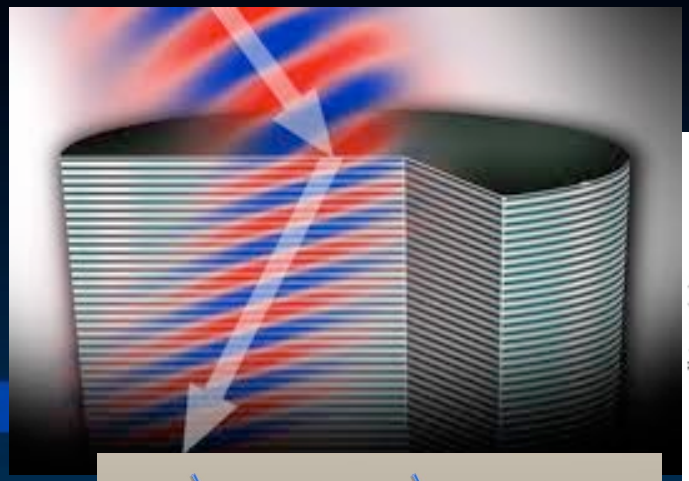
赤外吸収メタマテリアル

理化学研究所　田中メタマテリアル研究室
理化学研究所　光量子工学研究領域
東京工業大学

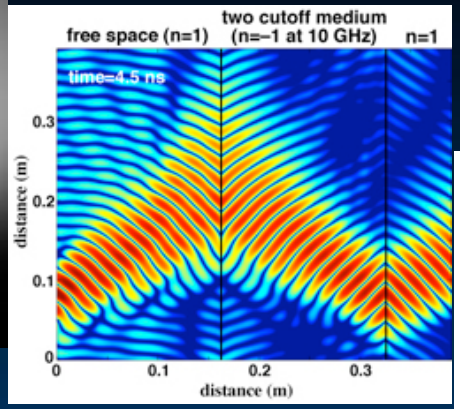
田中　拓男

第６回可視赤外線観測装置技術WS　November/24/’16　11:30-12:00

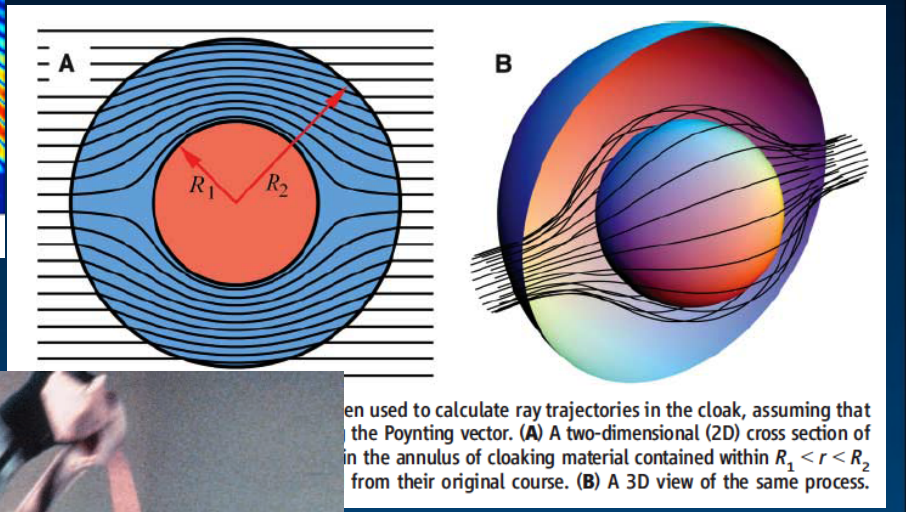
Metamaterials



negative index/negative refraction

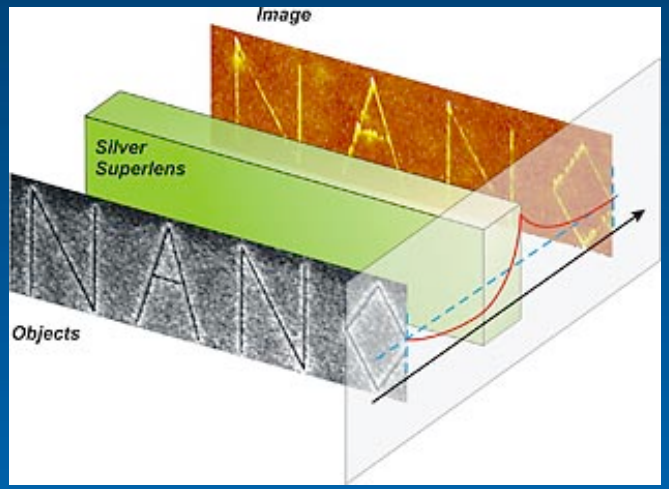


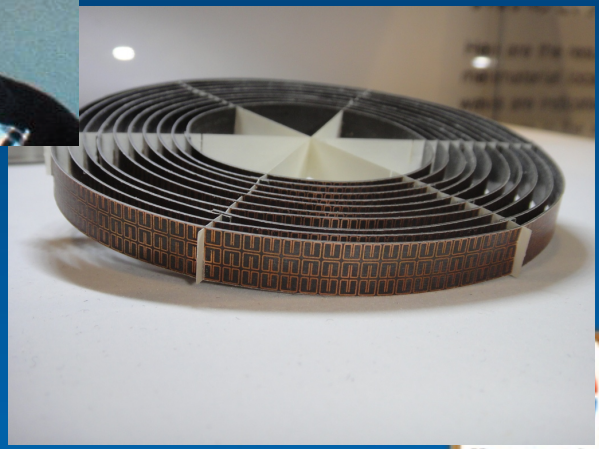
cloaking



en used to calculate ray trajectories in the cloak, assuming that the Poynting vector. **(A)** A two-dimensional (2D) cross section of in the annulus of cloaking material contained within $R_1 < r < R_2$ from their original course. **(B)** A 3D view of the same process.

perfect lens/ super lens

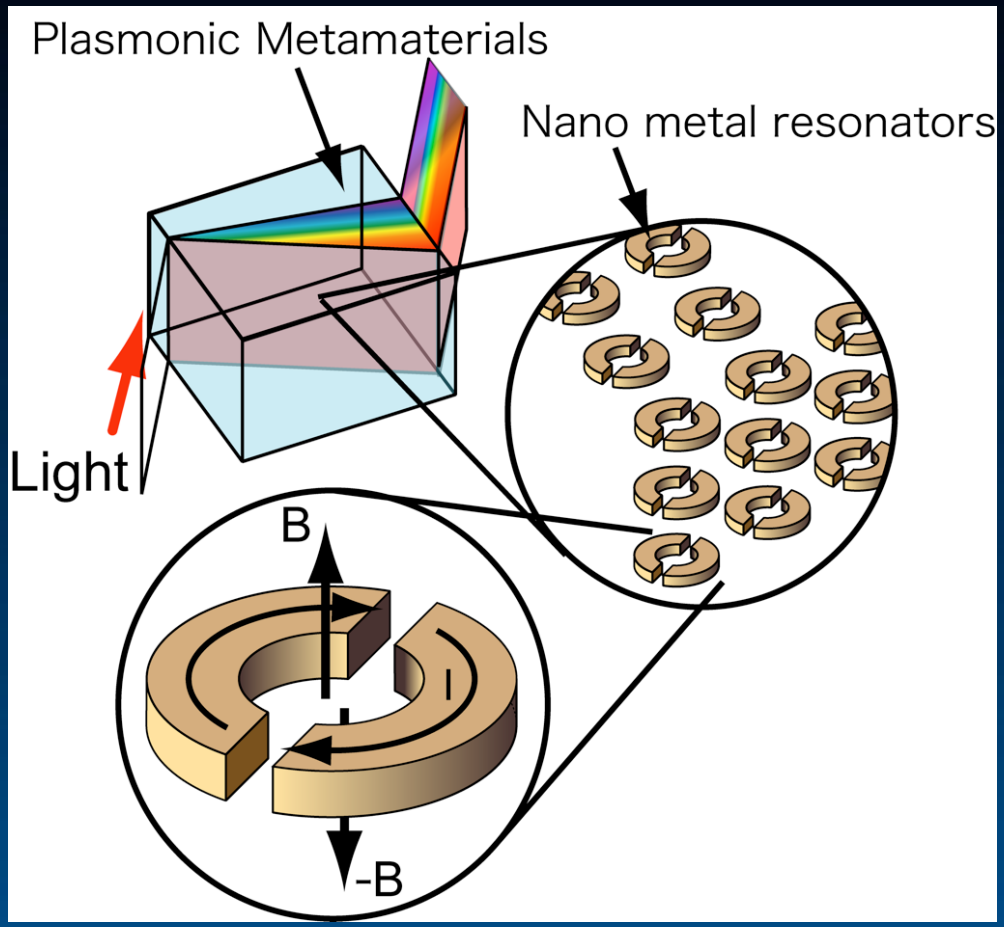




RIKEN

Metamaterials

メタマテリアル



波長より細かな（サブ波長）人工構造を用いて，
物質の光学特性(n，ε，μ)を操作した人工物質

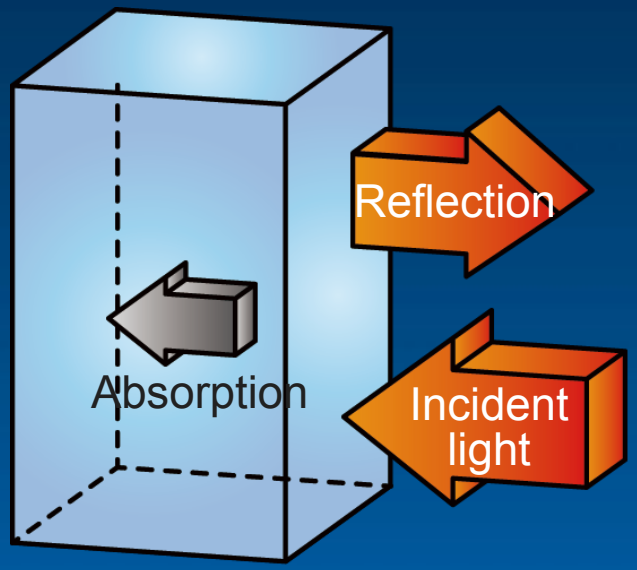
Discrepancy between extinction coefficient and absorption

Index of refraction $N = n - ik$

extinction coefficient

Fresnel formula (from air to matter.)

$$R = \left| \frac{N-1}{N+1} \right|^2$$



Highly absorptive material

↓

Large extinction coef.

↓ contradiction

Highly reflection

↓

Low absorption

e.g. : Au

N = 1.85 - 21.7i at λ = 3 μm

~ 98% reflection

Only 2% absorption

Gold

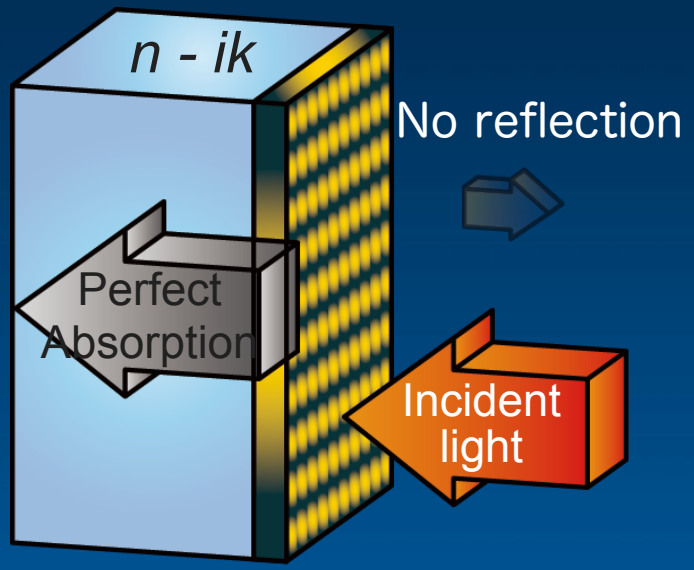
Sci. Rep. **5**, 12570 (2015).

Metamaterial absorber

- Nano scale structures on flat metal film (no transmission)
- Impedance matching for no reflection

➡ Perfect absorption by ultra-thin layer

~~反射~~＋~~透過~~＋吸収＋（~~散乱~~）=1.0



Outline

光を吸収する技術と，その応用

1．Non-radiative surface plasmon (Dark plasmon)

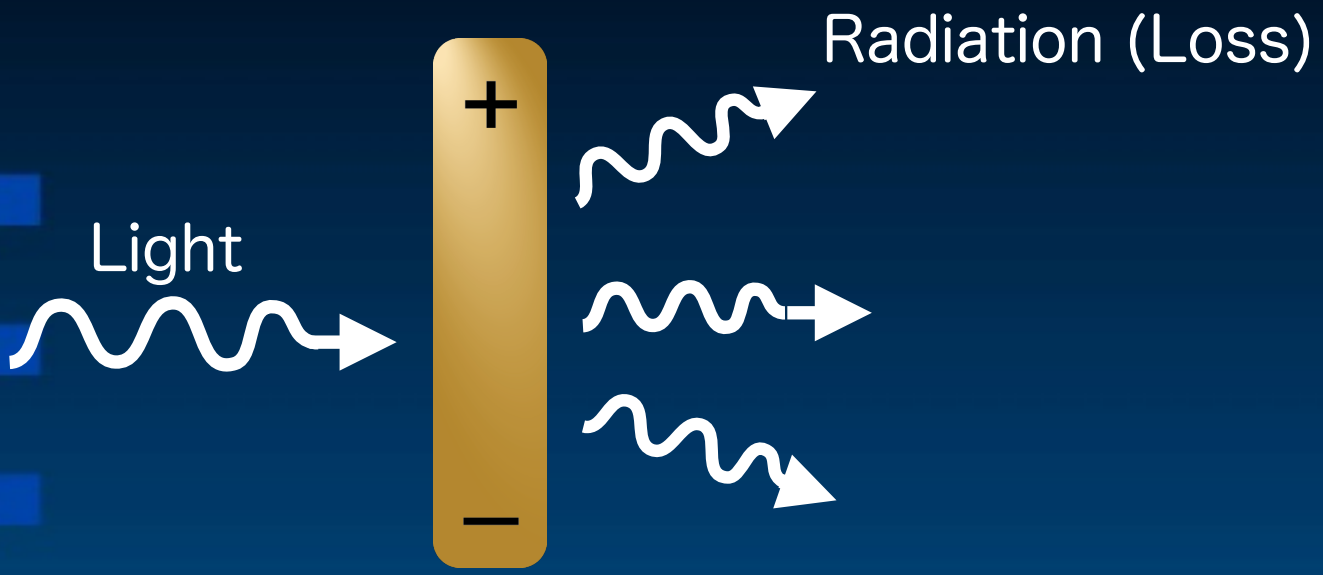
2．Dark plasmonic metamaterials for absorber

3．Application of dark metamaterials for molecular sensing

4．(Fabrication technique for 3D Metamterials)

Dark plasmons

1st mode
(dipole mode)

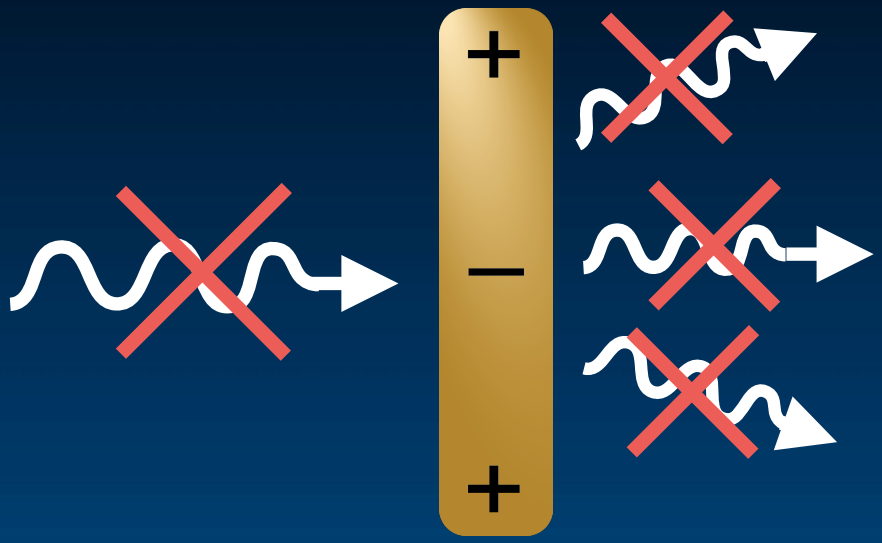


Excitation ○

Q-factor low

Dark plasmons

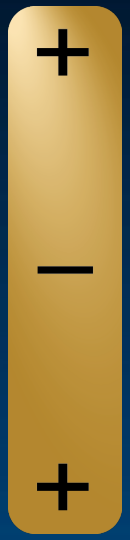
2nd mode



Excitation X

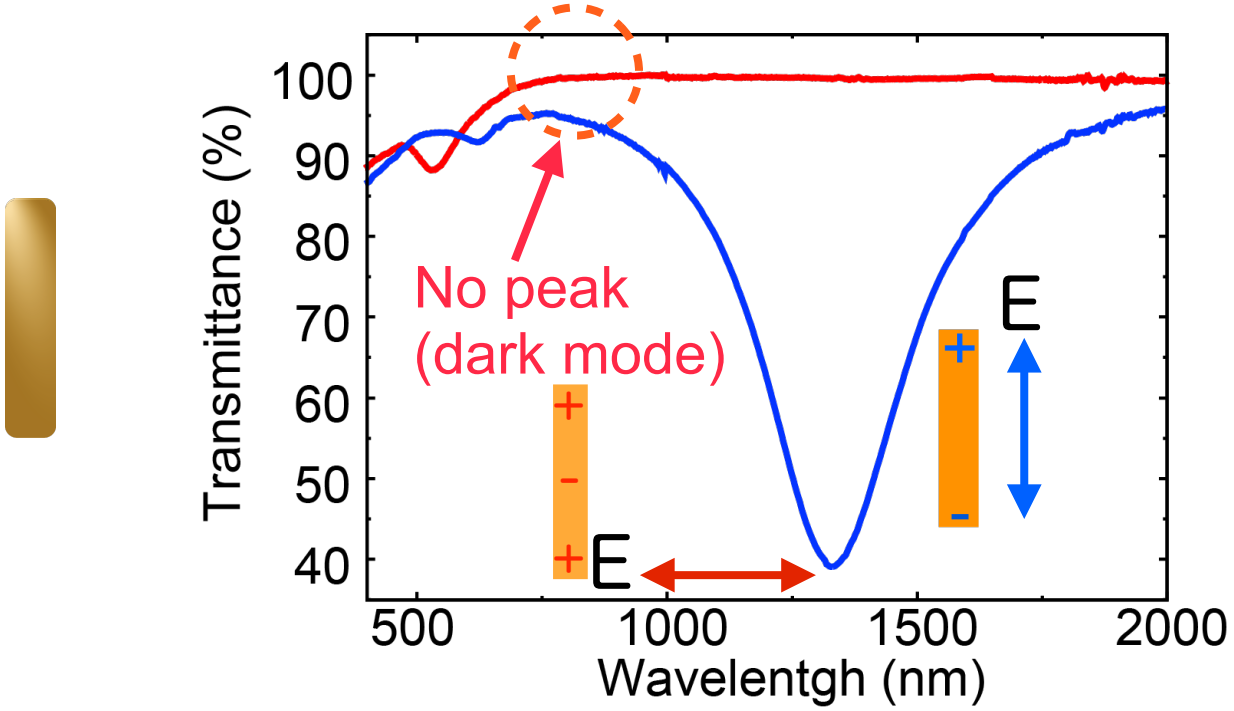
Q-factor high

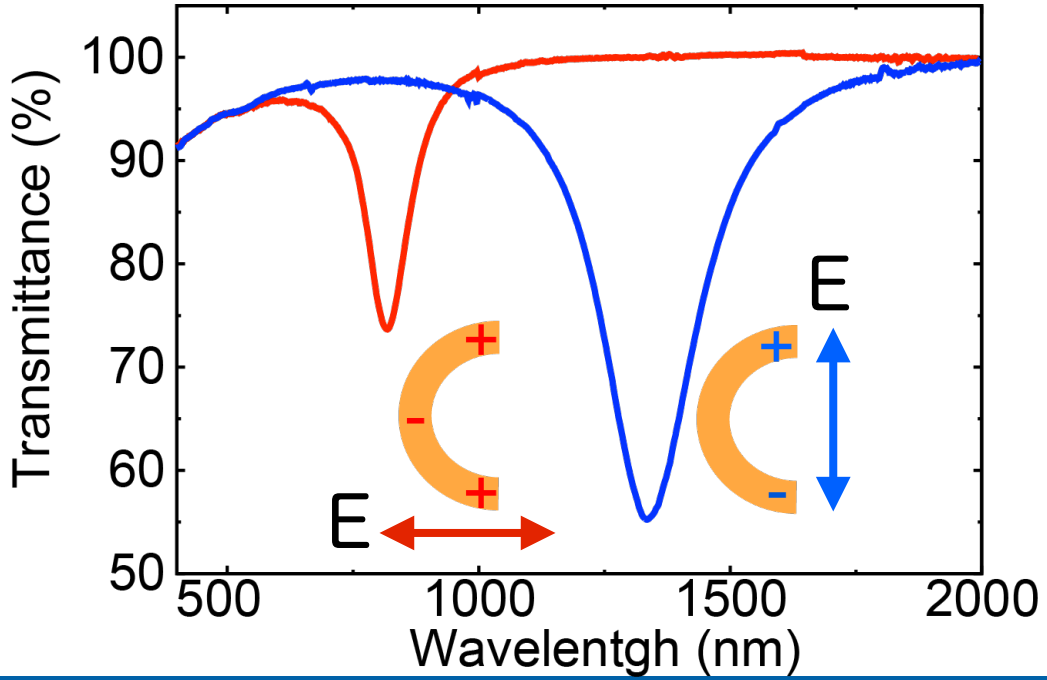
Dark plasmons

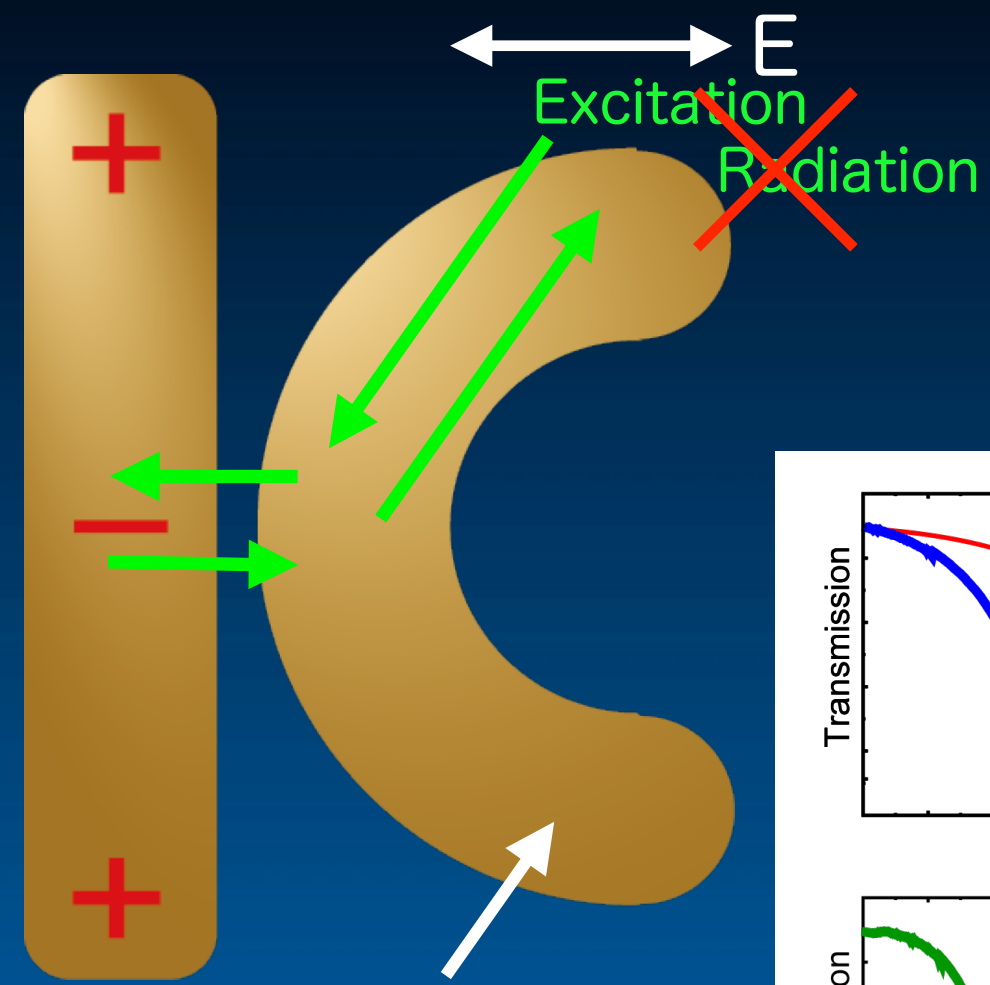
	Bright plasmons	Dark plasmons
		
Excitation	○	X (No radiation loss)
Q-factor	low	high

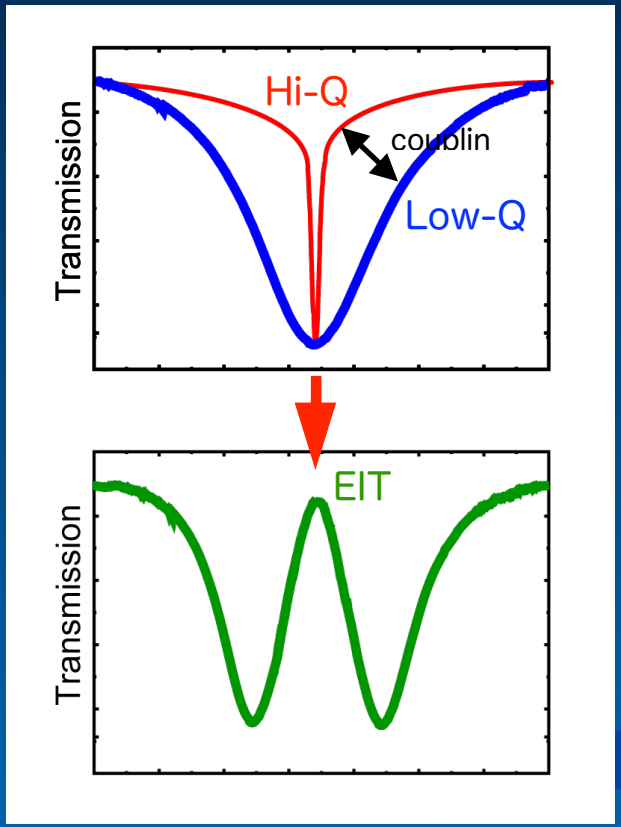
Outline

1. Non-radiative surface plasmon (Dark plasmon)
2. Dark plasmonic metamaterials for absorber
3. Application of dark metamaterials for molecular sensing
4. (Fabrication technique for 3D Metamterials)


Transmittance (%)
100
90
80
70
60
50
40
500
1000
1500
2000
Wavelentgh (nm)
No peak
(dark mode)
+
-
+
E
+
-
E


Transmittance (%)
100
90
80
70
60
50
500
1000
1500
2000
Wavelentgh (nm)
+
-
+
E
+
-
E





Plasmonic induced transparency

Fabrication of dark metamaterial

1.Substrate cleaning

Glass or sapphire substrate

2.Electron beam patterning

resist

3.Resist coat

4.Sputtering

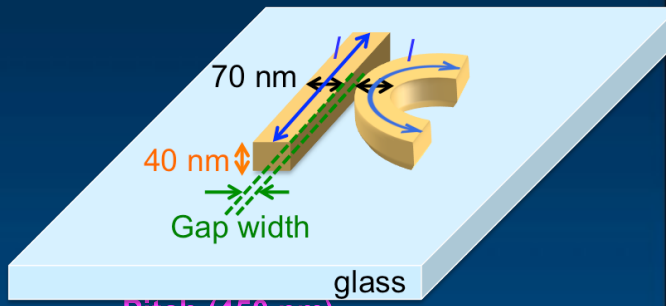
Au (40 nm)

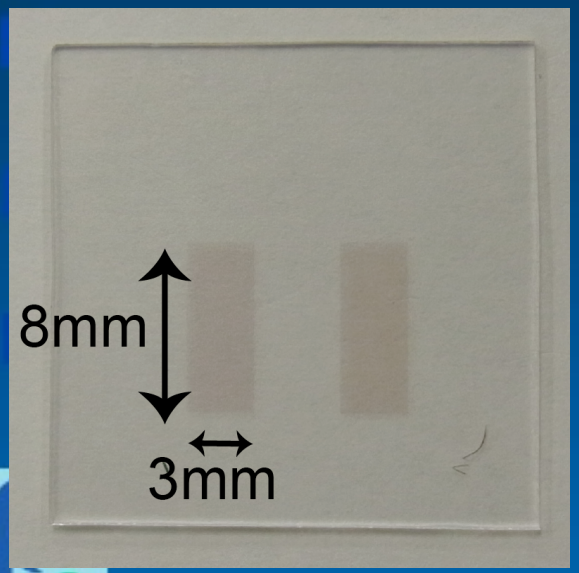
Cr (2 nm)

5.Lift-off

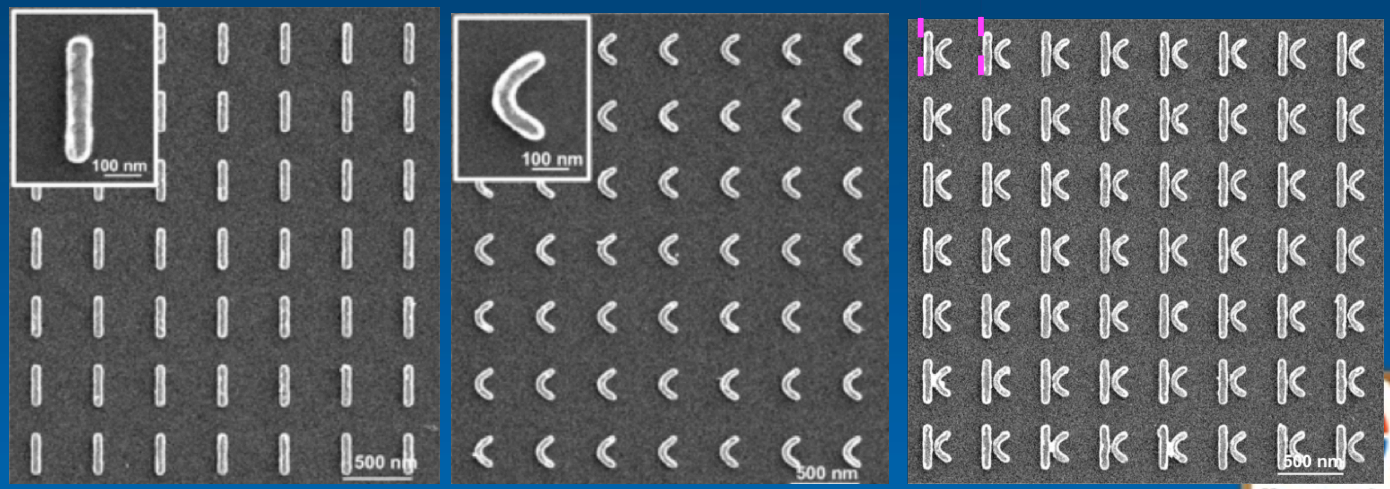
Au (40 nm)

Cr (2 nm)

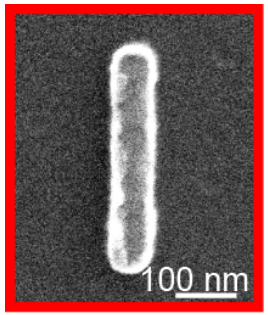




SEM images

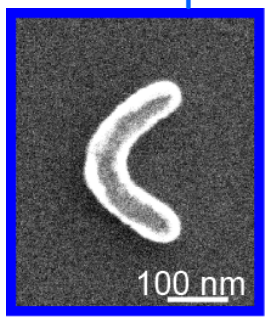


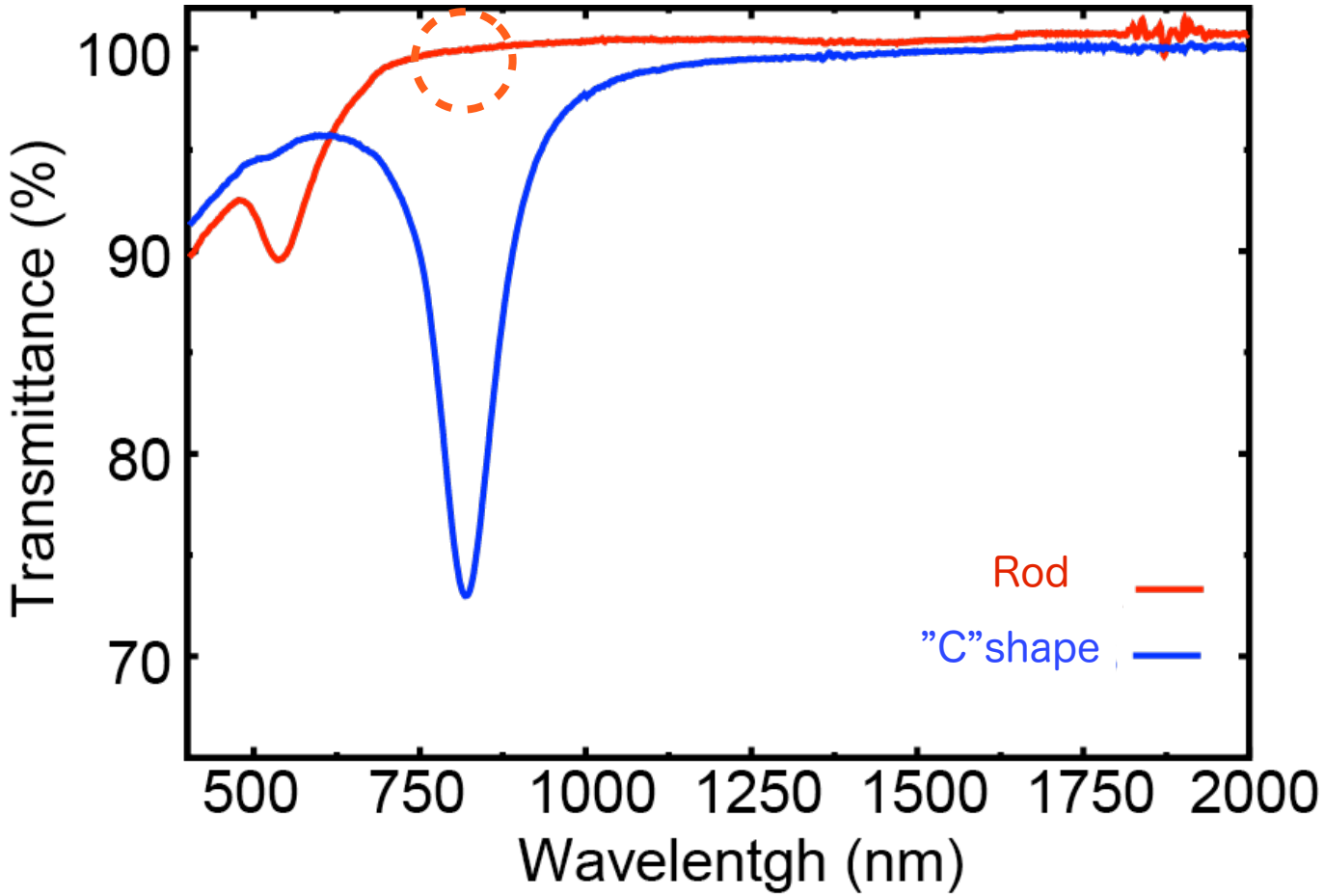
Rod


100 nm

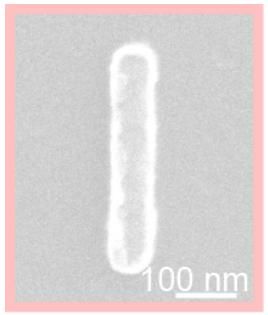
E

"C"shape

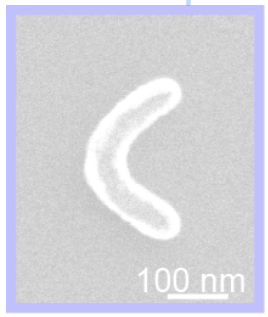

100 nm


Transmittance (%)
100
90
80
70
500
750
1000
1250
1500
1750
2000
Wavelentgh (nm)
Rod
"C"shape

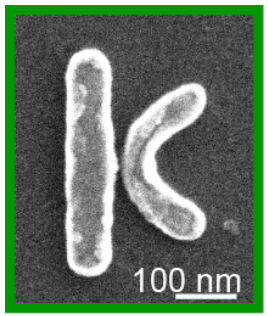
Rod

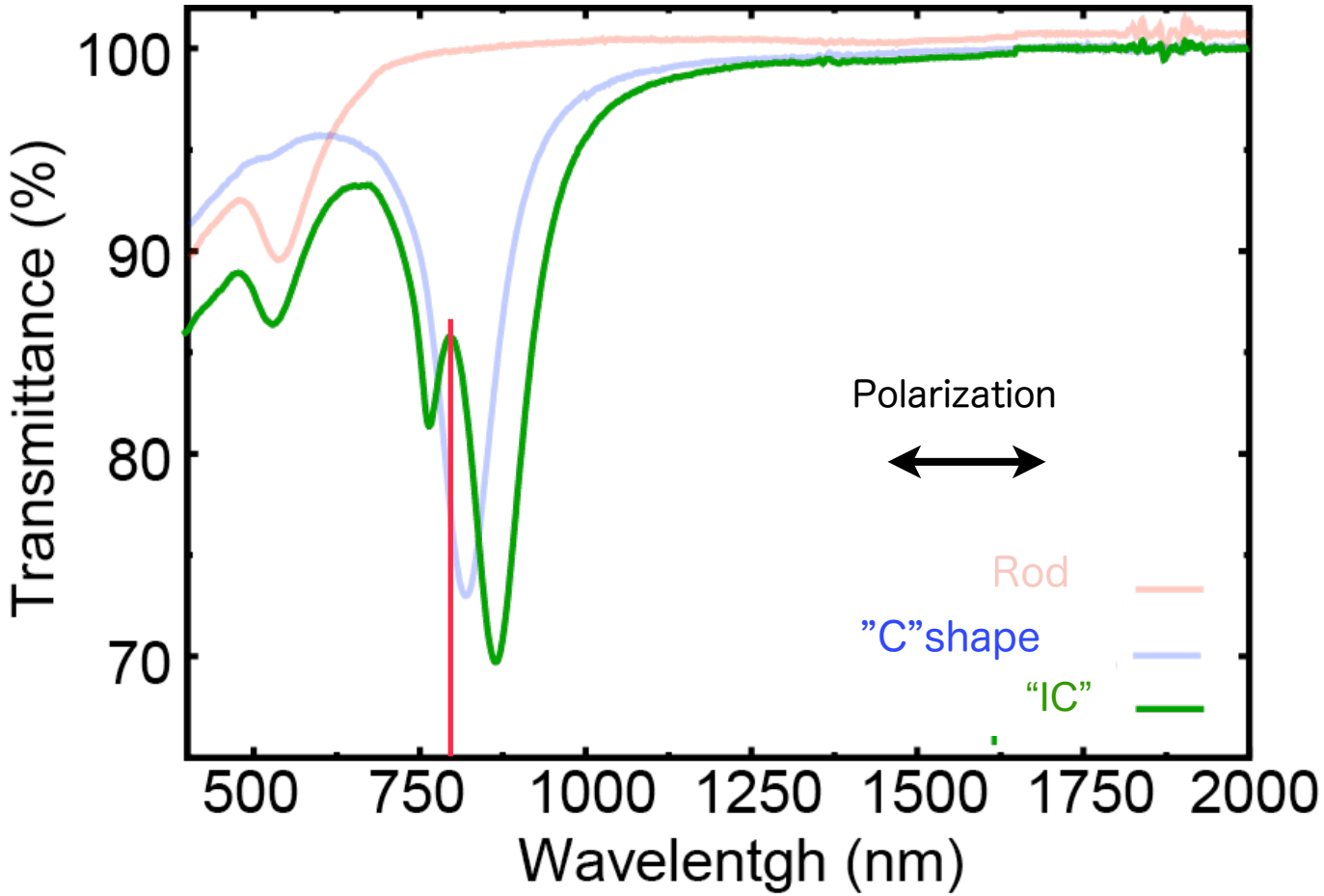


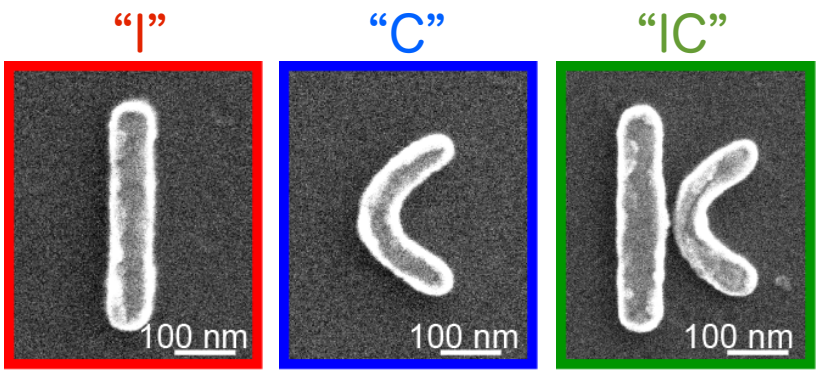
”C”shape

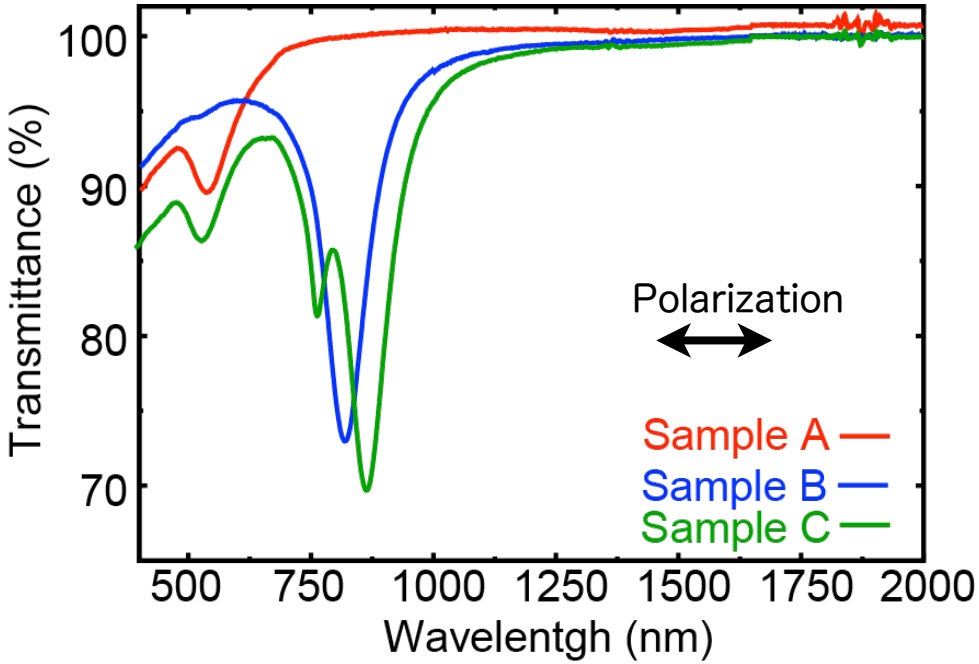


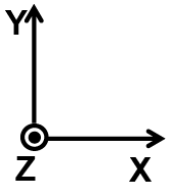
”IC”

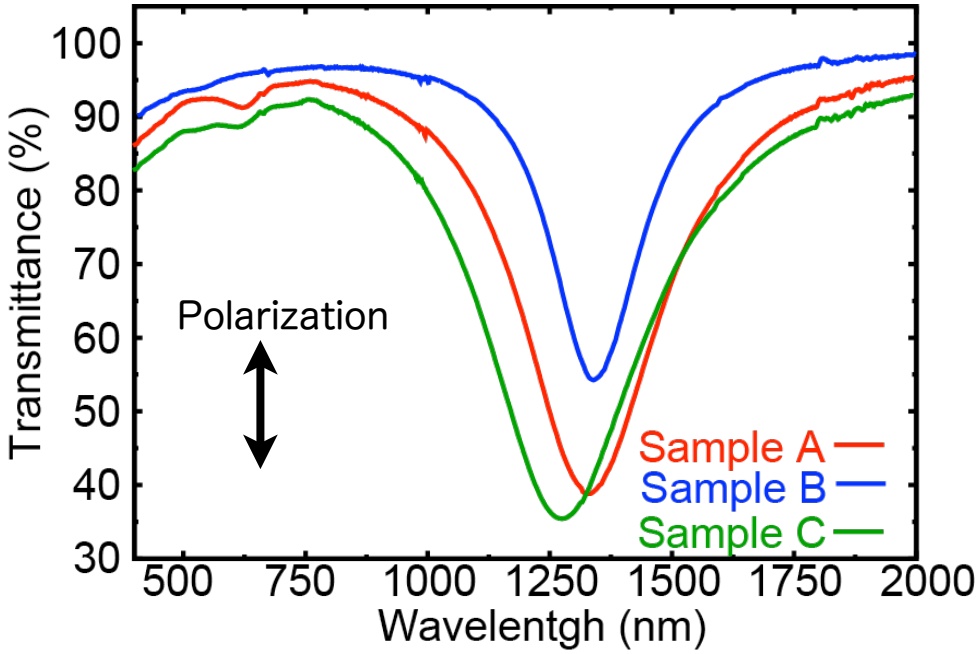


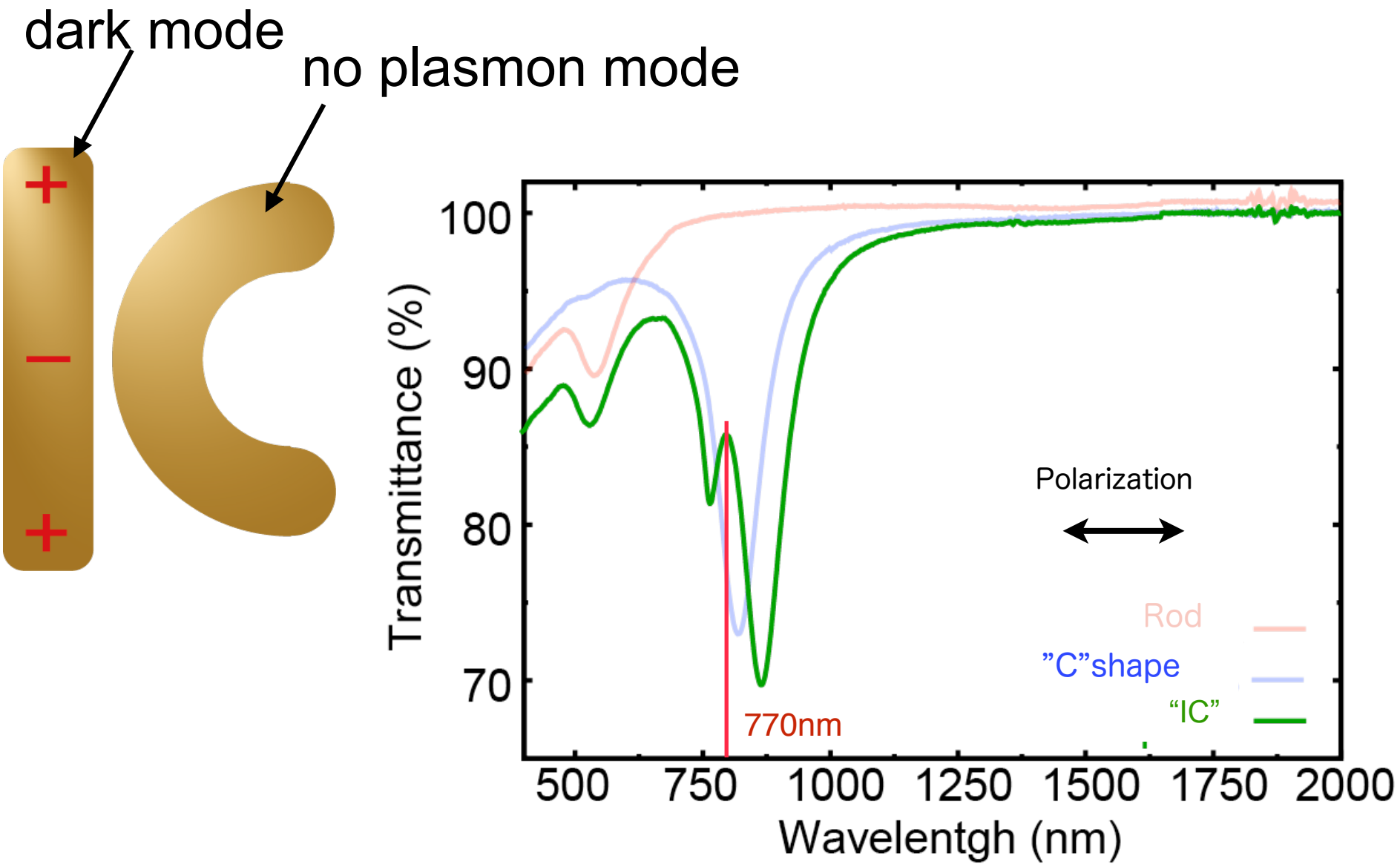



"I"
"C"
"IC"
100 nm
100 nm
100 nm

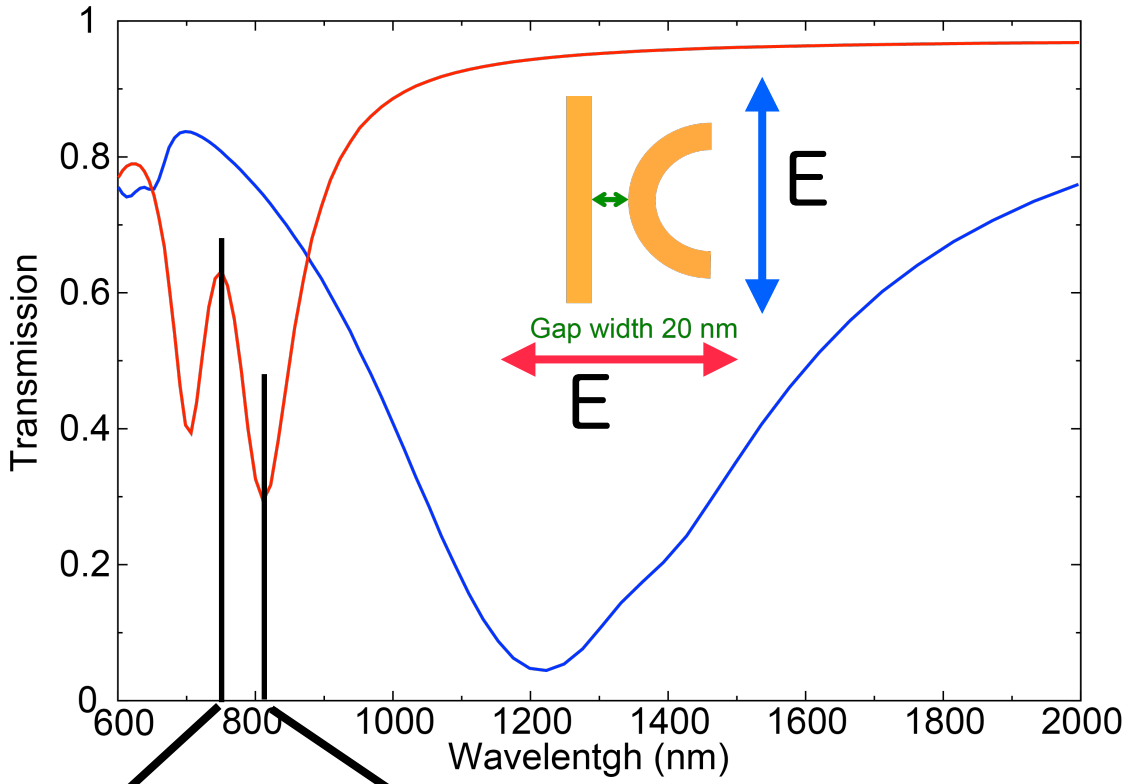

Transmittance (%)
100
90
80
70
Polarization
Sample A
Sample B
Sample C
500
750
1000
1250
1500
1750
2000
Wavelentgh (nm)


Y
Z
X

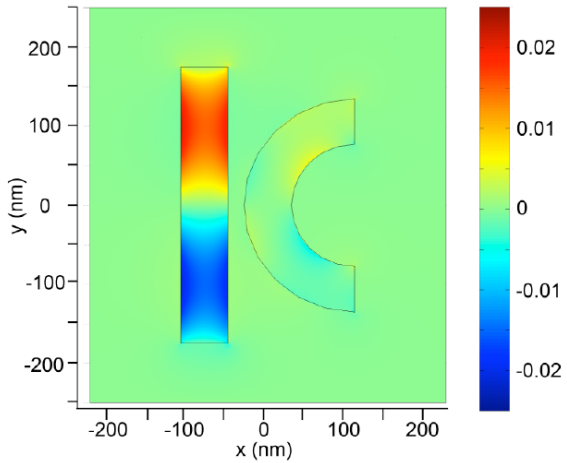

Transmittance (%)
100
90
80
70
60
50
40
30
Polarization
Sample A
Sample B
Sample C
500
750
1000
1250
1500
1750
2000
Wavelentgh (nm)


dark mode
no plasmon mode
+
−
+
Transmittance (%)
100
90
80
70
500
750
1000
1250
1500
1750
2000
Wavelentgh (nm)
770nm
Polarization
Rod
”C”shape
“IC”

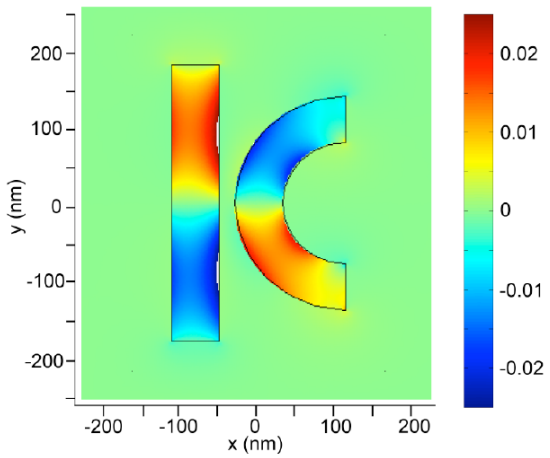
Numerical results (by FEM)



at 750nm

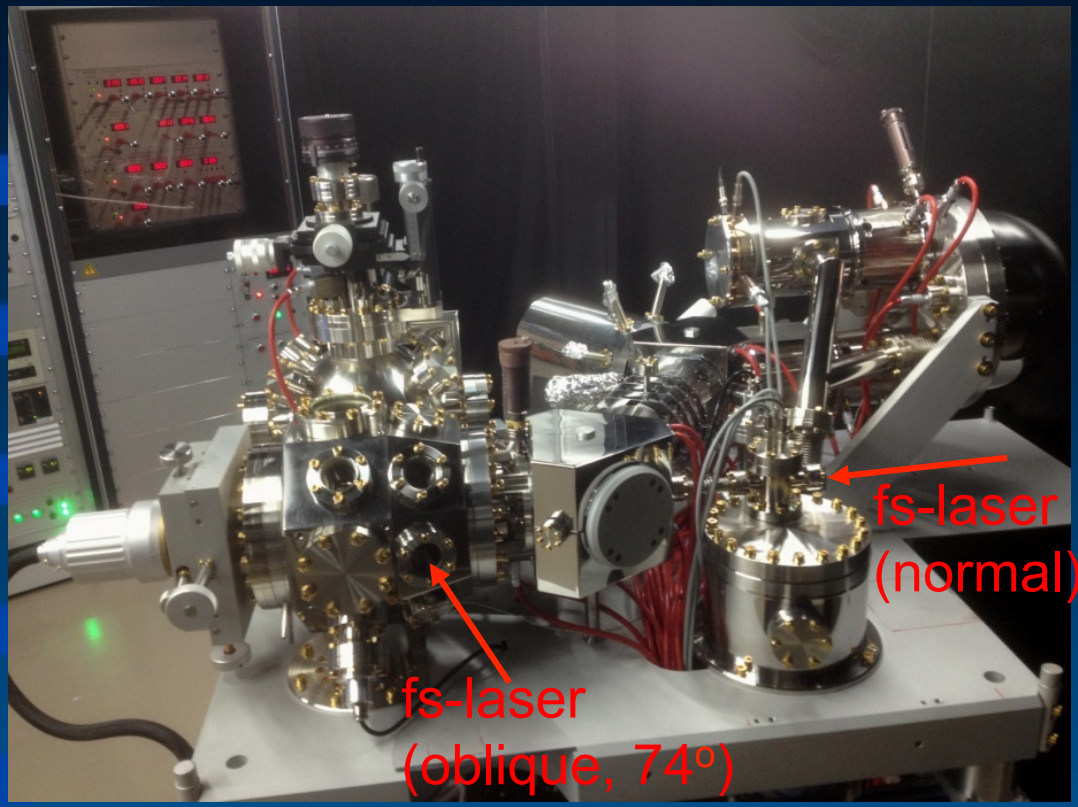


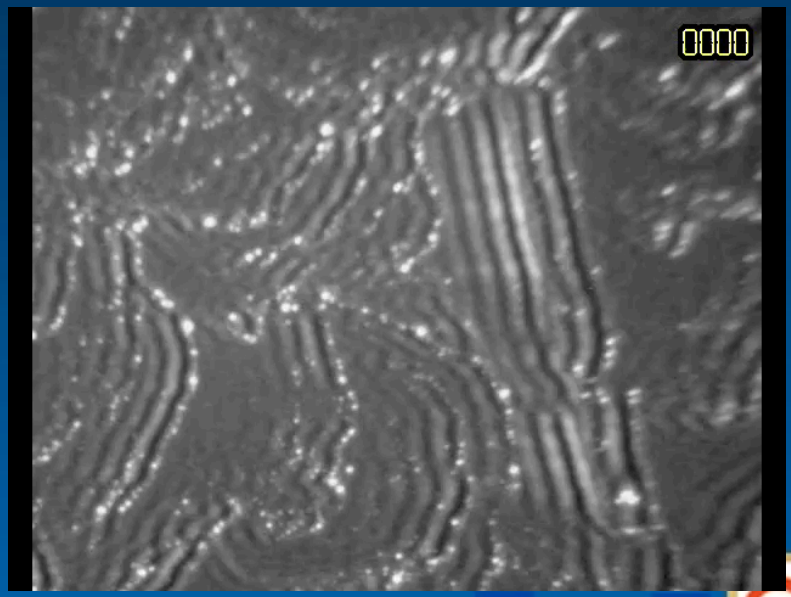
at 810nm



J

- Time resolved Photoemission Electron Microscopy

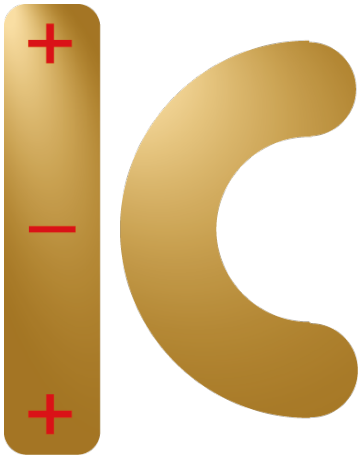




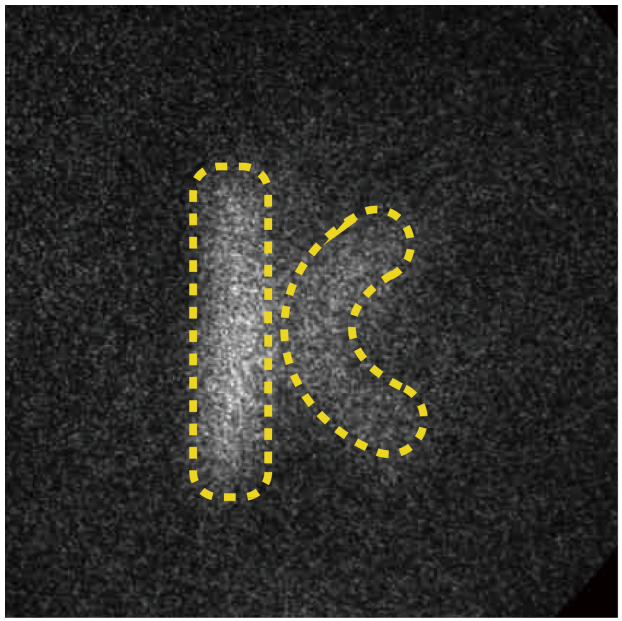
SP propagates on Au surface

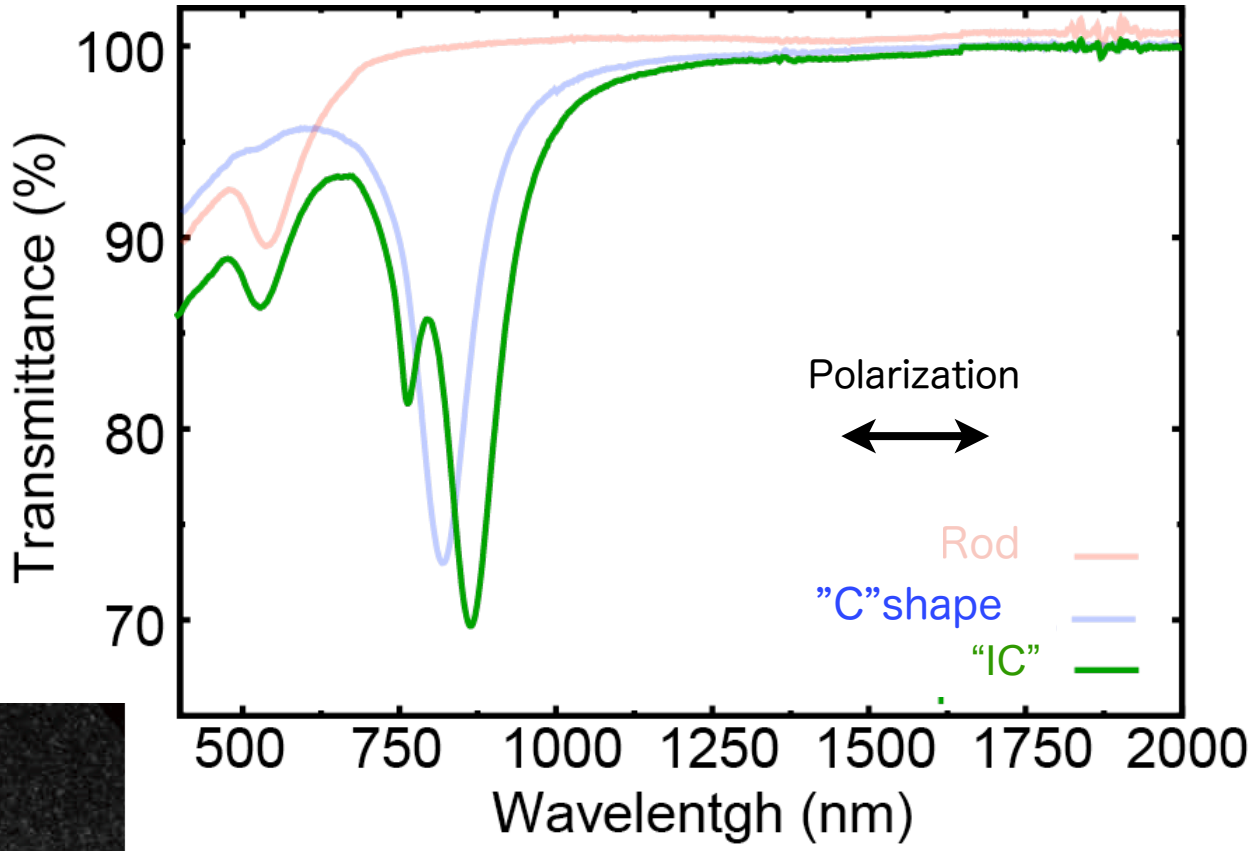
Collaboration with Prof. Misawa & Ueno
in Hokkaido Univ.

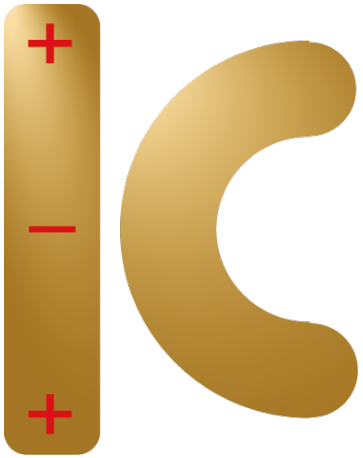
4nm spatial resolution


+
−
+

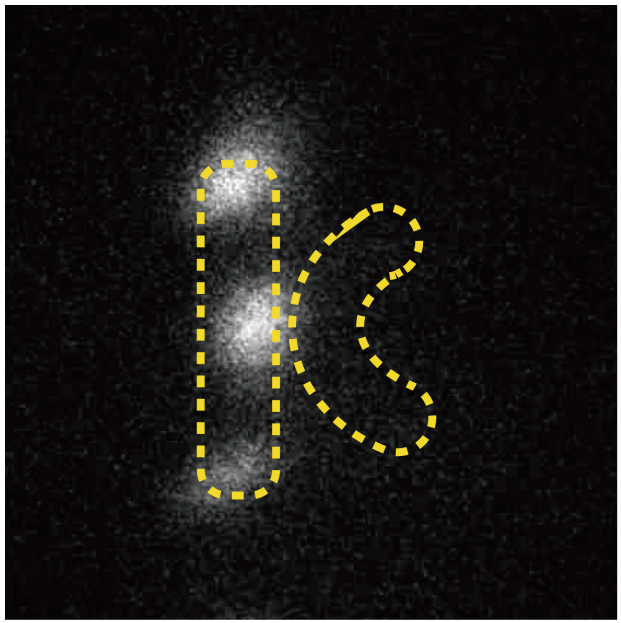
PEEM@UV

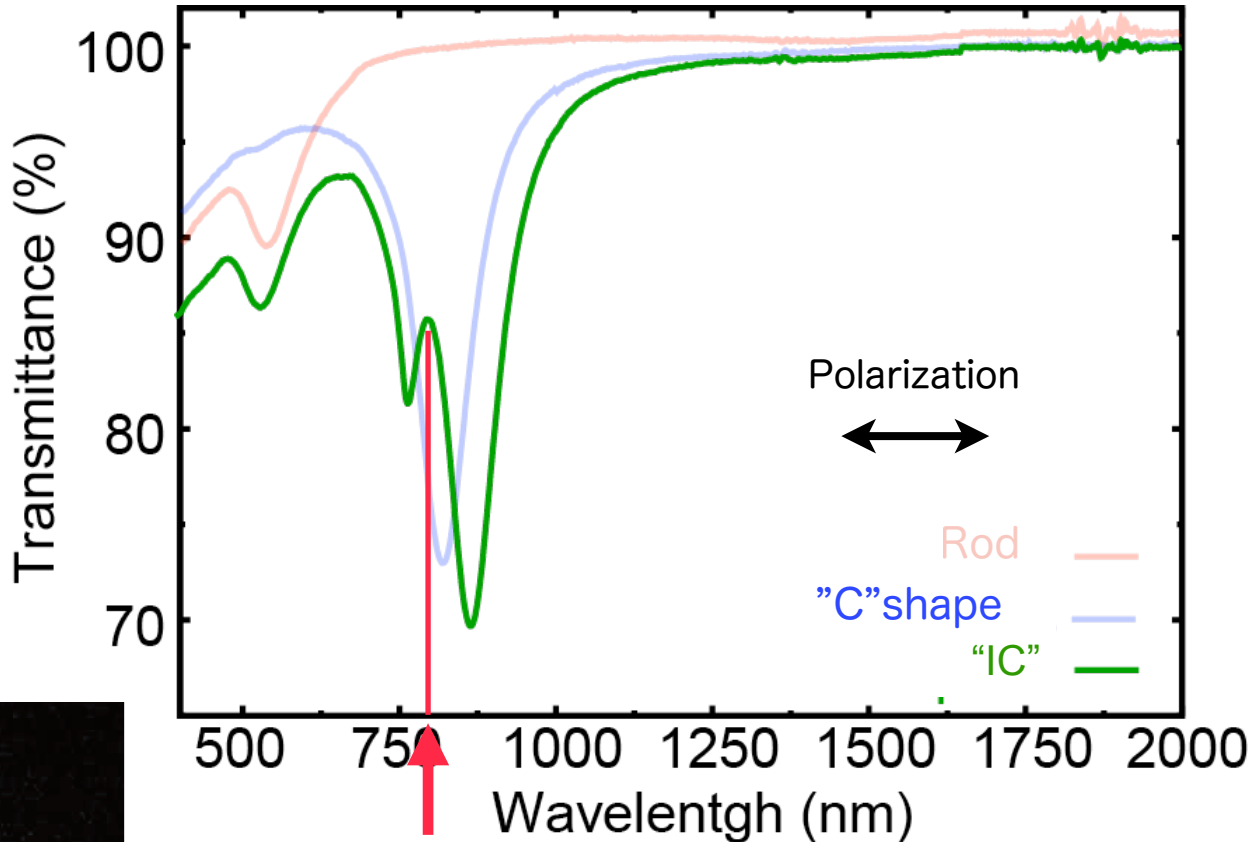



Transmittance (%)
100
90
80
70
500
750
1000
1250
1500
1750
2000
Wavelentgh (nm)
Polarization
Rod
”C”shape
“IC”



PEEM@770nm





(almost) No radiation

Outline

1. Non-radiative surface plasmon (Dark plasmon)
2. Dark plasmonic metamaterials for absorber
3. Application of dark metamaterials for molecular sensing
4. (Fabrication technique for 3D Metamterials)

Metamaterial absorber

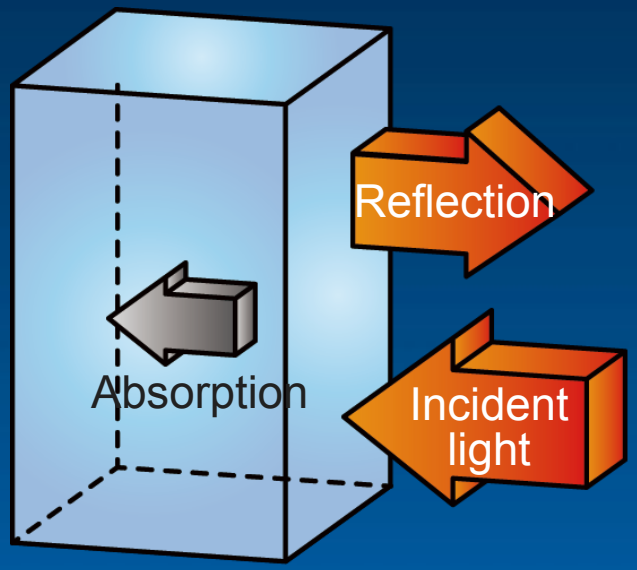
Discrepancy between extinction coefficient and absorption

Index of refraction $N = n - ik$

extinction coefficient

Fresnel formula (from air to matter.)

$$R = \left| \frac{N-1}{N+1} \right|^2$$



Highly absorptive material

↓

Large extinction coef.

↓ contradiction

Highly reflection

↓

Low absorption

e.g. : Au

N = 1.85 - 21.7i at λ = 3 μm

~ 98% reflection

Only 2% absorption

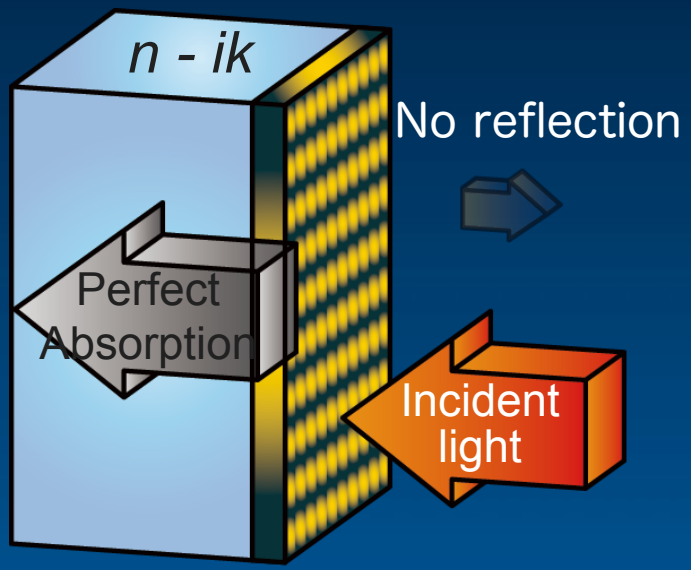
Gold

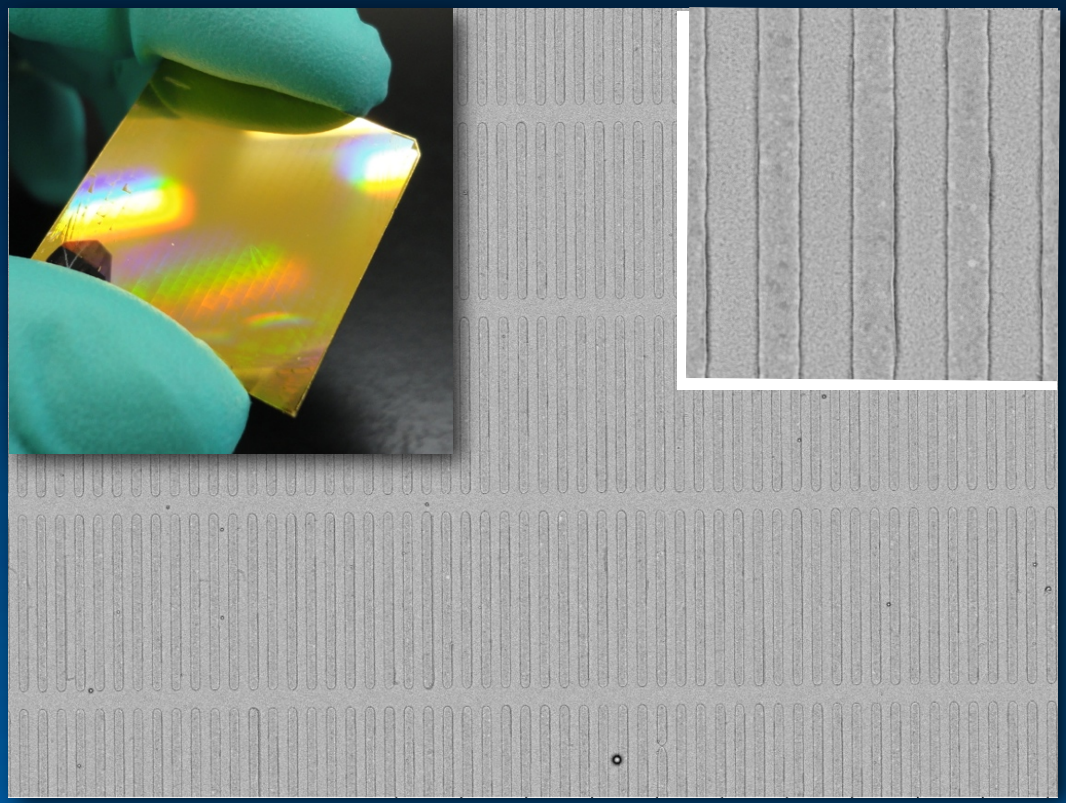
Sci. Rep. **5**, 12570 (2015).

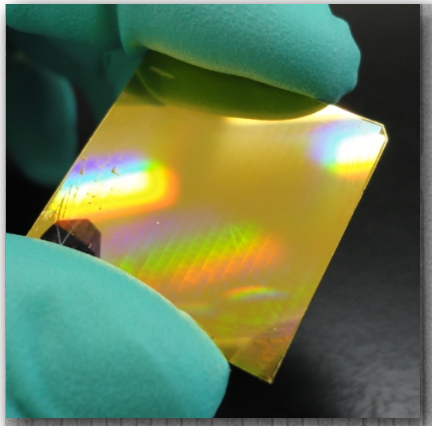
Metamaterial absorber

- Nano scale structures on flat metal film (no transmission)
- Impedance matching for no reflection

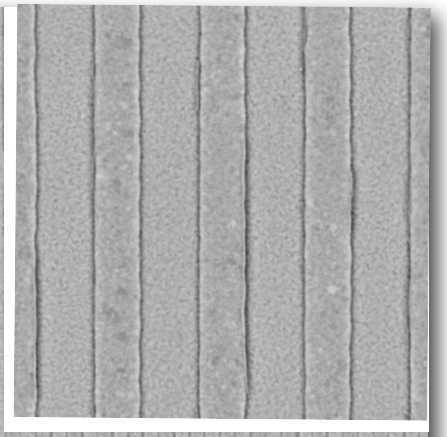
➡ Perfect absorption by ultra-thin layer







30 x 30mm²



w/Λ = 1.5/3.0µm

Au

MgF_2

Au

SiO_2

25µm

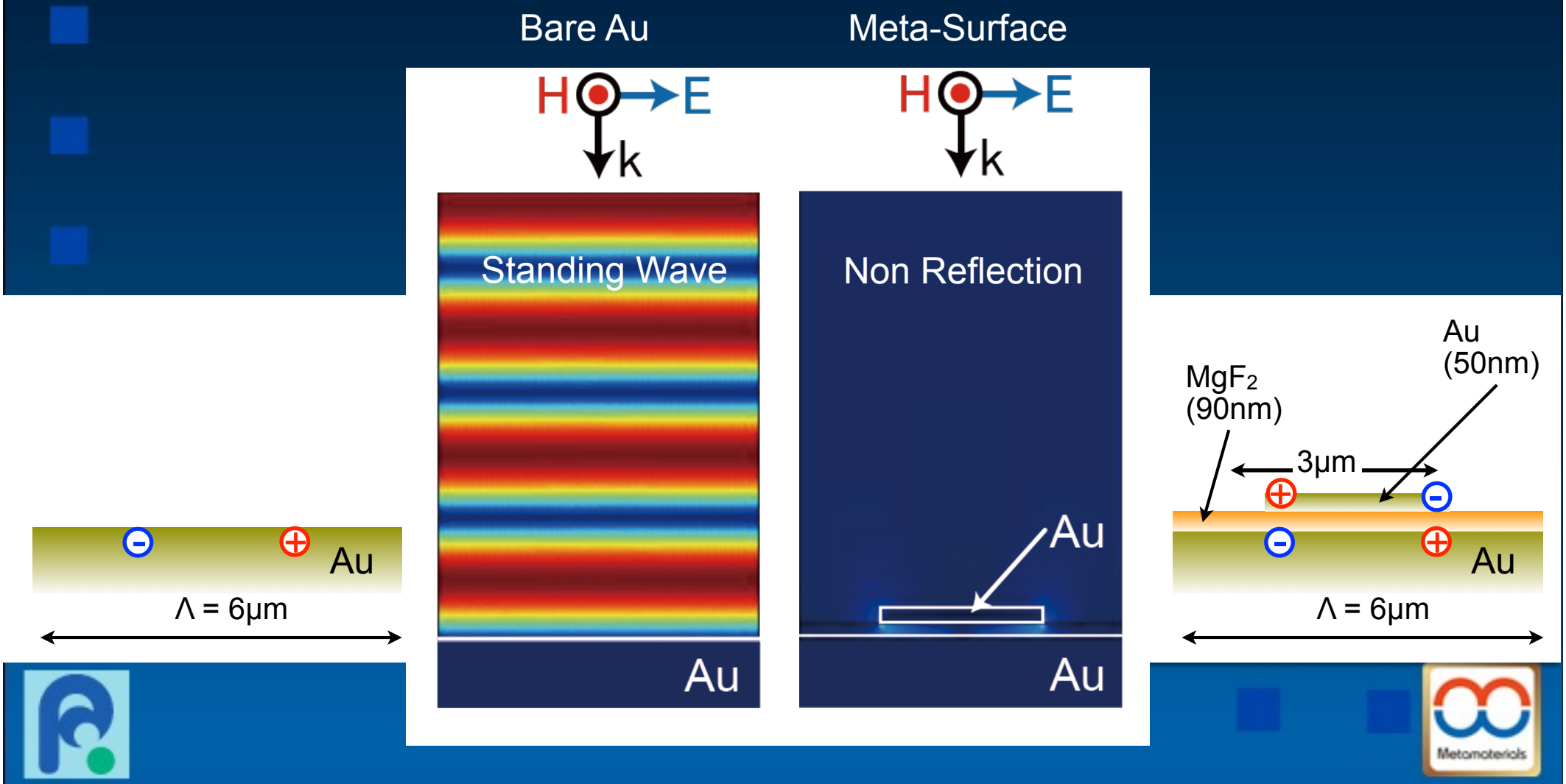
Sci. Rep. **5**, 12570 (2015).

Metamaterial absorber

Nano scale structures on flat metal film (no transmission)

Impedance matching for non-reflection

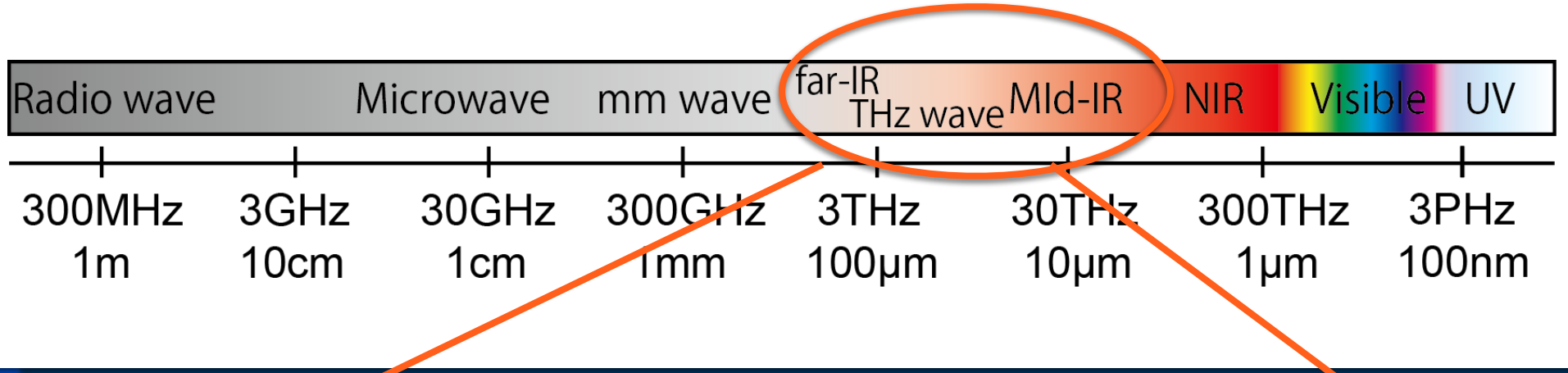
➡ Perfect absorption

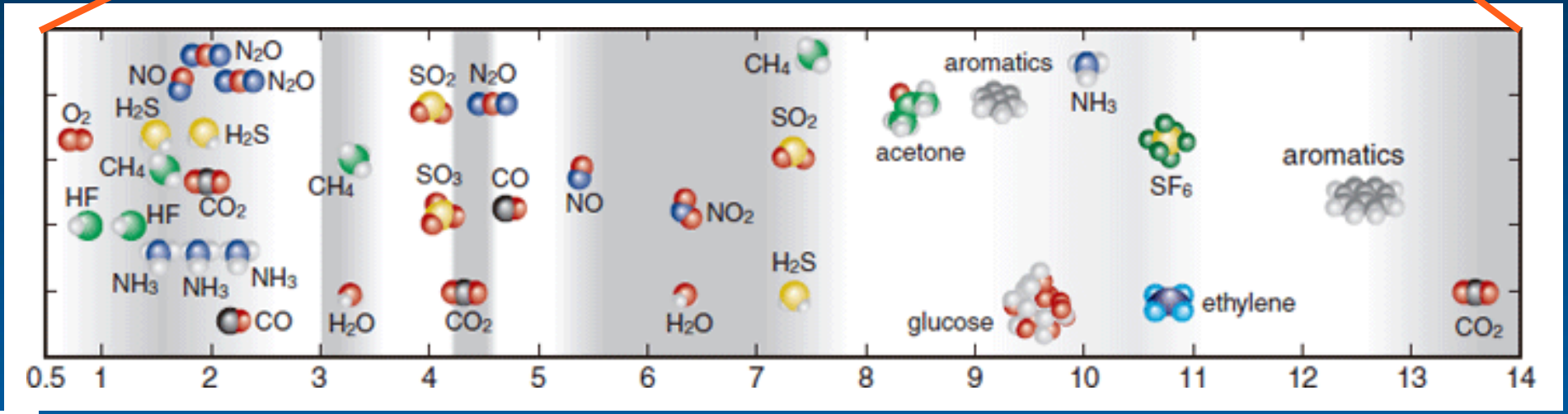


Outline

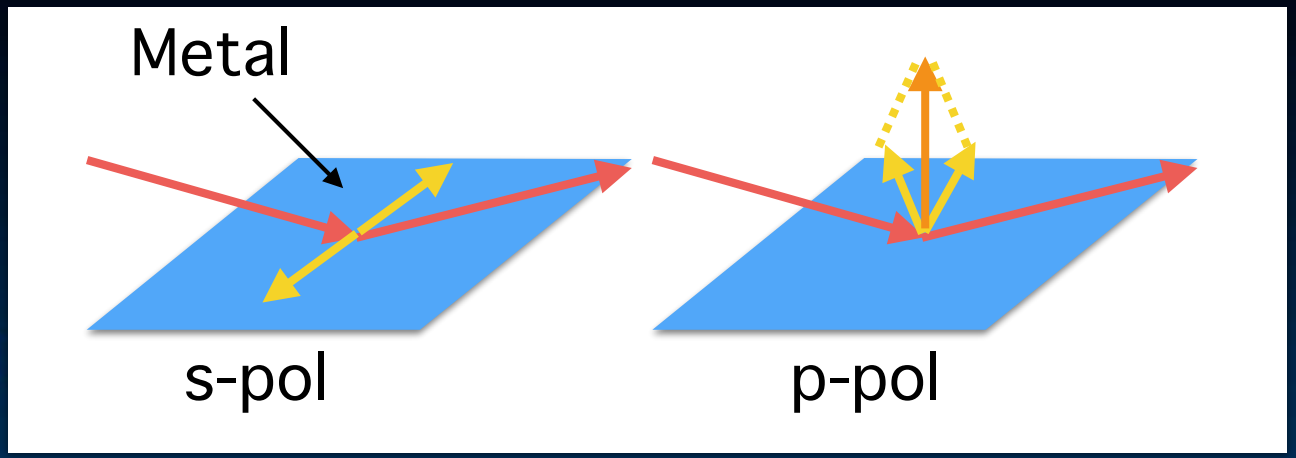
1. Non-radiative surface plasmon (Dark plasmon)
2. Dark plasmonic metamaterials for absorber
3. Application of dark metamaterials for molecular sensing
4. (Fabrication technique for 3D Metamterials)

Finger print region



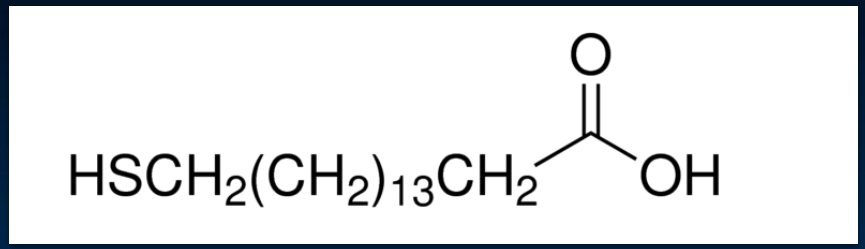


Reflection Absorption Spectrometry (RAS)



[1] A.Gole *et al.*, Langmuir **20**, 7117 (2004).

16-Mercaptohexadecanoic acid



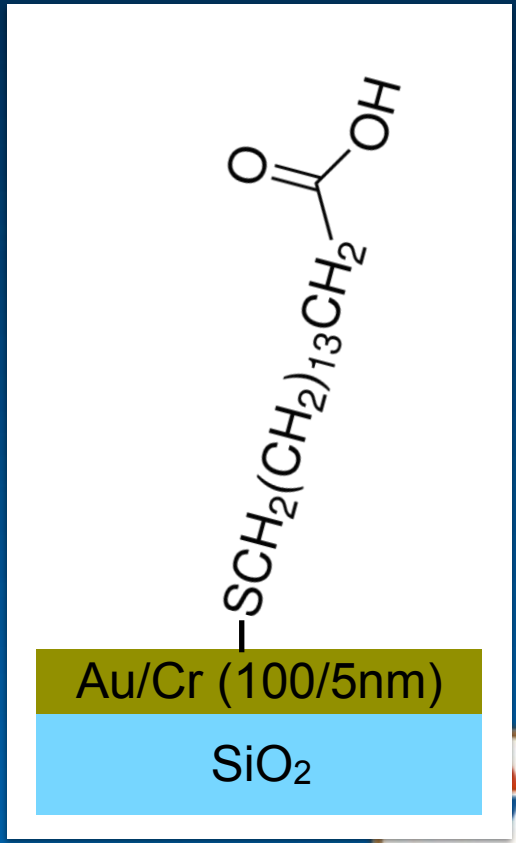
C–H stretching vibrational modes
2850–2863 cm^{-1} (symmetric)
2916–2936 cm^{-1} (asymmetric)

Recipe for 16-Mercaptohexadecanoic acid SAM

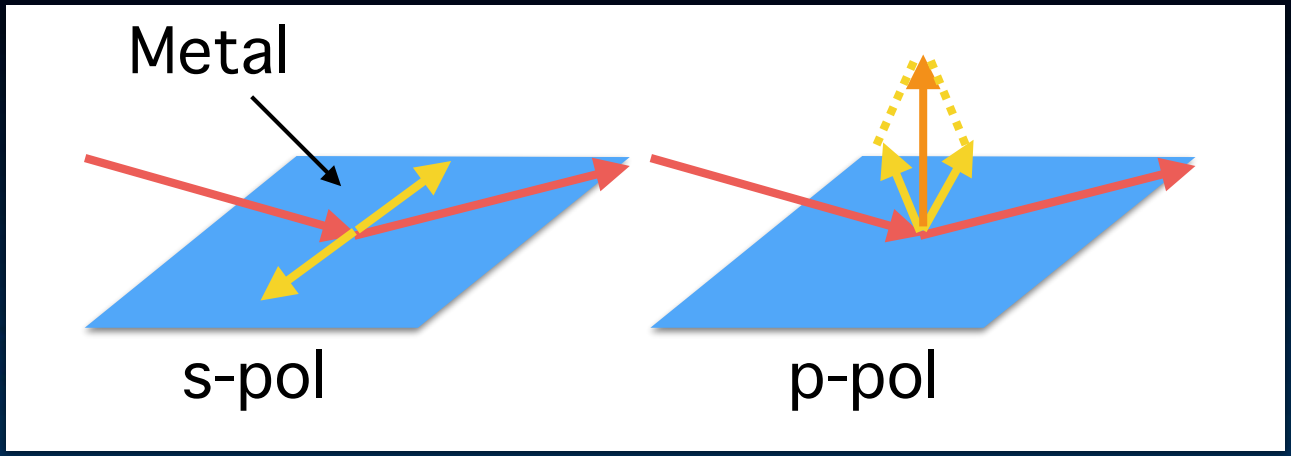
1) Au substrate immersed in a 10^{-3}M solution in ethanol for 36 h.

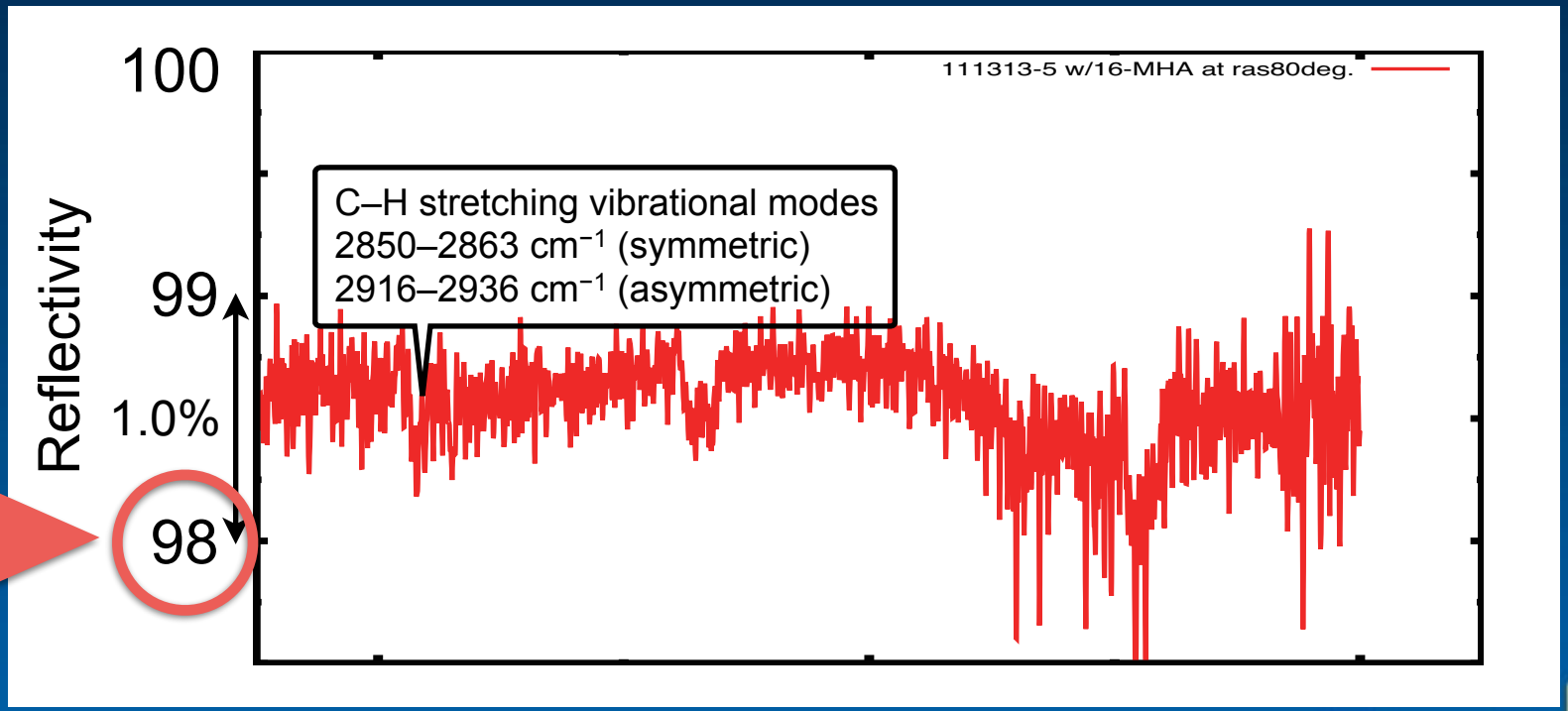
2) Then, rinsed in ethanol and dried with N_2 gas.

3) Characterized with FT-IR (Reflection Absorption Spectroscopy)



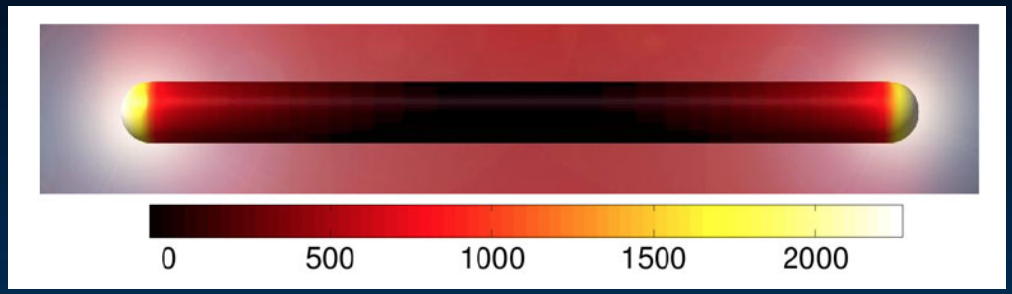
Reflection Absorption Spectrometry (RAS)

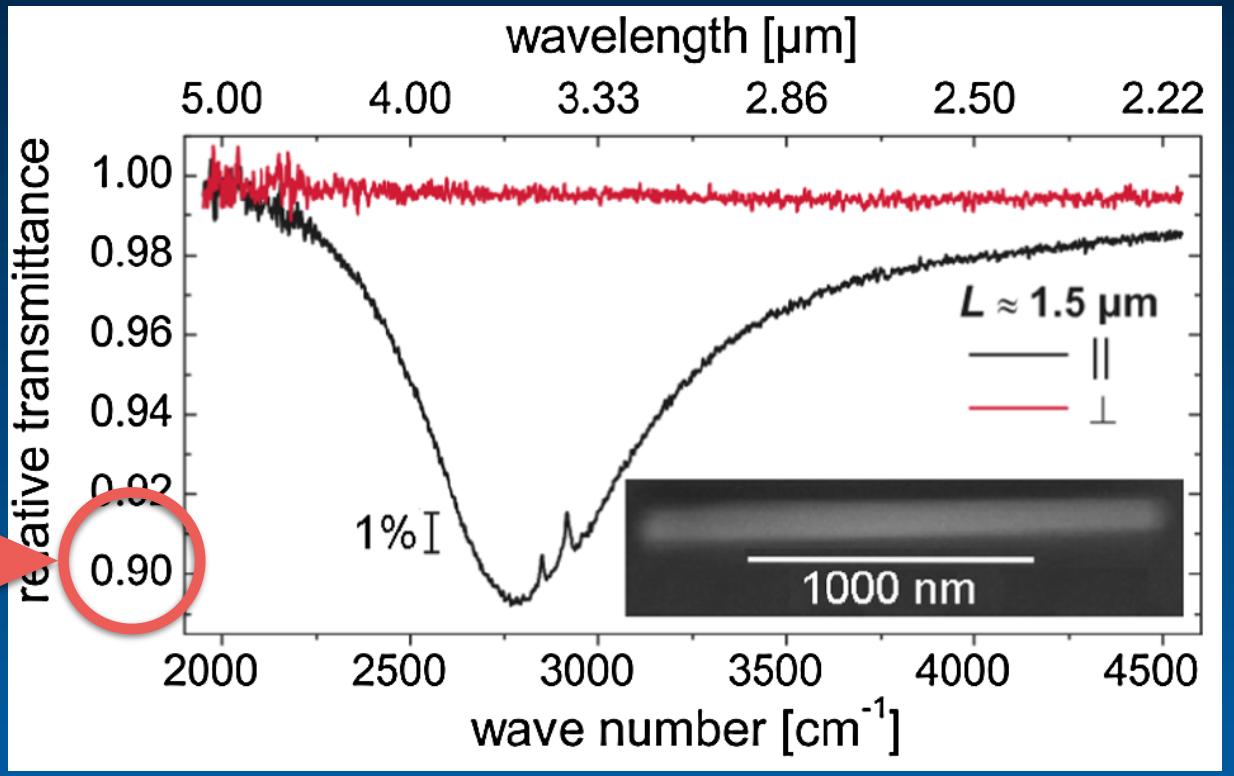




F. Neubrech et. al, Phys. Rev. Lett. 101, 157403 (2008)

SEIRA (Surface enhancement IR absorption) using coupling of plasmon with vibrational mode of molecule.





$CH_3(CH_3)_{16}CH_2SH$

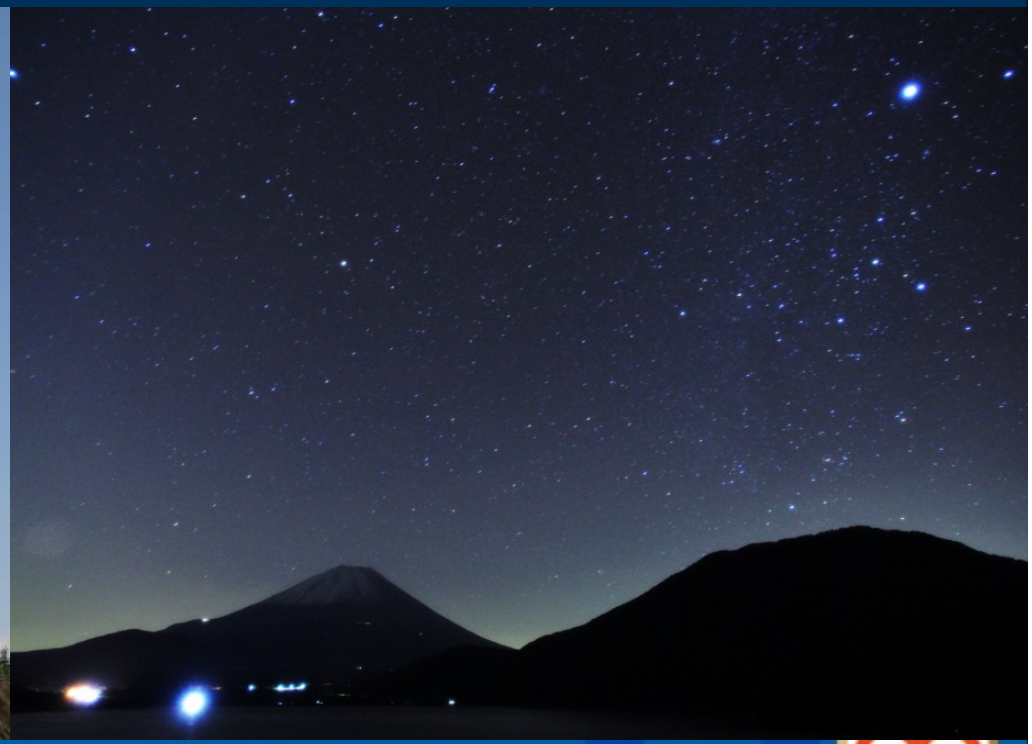
S/B = Signal / Background

Enhancement

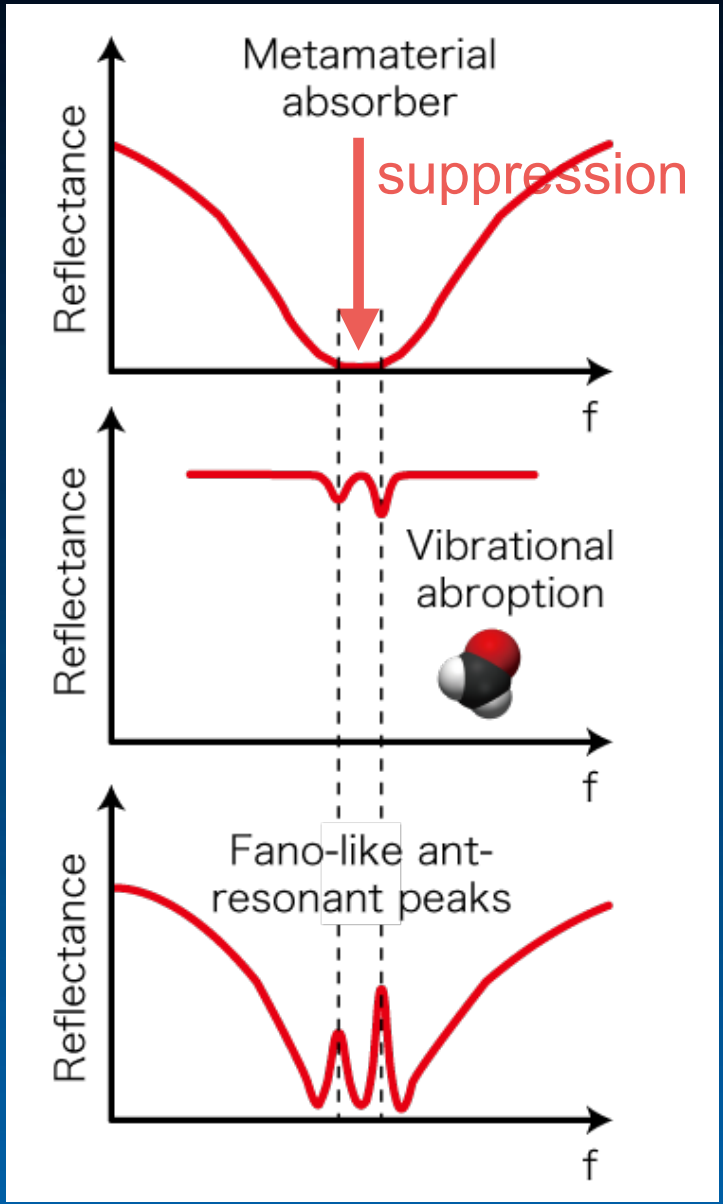
Suppression

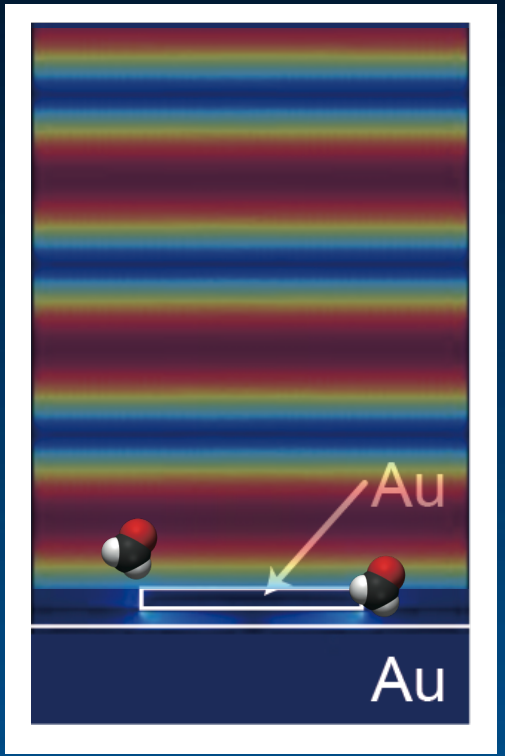
×

○



Resonant Coupling Absorption Spectroscopy





- Background suppression
- Resonantly enhanced signal
- Zepto-mol sensitivity

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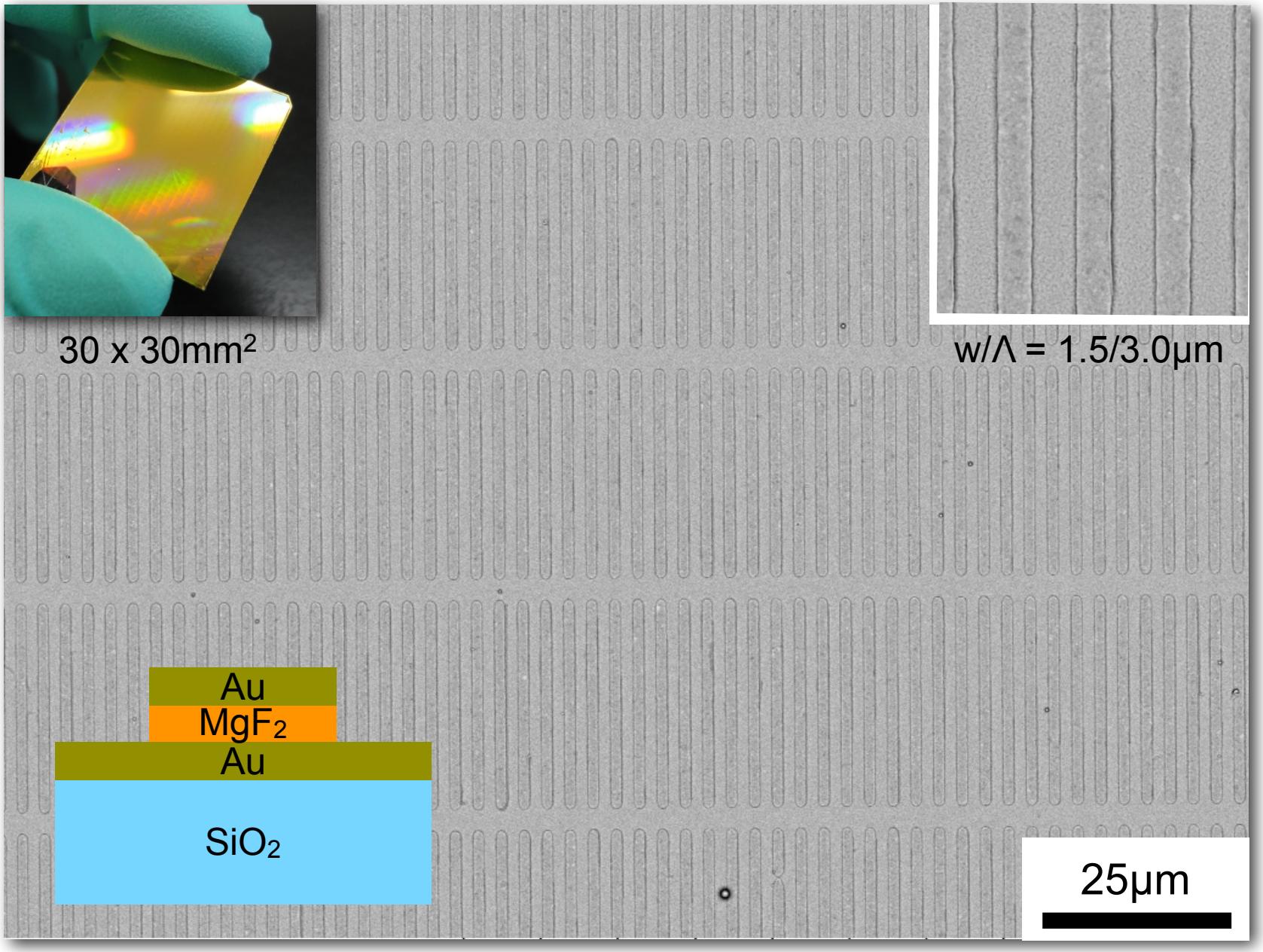
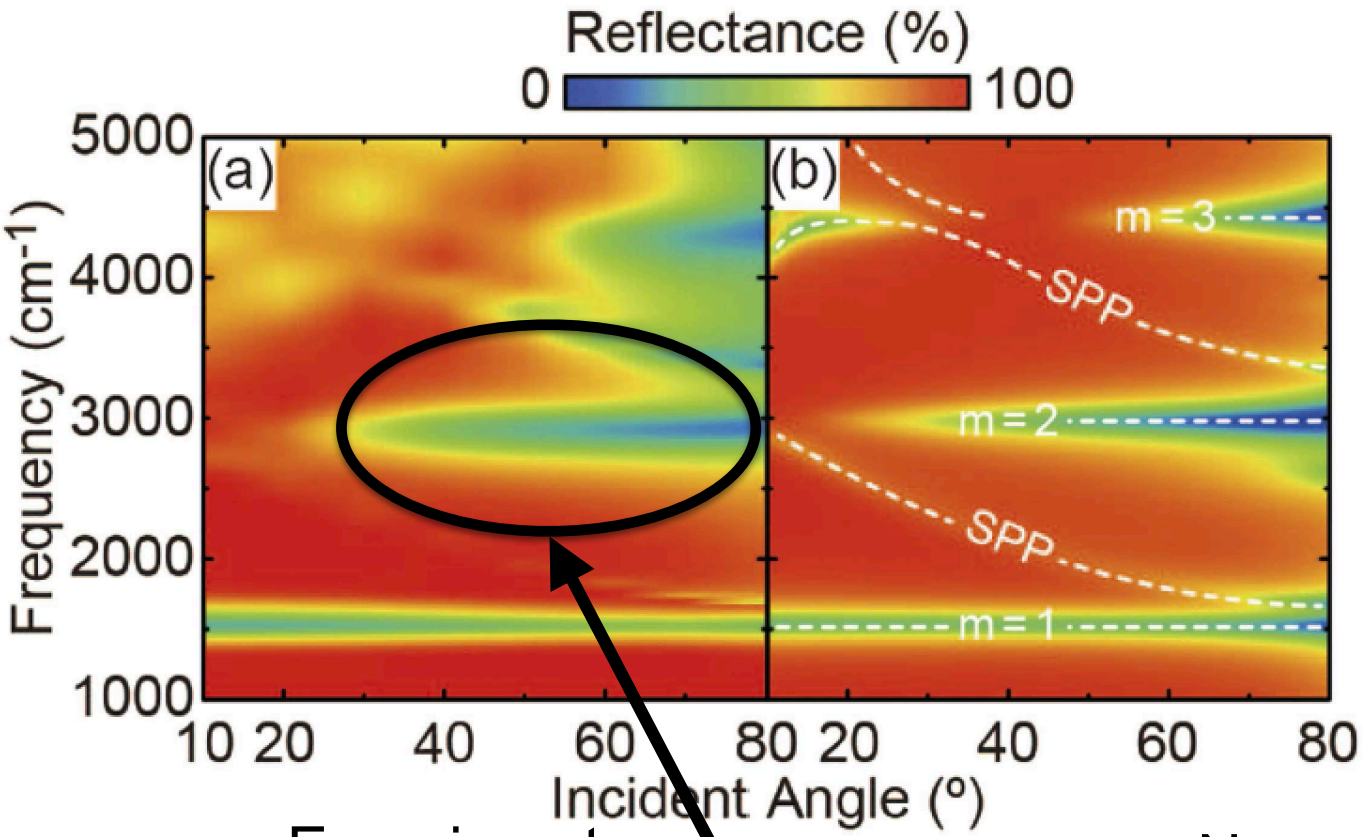
Metamaterials

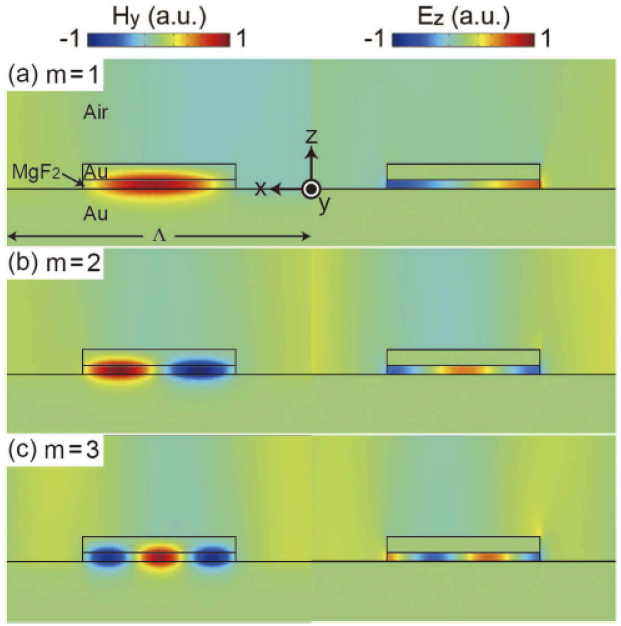
Sci. Rep. **5**, 12570 (2015).



IR characterization

Sci. Rep. **5**, 12570 (2015).



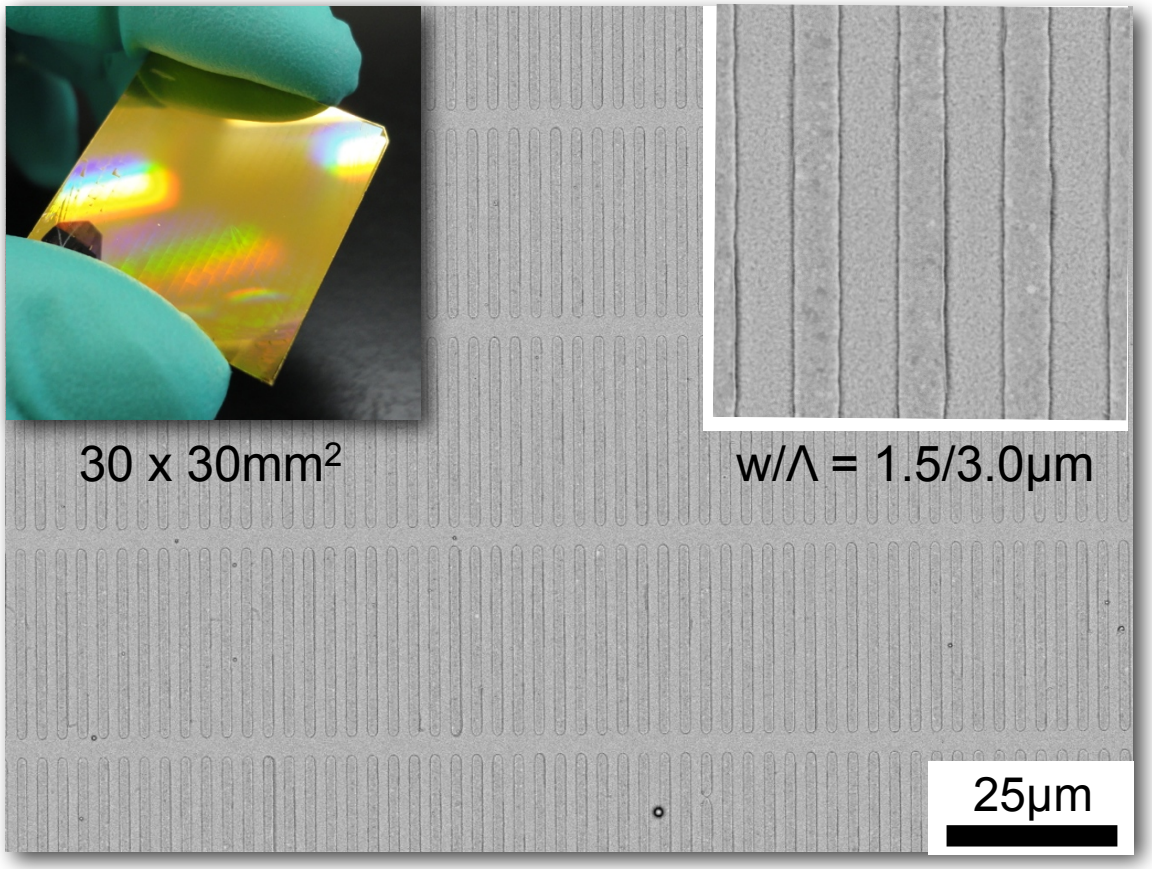


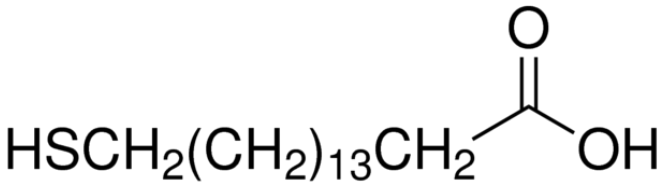
Experiment

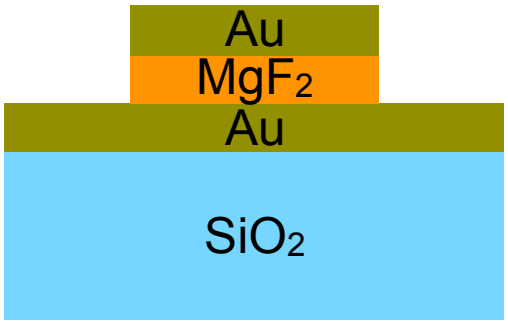
Numerical

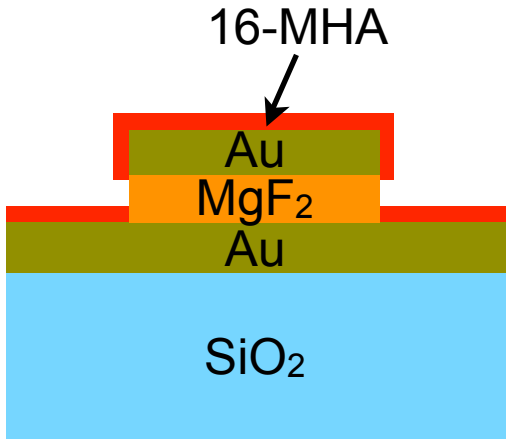
C–H stretching vibrational modes
2850–2863 cm^{-1} (symmetric)
2916–2936 cm^{-1} (asymmetric)

Sci. Rep. **5**, 12570 (2015).



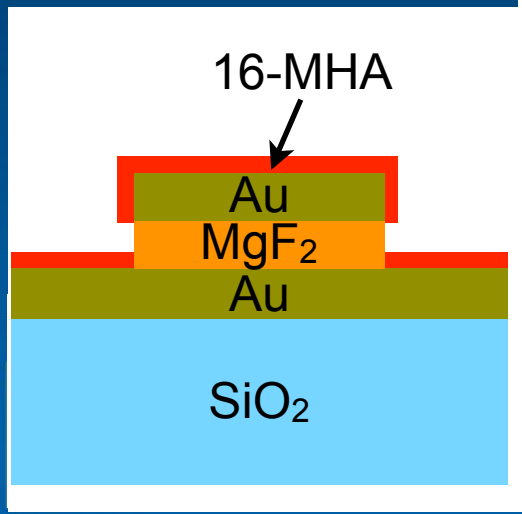


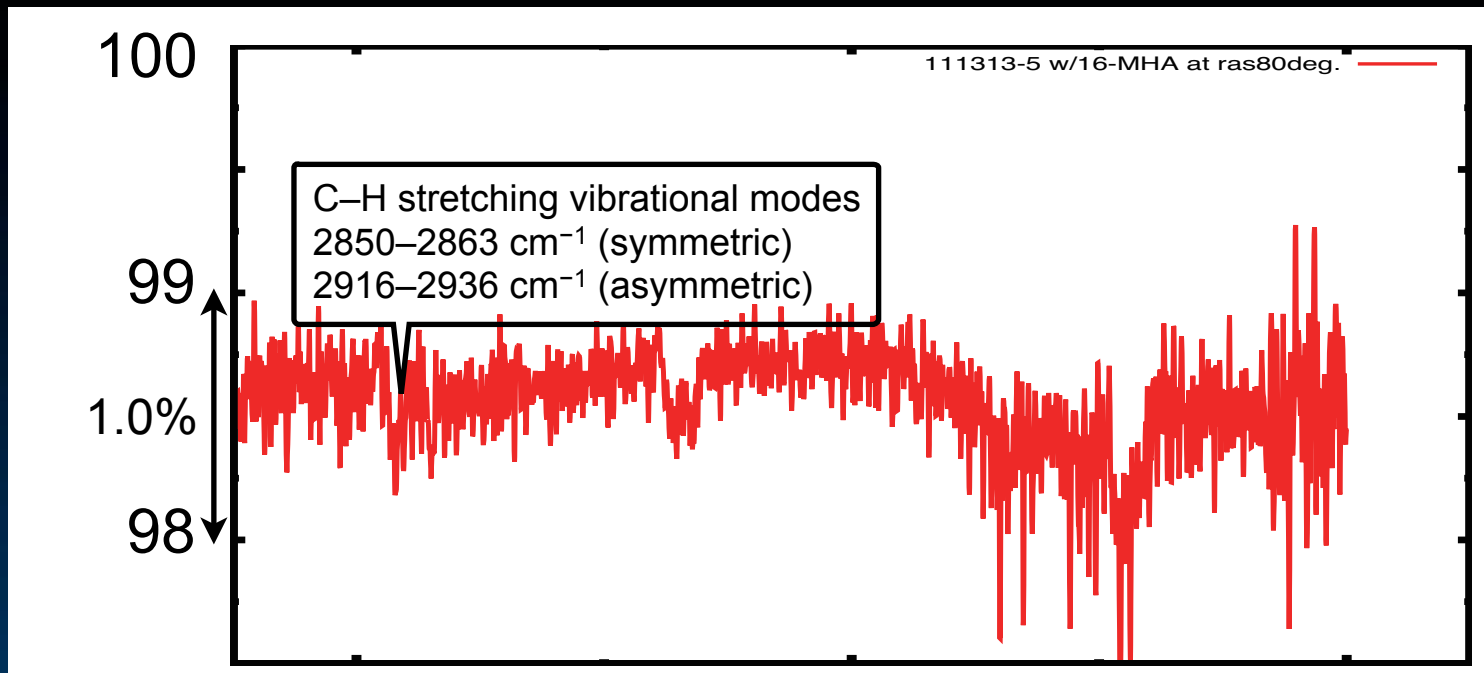


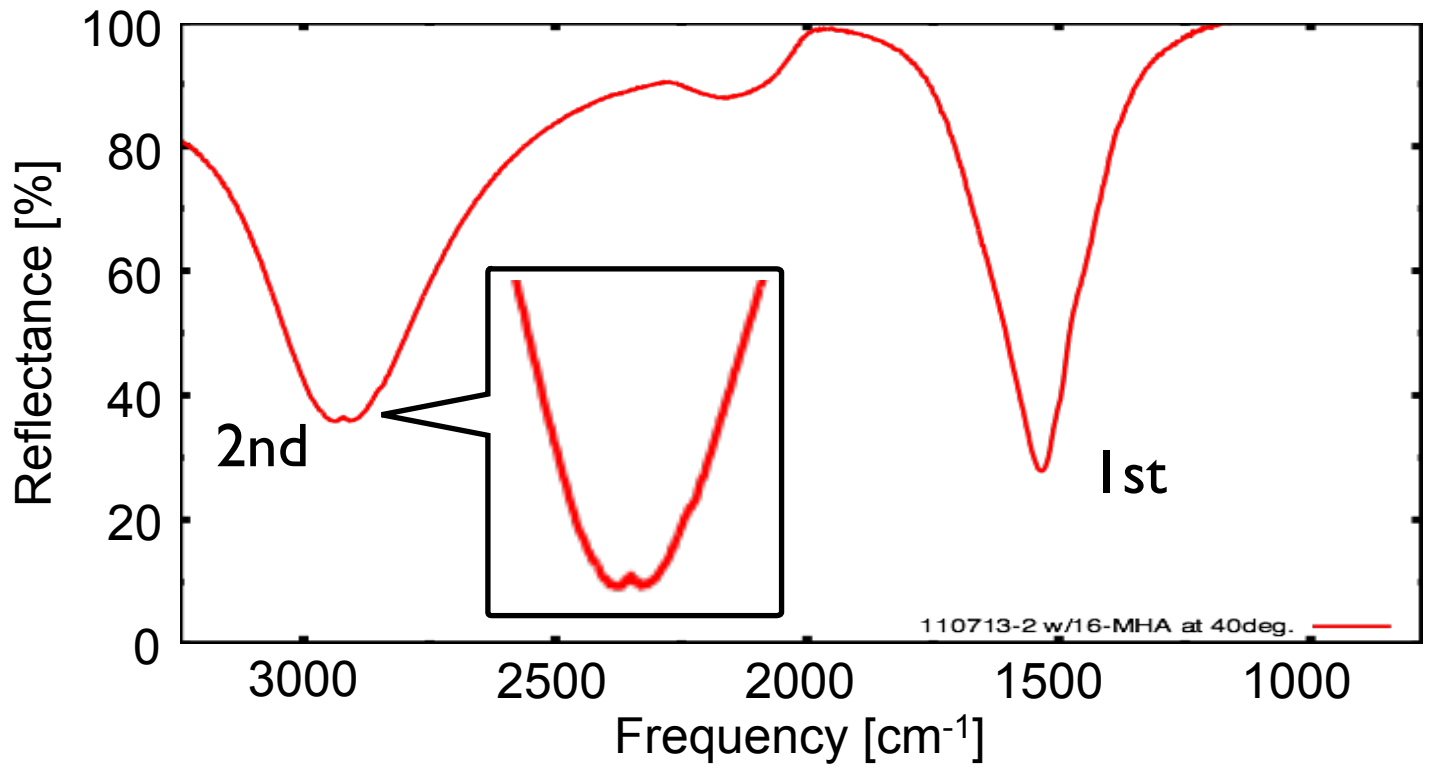


on bare Au

on meta-absorber

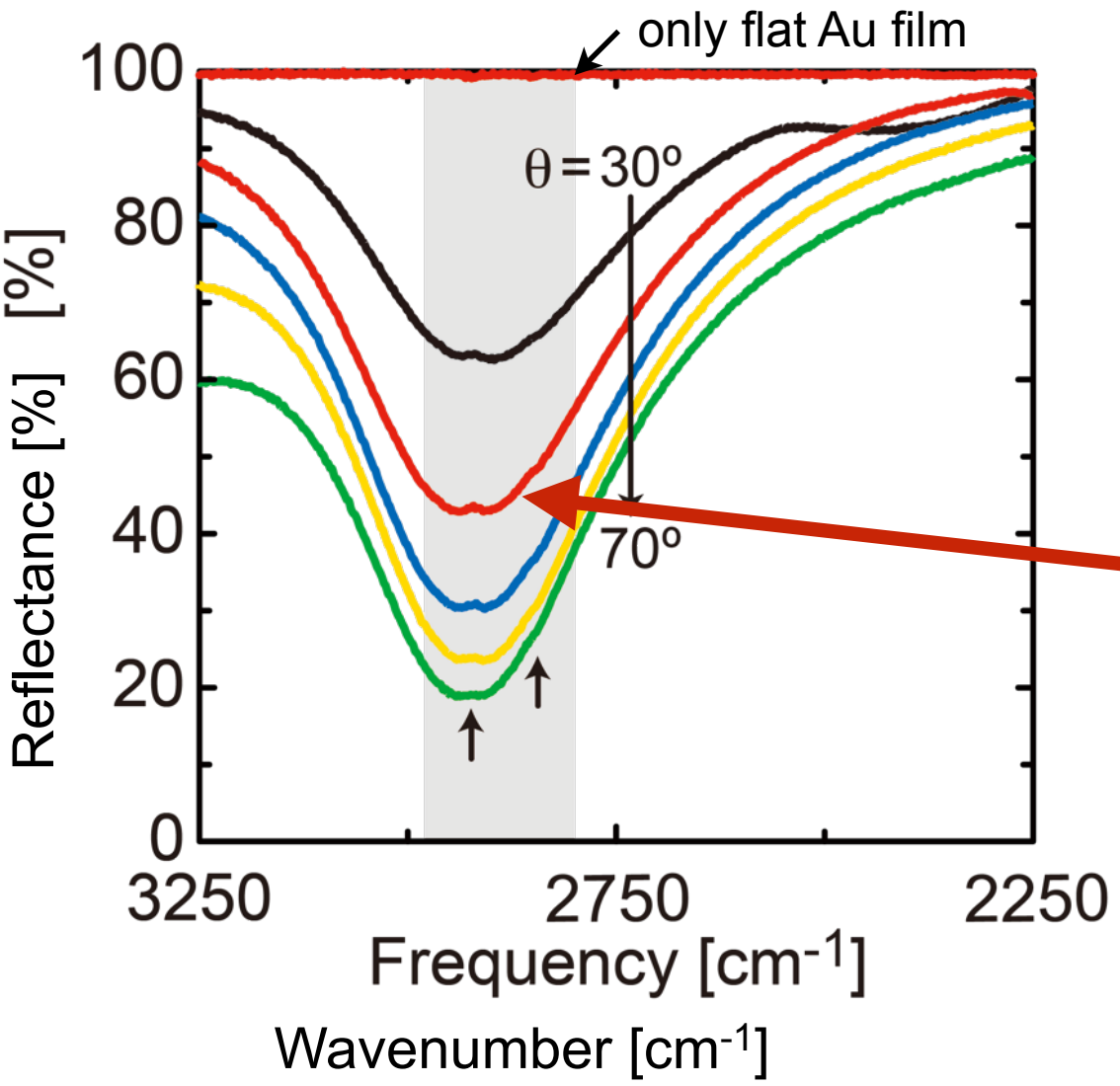


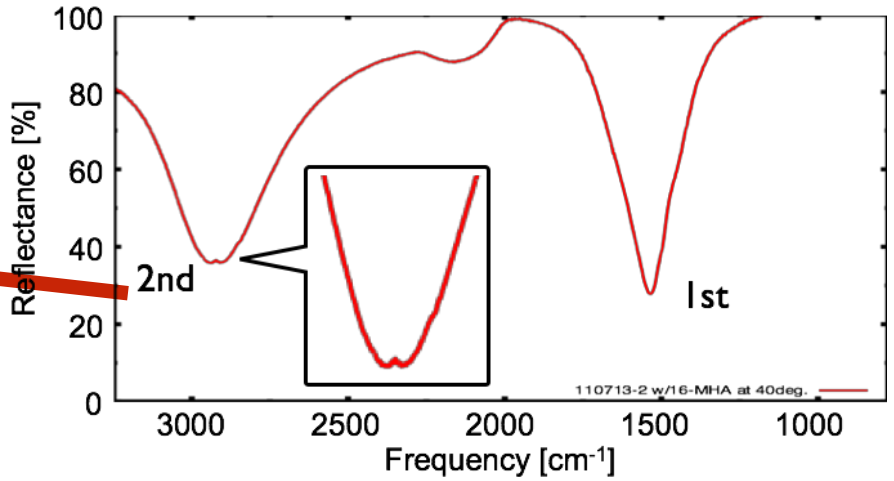




Sci. Rep. **5**, 12570 (2015).

Absorption Properties



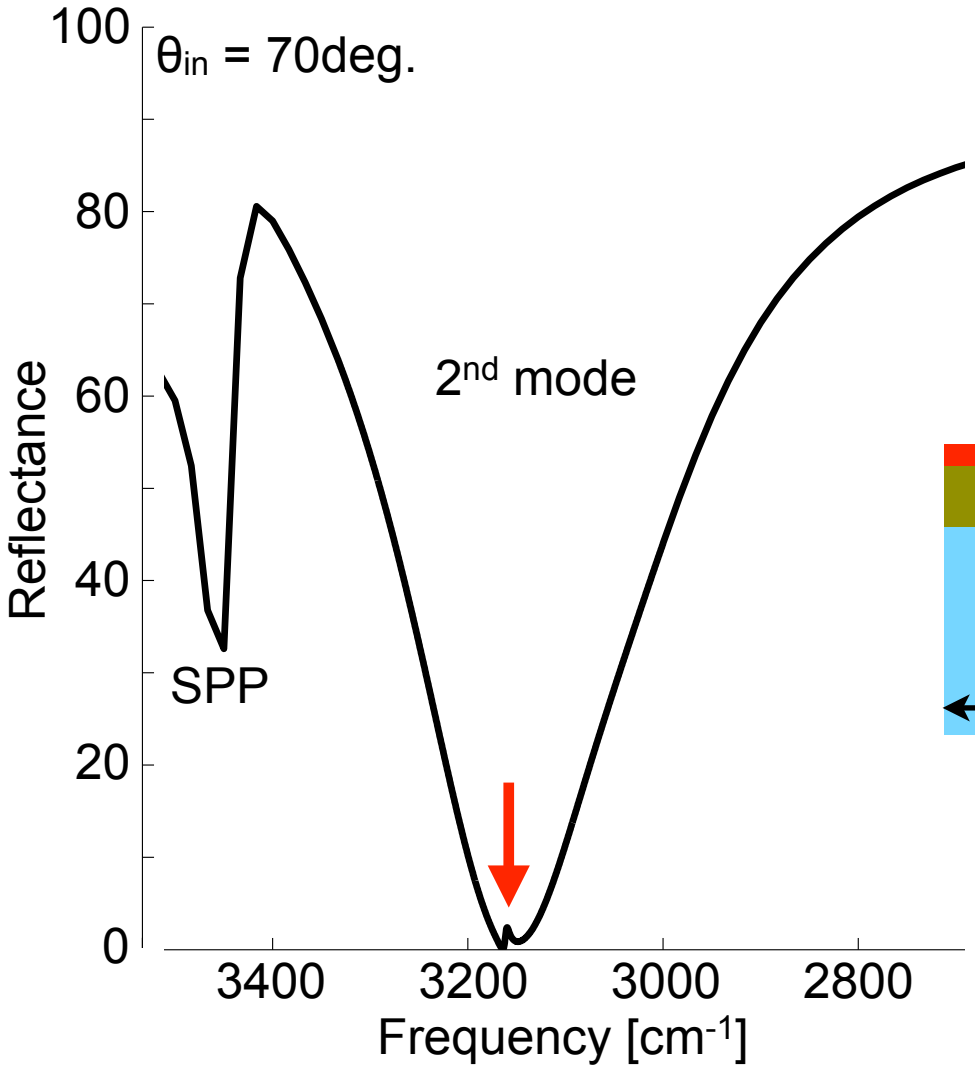


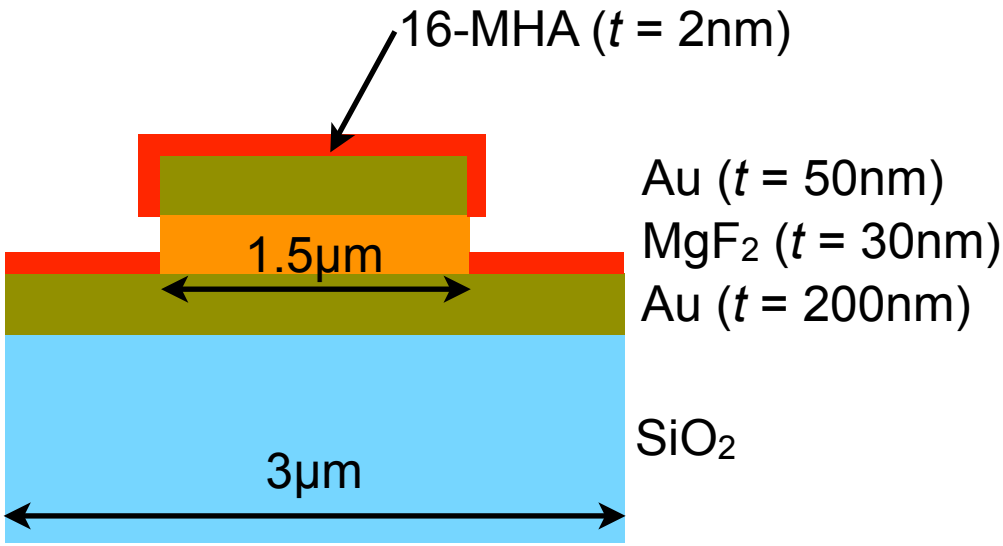
Sensitivity = 1.8atto(10^{-18}) mol in beam spot

Numerical calculation

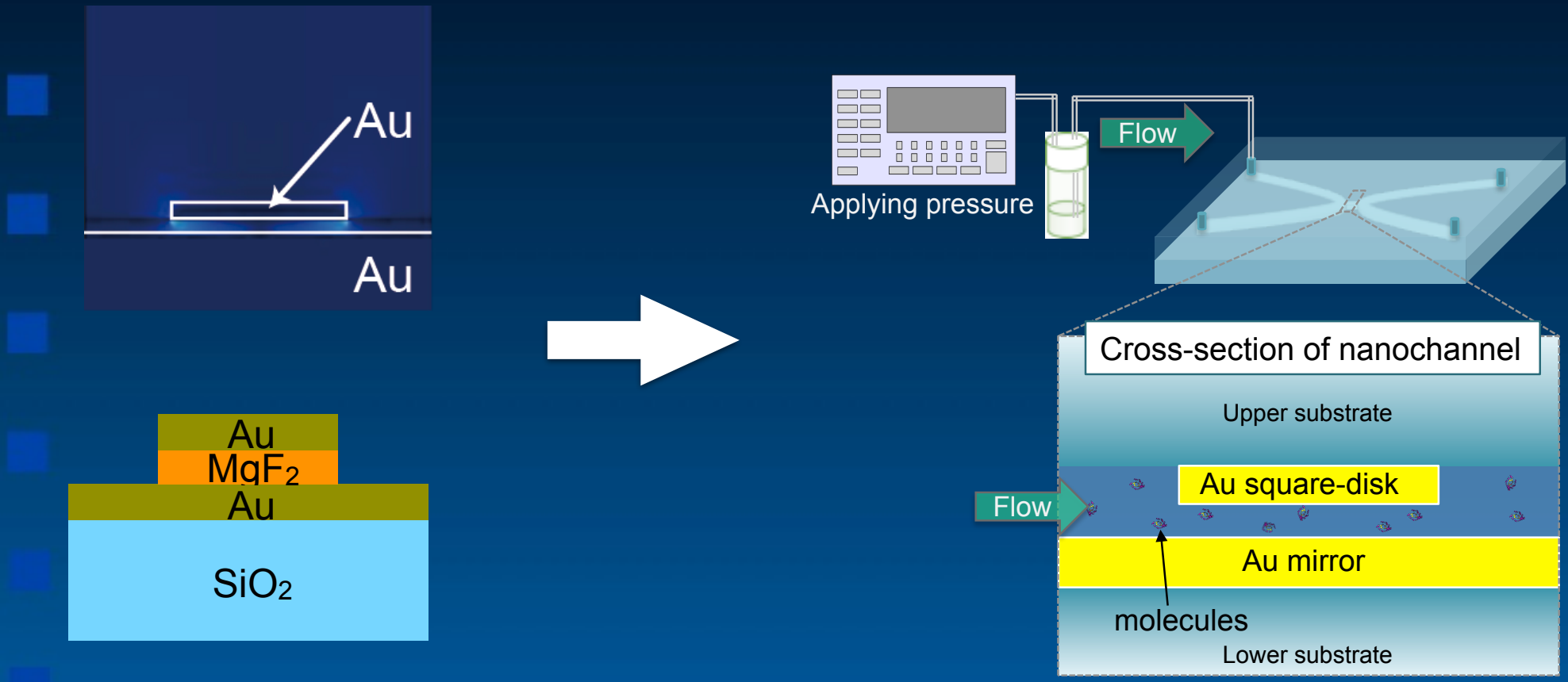
16-MHA modeled using Lorentz dielectric function: $\varepsilon(\omega) = \varepsilon_s \left(1 + \frac{\omega_p^2}{\omega_0^2 - \omega^2 + i2\gamma\omega}\right)$

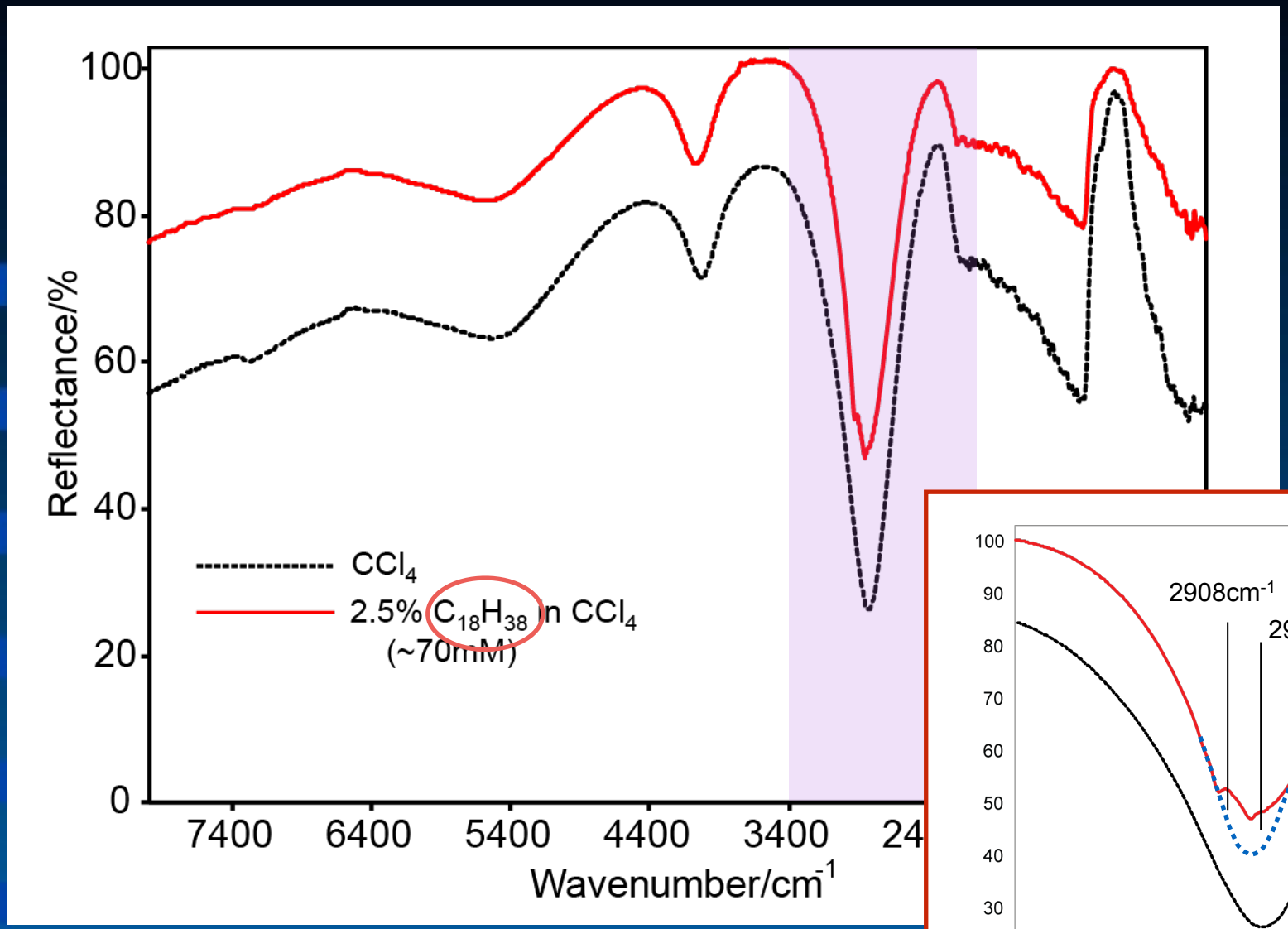
where ε_s = 1.0, ω_p = 300THz, ω_0 = 95THz (3166cm^{-1}), γ = 0.1THz

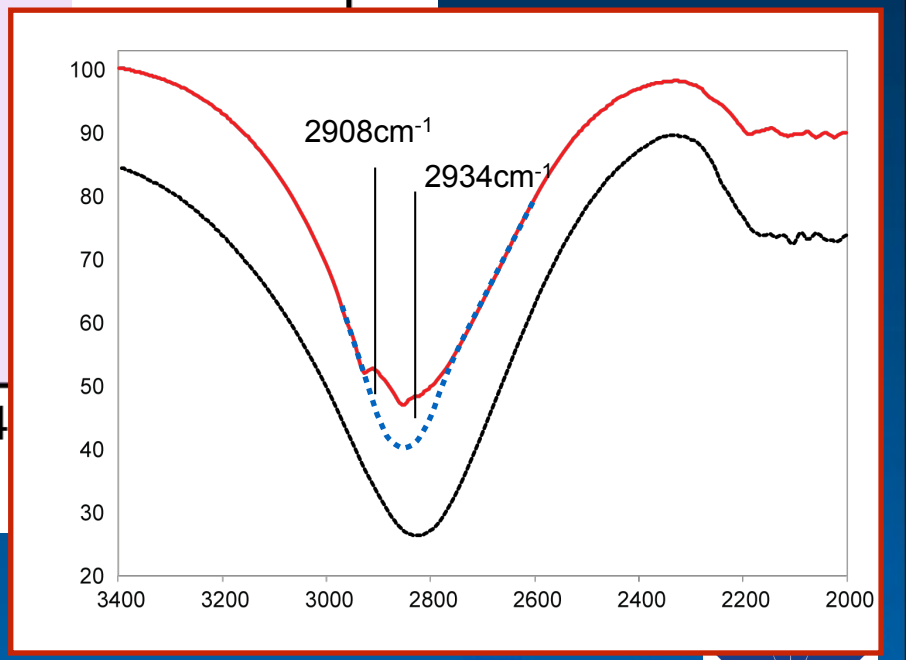


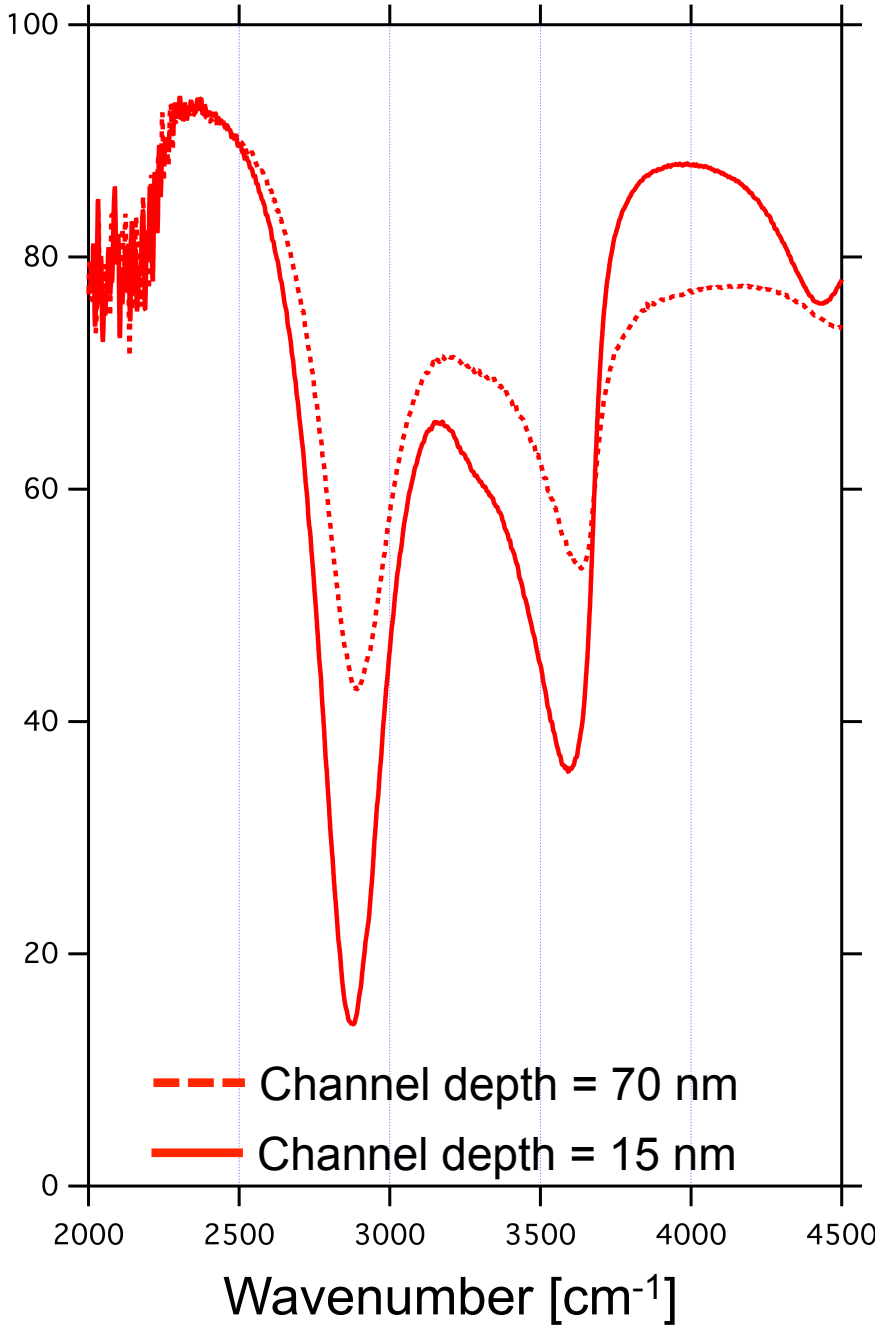


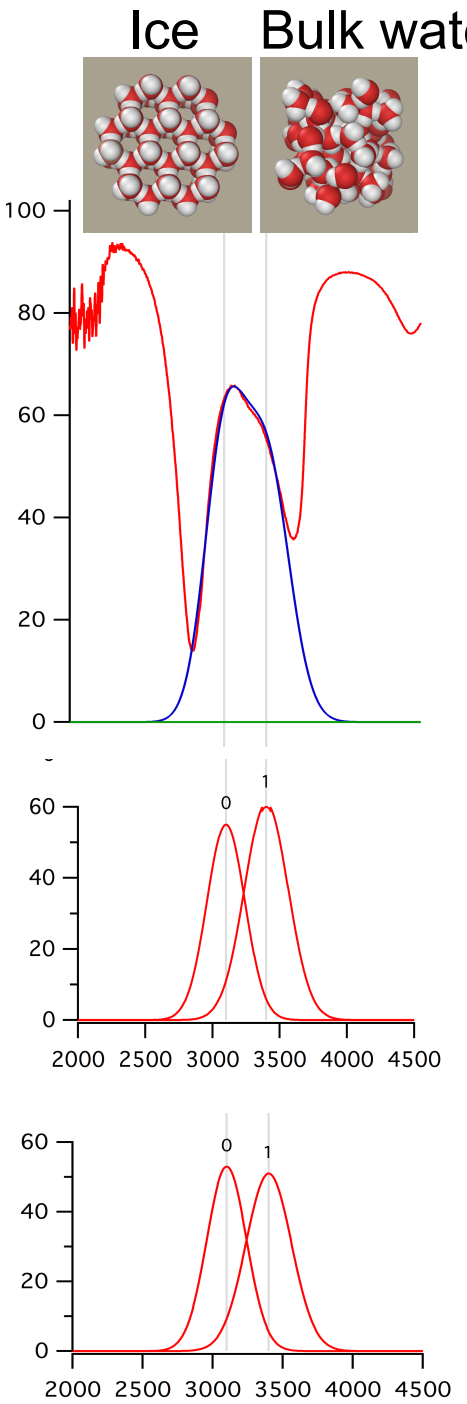
Nano fluidic device with metamaterial absorber




100
80
60
40
20
0
Reflectance/%
7400
6400
5400
4400
3400
Wavenumber/cm-1
CCl4
2.5% C18H38 in CCl4
(~70mM)


100
90
80
70
60
50
40
30
20
3400
3200
3000
2800
2600
2400
2200
2000
2908cm-1
2934cm-1

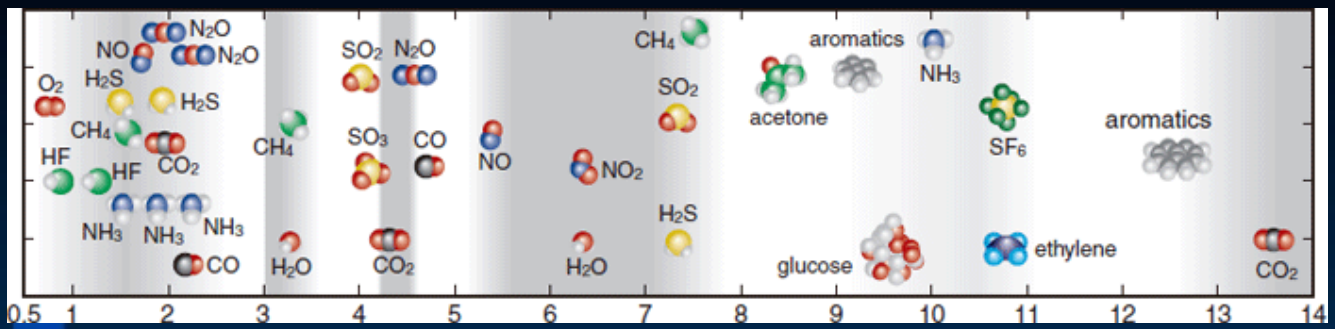

100
80
60
40
20
0
2000
2500
3000
3500
4000
4500
Wavenumber [cm-1]
Channel depth = 70 nm
Channel depth = 15 nm


Ice
Bulk water
100
80
60
40
20
0
0
1
2000 2500 3000 3500 4000 4500

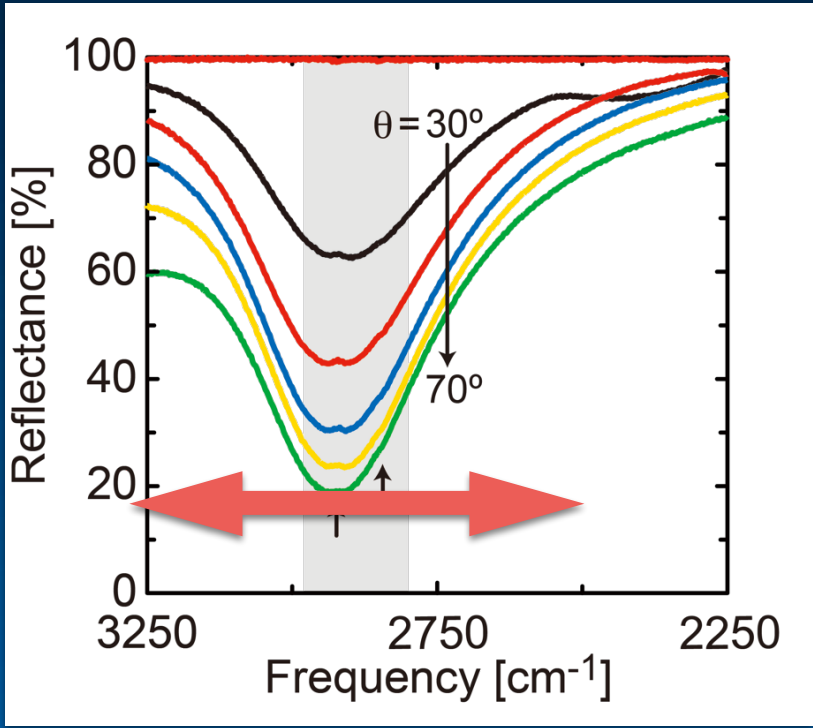
d=70nm

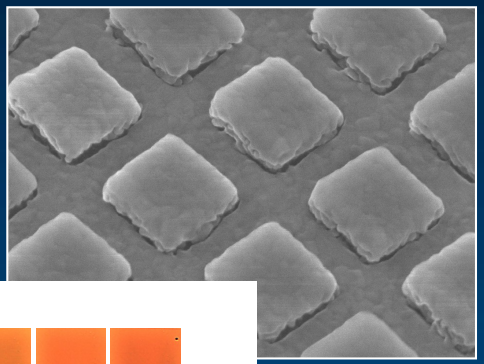
d=15nm

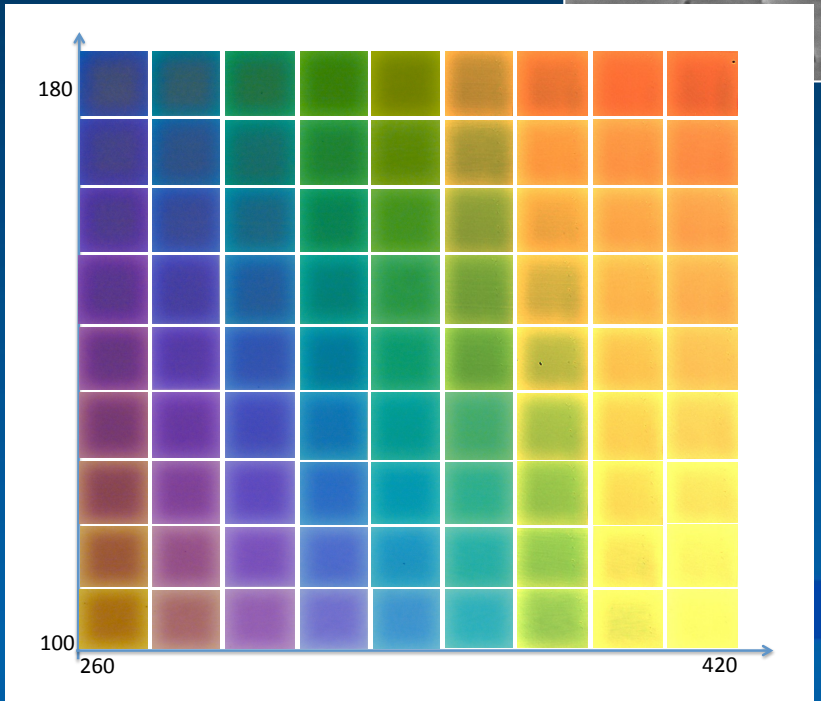
Absorption engineering by metal nano structures



Original Data By aluminum structure



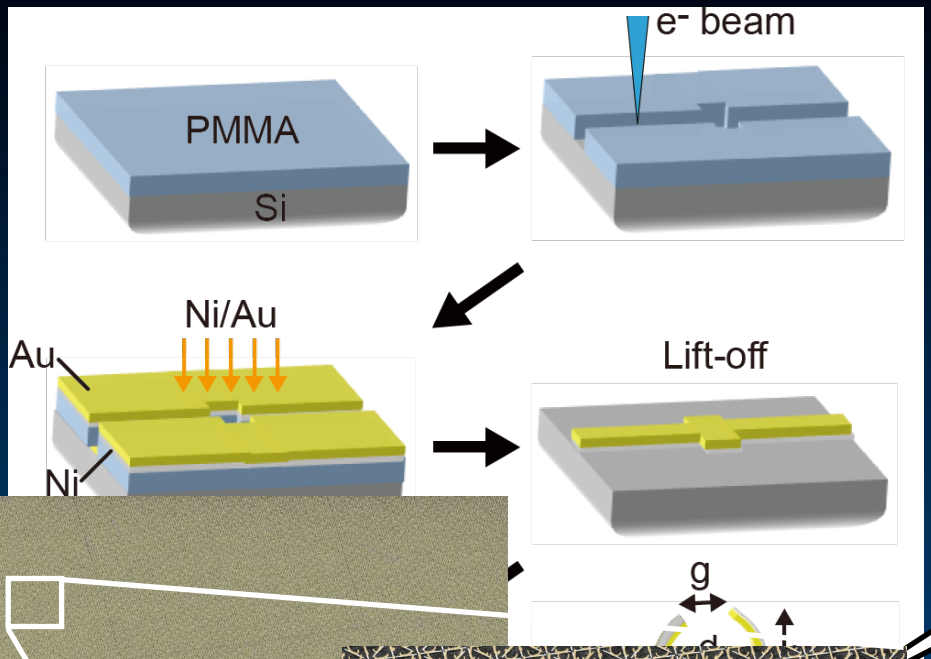


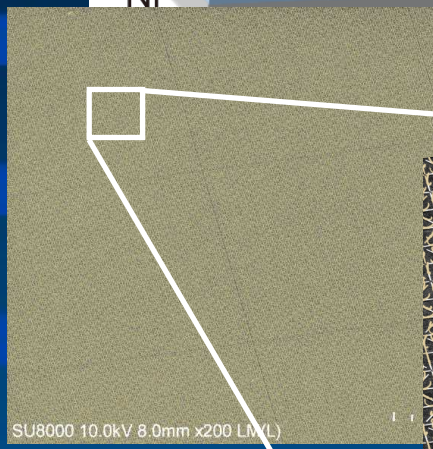


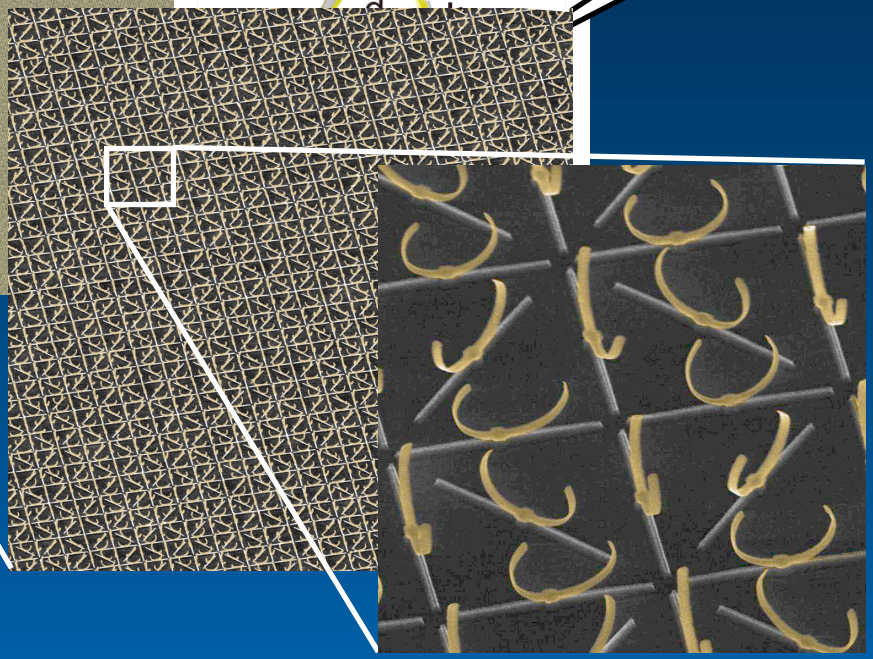
Outline

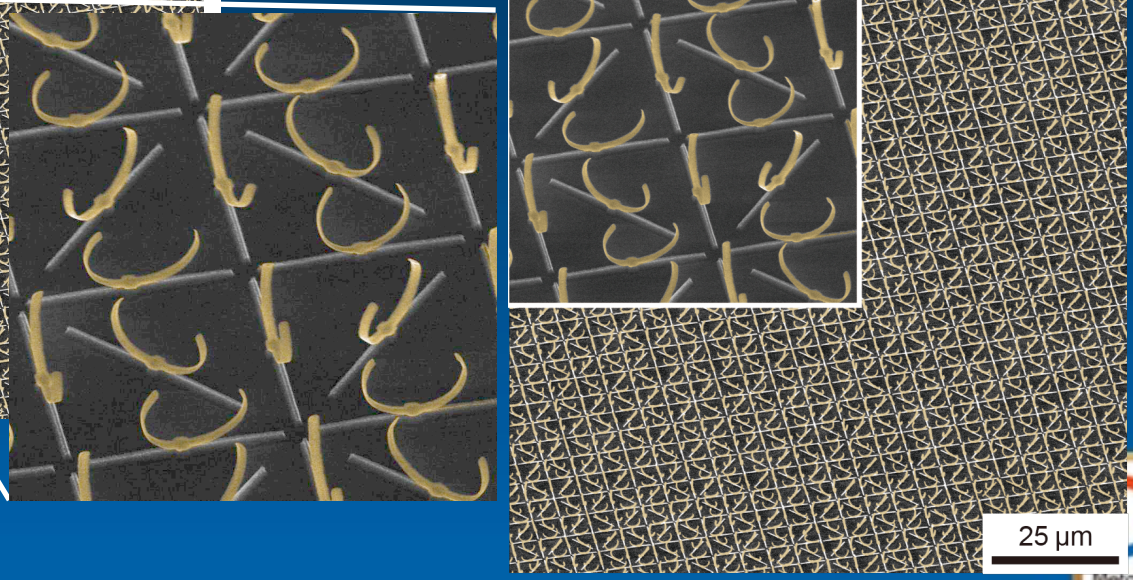
1. Non-radiative surface plasmon (Dark plasmon)
2. Dark plasmonic metamaterials for absorber
3. Application of dark metamaterials for molecular sensing
4. Fabrication technique for 3D Metamterials

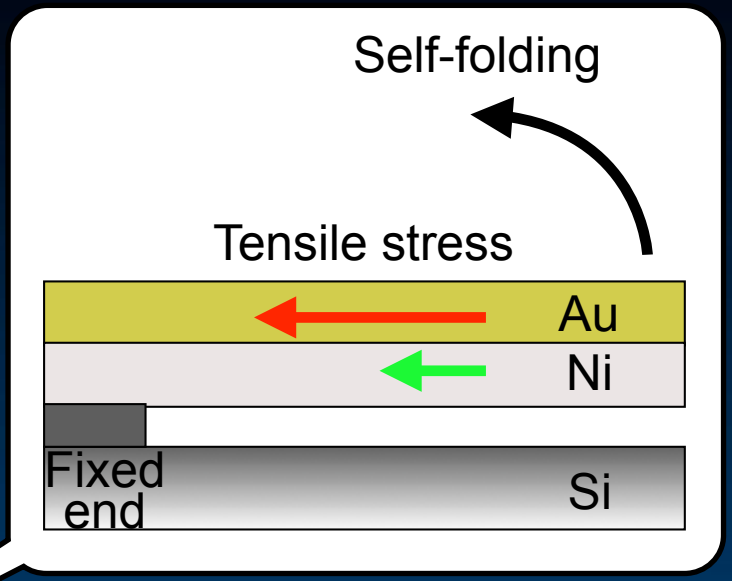
Self-folding of Bi-layer Metallic Structures





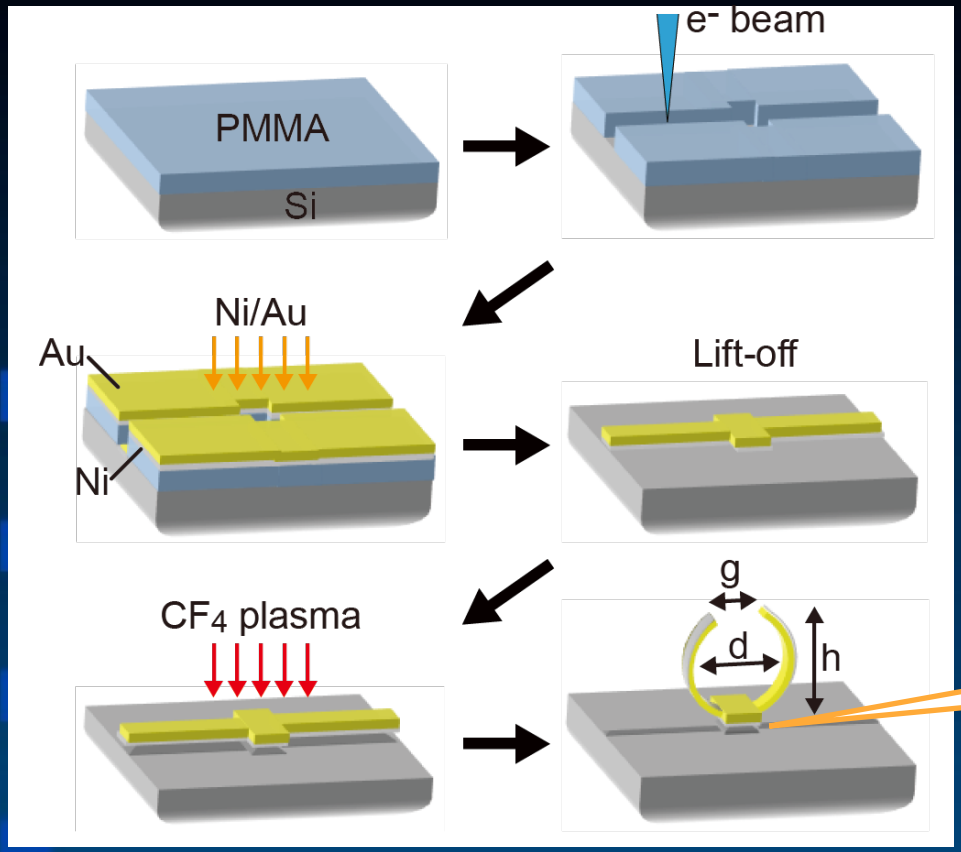


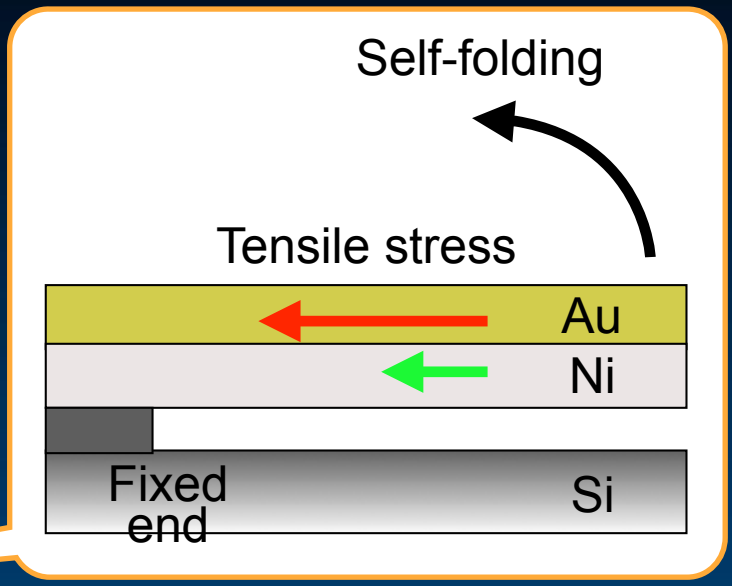




Self-folding of Bi-layer Metallic Structures

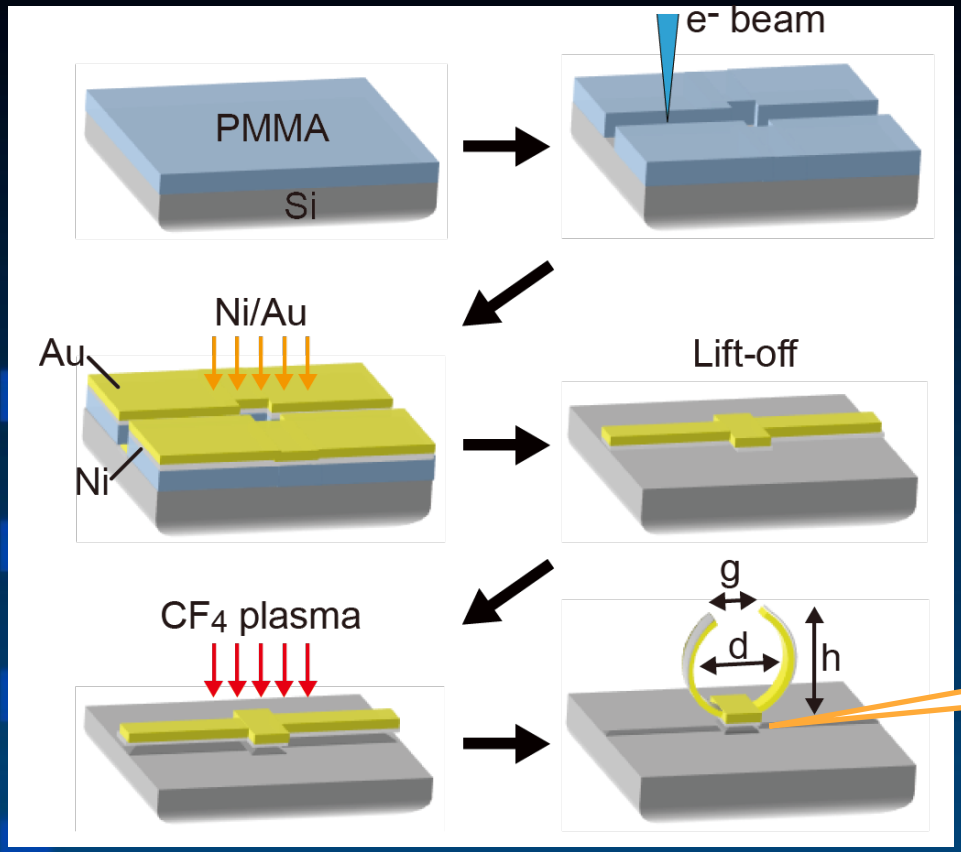
Adv. Opt. Mater 2014
nature "Research Highlights" **515**

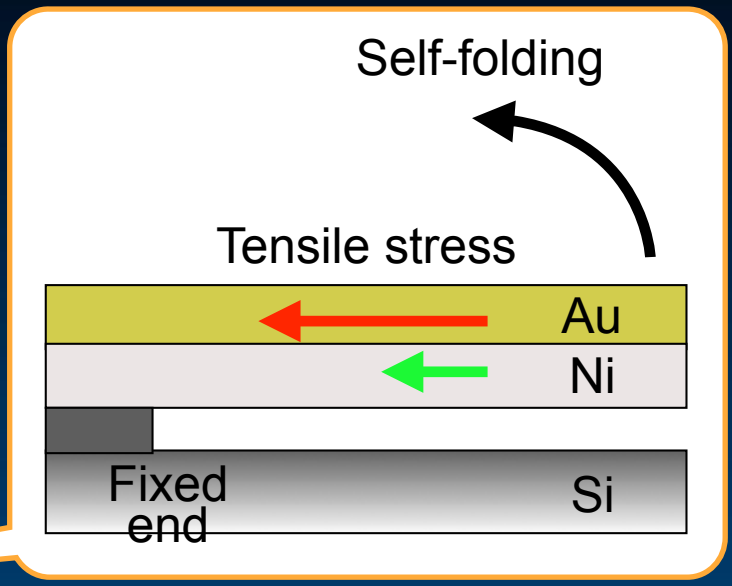


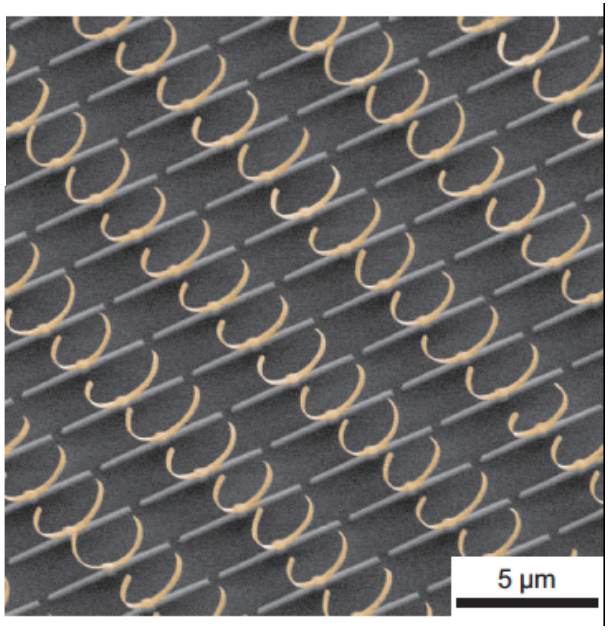


Self-folding of Bi-layer Metallic Structures

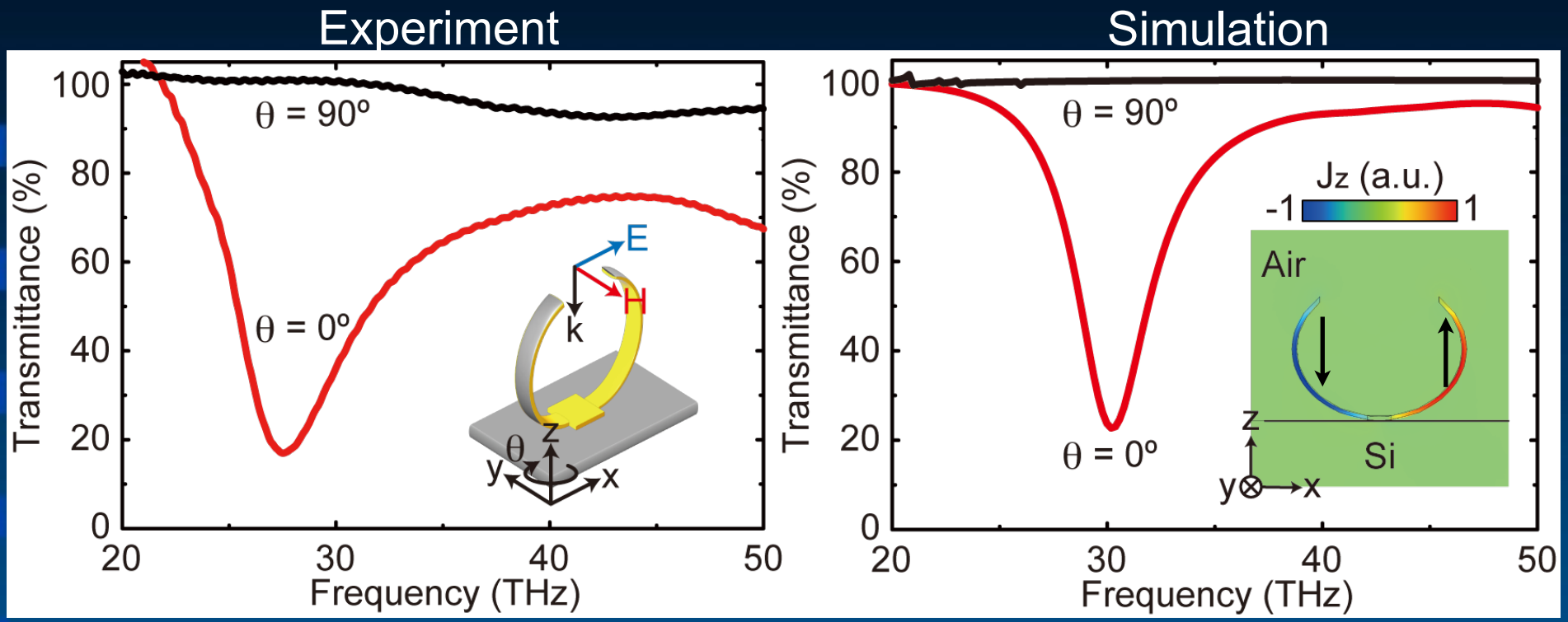
Adv. Opt. Mater 2014
nature "Research Highlights" **515**







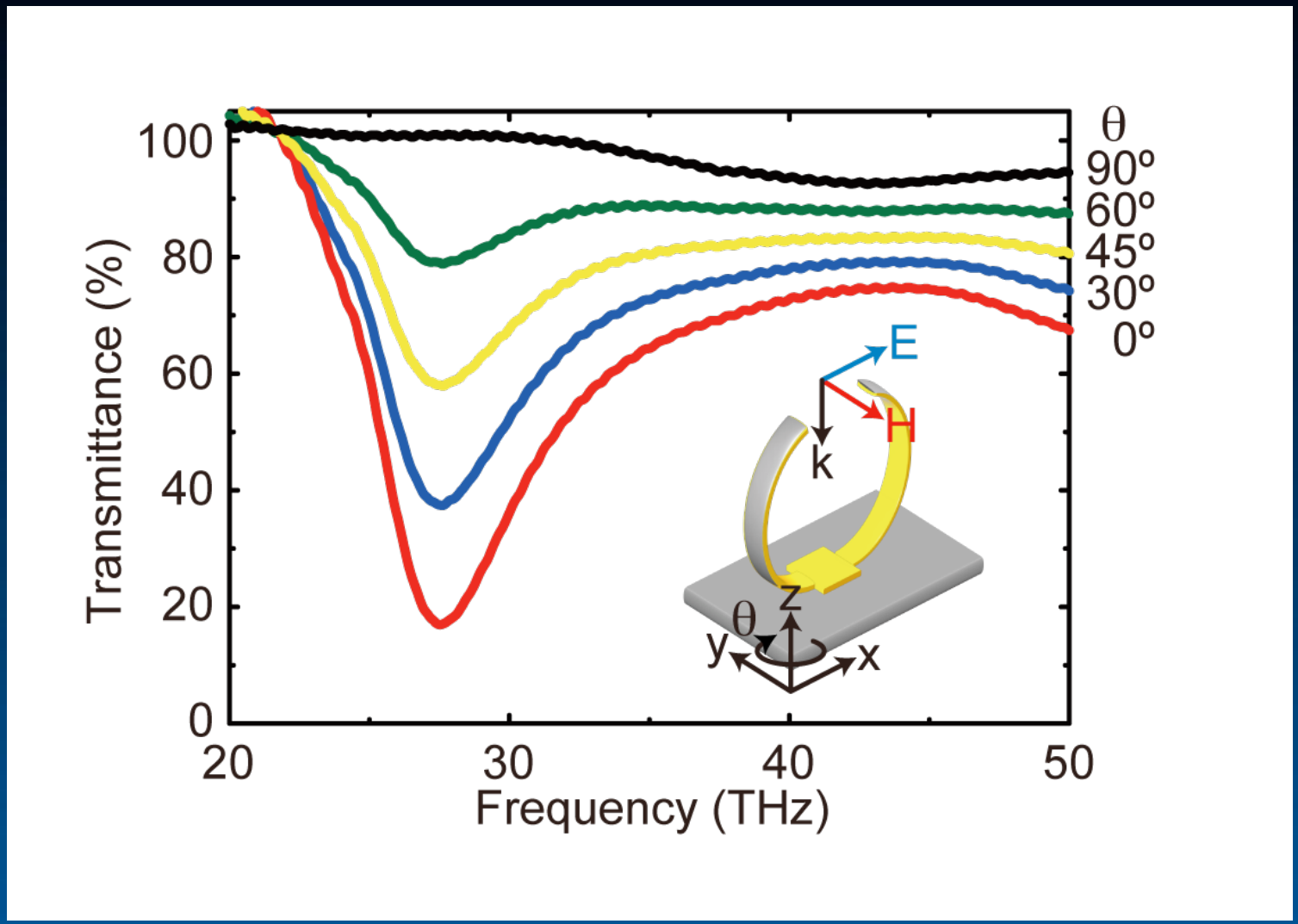
Optical Characterization



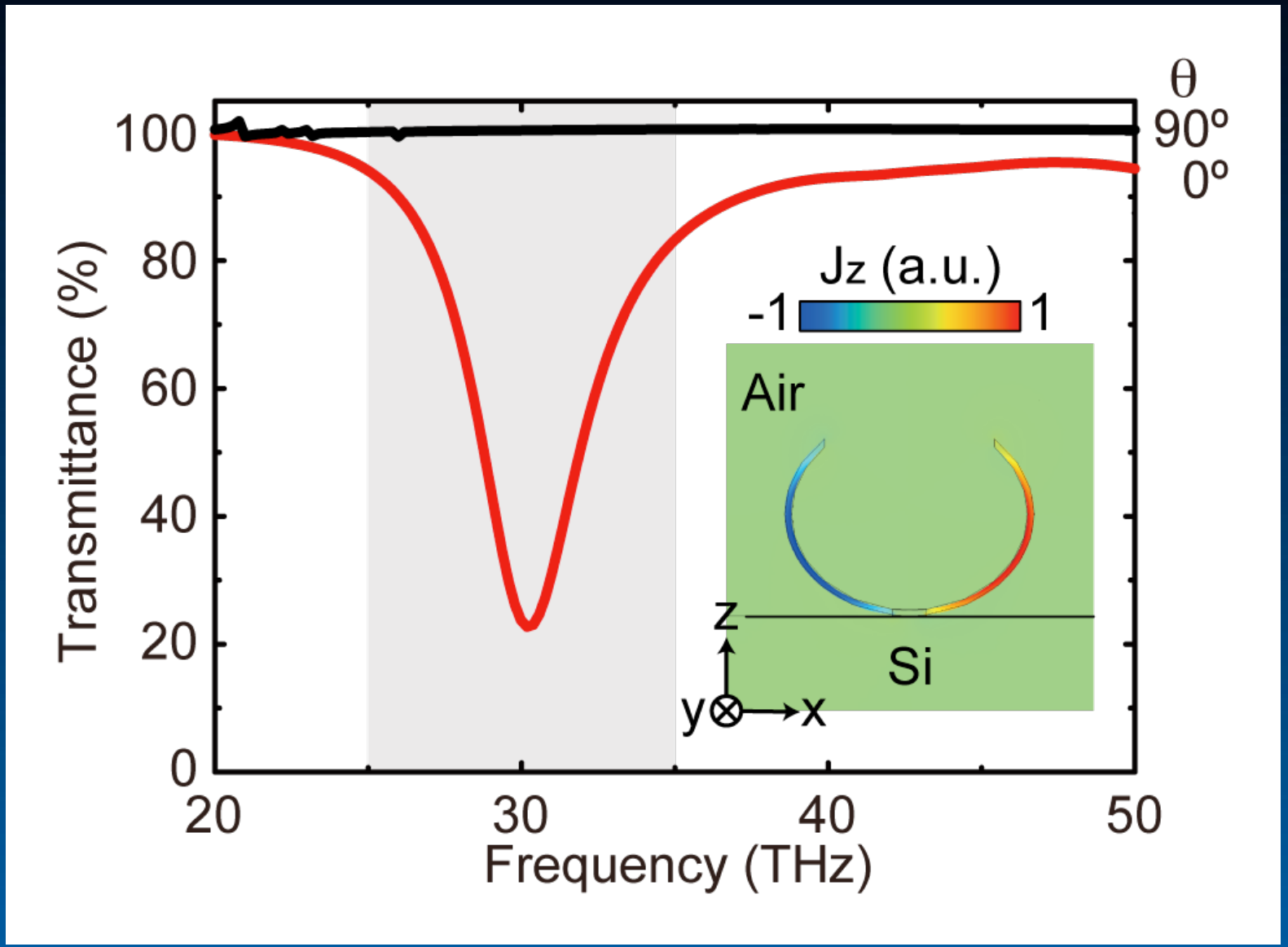
asymmetric mode

- Magnetic (LC) resonance at f = 30 THz (λ = 10 μm)
- Excellent agreement b/t the experiment & simulation

SRRアレイの異方性


Transmittance (%)
100
80
60
40
20
0
20
30
40
50
Frequency (THz)
θ
90°
60°
45°
30°
0°
E
H
k
z
θ
y
x

単一共振器の異方性

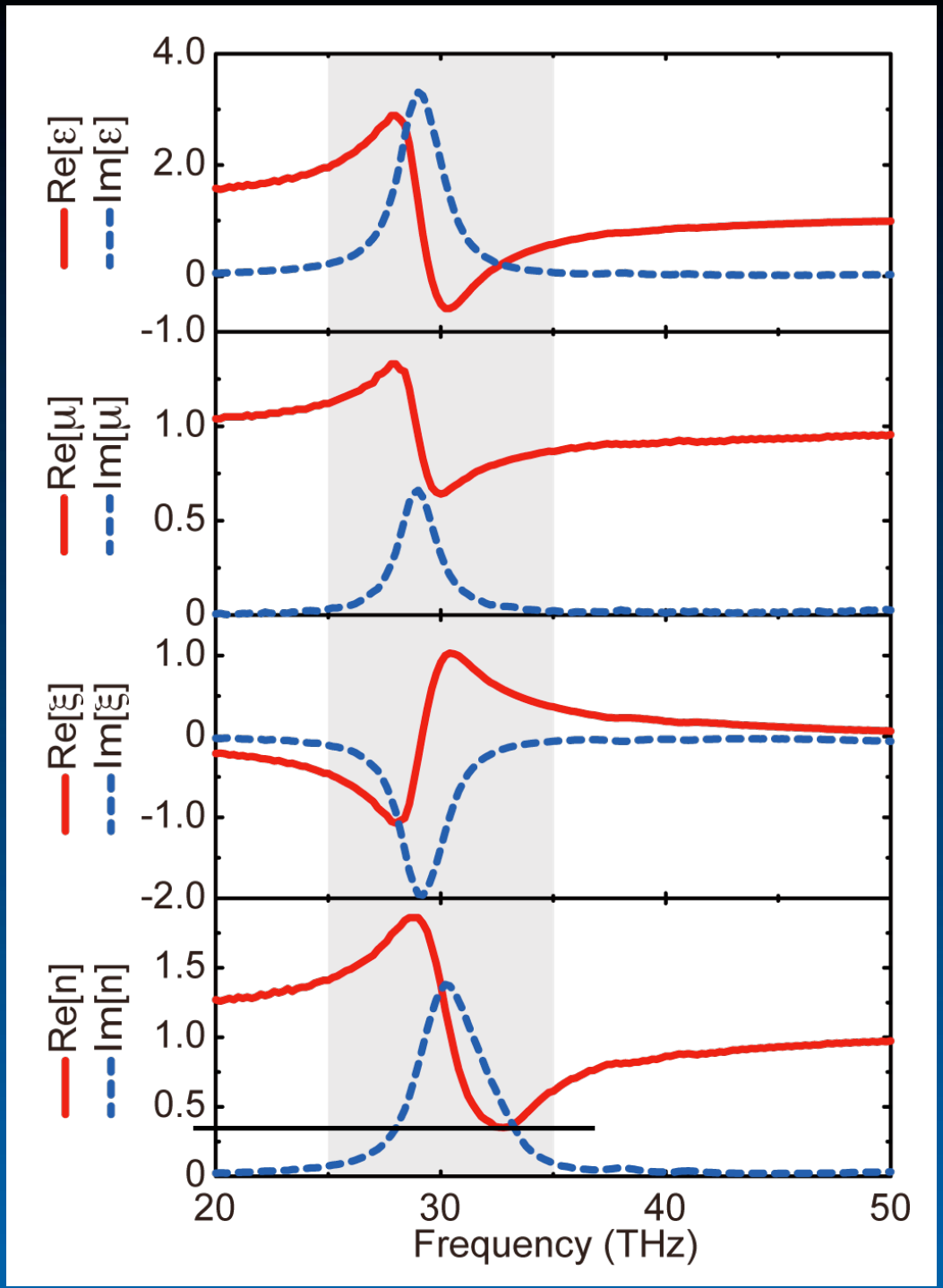

θ
90°
0°
100
80
60
40
20
0
Transmittance (%)
20
30
40
50
Frequency (THz)
Jz (a.u.)
-1
1
Air
Si
z
y
x

ε

μ

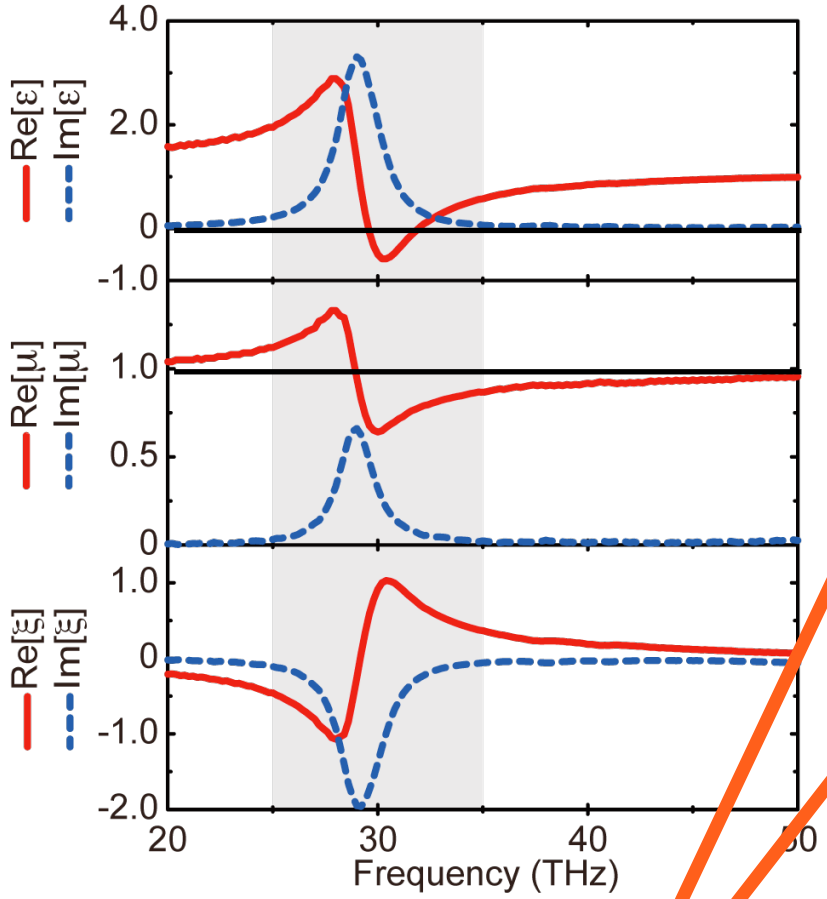
ξ

n

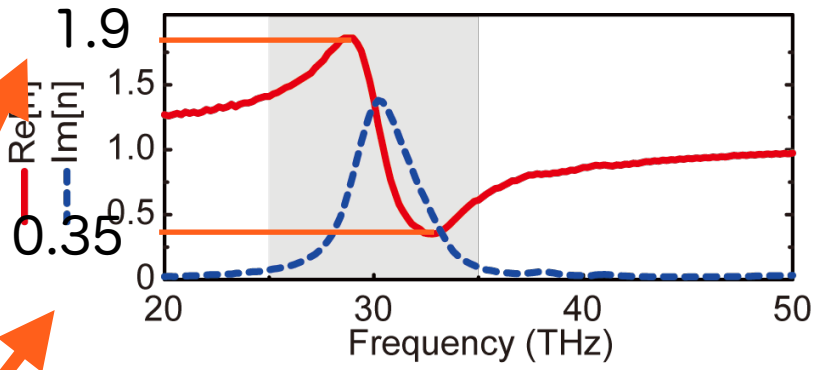

Re[ε]
Im[ε]
Re[μ]
Im[μ]
Re[ξ]
Im[ξ]
Re[n]
Im[n]
4.0
2.0
0
-1.0
1.0
0.5
0
1.0
0
-1.0
-2.0
1.5
1.0
0.5
0
20
30
40
50
Frequency (THz)

n~0.3

Retrieved Effective Optical Parameters



$$n^2 = \epsilon\mu - \xi^2$$



- Lorentz-like responses at f = 30 THz
- n largely swings from 0.35 to 1.9

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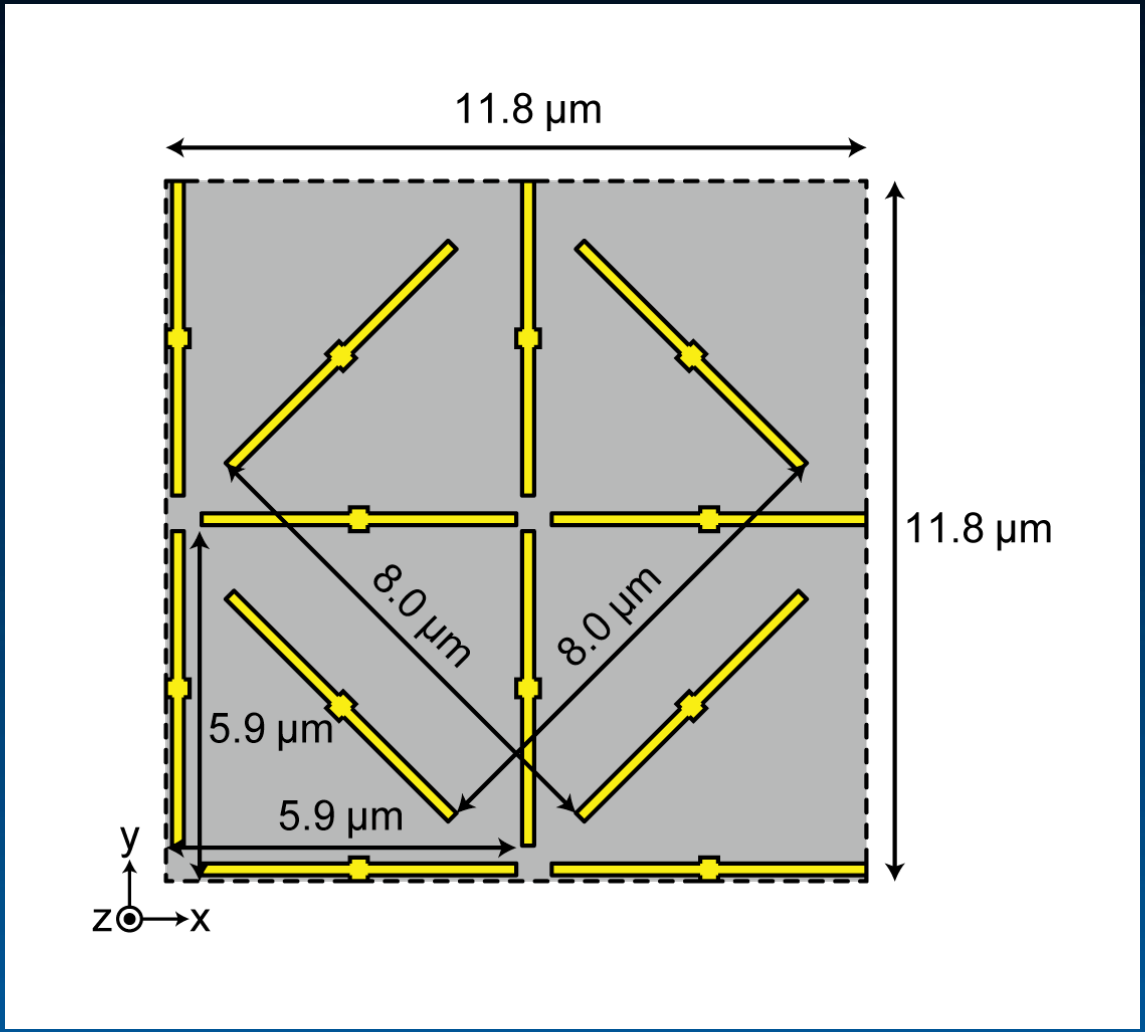
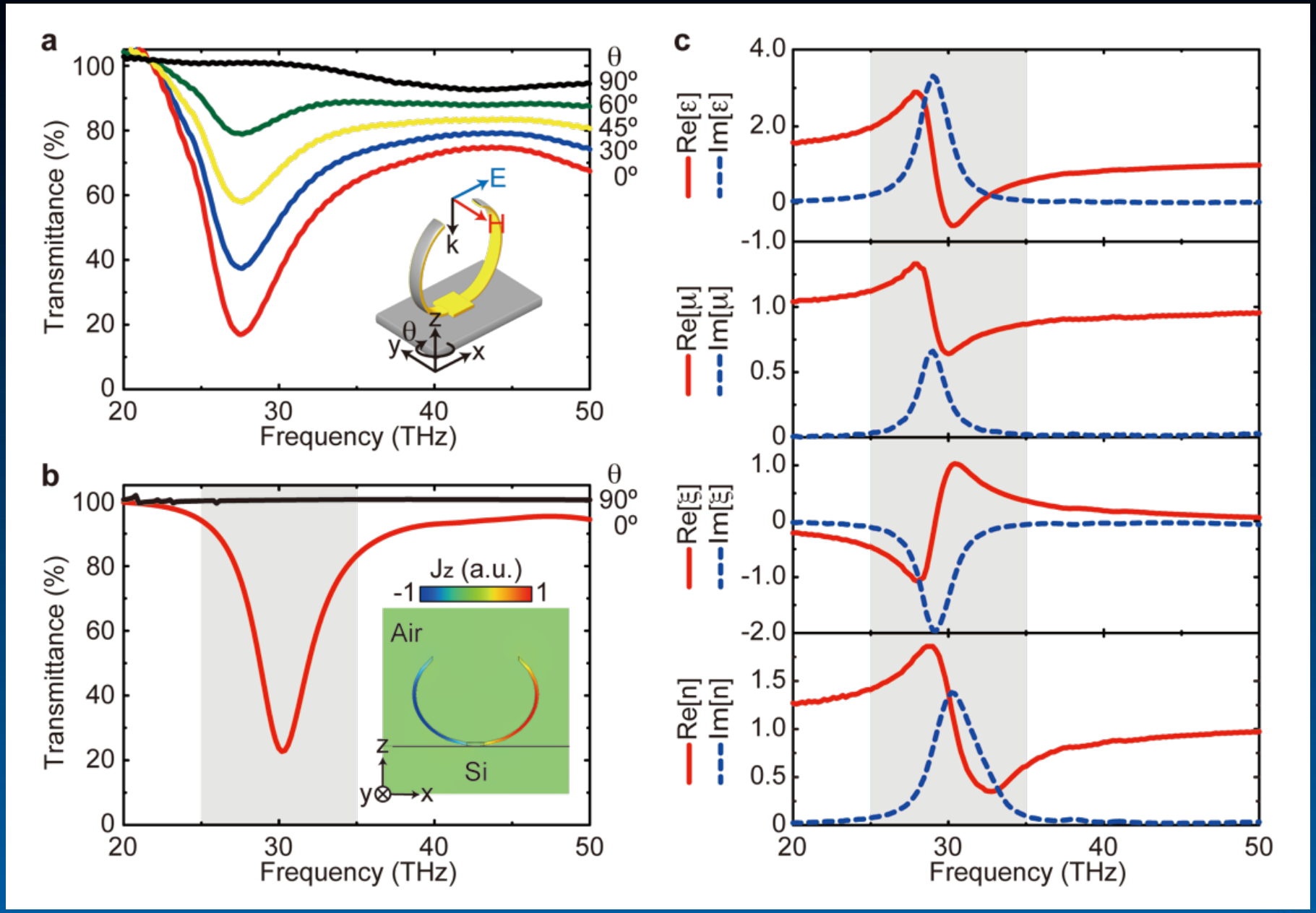
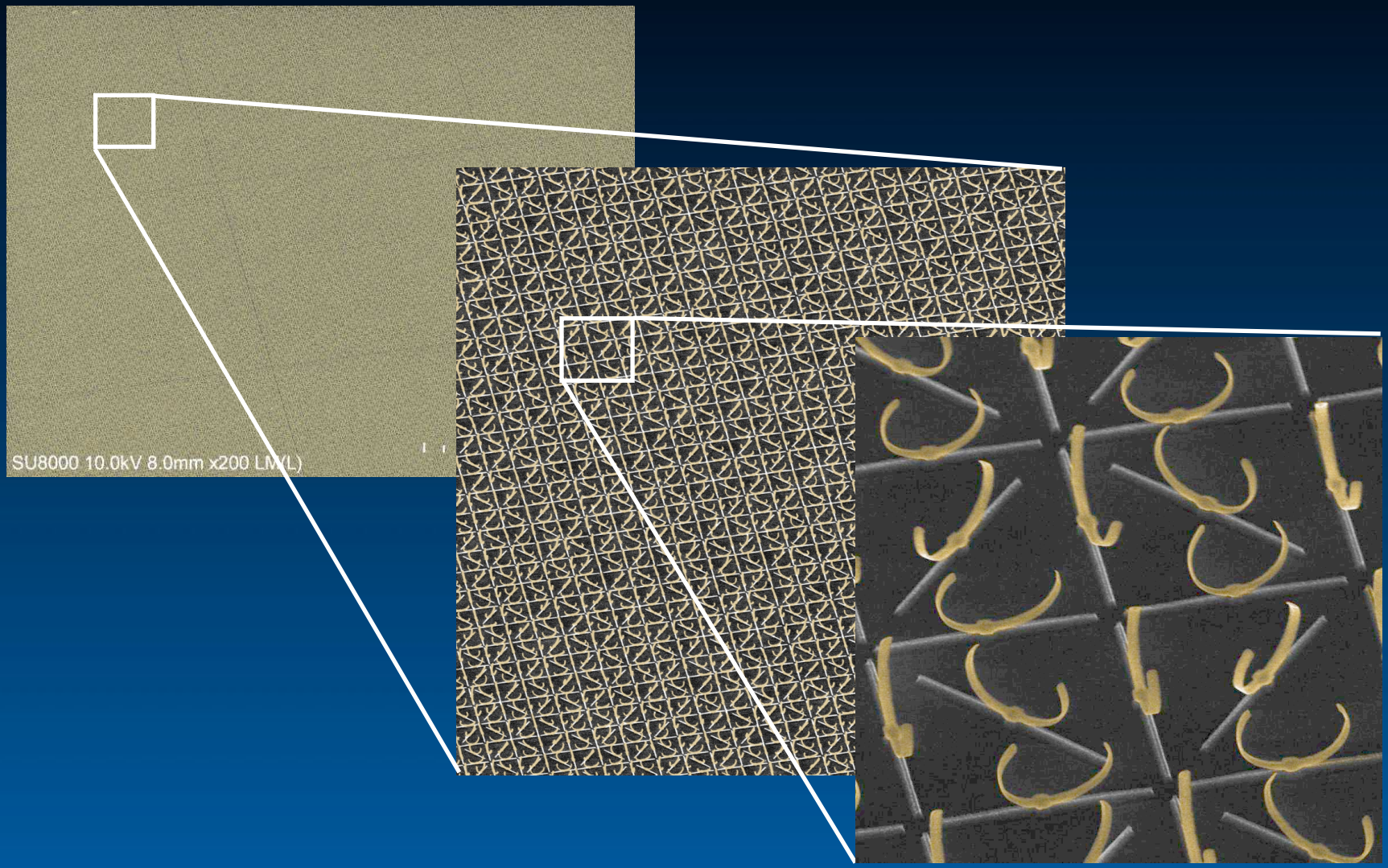
Metamaterials

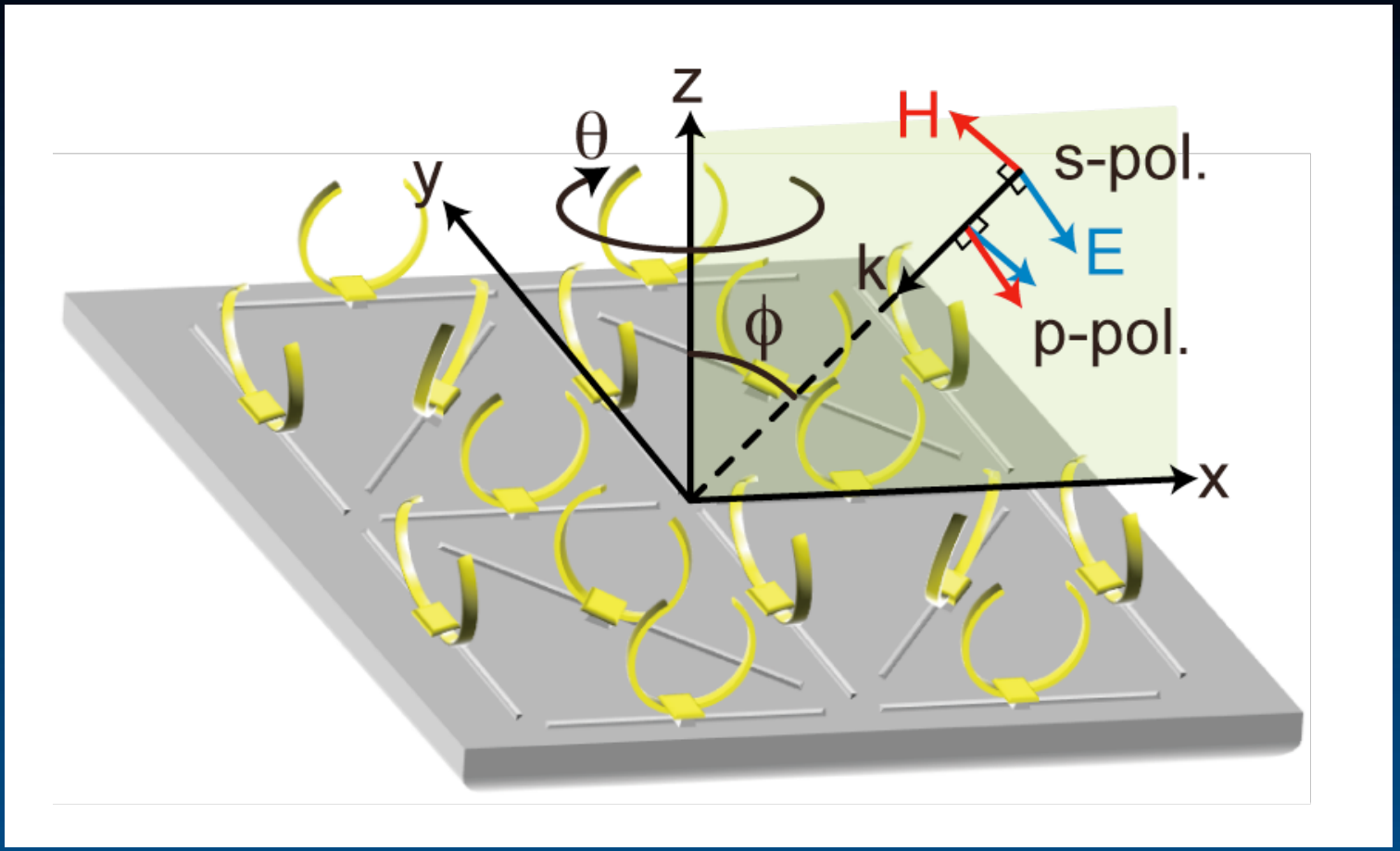

11.8 μm
11.8 μm
8.0 μm
8.0 μm
5.9 μm
5.9 μm
y
z
x


a
Transmittance (%)
Frequency (THz)
θ
90°
60°
45°
30°
0°
E
H
k
z
θ
y
x
b
Transmittance (%)
Frequency (THz)
θ
90°
0°
Jz (a.u.)
-1
1
Air
Si
z
y
x
c
Re[ε]
Im[ε]
Re[μ]
Im[μ]
Re[ξ]
Im[ξ]
Re[η]
Im[η]
Frequency (THz)

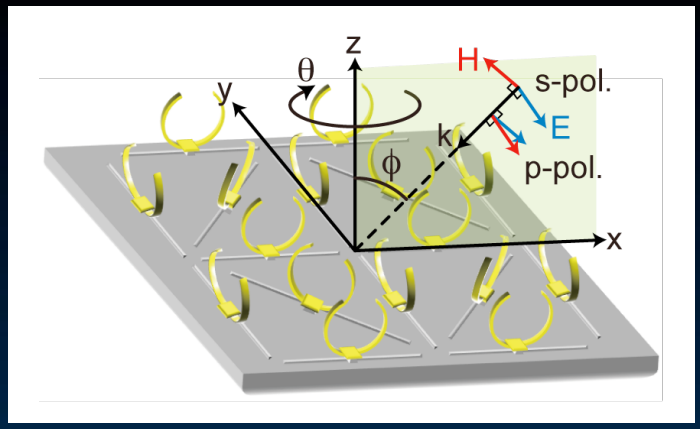
Isotropic metamaterial by stress tension assisted EB litho.

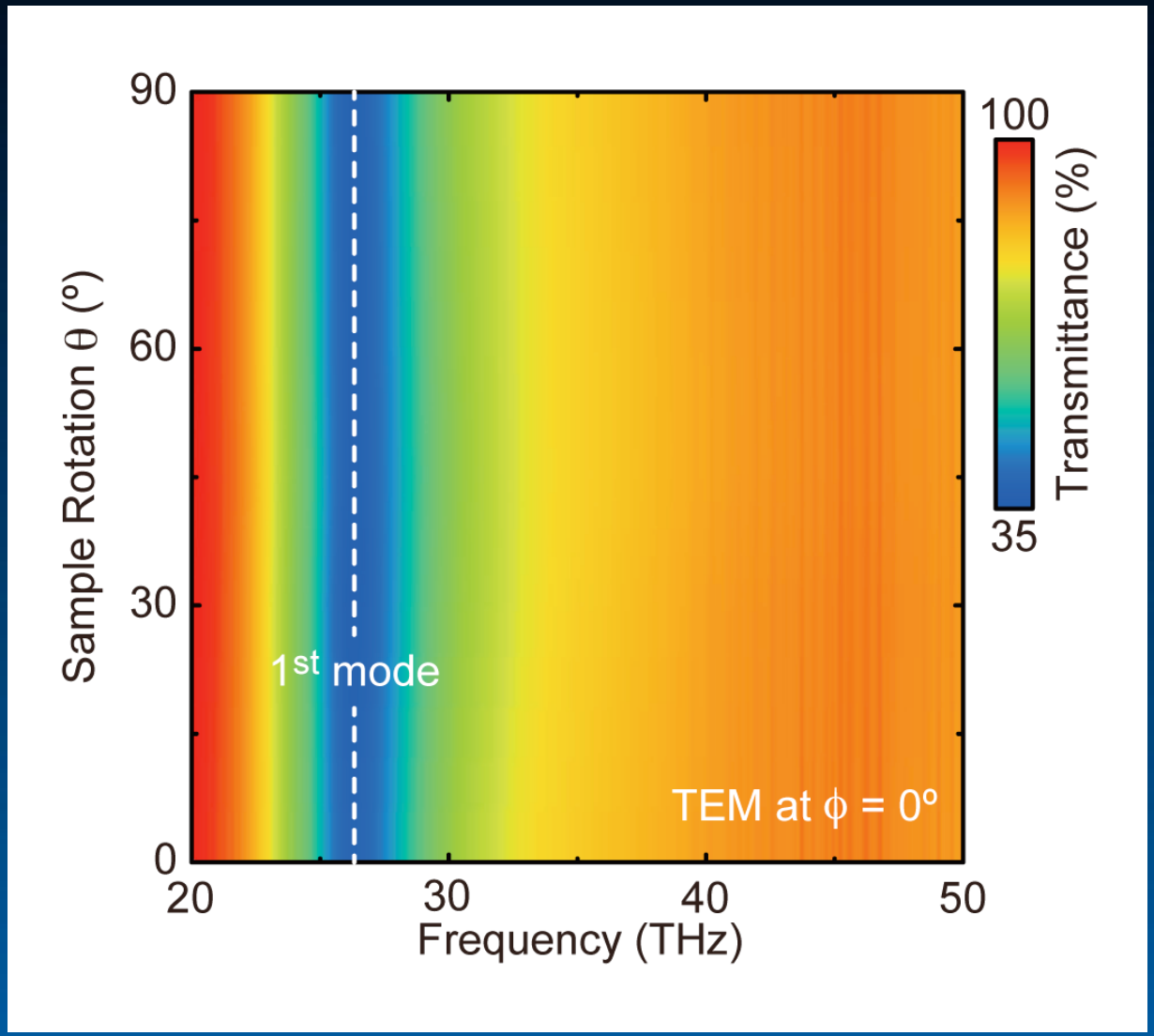
an example of combination of top-down and bottom-up



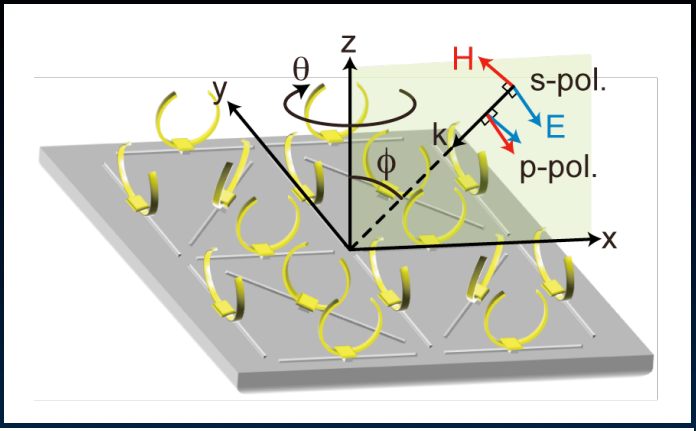

z
θ
y
H
s-pol.
E
k
ϕ
p-pol.
x

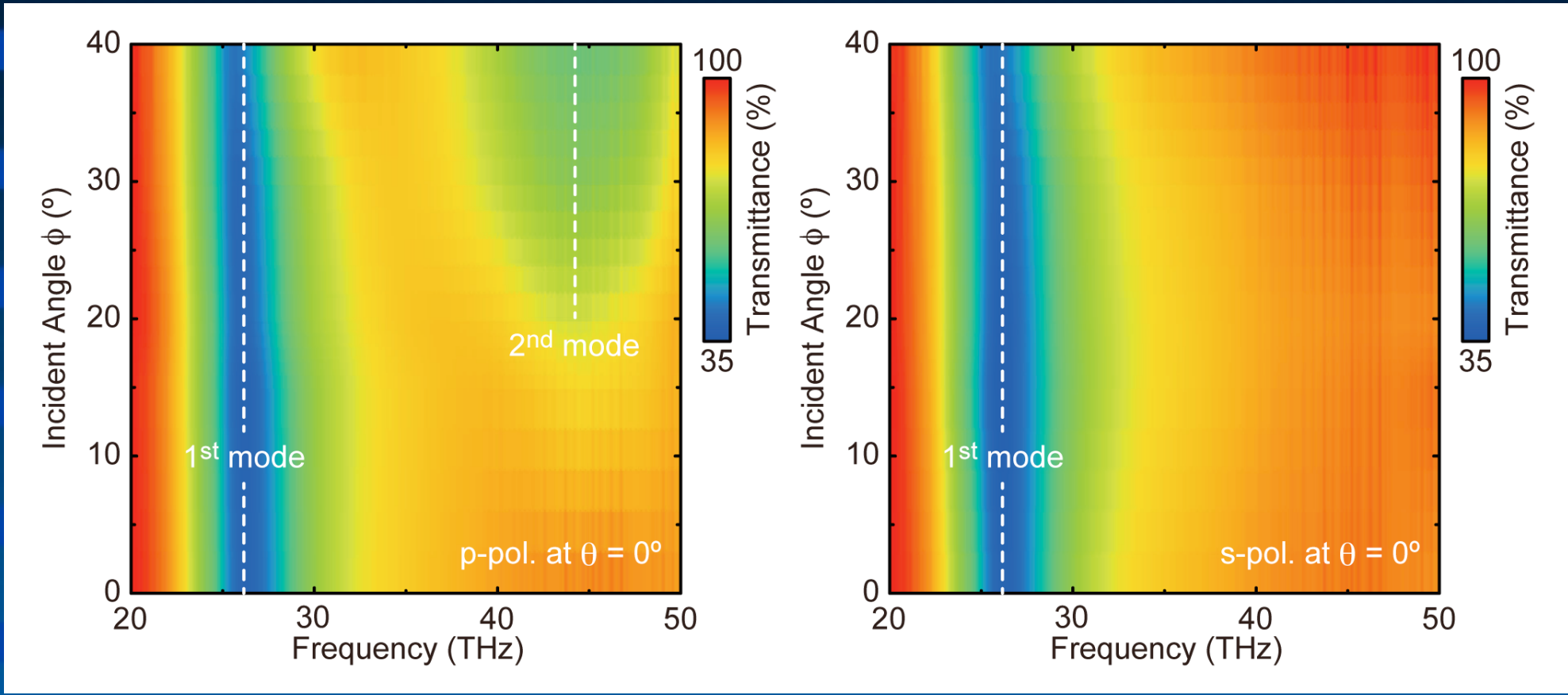
垂直入射，θ依存性


z
θ
y
H
s-pol.
E
k
ϕ
p-pol.
x


Sample Rotation θ (°)
90
60
30
0
20
30
40
50
Frequency (THz)
1st mode
TEM at ϕ = 0°
100
35
Transmittance (%)

入射(φ)角依存性


z
θ
y
H
s-pol.
E
k
p-pol.
ϕ
x


Incident Angle ϕ (°)
Frequency (THz)
Transmittance (%)
100
35
1st mode
2nd mode
p-pol. at θ = 0°
s-pol. at θ = 0°

Conclusion

1. Non-radiative surface plasmon (Dark plasmon)
2. Dark plasmonic metamaterials for absorber
3. Application of dark metamaterials for molecular sensing
4. (Fabrication technique for 3D Metamterials)