$ and $\vec{M}(\vec{E},\vec{H})$

   by $\overleftrightarrow{\varepsilon}$, $\overleftrightarrow{\mu}$, $\overleftrightarrow{\xi}$ and $\overleftrightarrow{\zeta}$

   $\rightarrow \vec{P} = (\overleftrightarrow{\varepsilon} - 1)\vec{E} + \overleftrightarrow{\xi}\,\vec{H} \quad ; \quad \vec{M} = (\overleftrightarrow{\mu} - 1)\vec{H} + \overleftrightarrow{\zeta}\,\vec{E}$

But: Only possible if we assume an eff. material!

λ (wavelength) >> ***a*** (unit cell)

$\boldsymbol{d}_{\text{propdir}}$ > ***λ*** (wavelength)

Metamaterials as effective media

Need for effective materials

=

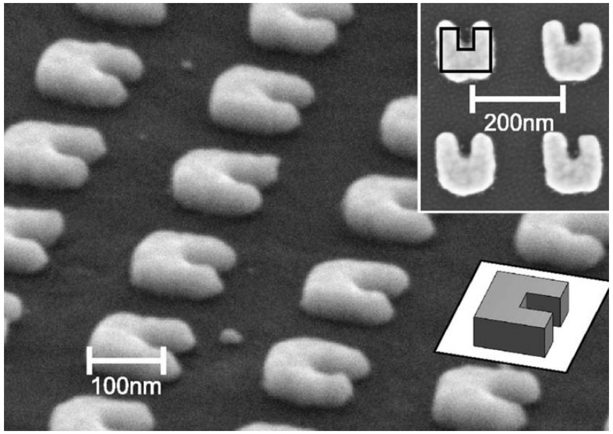
Motivation to fabricate
3D metamaterials

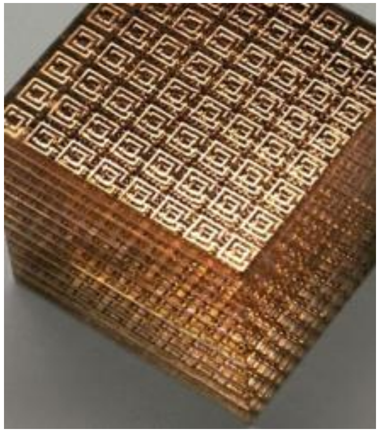
→ Is the formalism of bulk materials also applicable to “flat” structures?

Metamaterials as effective media

ANSWER: Sometimes!

Usually: Simple effective material models of electrically thin layers (height < λ) are limited in their physical meaning!





E. Seanz *et al.*, J. Appl. Phys. **101**, 114910 (2007)
M.W. Klein *et al.*, Opt. Lett. **31**, 1259 (2006)

Metamaterials as effective media

Let's assume that we have an eff. medium:

Isotropic media: · $\varepsilon , \mu \in \mathbb{C}$ · $\xi , \zeta = 0$	Anisotropic media: · ε , μ are tensors · $\xi , \zeta = 0$
Bi-isotropic media: · $\varepsilon , \mu \in \mathbb{C}$ · $\xi , \zeta \in \mathbb{C}$	Bi-anisotropic media: · ε , μ are tensors · ξ , ζ are tensors

Who cares?

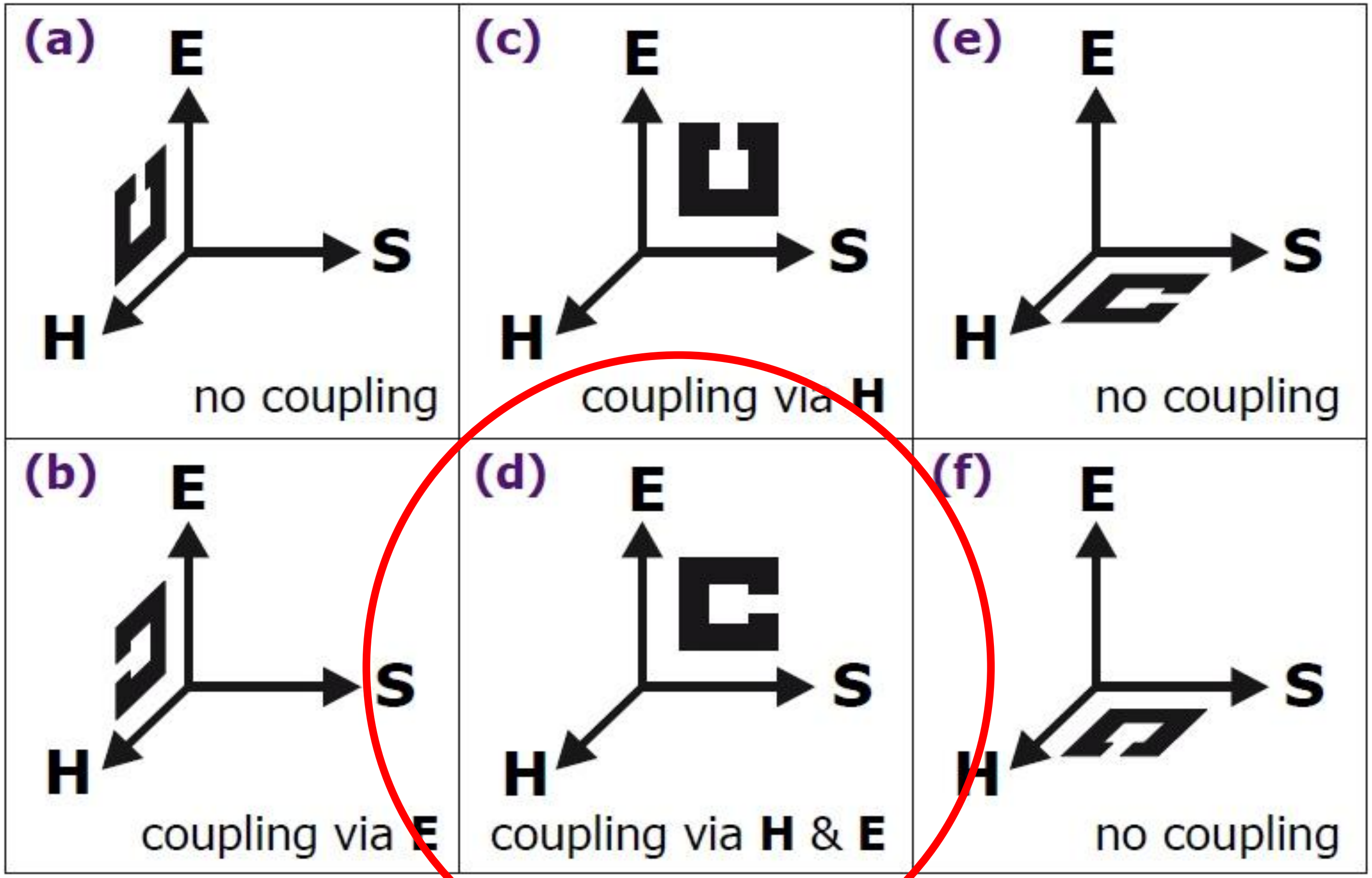
Metamaterials as effective media

Investigators of …

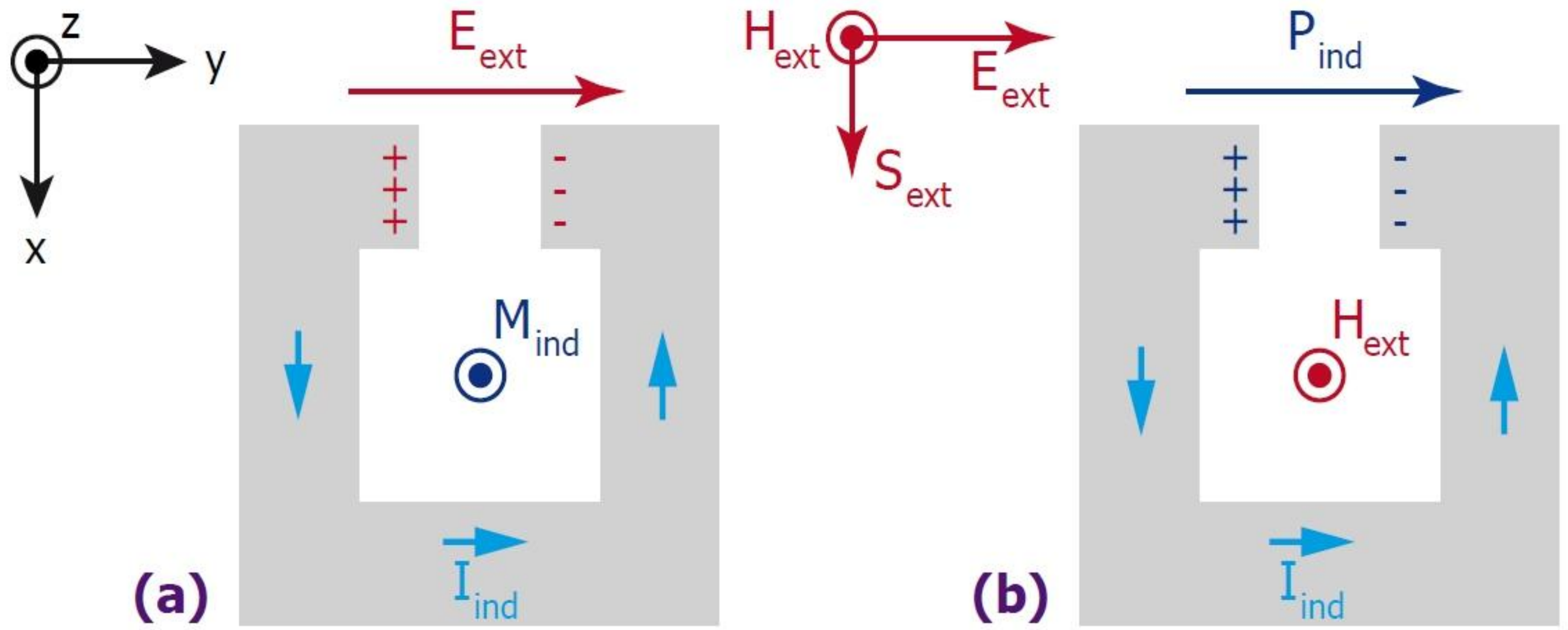
- Metallic sub-wavelength structures
  → **effective material** with $\mu \neq 1$

- **Non-symmetric atoms** along propagation direction
  → cross-coupling

Metamaterials as effective media



Metamaterials as effective media



C. Kriegler, diploma thesis (2008)

Metamaterials as effective media

Physical description:

$$\vec{D} = \varepsilon_0 \overleftrightarrow{\varepsilon} \vec{E} + c_0^{-1} \overleftrightarrow{\xi} \vec{H}$$

$$\vec{B} = c_0^{-1} \overleftrightarrow{\zeta} \vec{E} + \mu_0 \overleftrightarrow{\mu} \vec{H}$$

$$\overleftrightarrow{\varepsilon} = \begin{pmatrix} \varepsilon_x & 0 & 0 \\ 0 & \varepsilon_y & 0 \\ 0 & 0 & 1 \end{pmatrix}, \quad \overleftrightarrow{\mu} = \begin{pmatrix} 1 & 0 & 0 \\ 0 & 1 & 0 \\ 0 & 0 & \mu_z \end{pmatrix}$$

$$\overleftrightarrow{\xi} = \begin{pmatrix} 0 & 0 & 0 \\ 0 & 0 & -i\xi \\ 0 & 0 & 0 \end{pmatrix}, \quad \overleftrightarrow{\zeta} = \begin{pmatrix} 0 & 0 & 0 \\ 0 & 0 & 0 \\ 0 & i\xi & 0 \end{pmatrix}$$

- Reciprocal medium
  (exchange of currents and fields is possible / NO Faraday effect)
- Geometry as shown before

X. Chen *et al.*, Phys. Rev. E **71**, 046610 (2005)
R. Marqués *et al.*, Phys. Rev. B **65**, 144440 (2002)

Metamaterials as effective media

Long story short:

$$D_y = \varepsilon_0 \varepsilon_y E_y - i\xi c_0^{-1} H_z \quad ; \quad B_z = \mu_0 \mu_z H_z + i\xi c_0^{-1} E_y$$

After calculating the dispersion relation with an harmonic ansatz (as "usual"), we finally derive:

$$n = \sqrt{\varepsilon_y \mu_z - \xi^2} \; ; \qquad z_\pm \equiv \frac{Z_\pm}{Z_{\mathrm{vac}}} = \frac{\mu_z}{\pm n - i\xi}$$

→ direction dependant

Metamaterials as effective media

How can we determine *n* and $z_{\pm}$?

- Effective material parameters are not observable.
- Easiest way: measure T and $R_{\pm}$
- Problem: Lossy material ➔ $\varepsilon, \mu, \xi \in \mathbb{C}$
  (6 unknown variables)

- **Solution:** Simulate $t, r_{\pm} \in \mathbb{C}$ and compare with experimental data

We already noticed that Z_+ is the impedance of the bianisotropic medium in the (+)-direction and Z_- is the opposite of the impedance in the (−)-direction. This accounts for (A.18). Yet, it is not clear a priori whether z_+ and z_- defined in (A.17) fulfil the condition (A.18). We decided to define $z_\pm$ as in (A.17) because it turned out to fulfil (A.18) in all the cases we studied, but the condition (A.18) should be tested in every case.

To find the refractive index, we can rewrite (A.13) and (A.14) as:

$$\begin{aligned} t_+ &= \frac{e^{\imath n k_0 d}}{\left(1-\frac{z_-}{z_t}\right)}\left(\left(1-\frac{z_-}{z_t}\right)+r_+\left(1+\frac{z_-}{z_t}\right)\right) \\ t_+ &= \frac{e^{-\imath n k_0 d}}{\left(1-\frac{z_+}{z_t}\right)}\left(\left(1-\frac{z_+}{z_t}\right)+r_+\left(1+\frac{z_+}{z_t}\right)\right) \end{aligned}$$

Finally, we get an implicit expression for the refractive index

$$\boxed{\cos n k_0 d = \frac{t_+}{2}\left(\frac{1-\frac{z_-}{z_t}}{\left(1-\frac{z_-}{z_t}\right)+r_+\left(1+\frac{z_-}{z_t}\right)}+\frac{1-\frac{z_+}{z_t}}{\left(1-\frac{z_+}{z_t}\right)+r_+\left(1+\frac{z_+}{z_t}\right)}\right)}. \tag{A.19}$$

A.3 Retrieval of the Effective Parameters

(A.19) has an infinity of solutions for n due to the inverse cosine. We proceed as in the symmetric case to choose the correct branch [Enk05].

Once $z_\pm$ and n are calculated, we deduce ϵ, μ and ξ from (A.19) and (A.17):

$$\begin{aligned} \xi &= \imath n\left(\frac{z_-+z_+}{z_--z_+}\right) \\ \epsilon &= \frac{n+\imath\xi}{z_+} \\ \mu &= z_+\left(n-\imath\xi\right) \end{aligned}$$

A.3. RETRIEVAL OF THE EFFECTIVE PARAMETERS

Metamaterials as effective media

$$z_{\pm} = \left(-b \mp \sqrt{b^2 - 4ac}\right)/(2a), \text{ with}$$

$$a = t_{\text{air}} t_{\text{sub}} - (1 - r_{\text{air}})\,(1 - r_{\text{sub}})\,,$$

$$b = (z_{\text{air}} - z_{\text{sub}})\,(t_{\text{air}} t_{\text{sub}} + 1 - r_{\text{air}} r_{\text{sub}}) + (z_{\text{air}} + z_{\text{sub}})\,(r_{\text{air}} - r_{\text{sub}})\,,$$

$$c = z_{\text{air}} z_{\text{sub}}\,(-t_{\text{air}} t_{\text{sub}} + (1 + r_{\text{air}})\,(1 + r_{\text{sub}}))\,,$$

$$\cos(nk_0 d) = \frac{t_{\text{air}}}{2}\left[\frac{\left(1 - \frac{z_-}{z_{\text{sub}}}\right)}{\left(1 - \frac{z_-}{z_{\text{air}}}\right) + r_{\text{air}}\left(1 + \frac{z_-}{z_{\text{air}}}\right)} + \frac{\left(1 - \frac{z_+}{z_{\text{sub}}}\right)}{\left(1 - \frac{z_+}{z_{\text{air}}}\right) + r_{\text{air}}\left(1 + \frac{z_+}{z_{\text{air}}}\right)}\right].$$

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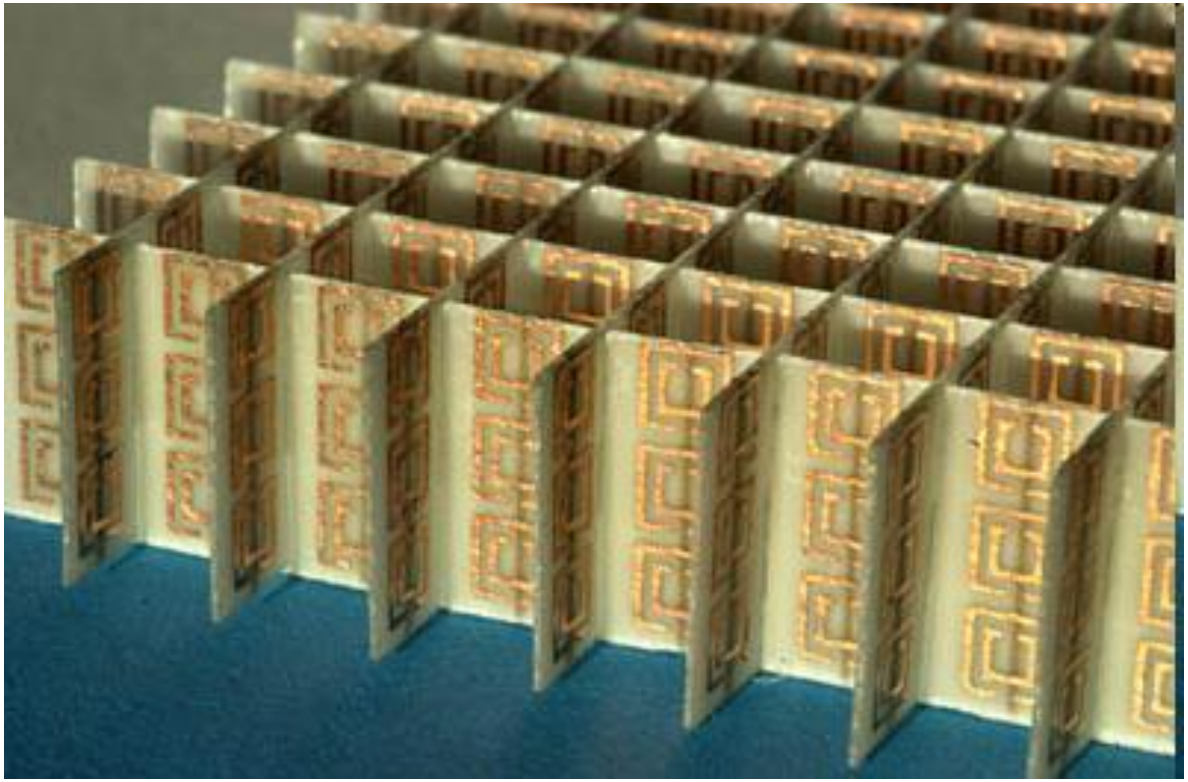
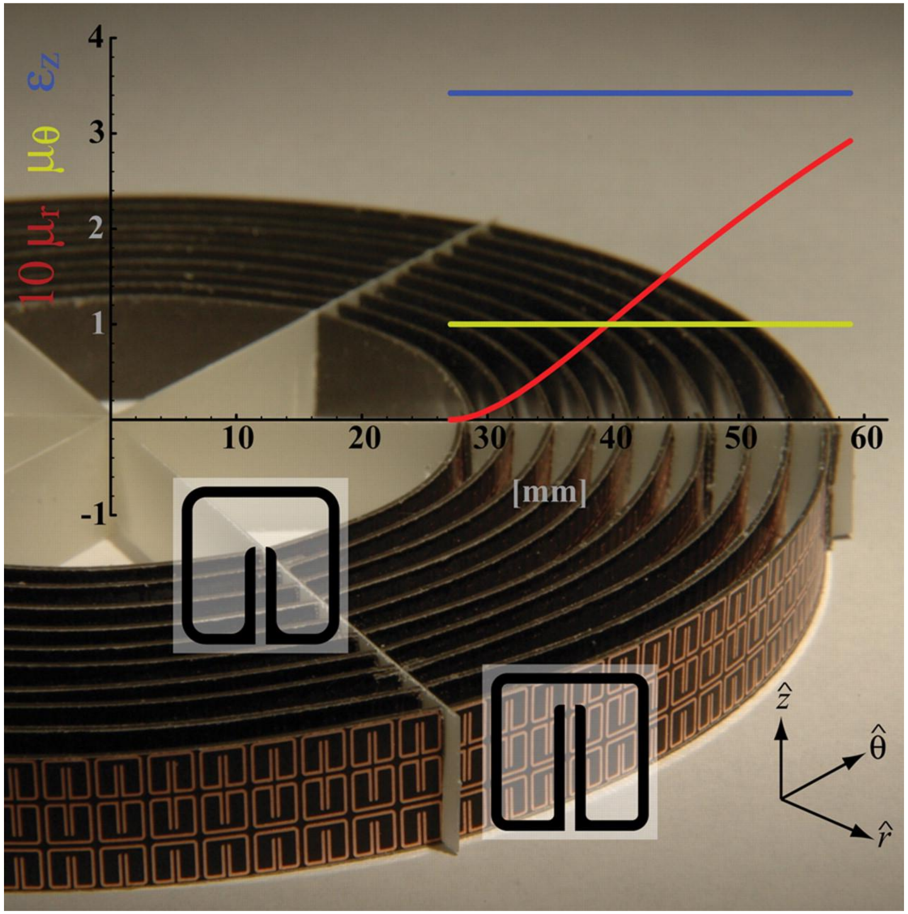
Hitherto existing realizations of 3D photonic metamaterials

"Experimental verification of a negative index of refraction"



R.A. Shelby *et al.*, Science **292**, 77 (2001)

Hitherto existing realizations of 3D photonic metamaterials

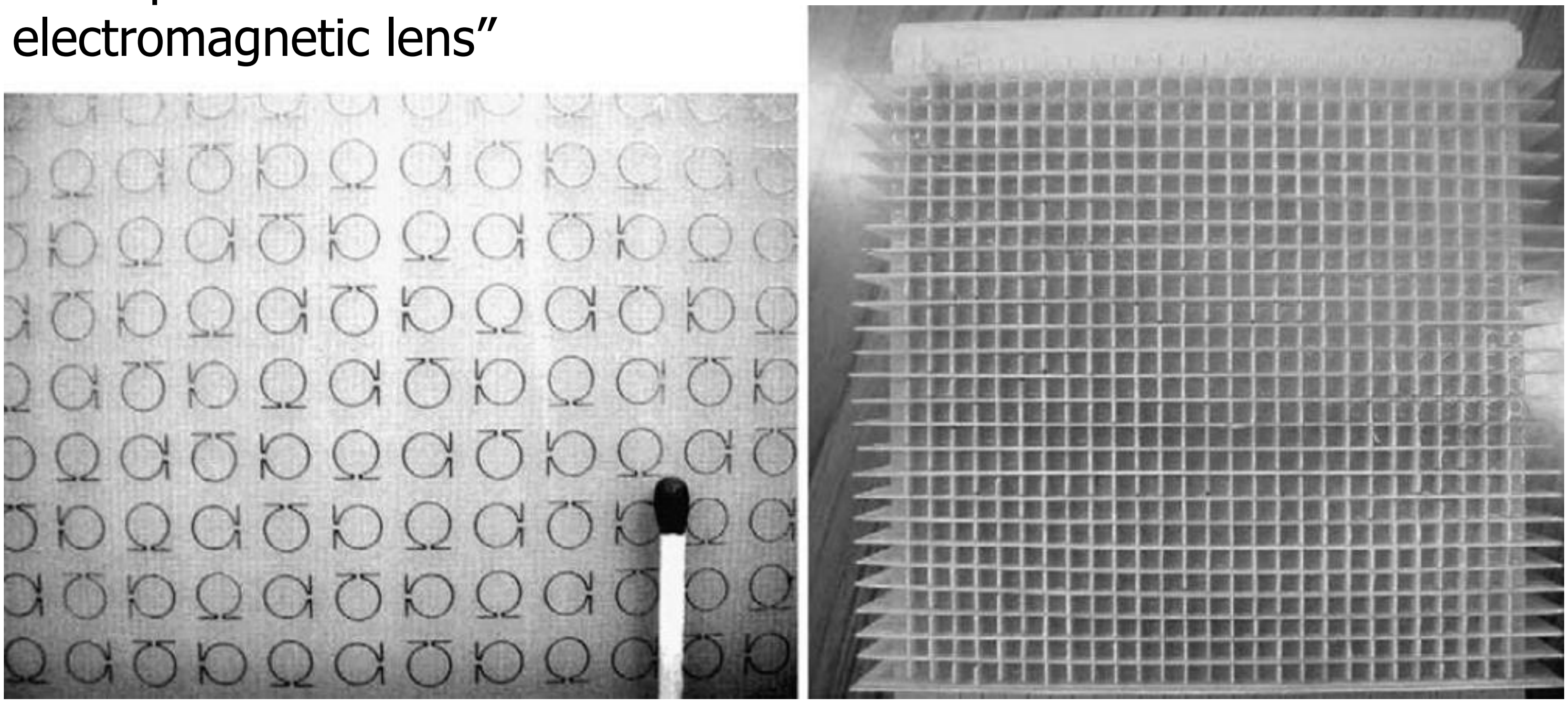


"3D metamaterial electromagnetic cloak at microwave frequencies"

D. Schurig *et al.*, Science **314**, 977 (2006)

Hitherto existing realizations of 3D photonic metamaterials

"Isotropic metamaterial electromagnetic lens"



E. Verney *et al.*, Phys. Lett. A **331**, 244 (2004)

Hitherto existing realizations of 3D photonic metamaterials

A real 3D metamaterial for the NIR or VIS optical range (regarding $\lambda < d_{\mathrm{propdir}}$) has not been reported, yet!

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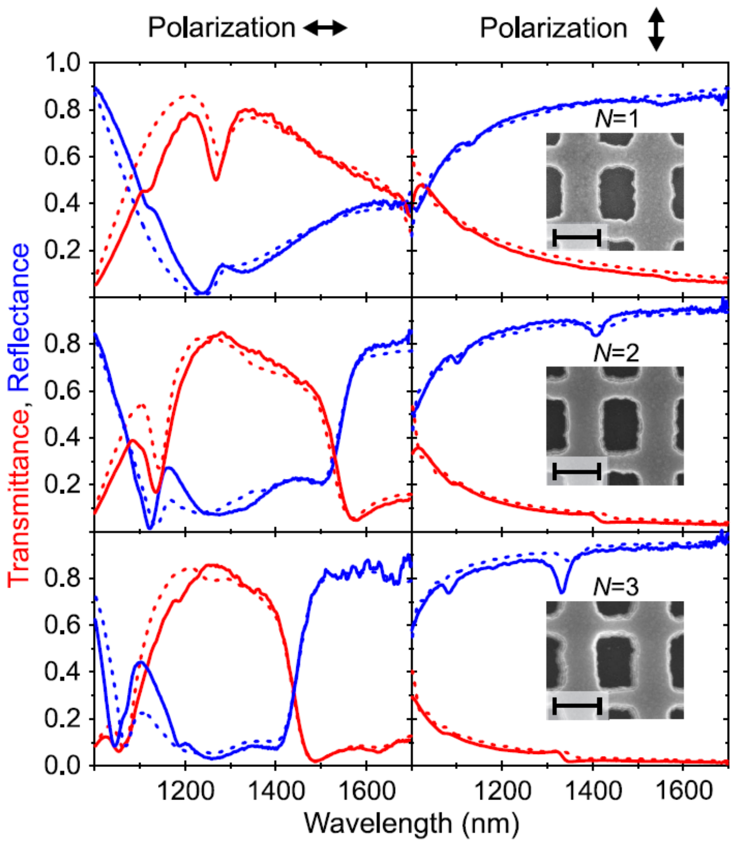
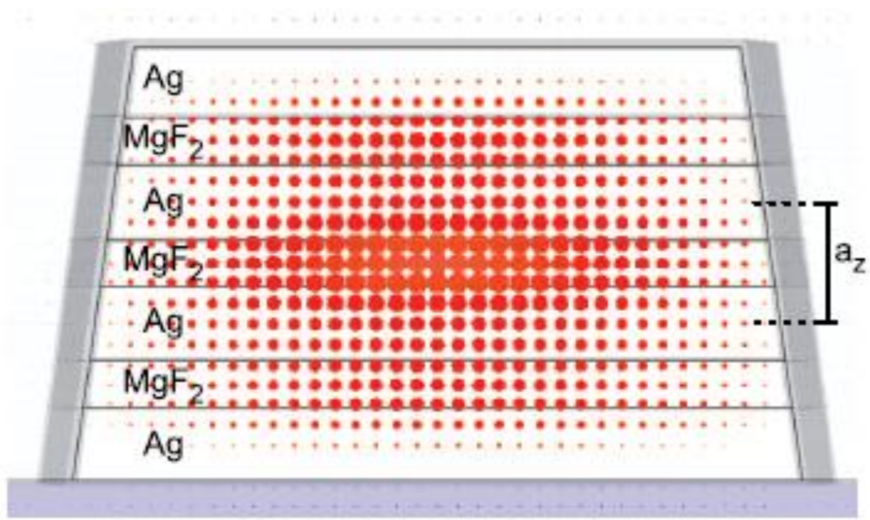
Fabricational approaches for the NIR:
Simple stacking (layer-by-layer)





G. Dolling *et al.*, Opt. Lett. **32**, 551 (2007)

Fabricational approaches for the NIR: Simple stacking (layer-by-layer)

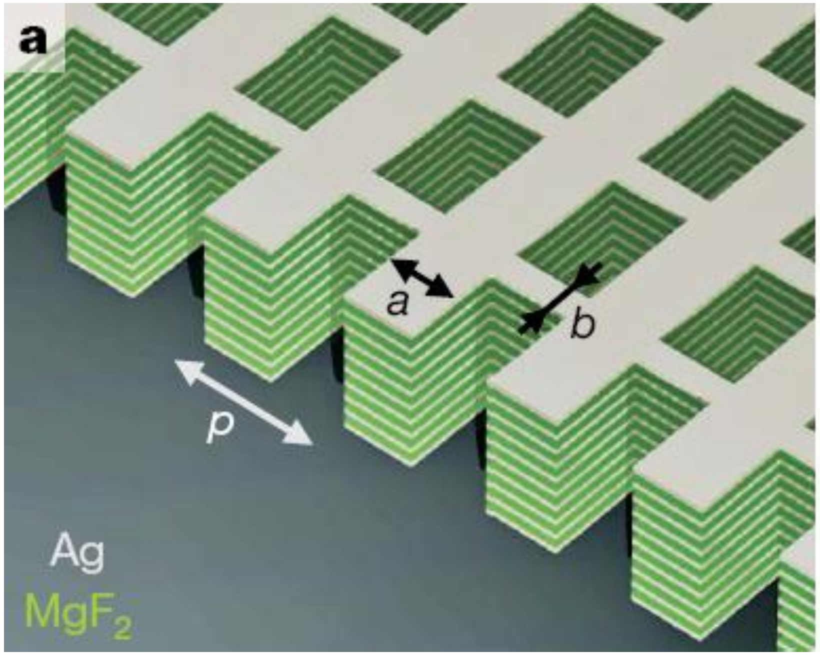
PROs:

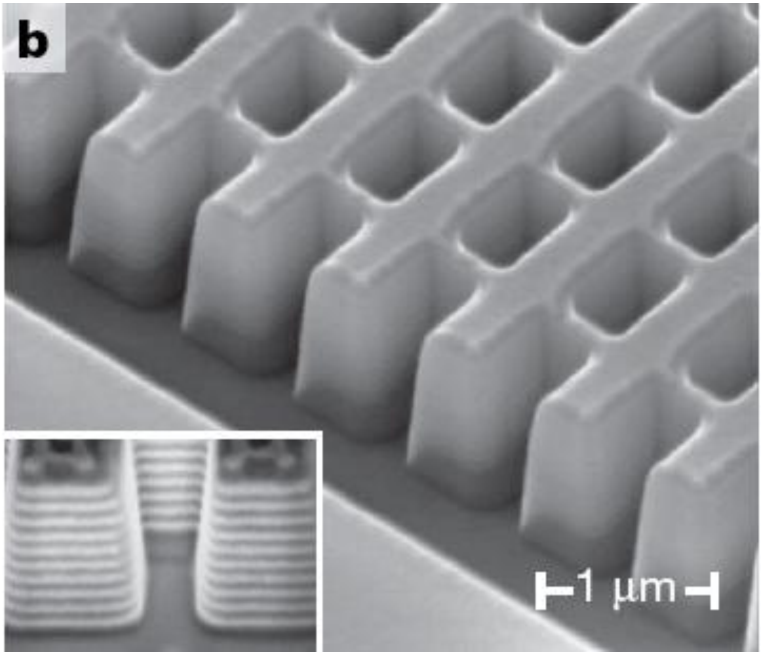
- No special fabrication techniques needed
- Structuring needs just one process step

CONs:

- Stacking of more than 4 functional layers not possible due to lift-off problems
- Tapering

Fabricational approaches for the NIR:
FIB-milling from the solid





J. Valentine *et al.*, Nature **455**, 376 (2008)

Fabricational approaches for the NIR:
FIB-milling from the solid

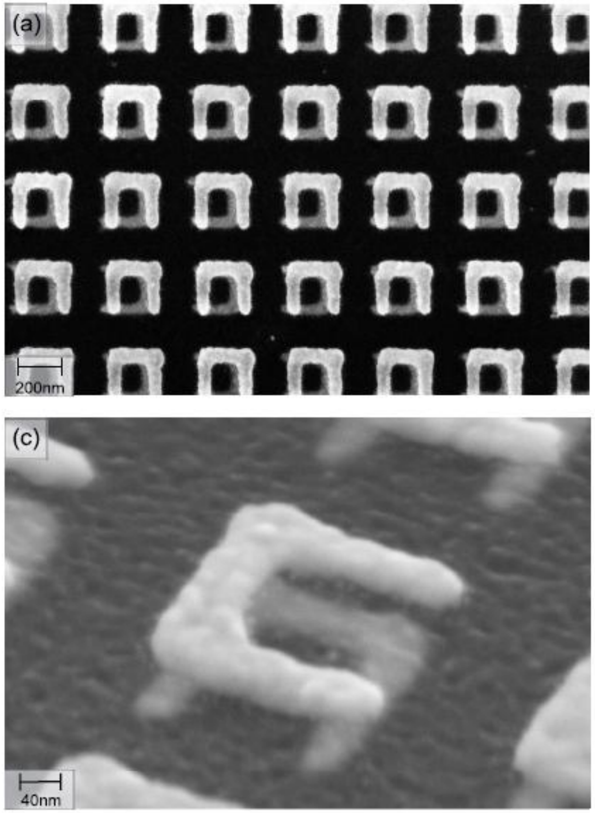
PROs:

- No special fabrication techniques needed
- Structuring needs just one process step

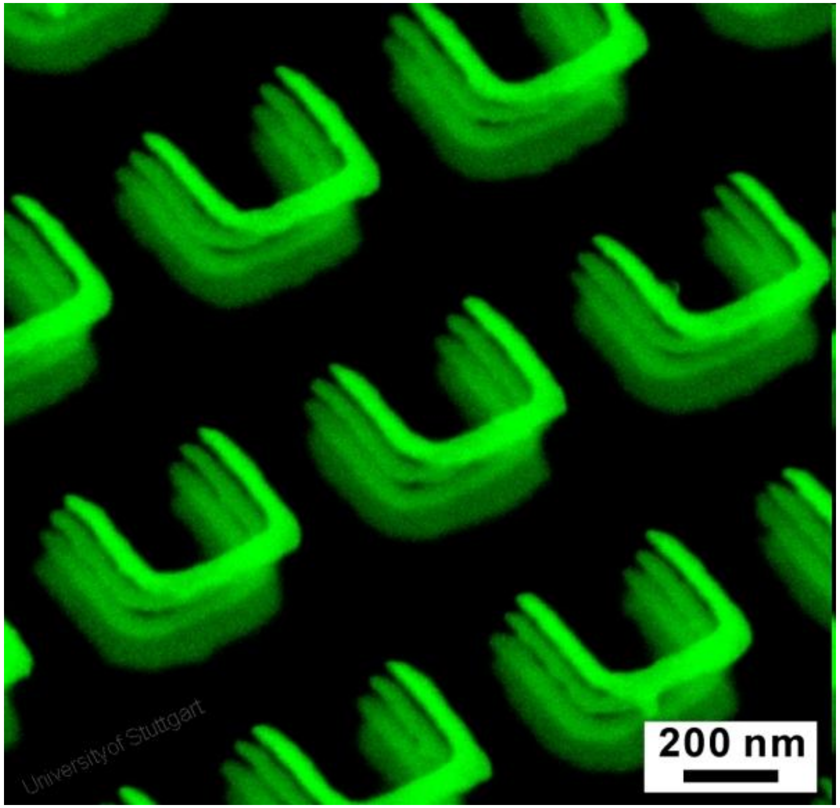
CONs:

- Tapering (max. 10 functional layers)
- Ga contamination due to FIB-milling
- Restriction to planar structure designs

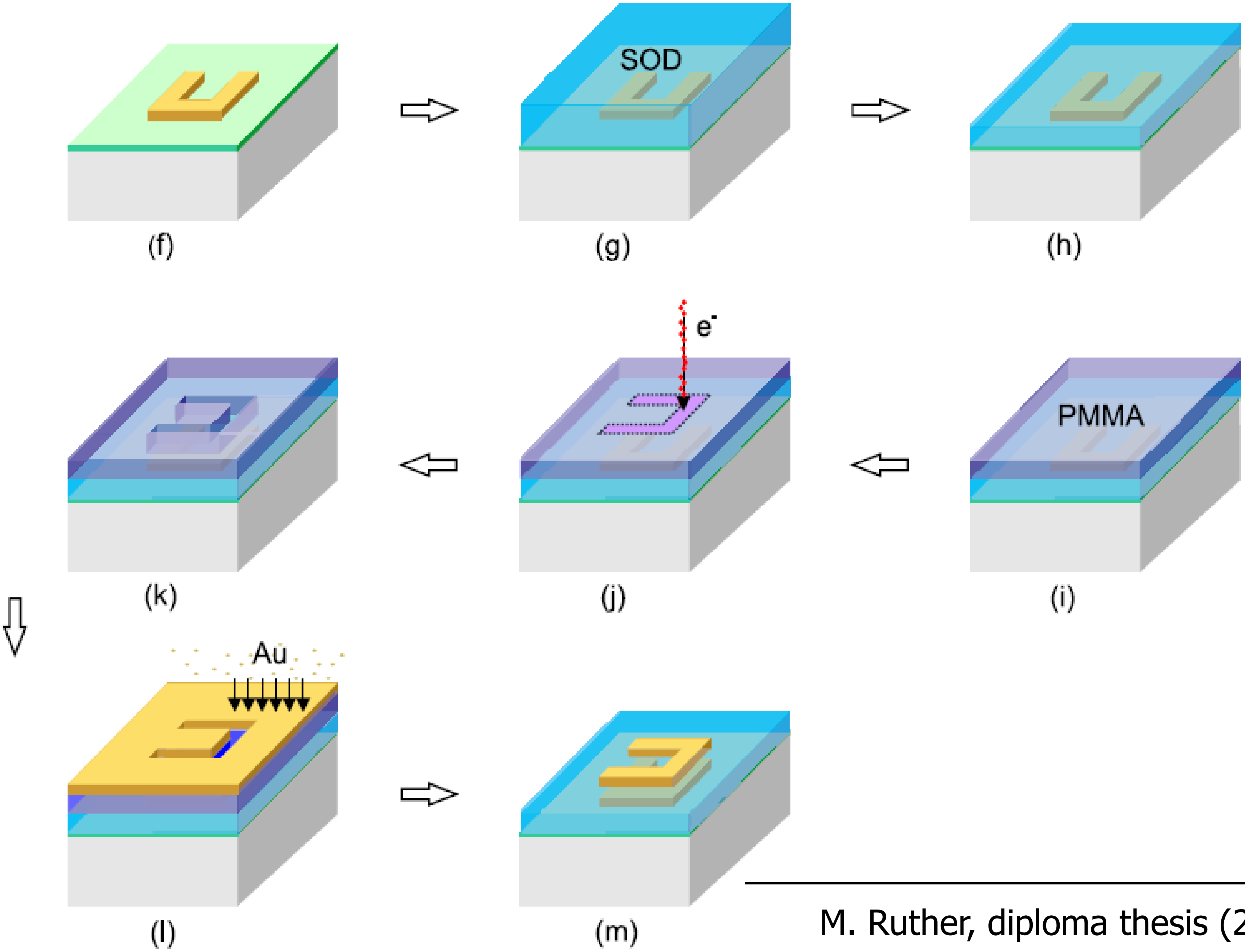
Fabricational approaches for the NIR: Planarization method (layer-by-layer)



M. Ruther, diploma thesis (2007)



N. Liu *et al.*, Nature Mater. **7**, 31 (2008)



M. Ruther, diploma thesis (2007)

Fabricational approaches for the NIR: Planarization method (layer-by-layer)

PROs:

- Spin-on-dielectric (IC2-200® – Polysiloxane compound) acts as a capacitance
- Arbitrarily scalable

CONs:

- Very time-consuming (many process steps) → approx. 1 day per unit cell
- Re-alignment before e-beam lithography
- Restriction to planar structure designs

Fabricational approaches for the NIR: Intermediate conclusions

What do all these approaches have in common?

- Restriction to planar (2D) designs
  → unsuitable for isotropic structures
  → very hard to achieve heights $d > \lambda$
- Time-consuming (serial processing)

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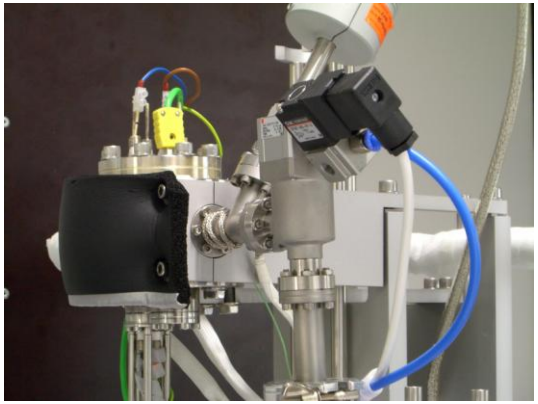
Our fabrication approach
Overview

1. Fabrication of dielectric template by direct laser writing

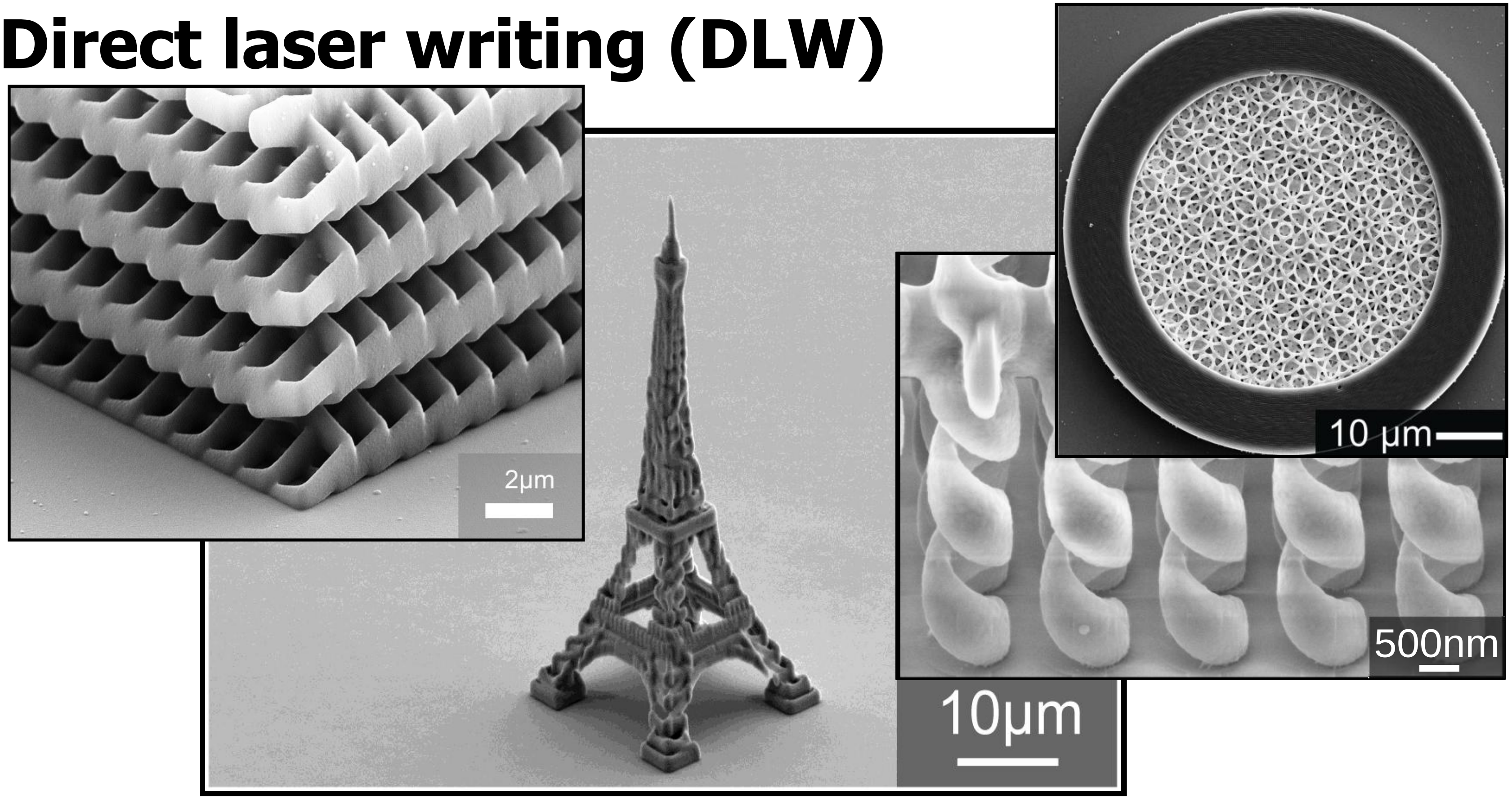
2. Protection layer by SiO_2 or TiO_2 atomic layer deposition

3. Isotropic metallization



Our fabrication approach
Step 1: Direct laser writing

Direct laser writing (DLW)



M. Thiel, diploma thesis
A. Ledermann *et al.*, Nature Mater. **5**, 942 (2006)

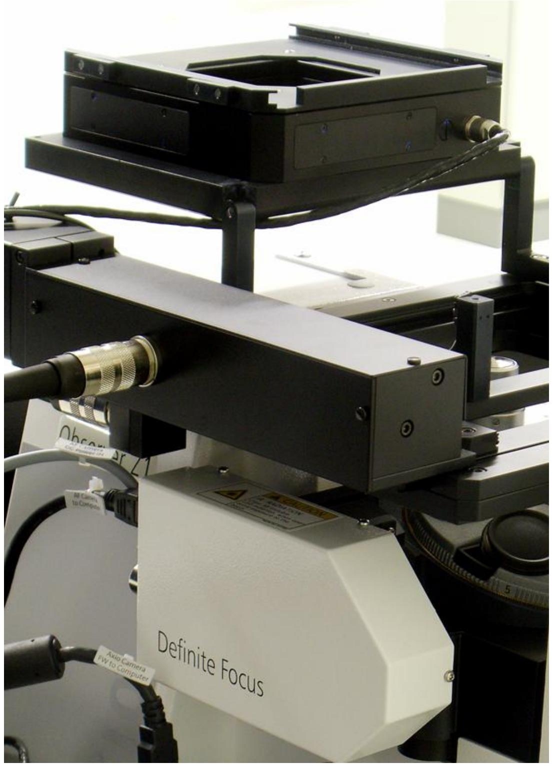
Our fabrication approach
Step 1: Direct laser writing

PROs:

- Feature sizes down to 100 nm (depends mainly on photo resist)
- Sample volume: 300 µm x 300 µm x 80 µm
- Easy fabrication (CAD support, WYSIWYHTG)
- Chemically and mechanically robust

CONs:

- Always connected structures
- Sensitive to high temperatures (> 150 °C)
- Proximity effect
- Aspect ratio of voxel (2.8:1 for SU-8)



Our fabrication approach
Step 2: Protection layer

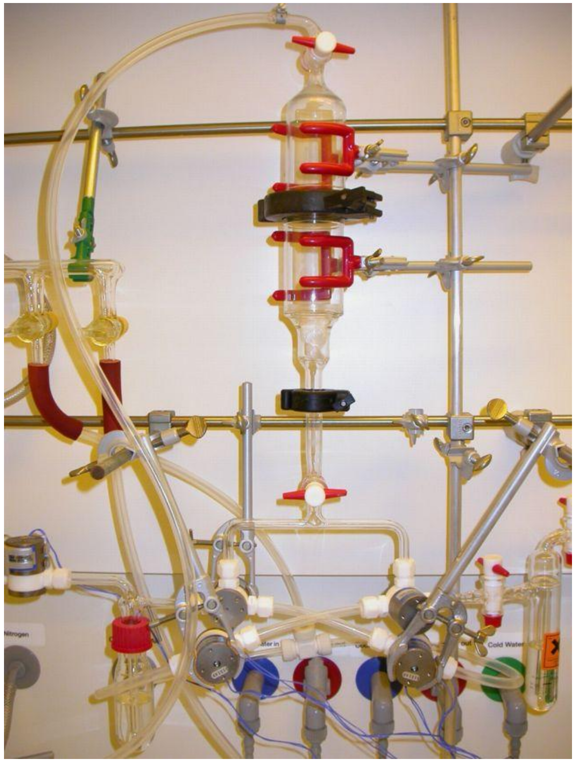
Protection coating before metallization

- **Reason:**
  Deformation of template @ 160˚C

- **SiO_2 PLD Process:**
  - Alternating pulses of H_2O and $SiCl_4$:

    $$SiCl_4 + H_2O \rightarrow SiO_2 + HCl$$

  - Purging with N_2 in between
  - Process @ room temperature and atmospheric pressure



E. J. Ehrlich *et al.*, Appl. Phys. Lett. **58**, 2675 (1991)

Our fabrication approach
Step 2: Protection layer

Protection coating before metallization

- **TiO_2 ALD Process:**
  - Alternating pulses of H_2O and $TiCl_4$:

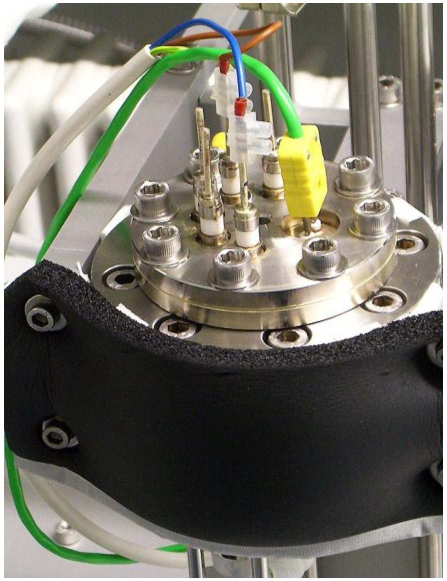
    $$\mathbf{TiCl_4 + H_2O \rightarrow TiO_2 + HCl}$$

  - Purging with N_2 in between
  - Process @ 110°C
  - Self-saturazing (1 monolayer per cycle)

http://www.cambridgenanotech.com

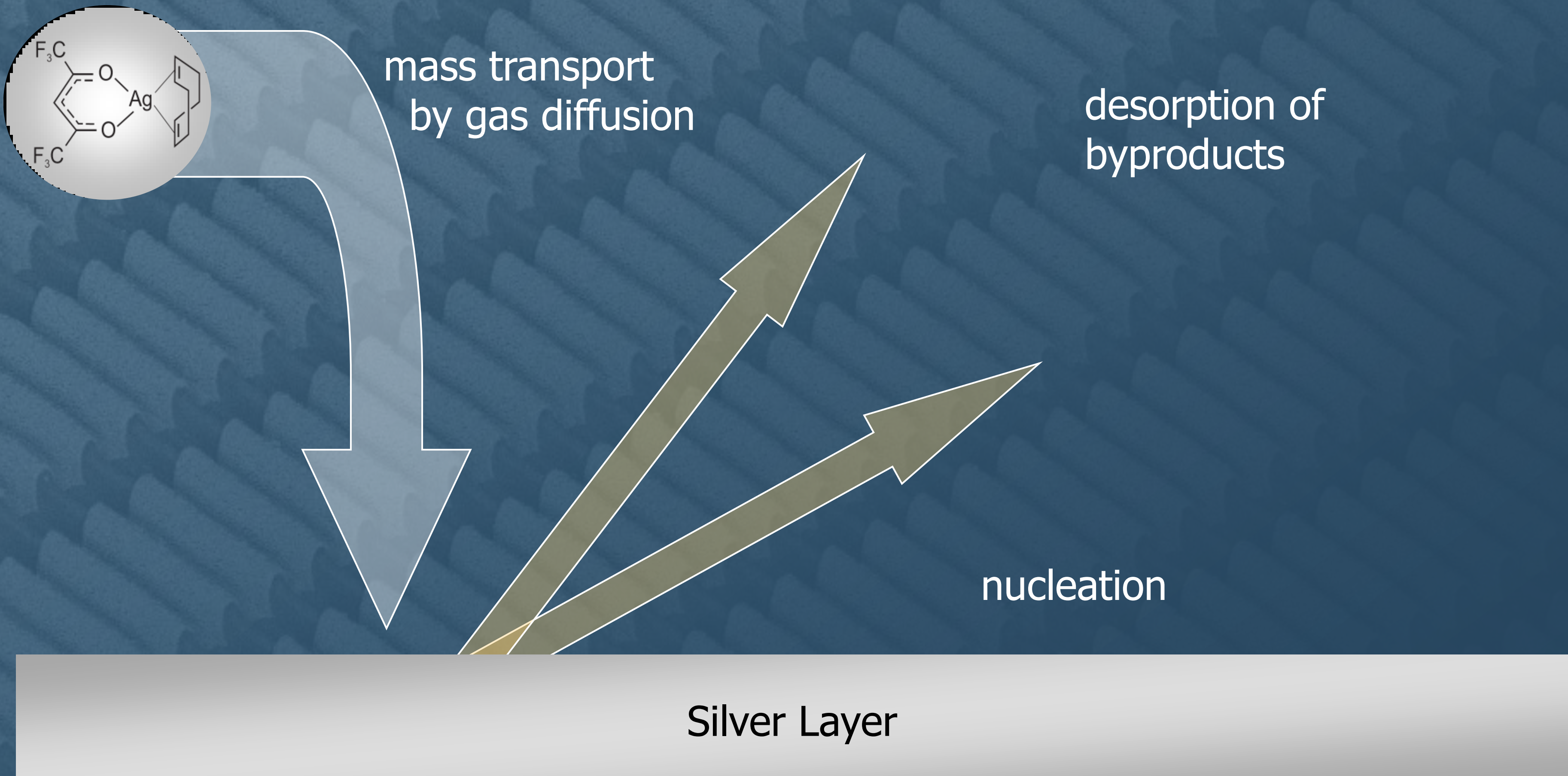
Our fabrication approach
Step 3: Metallization

- Sputtering processes not suitable (only 2D)
- **Useful 3D metallization processes:**
  - Electrochemical deposition (ECD)
    e.g., $Na_3[O_3S\text{-}Au^{(1)}\text{-}SO_3]$ (aqueous solution)
  - Atomic layer deposition (ALD)
    e.g., $[Cu(^sBu\text{-amd})]_2 + NH_3 \rightarrow Cu_3N + \text{lig.}$
    $2\ Cu_3N + 3\ H_2 \rightarrow 6\ Cu + 2\ NH_3$
  - Chemical vapor deposition (CVD)
    e.g., $(COD)(hfac)Ag^{(1)}$



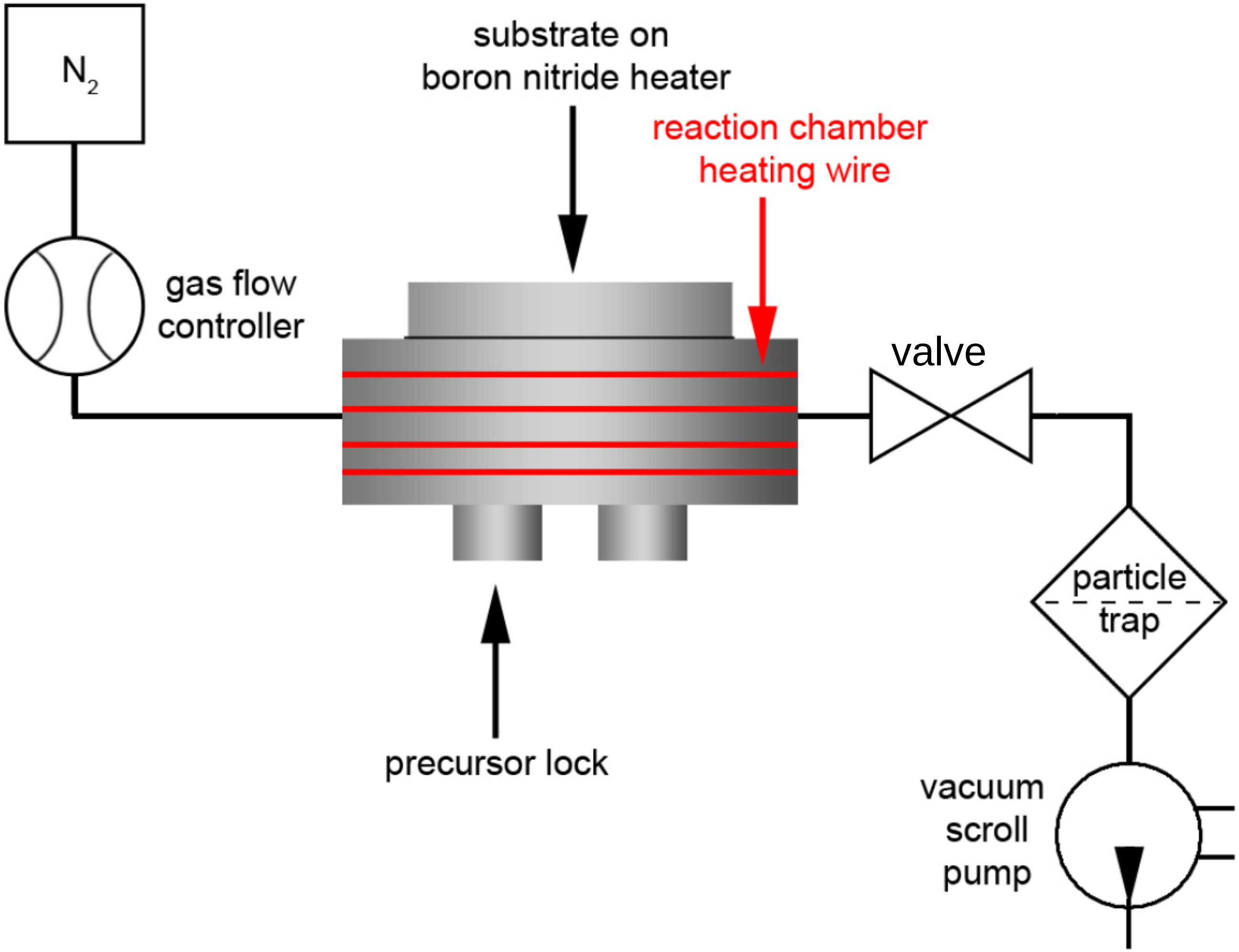
Me, Myself and I / A. Gänsel, unpublished

Our fabrication approach
Step 3: Metallization

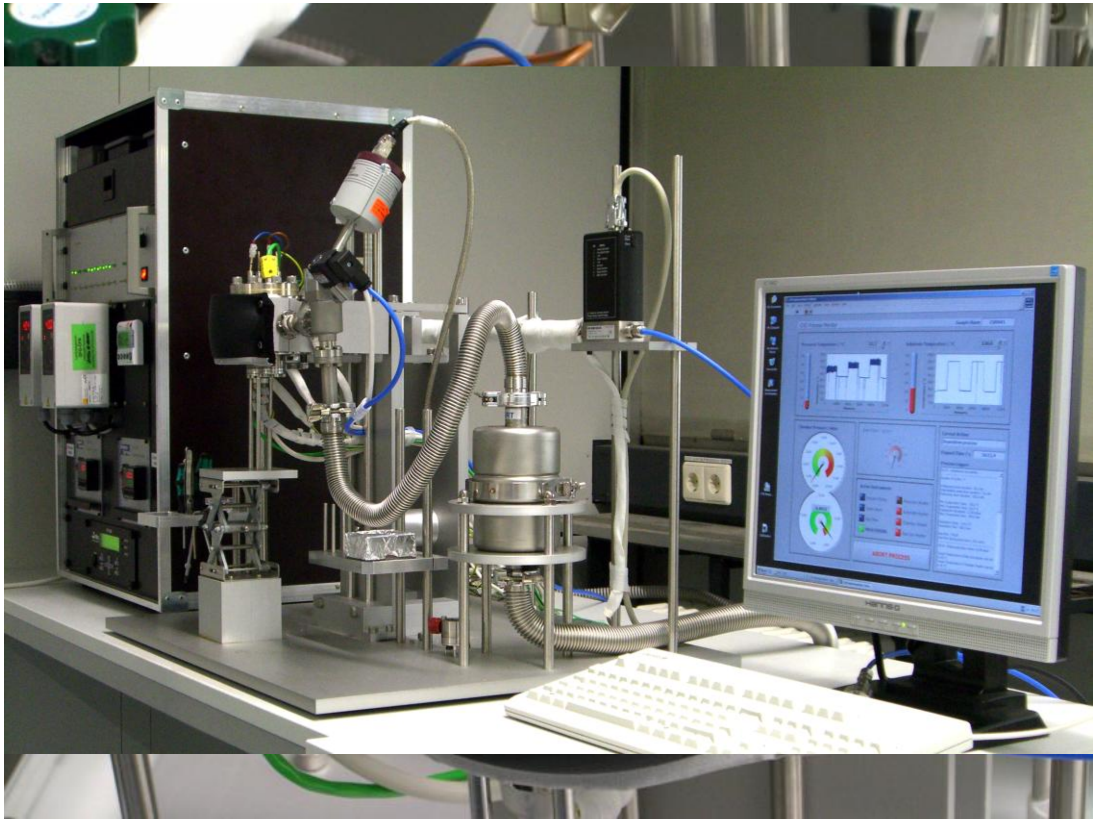


E. Eisenbraun *et al.*, J. Vac. Sci. Technol. B **19**, 585 (2001)

Our fabrication approach
Step 3: Metallization



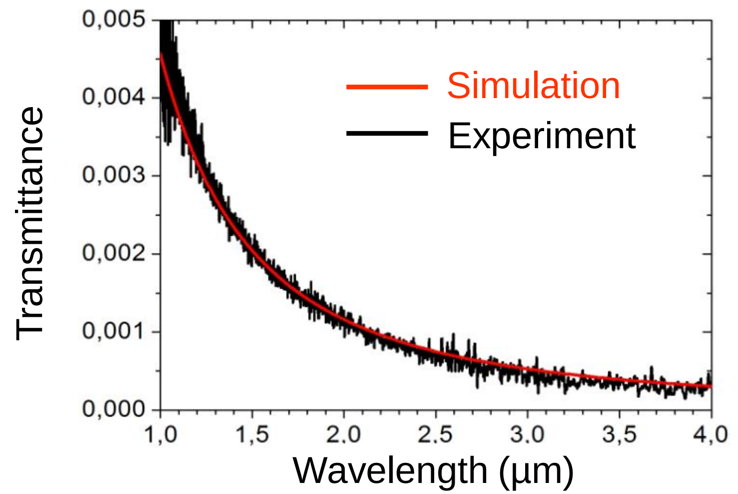
Our fabrication approach
Step 3: Metallization

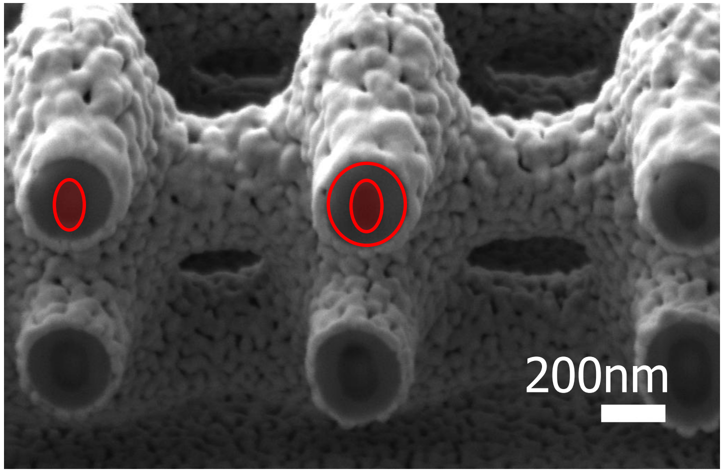


- Custom design
- Complete automation
  - reproducable
  - long-time deposition
- Several running modes possible
  - dynamic
  - static
  - pulsed

Our fabrication approach
Step 3: Metallization

- **PROs:**
  - Connected films (no particle plasmons)
  - Pure silver coating
  - Drude model fits well
  - Isotropic coating of 3D templates
- **CONs:**
  - Whole surface is metallized (even the substrate!)
  - Deep trenches and small holes cause problems

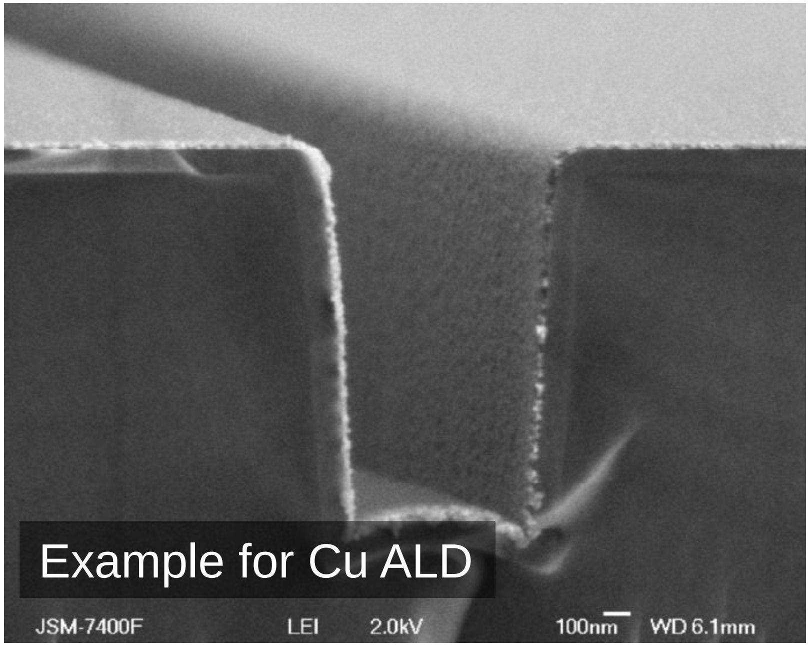




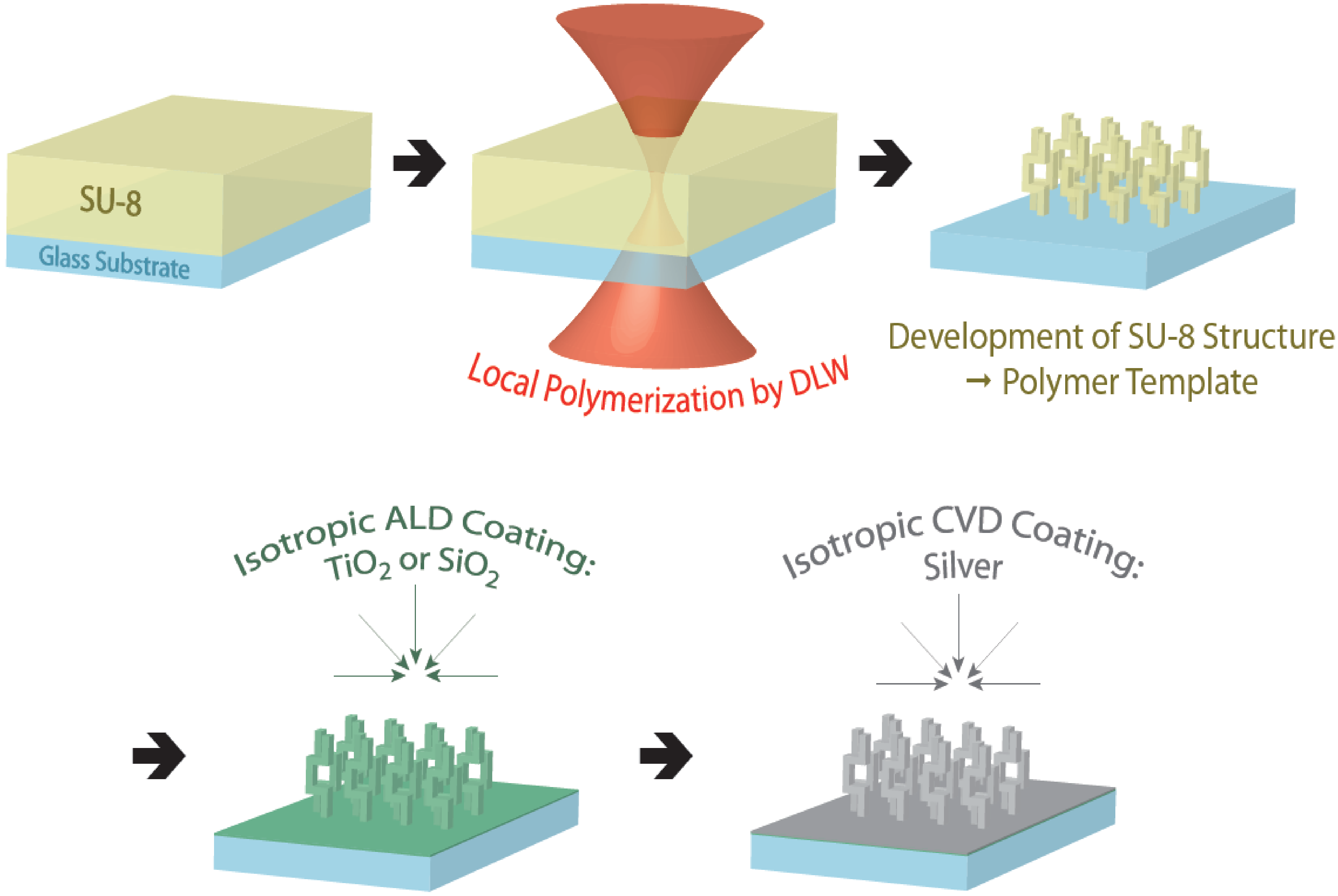
Our fabrication approach
Step 3: Metallization

- Solution for metallization of substrate:
  Lift-off process with soluable photo resist

- Metallization of deep trenches and high filling fractions:
  Switch to ALD processes



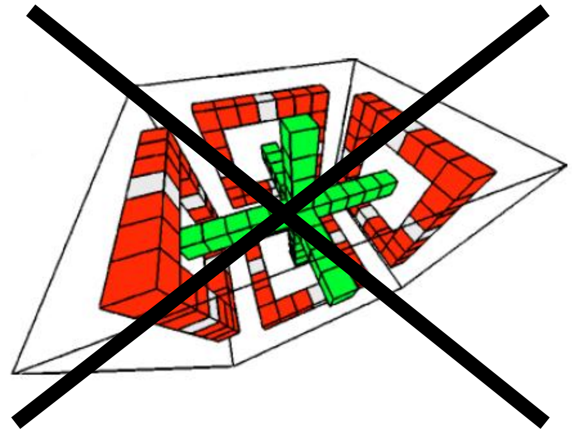
Niskanen *et al.*,
J. Elec. Chem. Soc. **152**, G25 (2005)



SU-8
Glass Substrate
Local Polymerization by DLW
Development of SU-8 Structure
→ Polymer Template
Isotropic ALD Coating:
TiO_2 or SiO_2
Isotropic CVD Coating:
Silver

Our fabrication approach
Boundary conditions

- **Connected** template structure (no "flying" features)
- **Whole surface** is metallized (no locally defined coatings)
- **Minimum periodicity** defined by direct laser writing → suitable for near-IR



T. Koschny *et al.*, PRB **71**, 121103 (2005)

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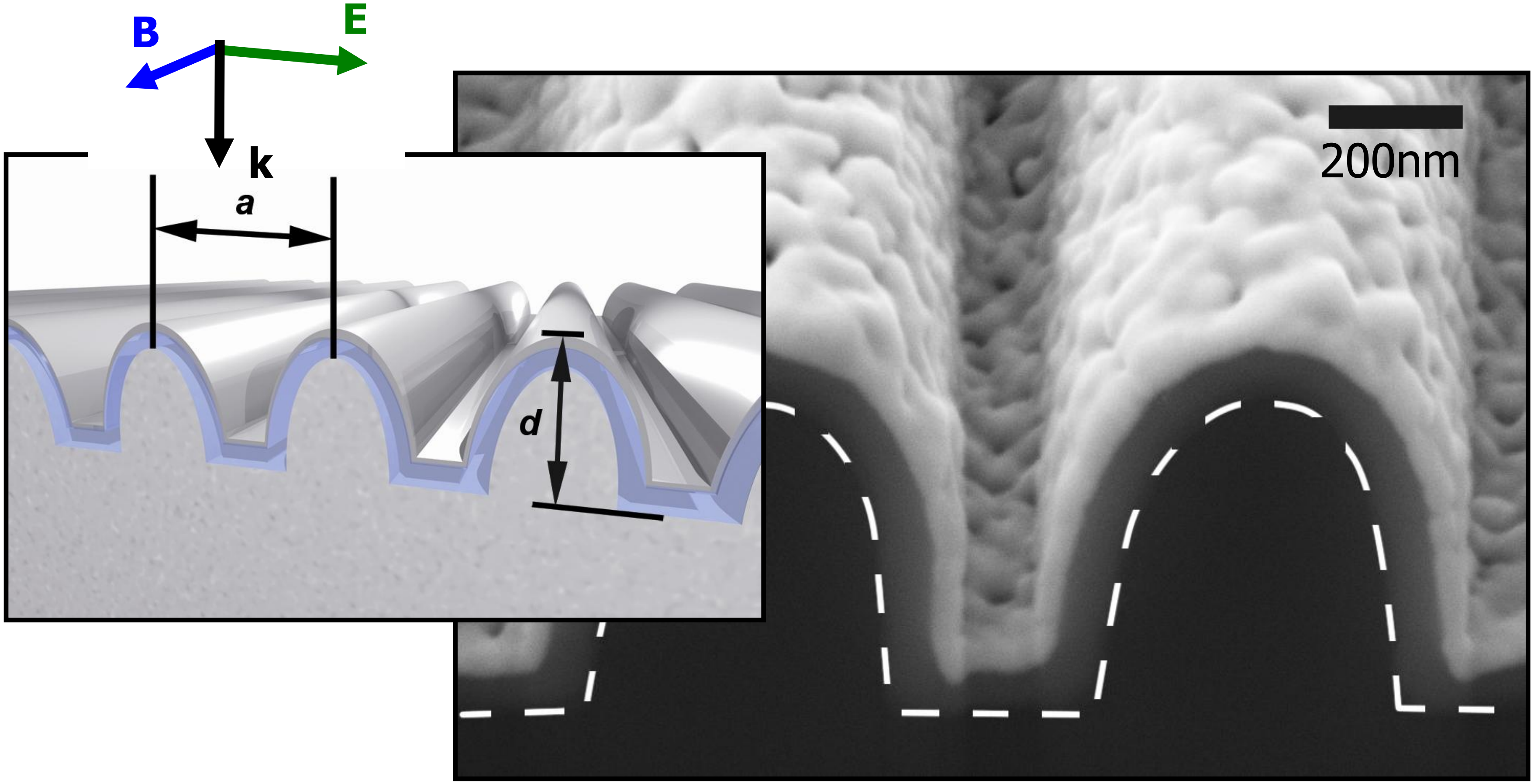
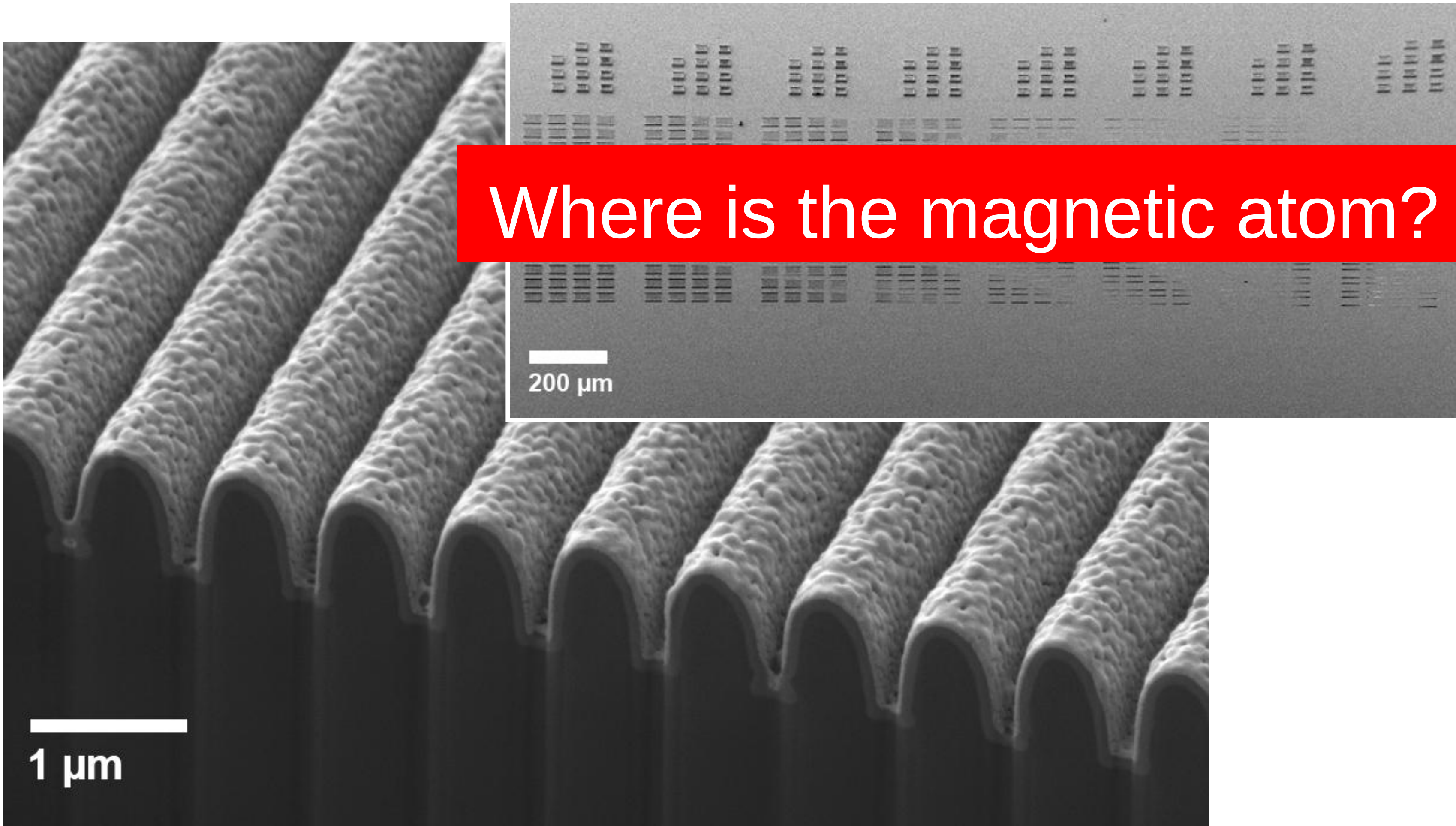
Recent results
Corrugated surface structure

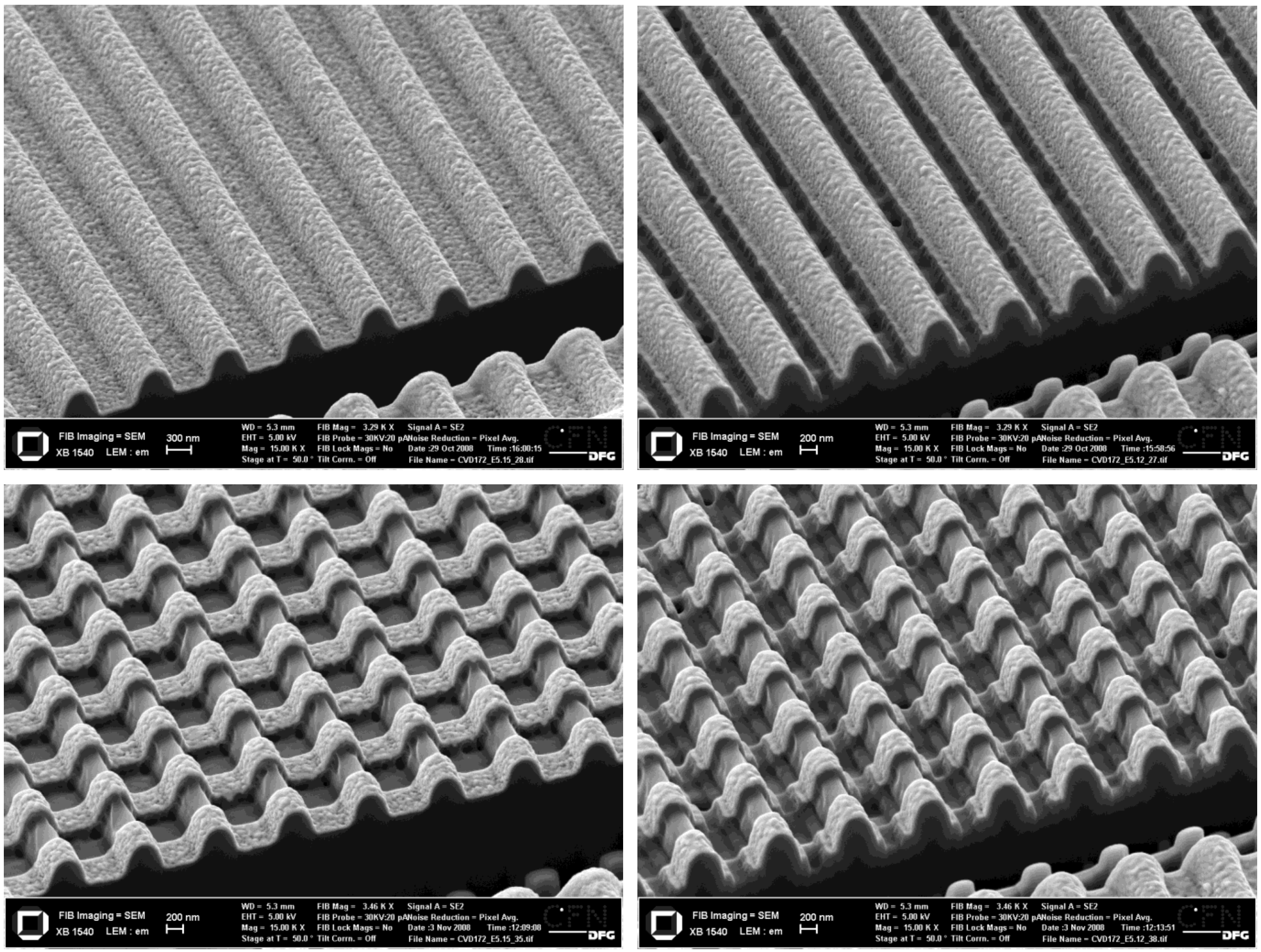


M.S. Rill *et al.*, Nature Mater. **7**, 543 (2008)

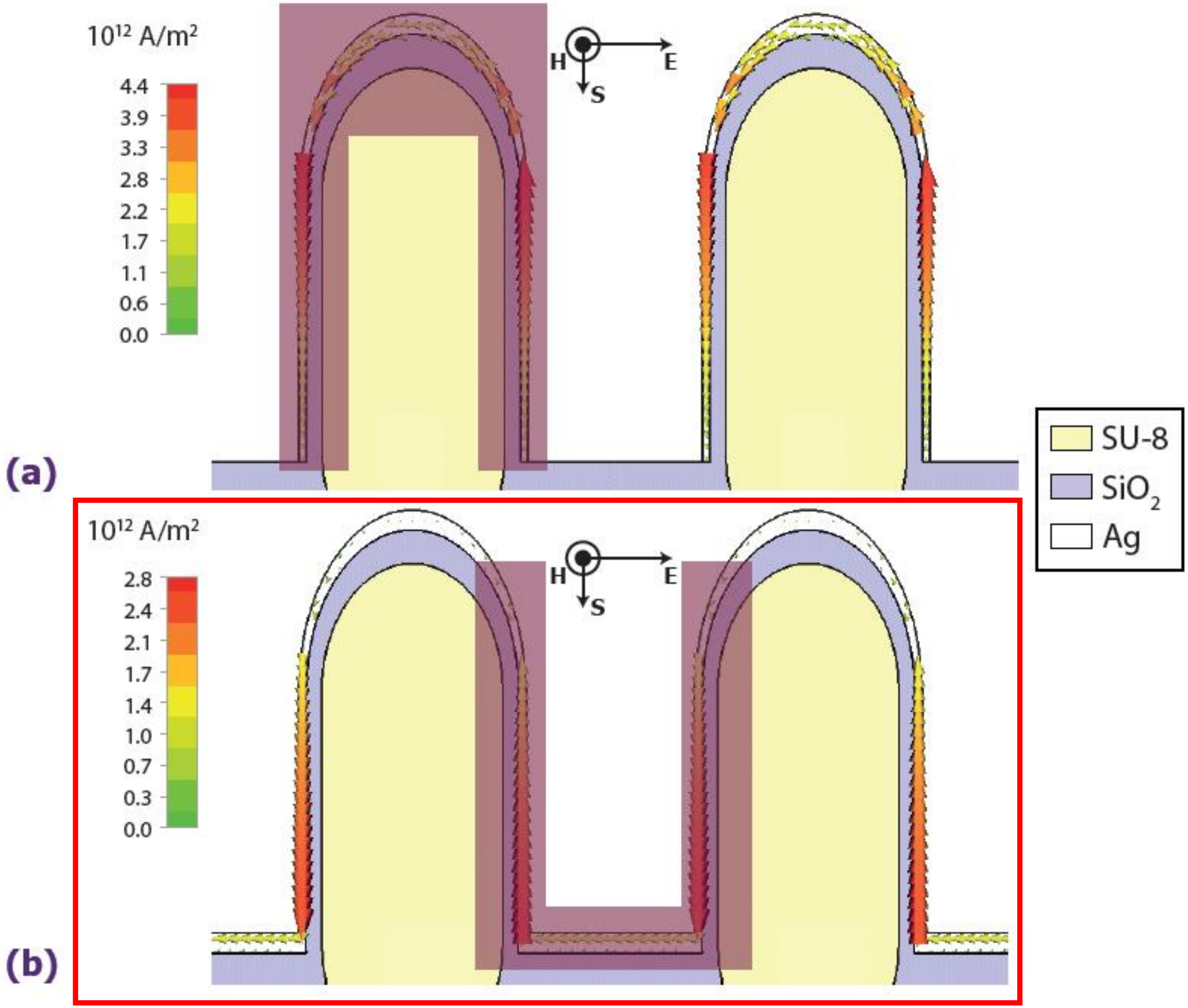
Recent results
Corrugated surface structure



Recent results
Corrugated surface structure

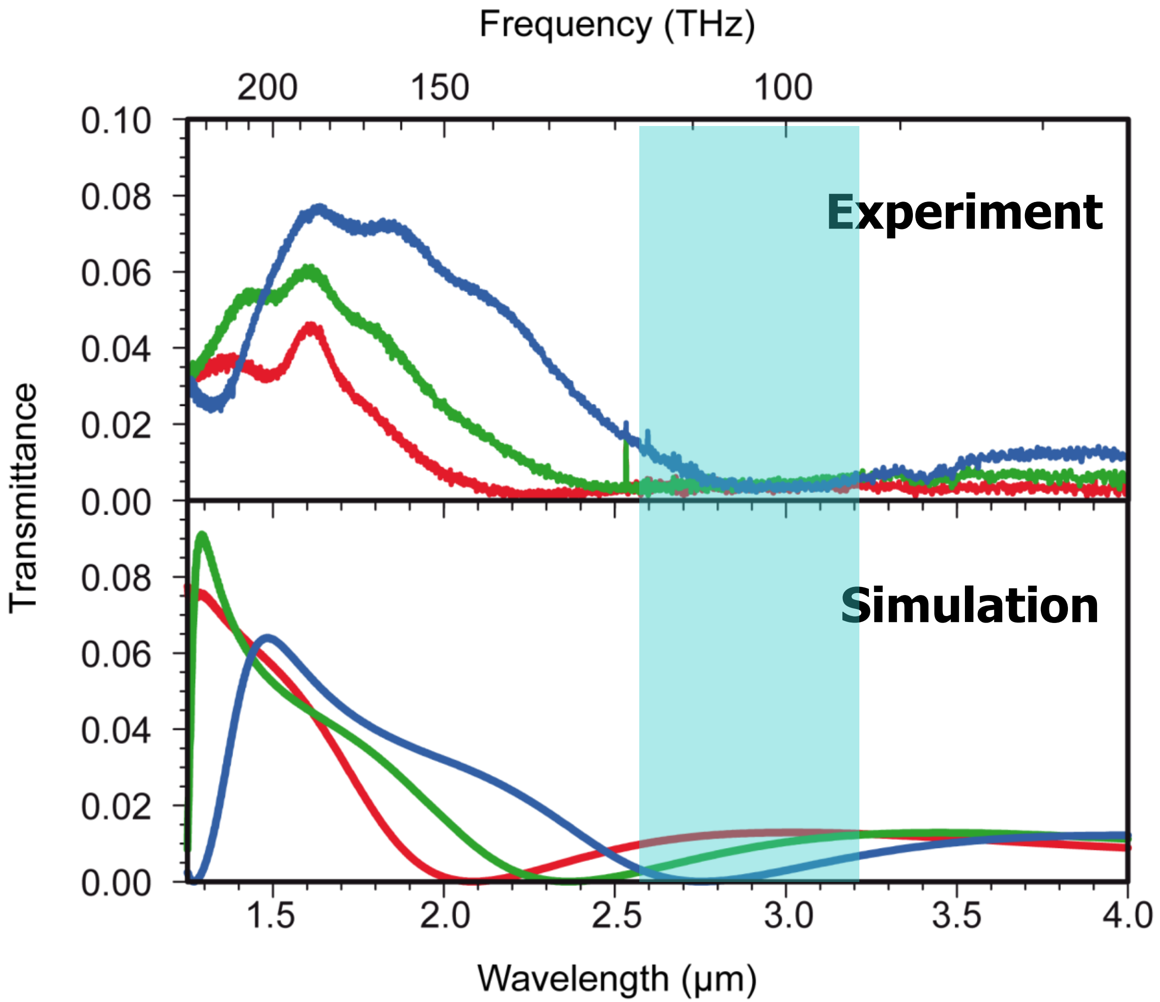


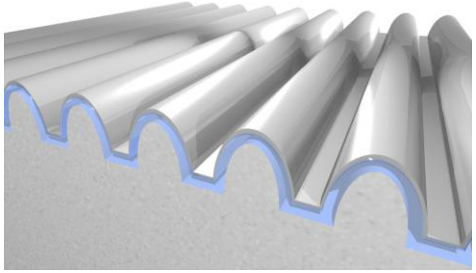
C. Kriegler *et al.*,
in preparation



(a)
10^{12} A/m^2
4.4
3.9
3.3
2.8
2.2
1.7
1.1
0.6
0.0
H
E
S
(b)
10^{12} A/m^2
2.8
2.4
2.1
1.7
1.4
1.0
0.7
0.3
0.0
H
E
S
SU-8
SiO_2
Ag

Recent results
Corrugated surface structure



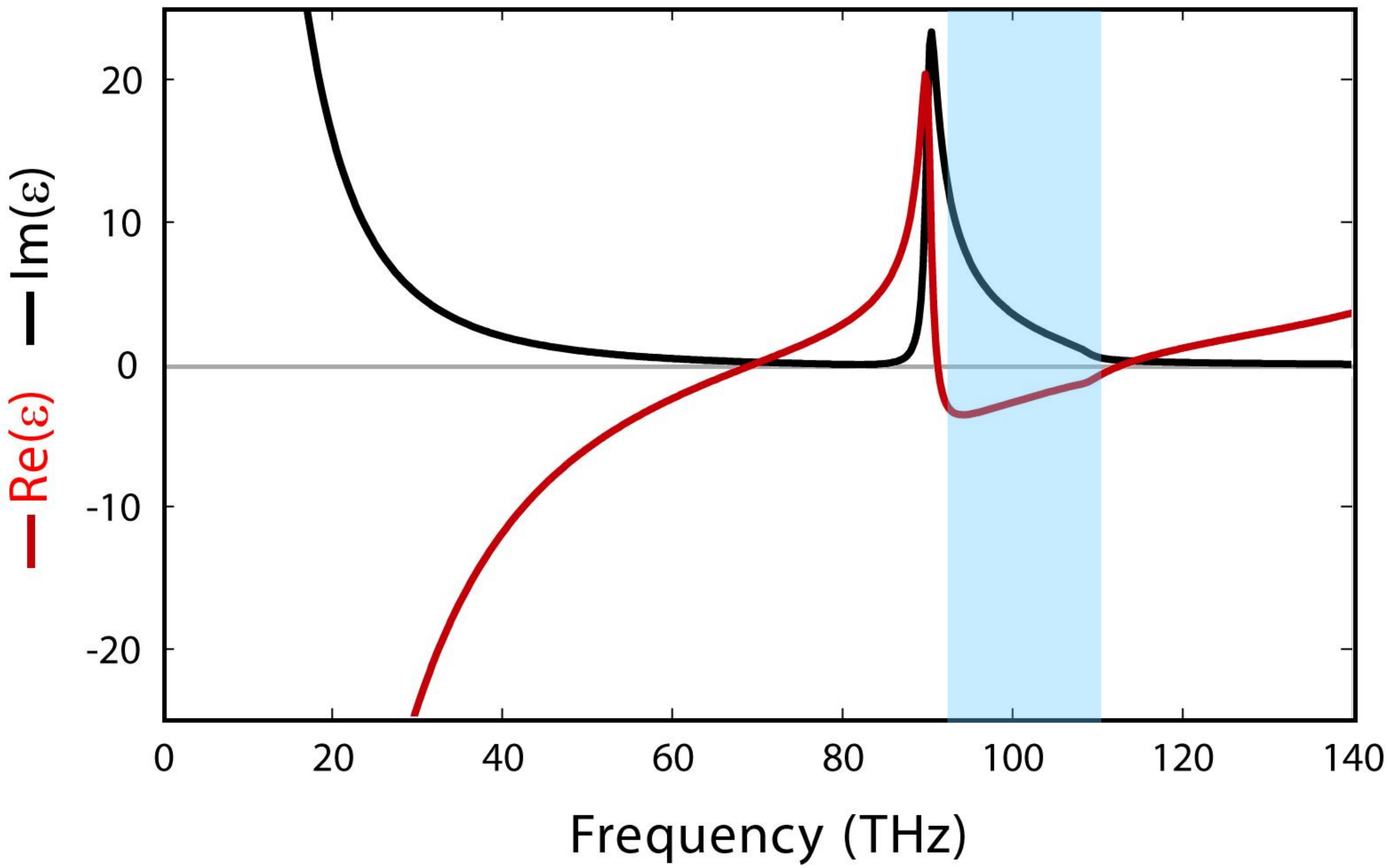


- d = 740nm
- d = 640nm
- d = 580nm

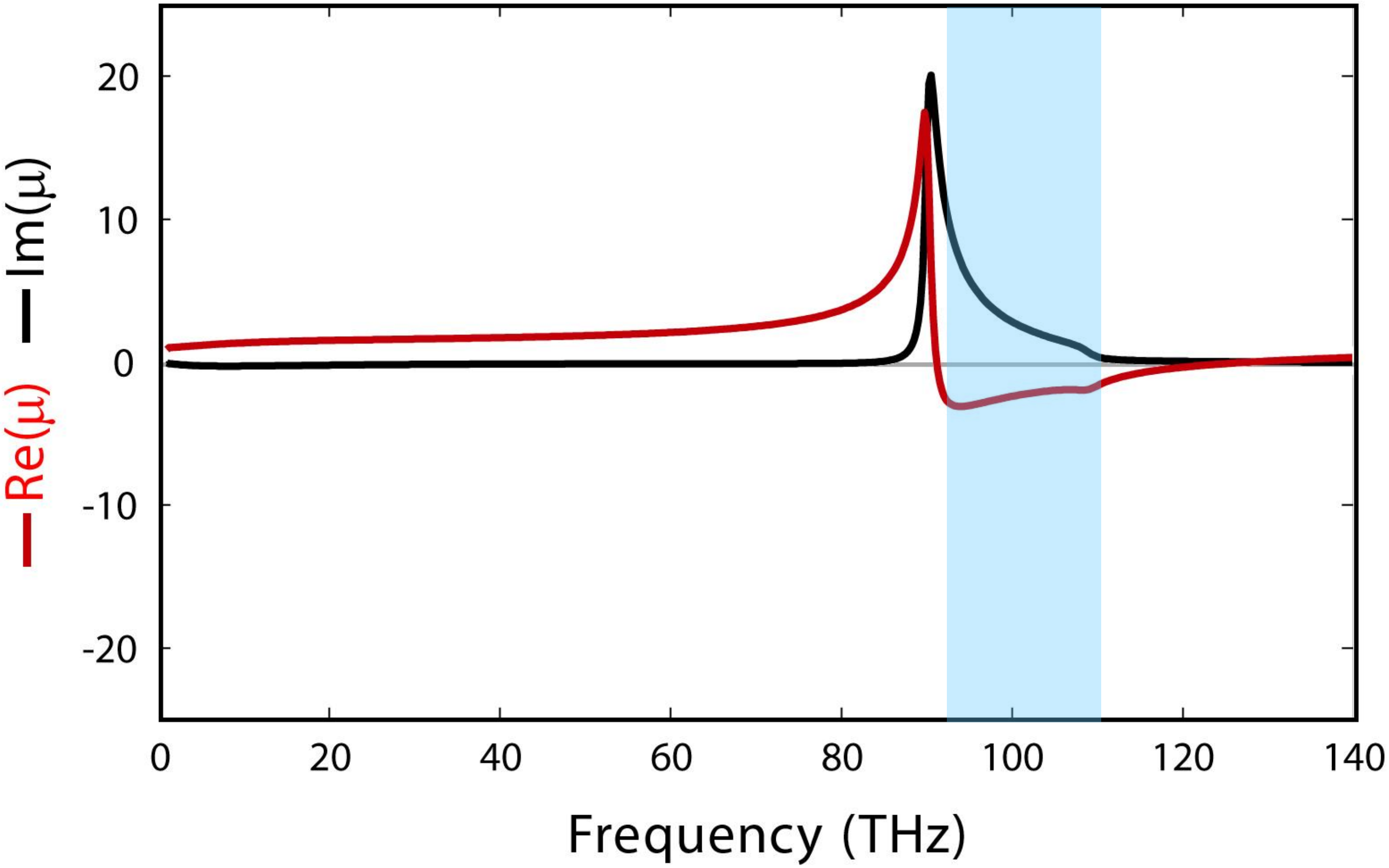
***LC* – eigenfreq.:**

$$\nu_0 \sim \frac{1}{\sqrt{LC}}$$

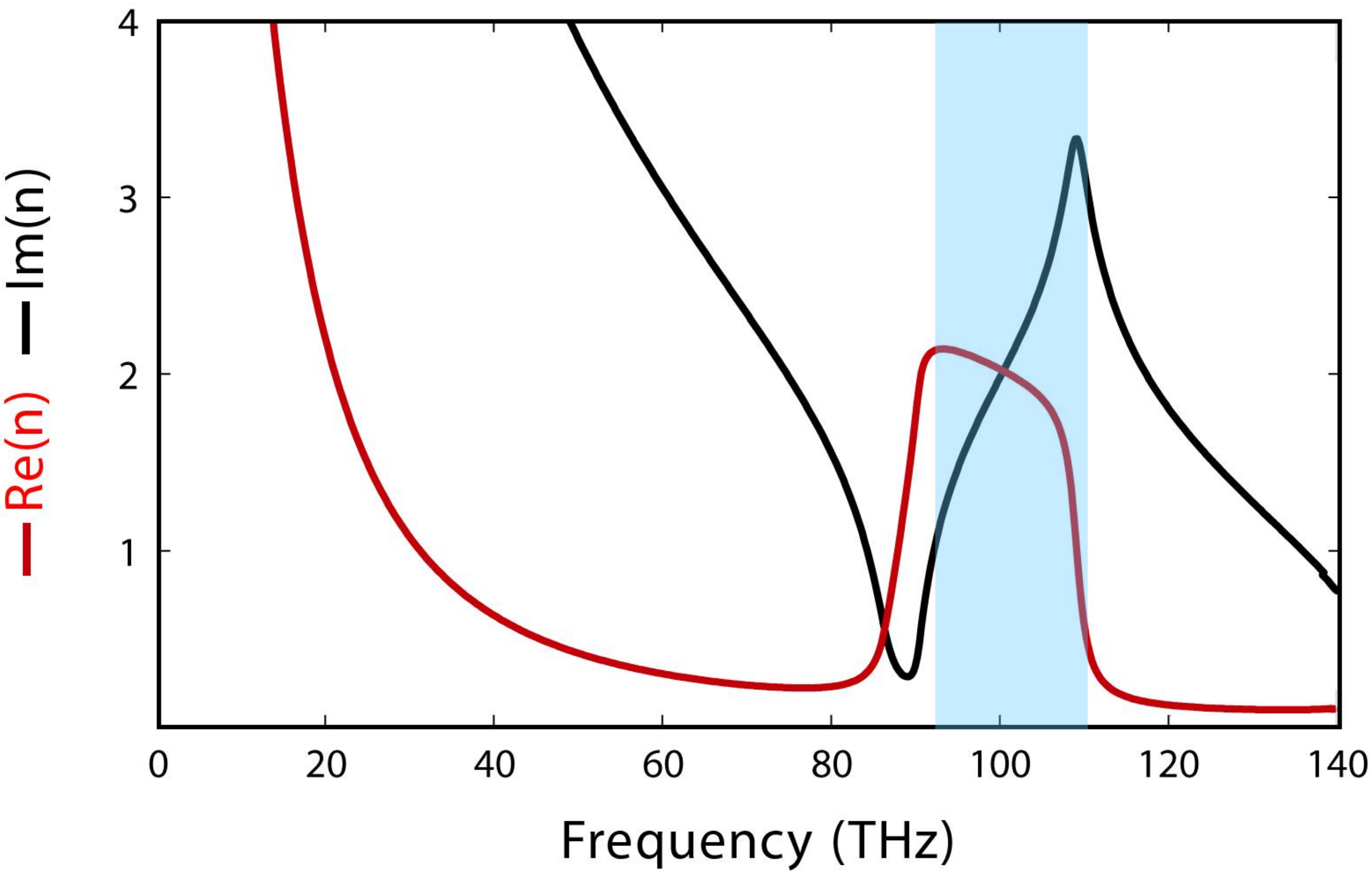
Recent results
Corrugated surface structure



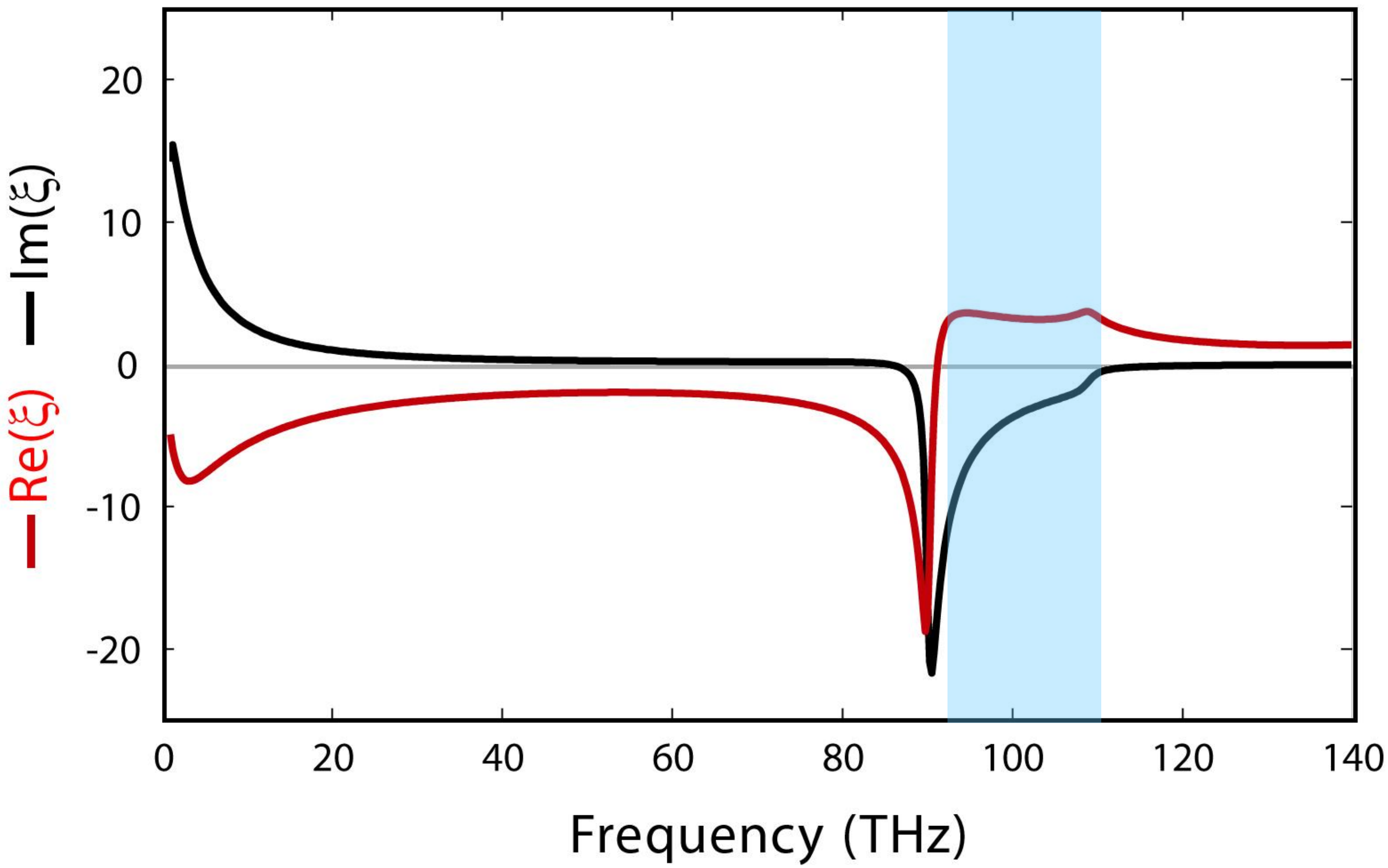
Recent results
Corrugated surface structure



Recent results
Corrugated surface structure



Recent results
Corrugated surface structure

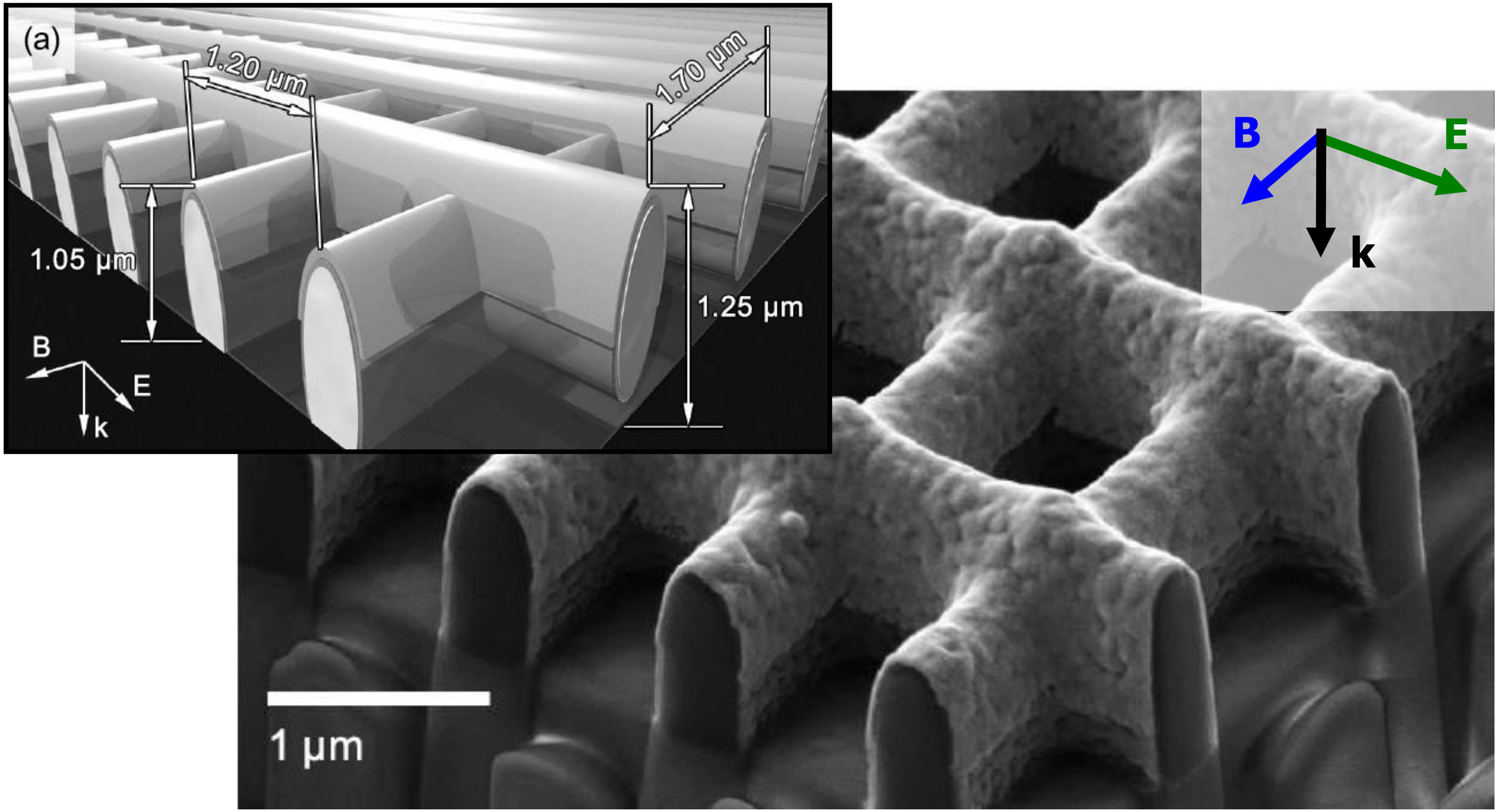


Recent results

Considering these results could bring us to the question:

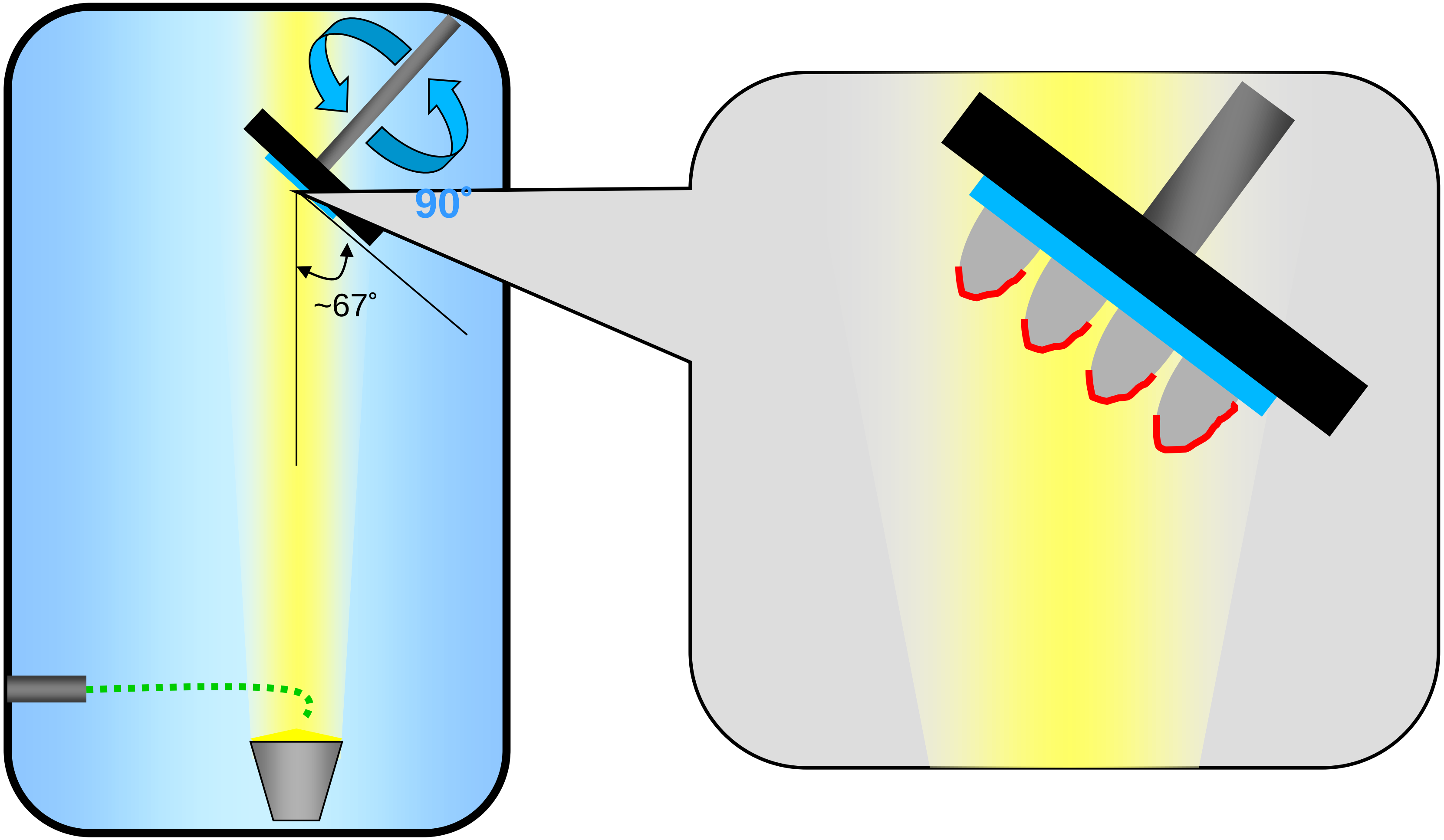
“Is bi-anisotropy a killer for negative refractive indices?”

Recent results
2-Layer negative-index metamaterial



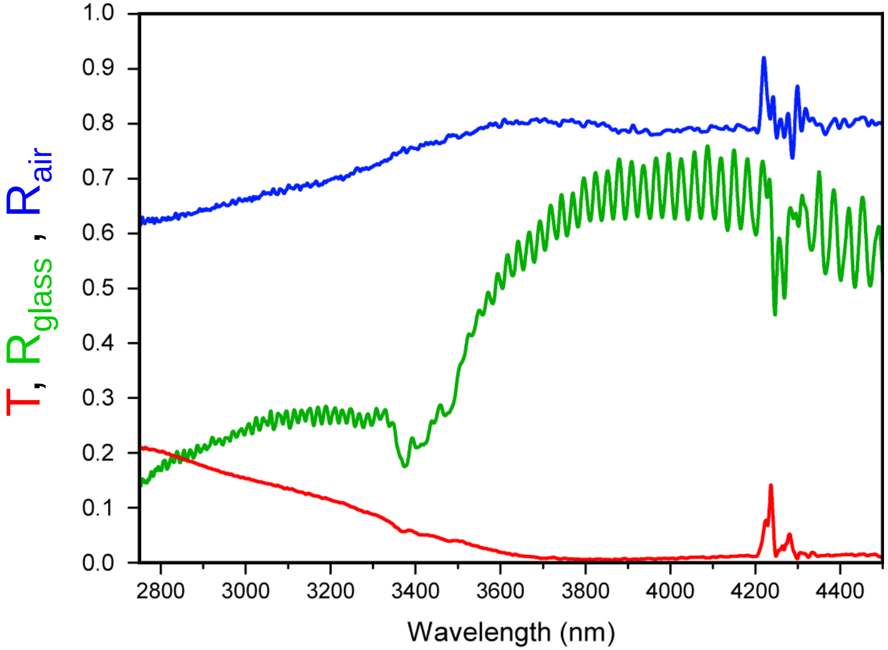
M.S. Rill *et al.*, arXiv:**0809**.2207v1

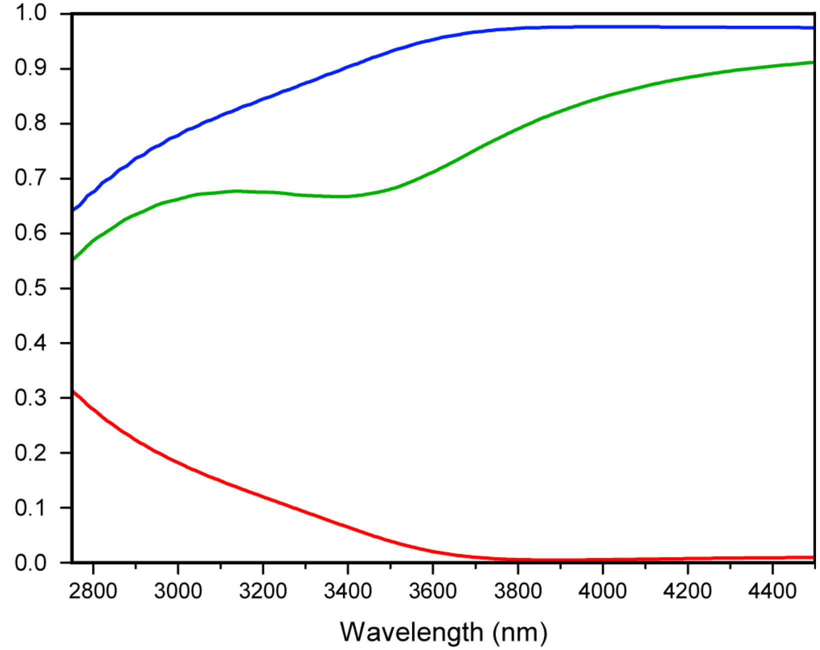
Recent results
2-Layer negative-index metamaterial



Recent results

2-Layer negative-index metamaterial

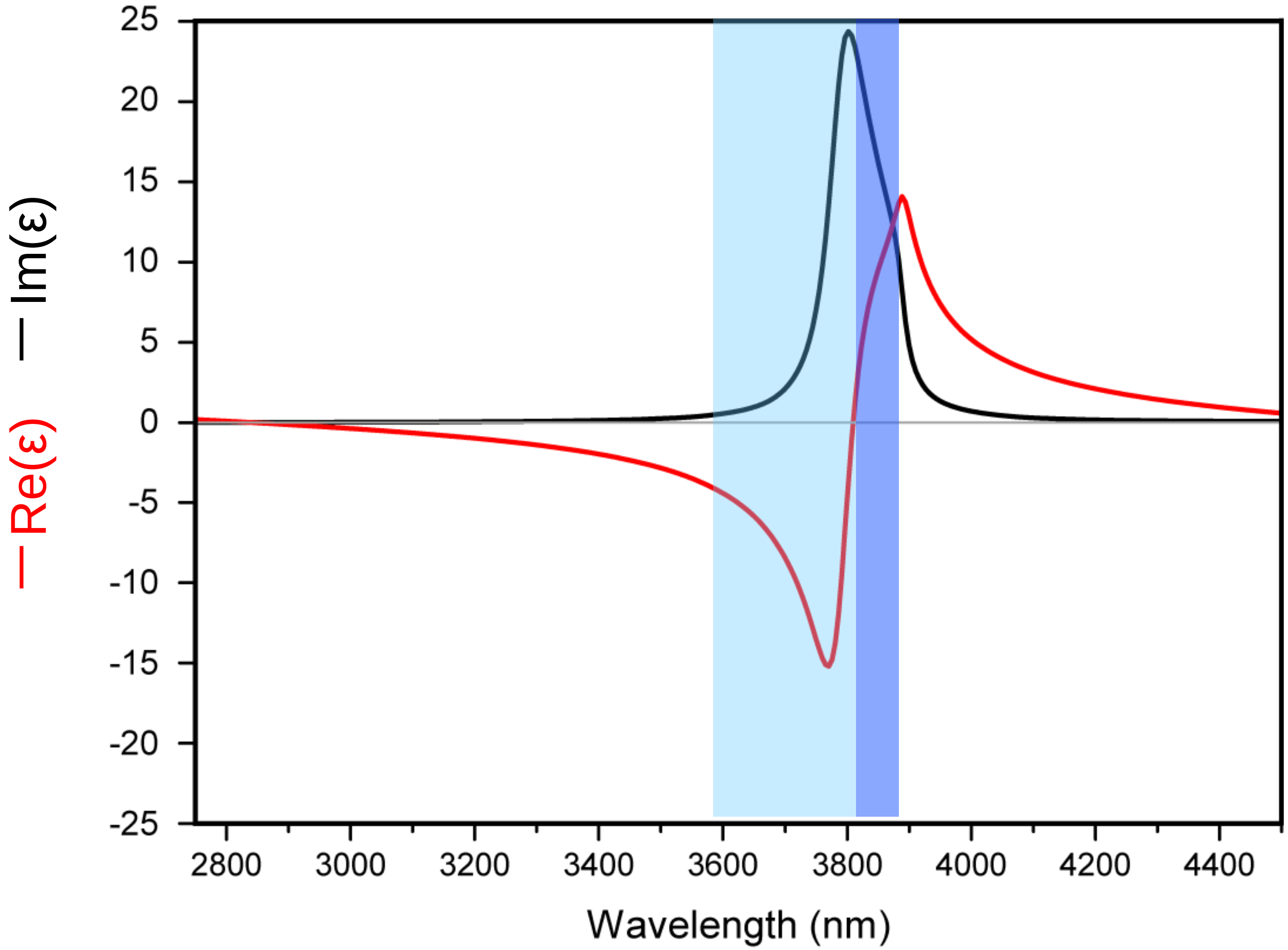




Experiment
(fringes are due to Fabry-Pérot resonances of the glass substrate)

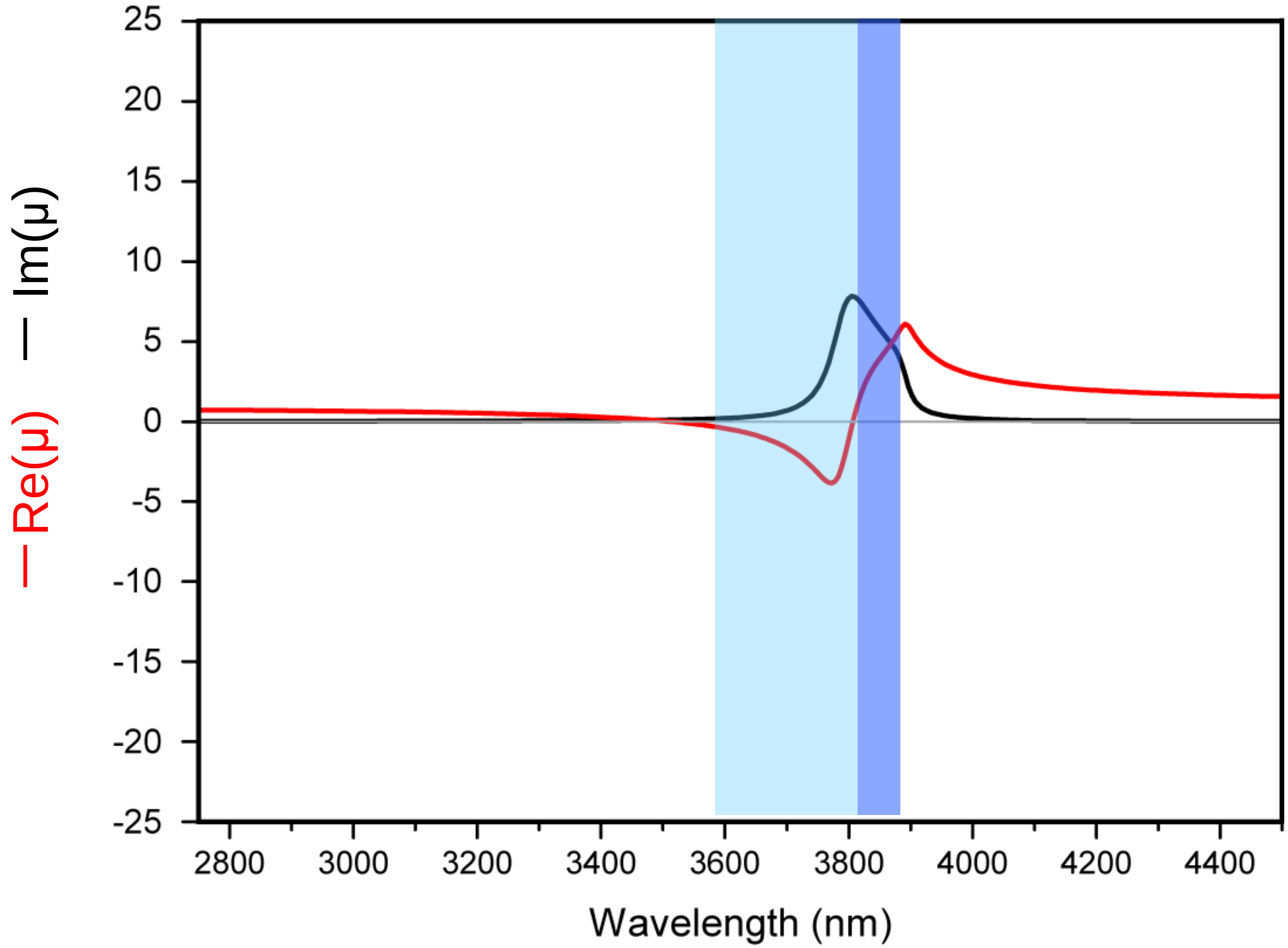
Theory
(calculations performed with CST Microwave Studio)

Recent results
2-Layer negative-index metamaterial

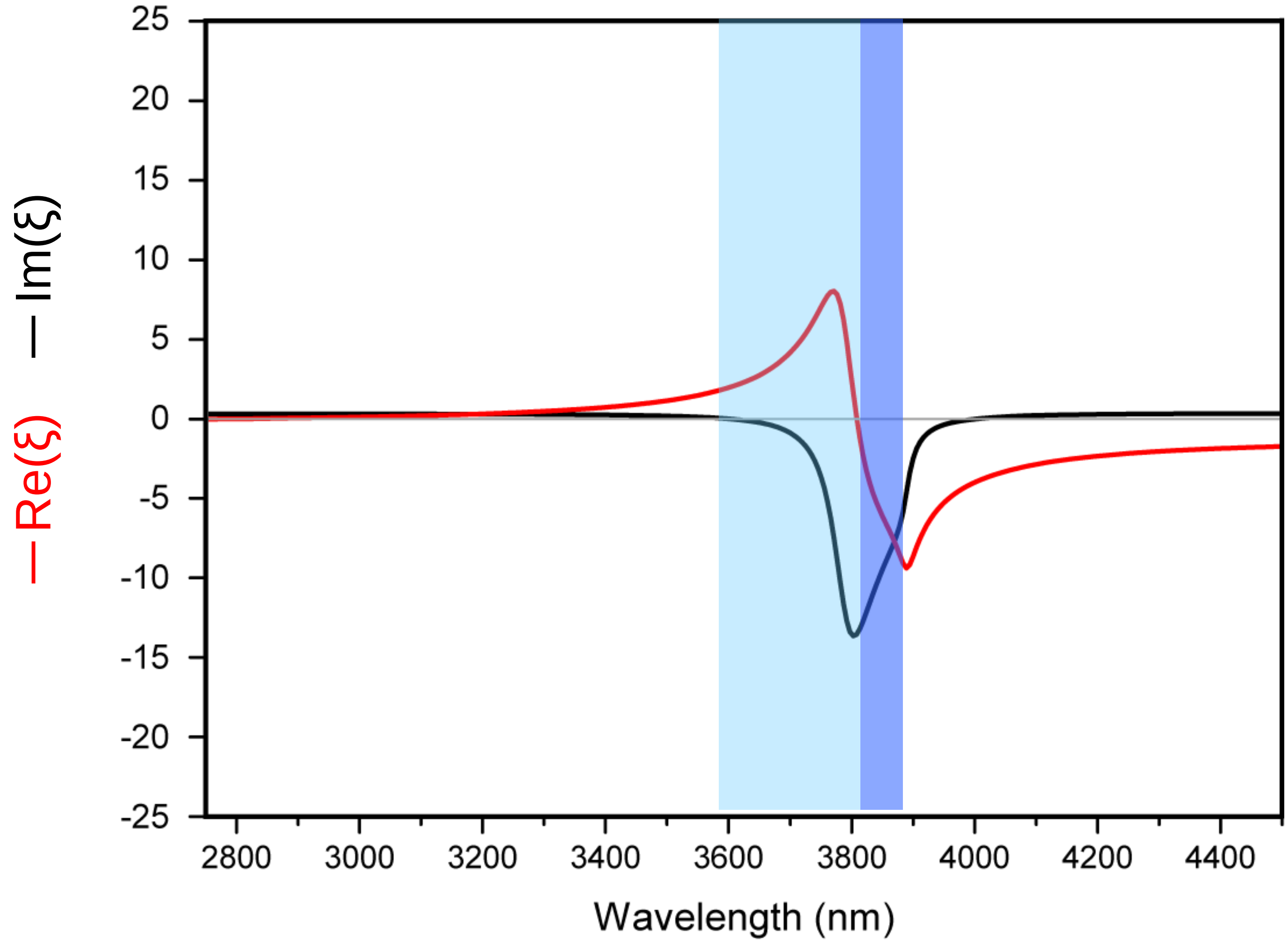


Recent results

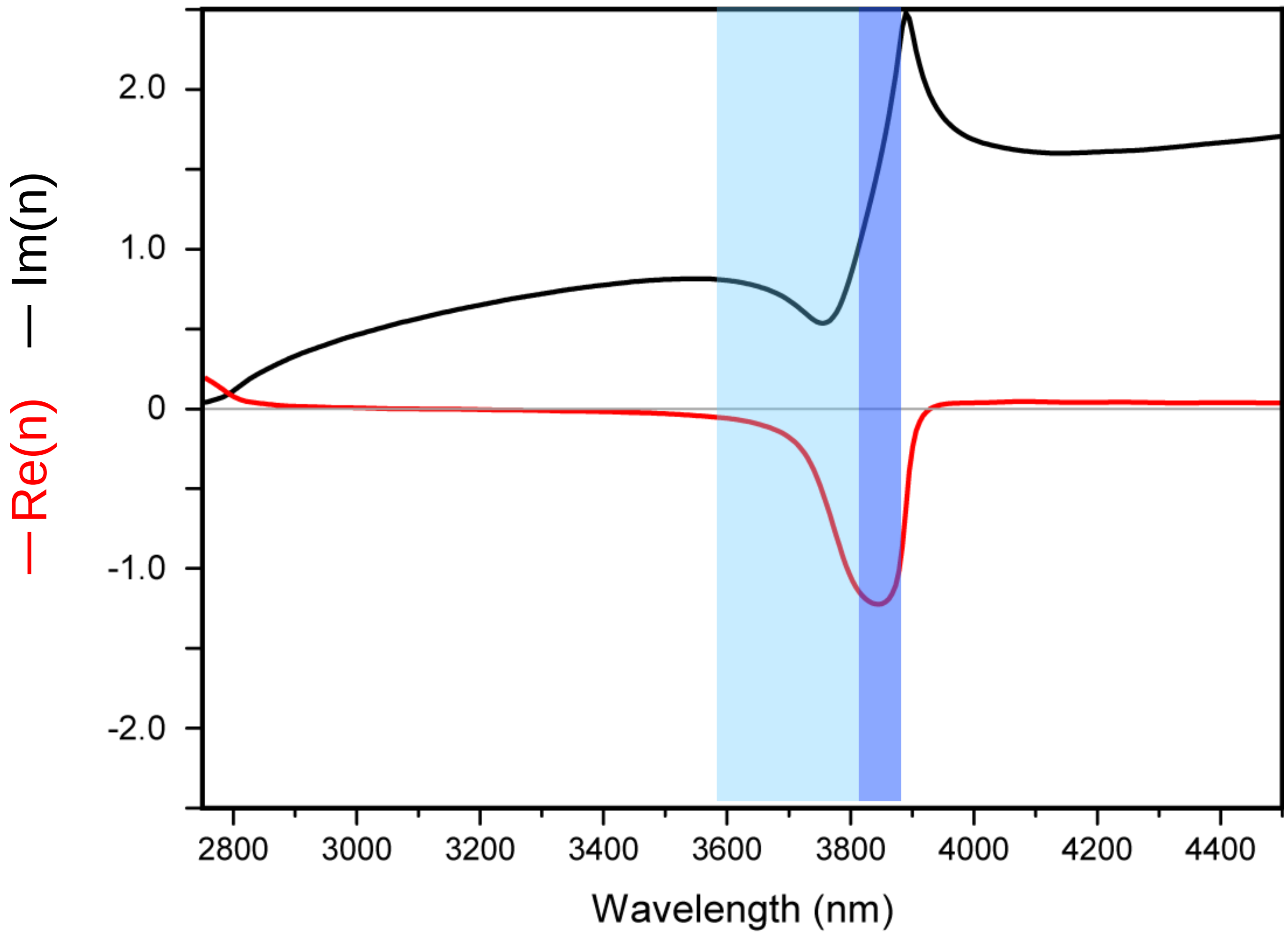
2-Layer negative-index metamaterial



Recent results
2-Layer negative-index metamaterial

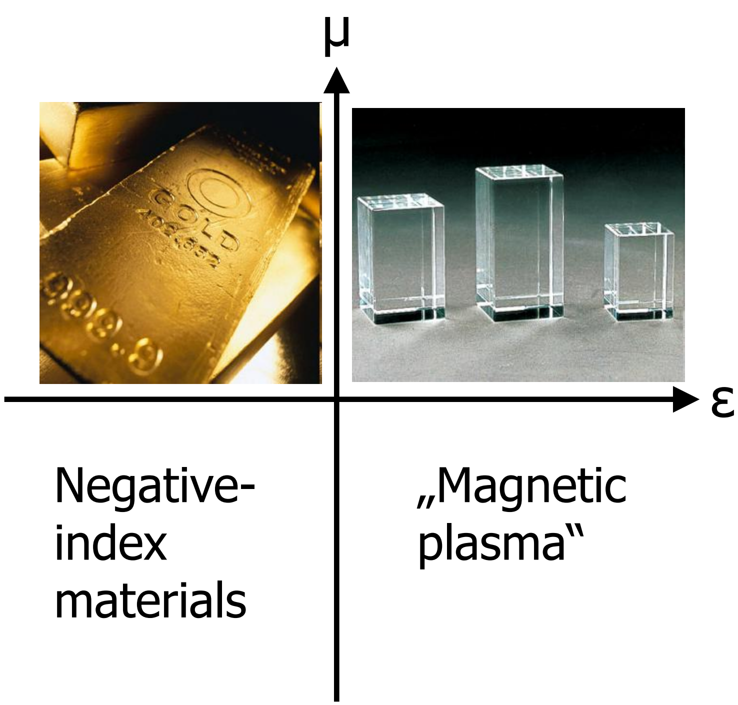


Recent results
2-Layer negative-index metamaterial

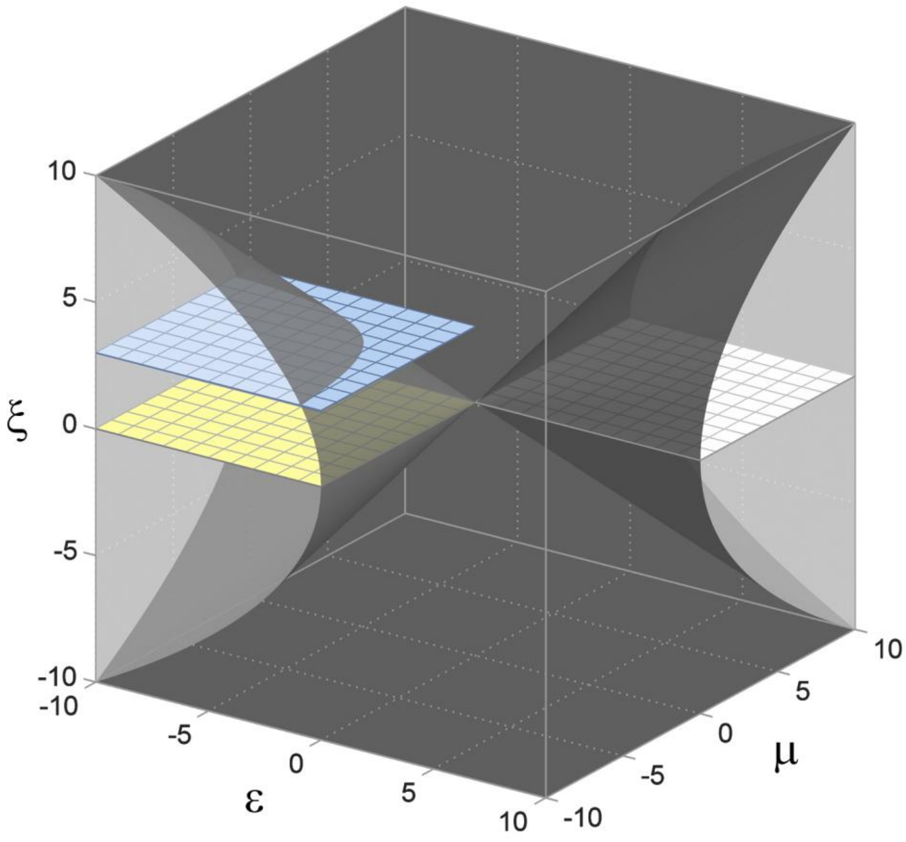


Recent results
2-Layer negative-index metamaterial

isotropic



bi-anisotropic

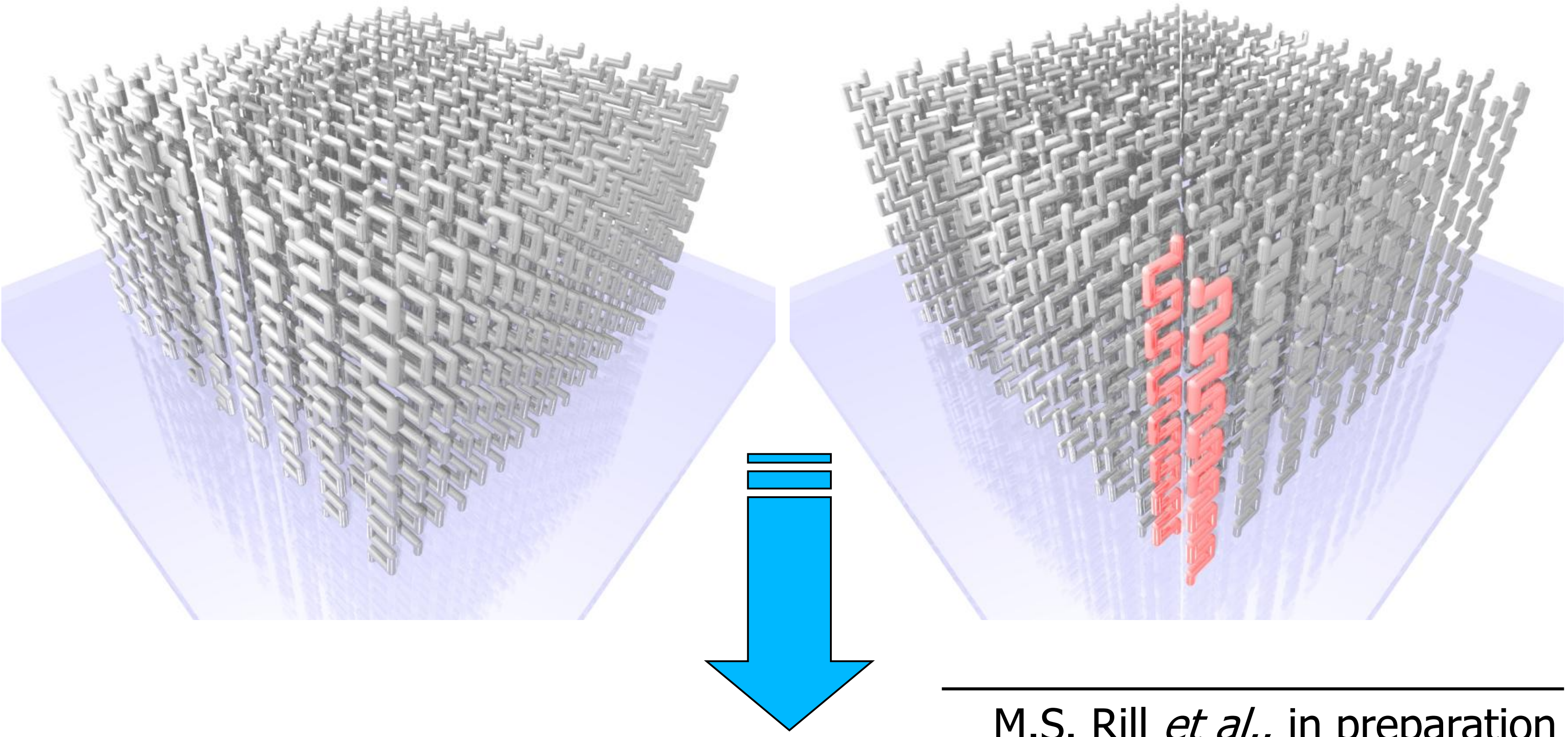


Outline

- Metamaterials as effective media
- Hitherto existing realizations of 3D metamaterials
- Fabricational approaches for the NIR
- Our fabricational approach
- Recent results
- **Current projects**
- Conclusions and outlook

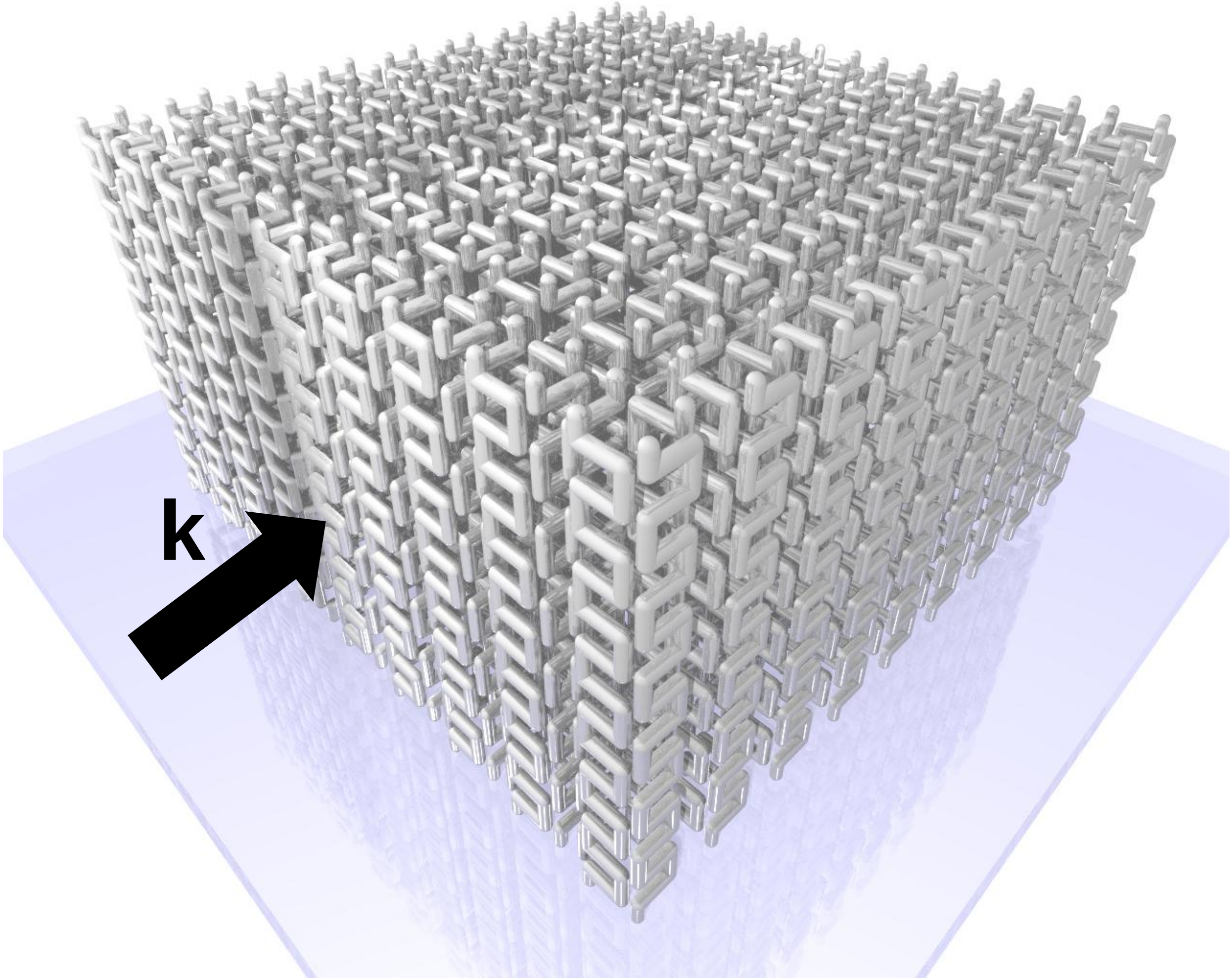
Current projects
2D isotropic metamaterial

Based on idea of **D. Güney** (Soukoulis group):
D.Ö. Güney *et al.*, arXiv:**0807**.4560

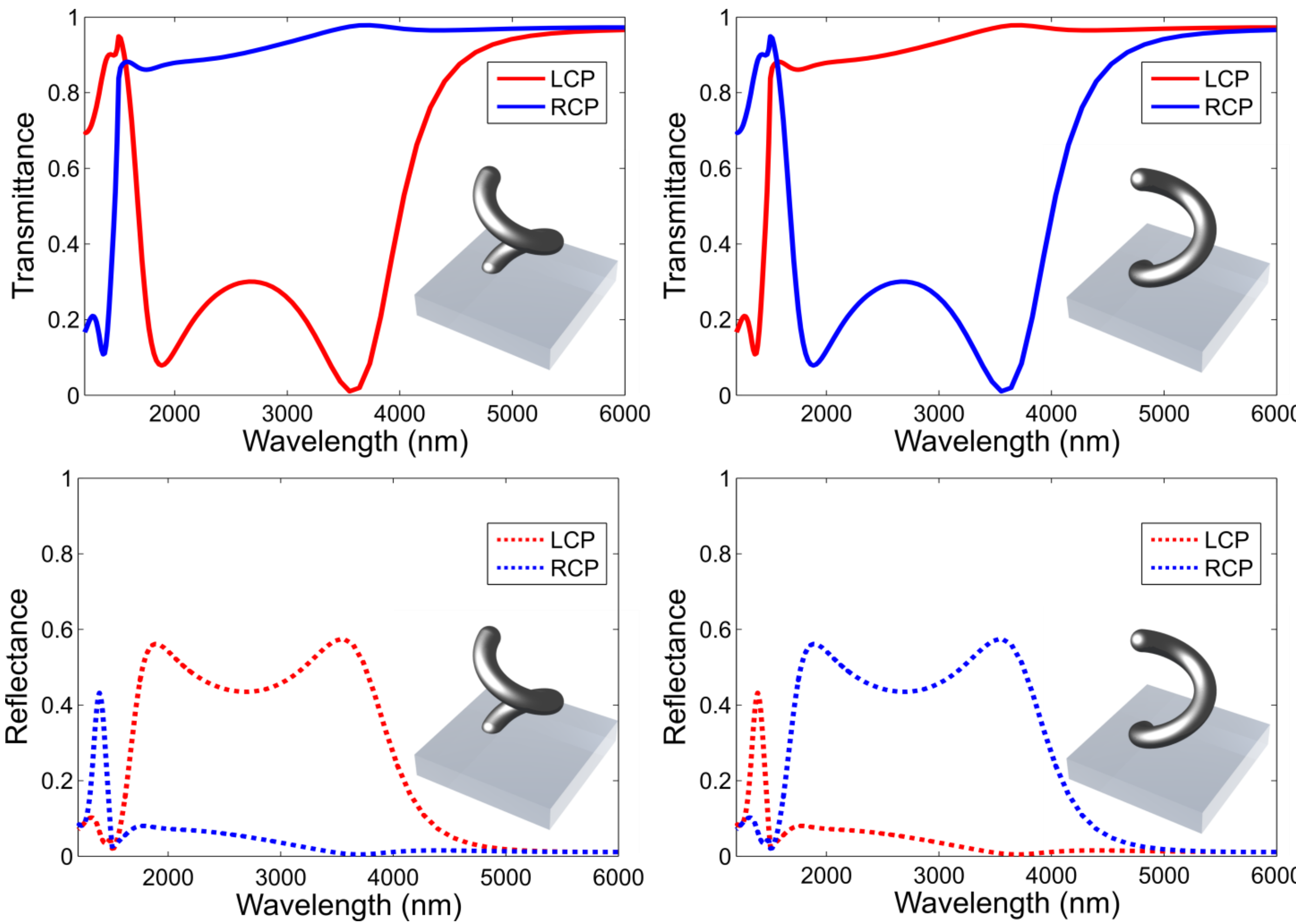


M.S. Rill *et al.*, in preparation

Current projects
2D isotropic metamaterial

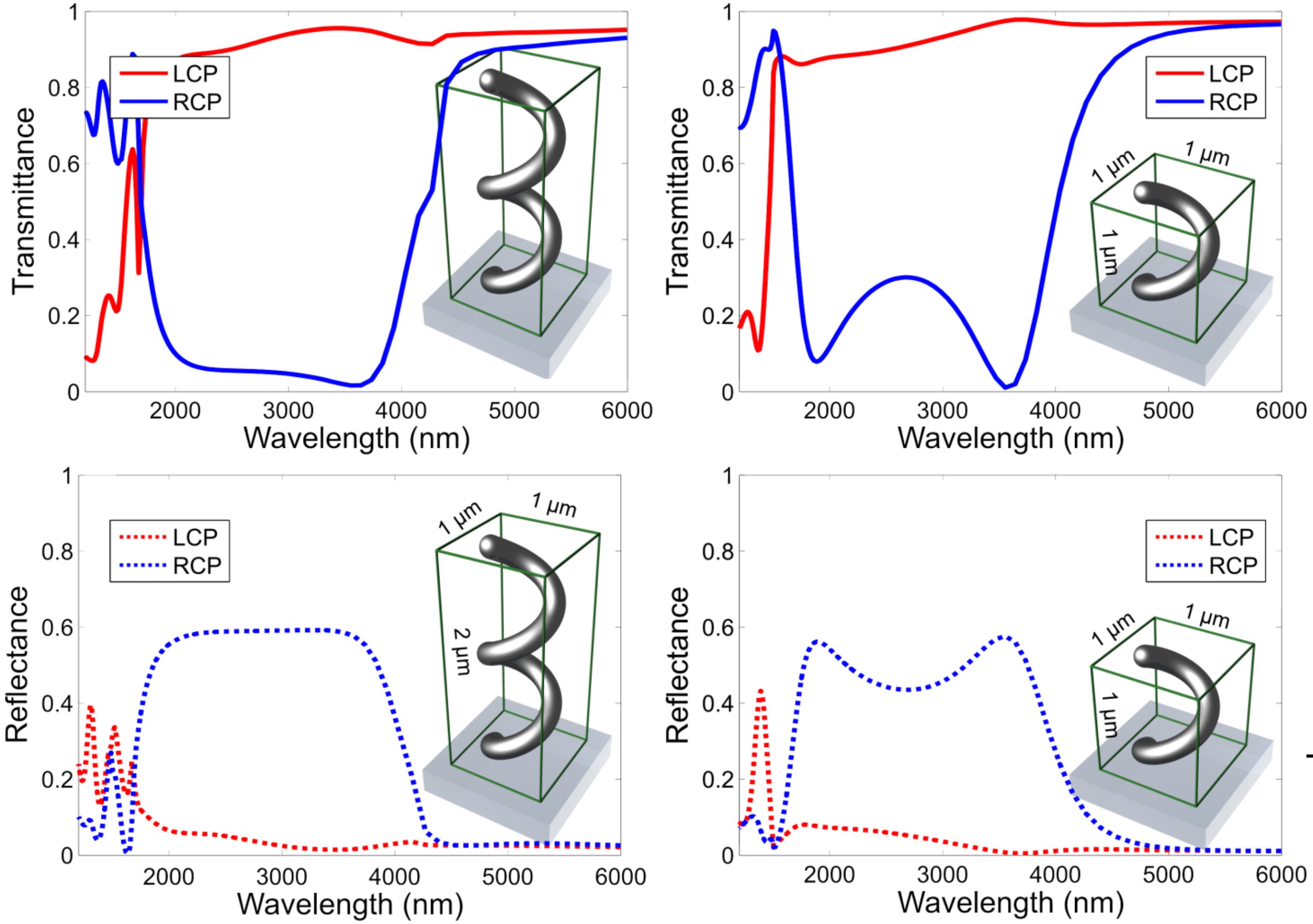


Current projects
Chiral 3D metamaterials



M. Thiel *et al.*,
in preparation

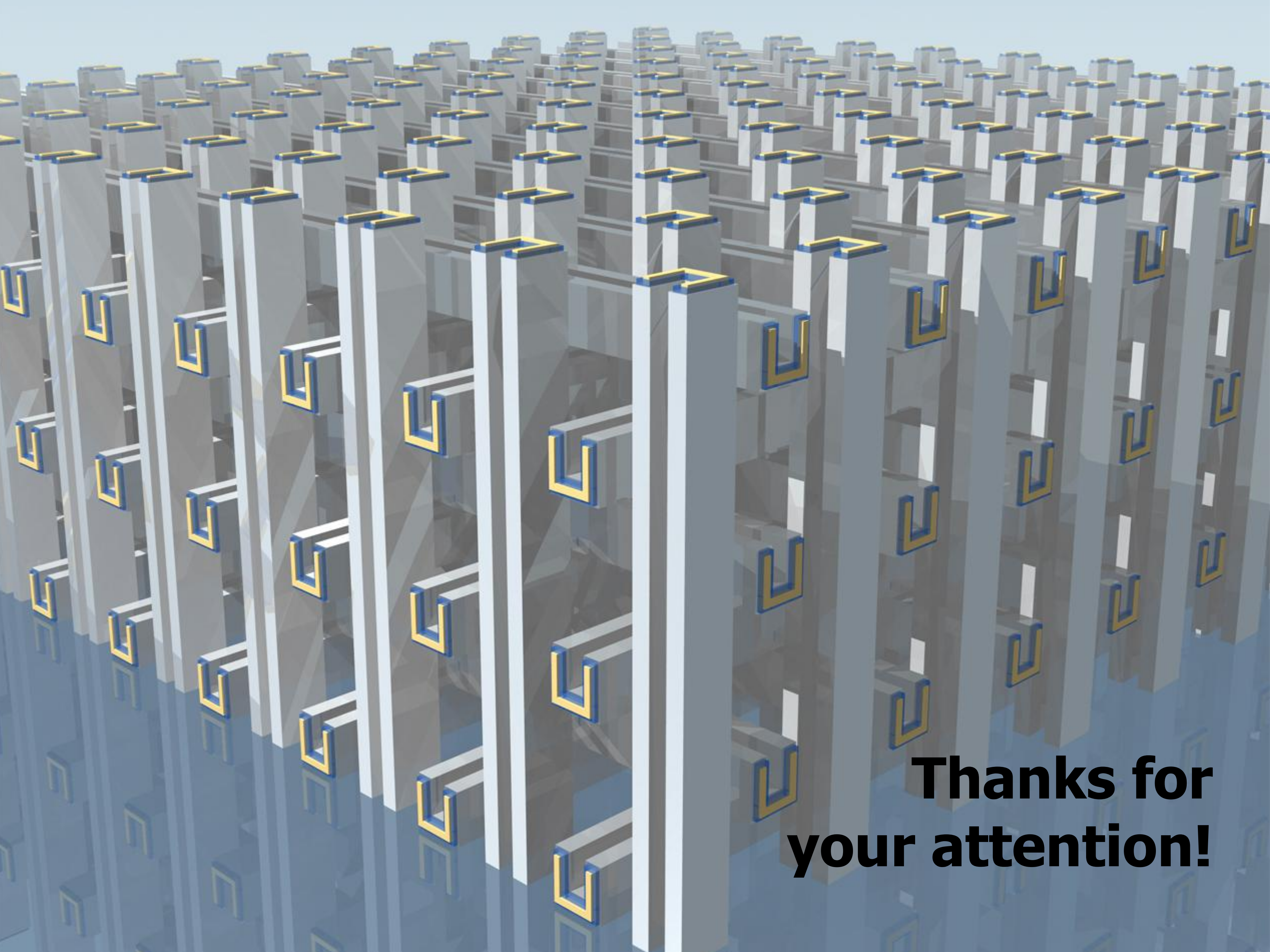
Current projects
Chiral 3D metamaterials

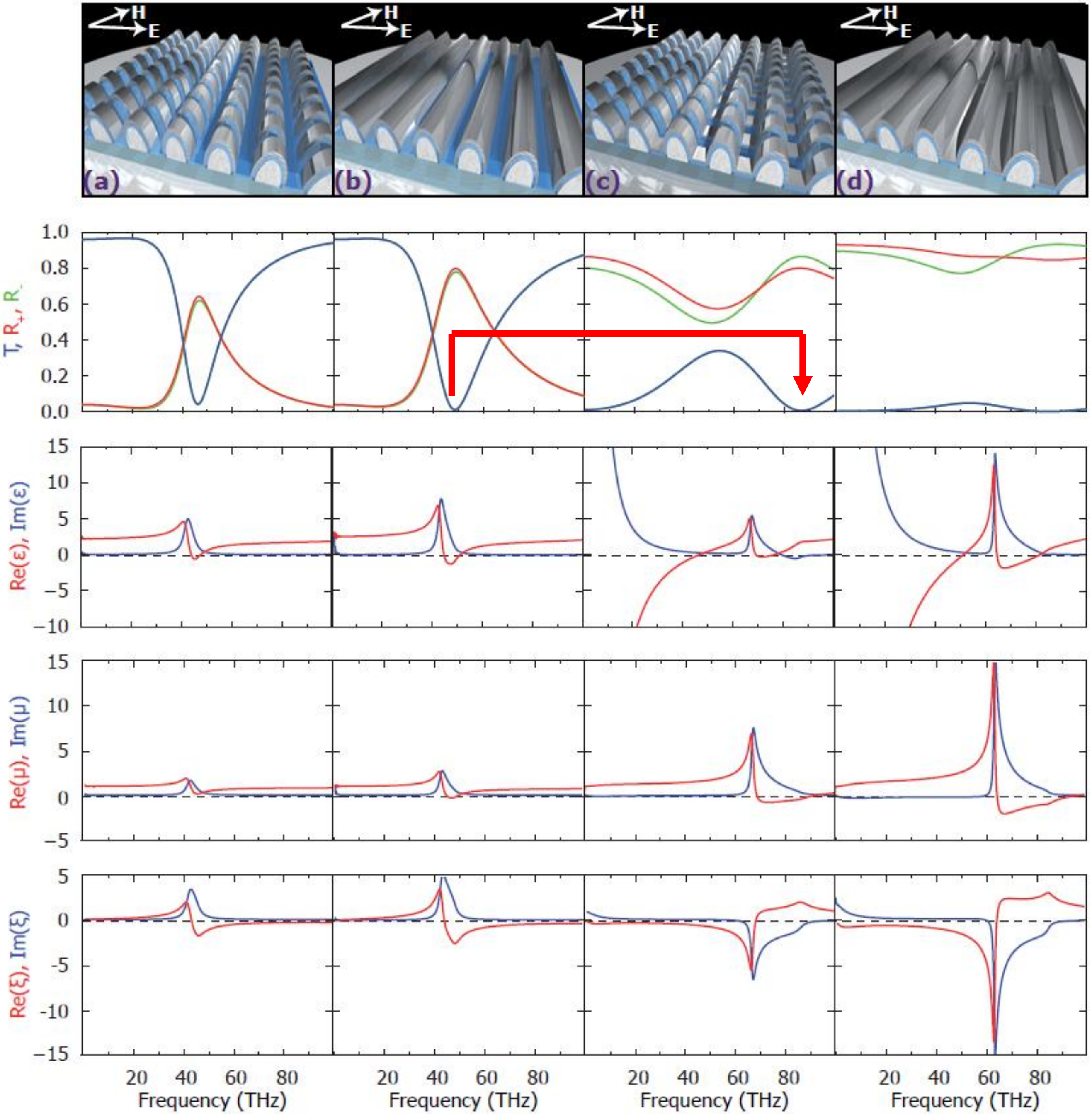


Simulations by
S. Linden

Conclusions

- Theory to describe even complicated structures (→ bi-anisotropy)
  BUT: Structure has to be an **effective material**!
- Technique which is suitable to fabricate 3D metamaterials for NIR optical range
  (→ We have not fabricated it, yet!)
- Found promising design which can be fabricated by our proposed method


Thanks for
your attention!



C. Kriegler *et al.*,
in preparation