Selected Topics in Advanced Optics

Week 12 – part 1

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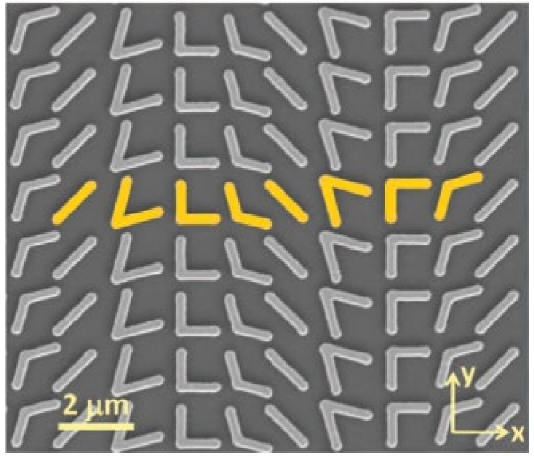
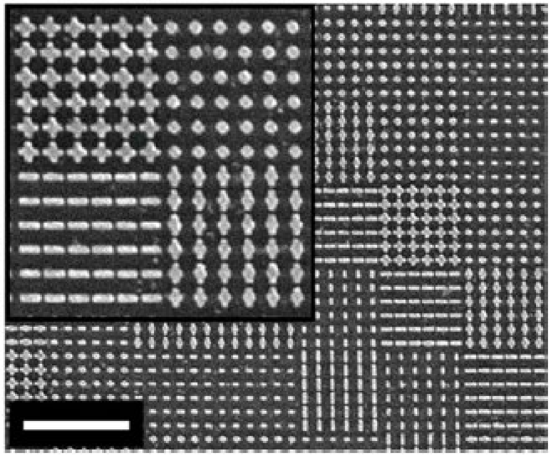
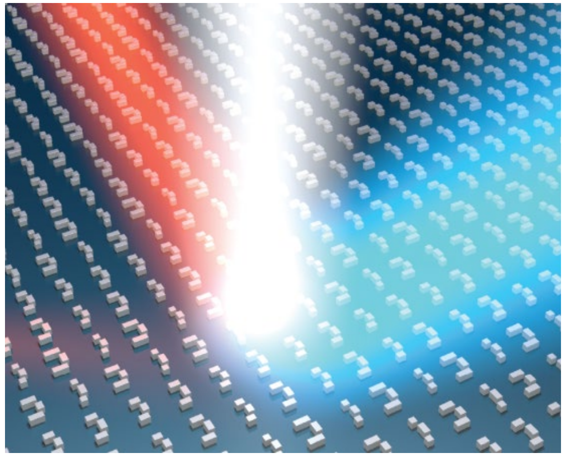
Metasurfaces

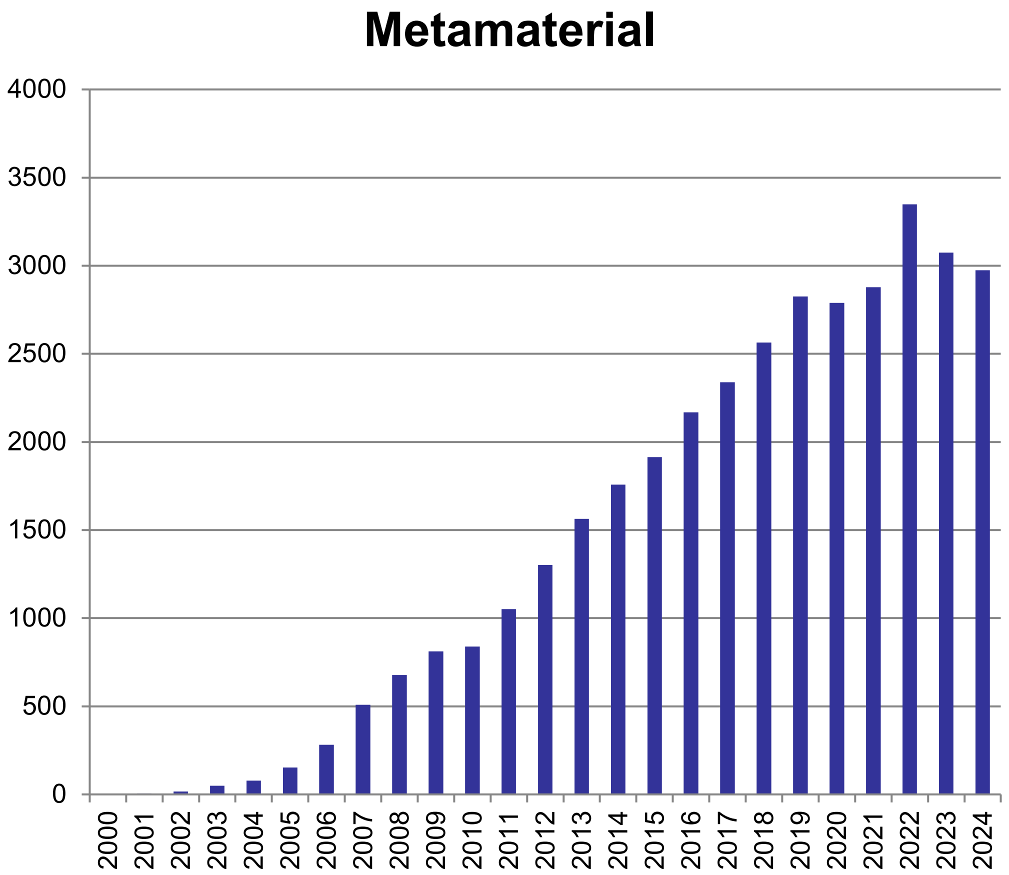
- It is quite challenging to fabricate a true 3D metamaterial, especially at optical frequencies
- A surface, structured with nanostructures, can already provide very interesting and useful features

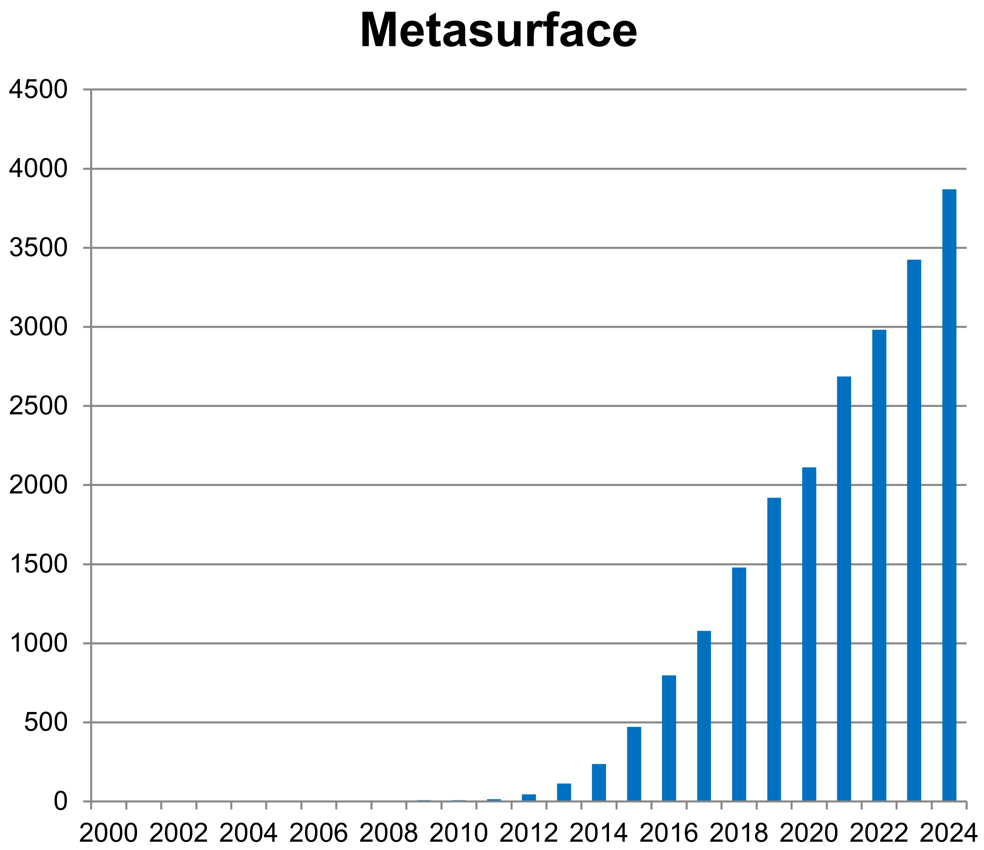






Metamaterials and metasurfaces





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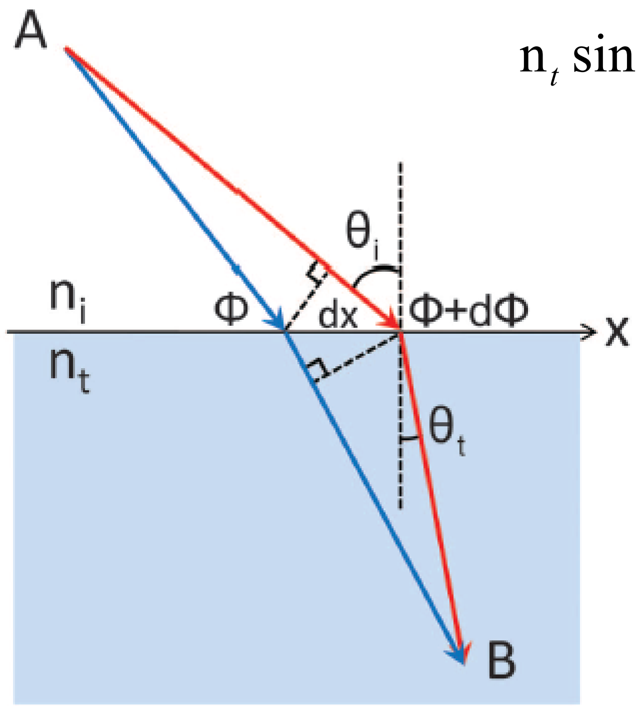
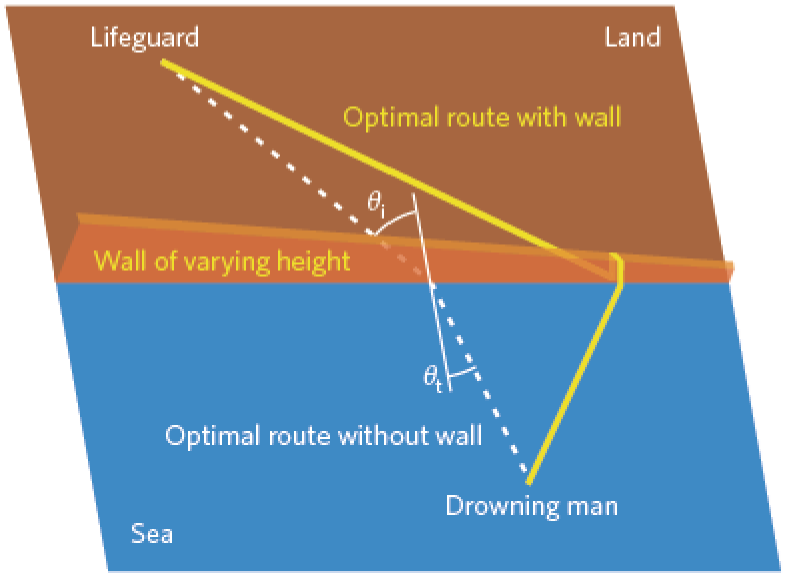
Generalized Snell's law

- The interface between the two media is artificially structured in order to introduce an abrupt phase shift ϕ in the light path, which is a function of the position x along the interface; this phase shift must be taken into the Snell law:

$$n_t \sin\theta_t - n_i \sin\theta_i = 0$$ Snell's law

$$n_t \sin\theta_t - n_i \sin\theta_i = \frac{\lambda_0}{2\pi}\frac{d\Phi}{dx}$$ Generalized Snell's law





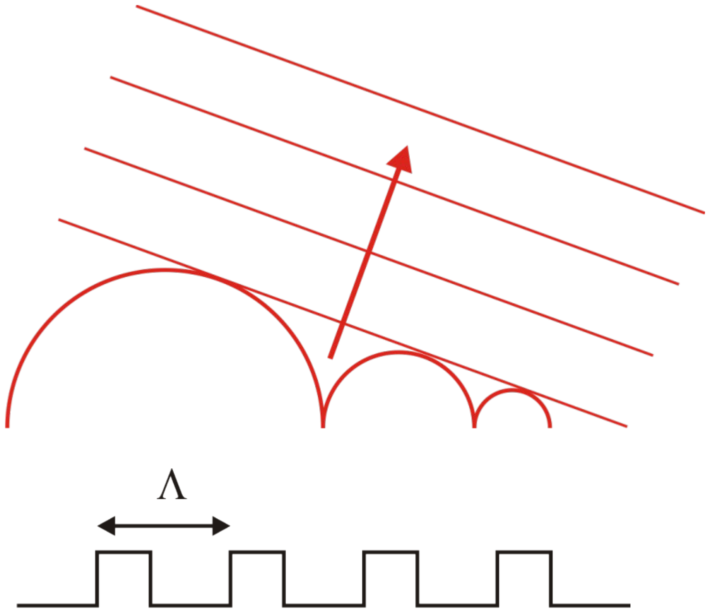
Science vol. 334, p. 333 (2011)

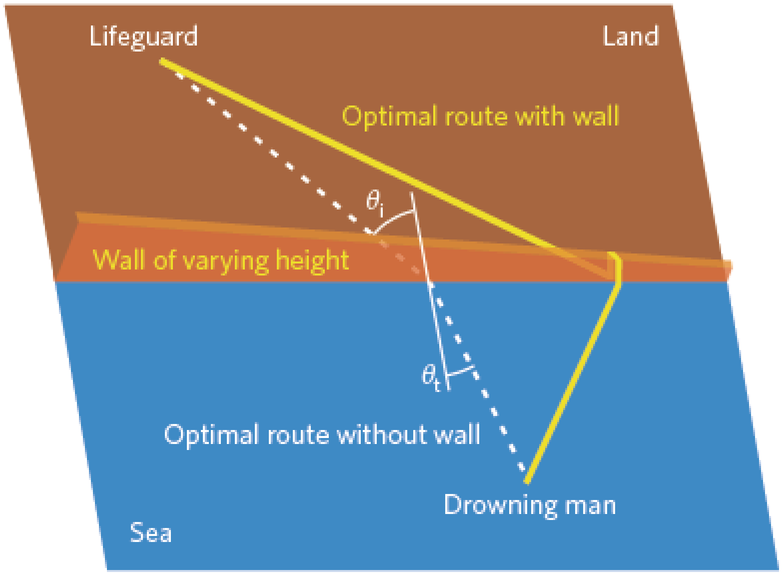
Generalized Snell's law

- A grating provides a monotonously growing phase along the interface:

$$n_t \sin\theta_t - n_i \sin\theta_i = \frac{\lambda_0}{2\pi}\frac{2\pi}{\Lambda} \qquad \text{Grating law}$$

$$n_t \sin\theta_t - n_i \sin\theta_i = \frac{\lambda_0}{2\pi}\frac{d\Phi}{dx} \qquad \text{Generalized Snell's law}$$

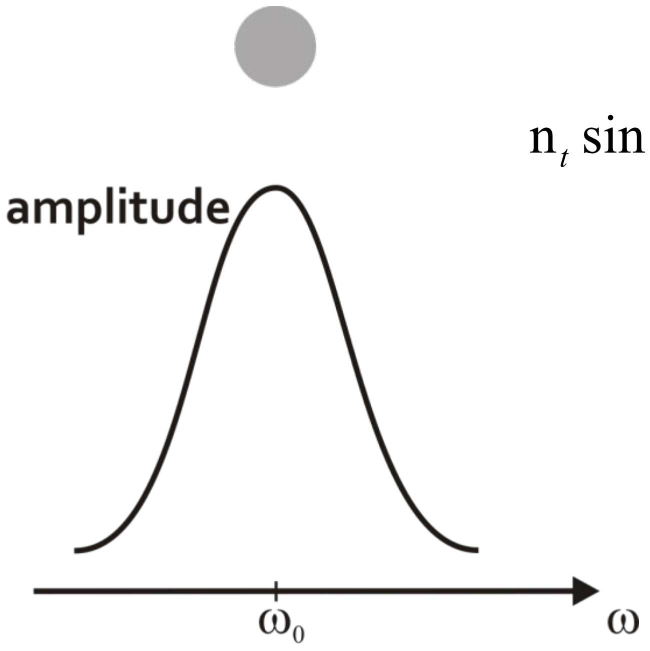




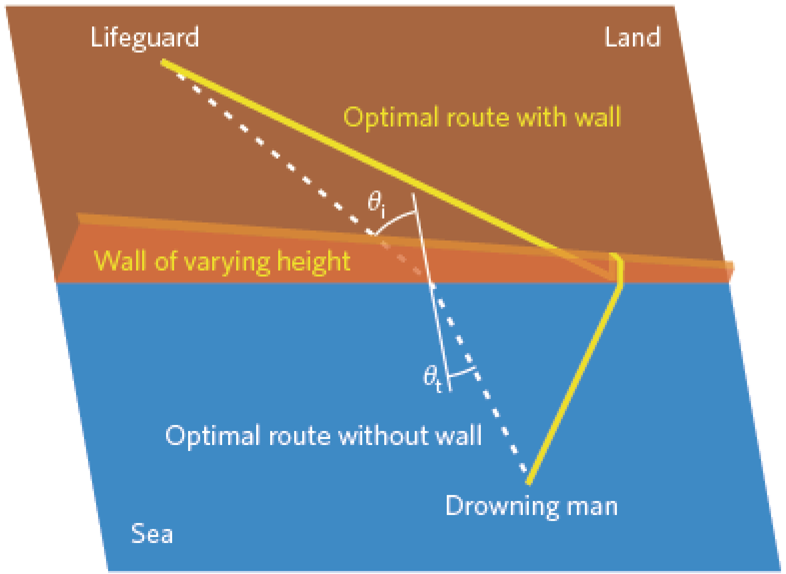
Science vol. 334, p. 333 (2011)

Generalized Snell's law

- A plasmonic nanostructure also produces a phase shift (which value depends on the wavelength, phase and material):



$$n_t \sin\theta_t - n_i \sin\theta_i = \frac{\lambda_0}{2\pi}\frac{d\Phi}{dx}$$ Generalized Snell's law



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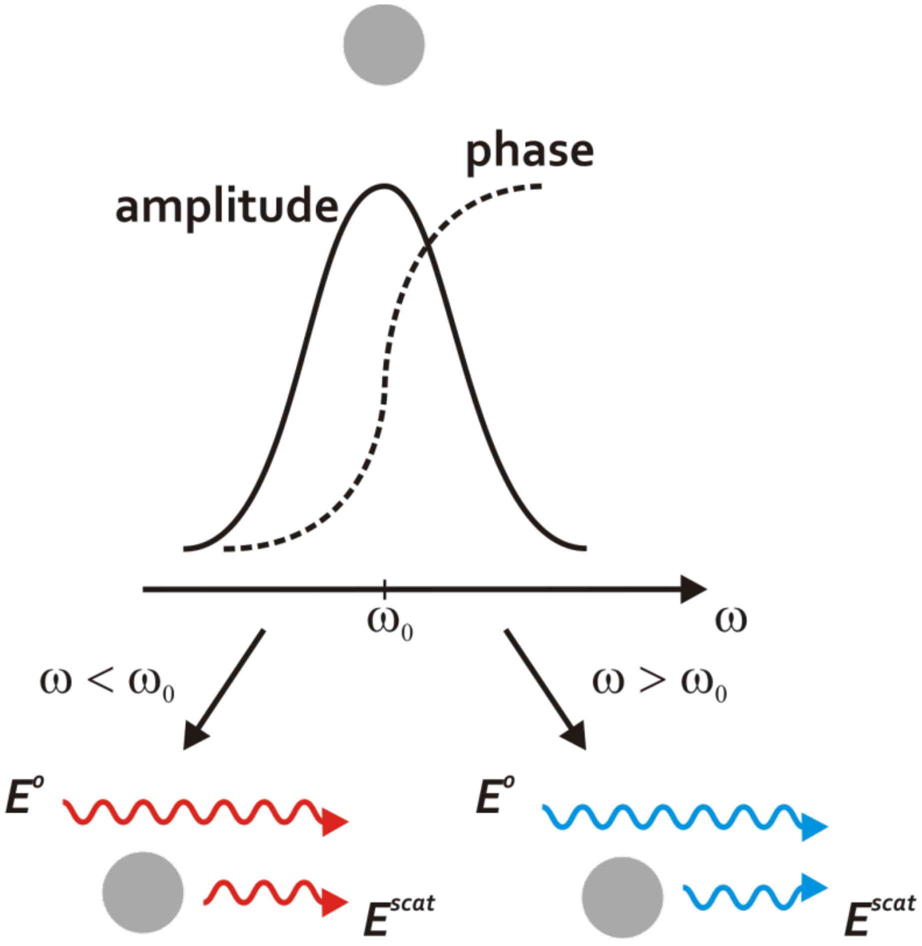
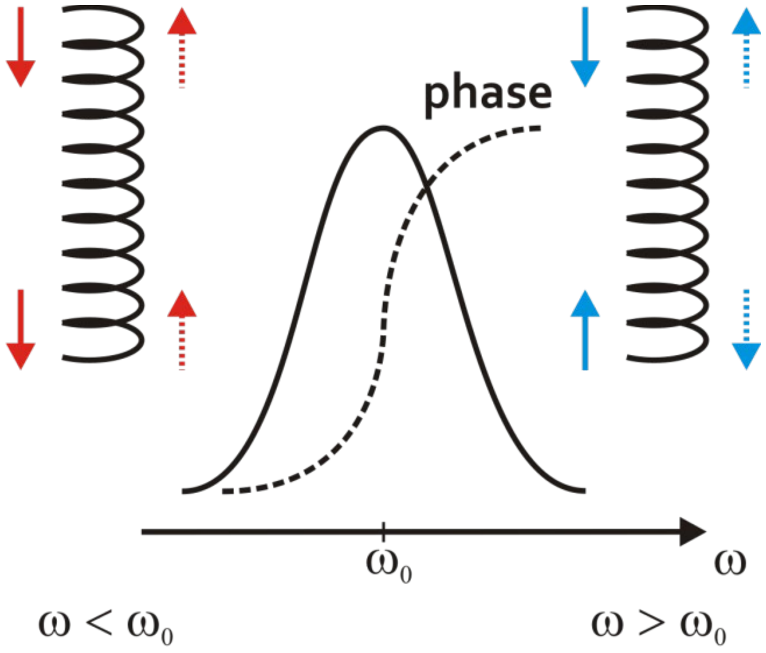
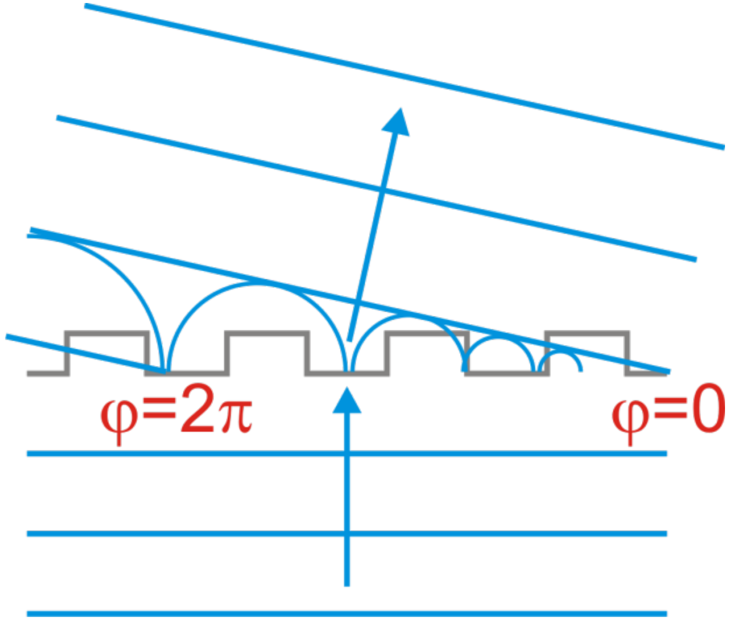


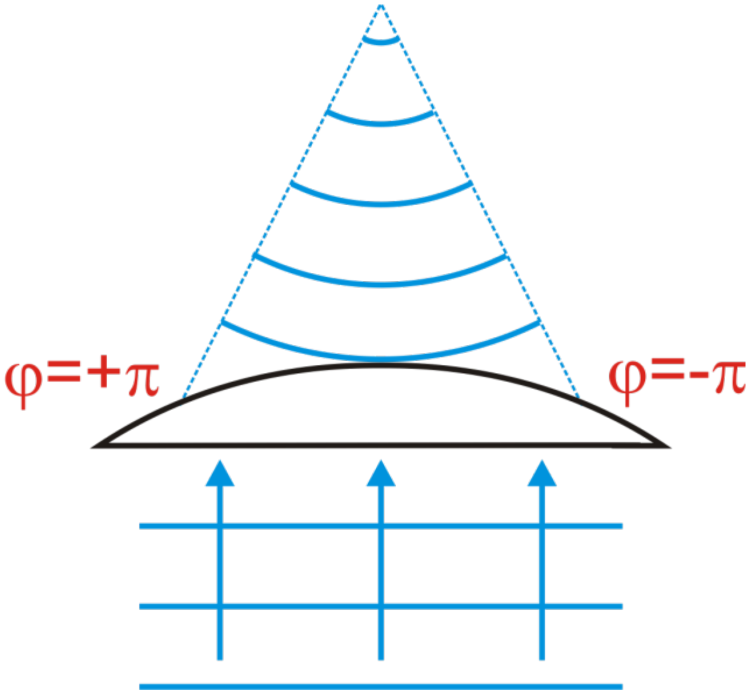


All optical components manipulate the phase of light

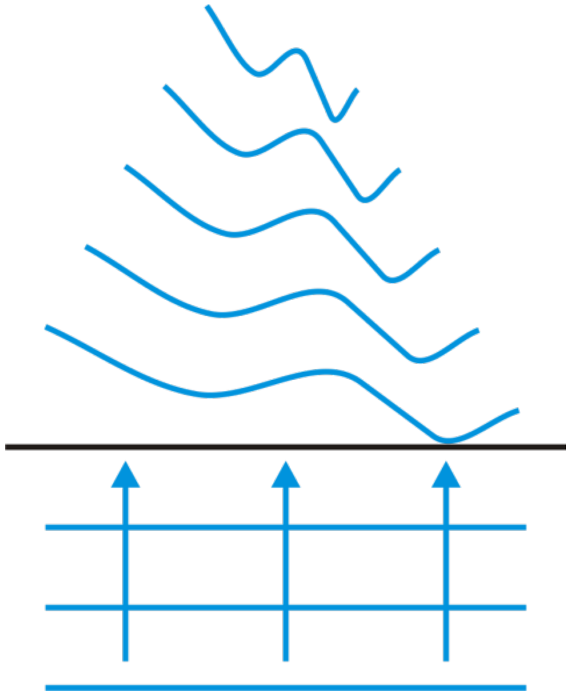
- **Grating:**



- **Lens:**



- **Hologram:**



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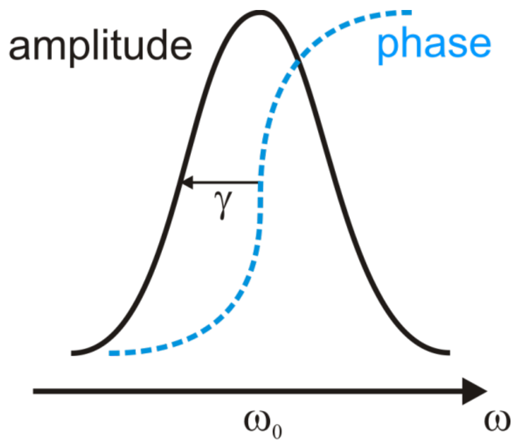

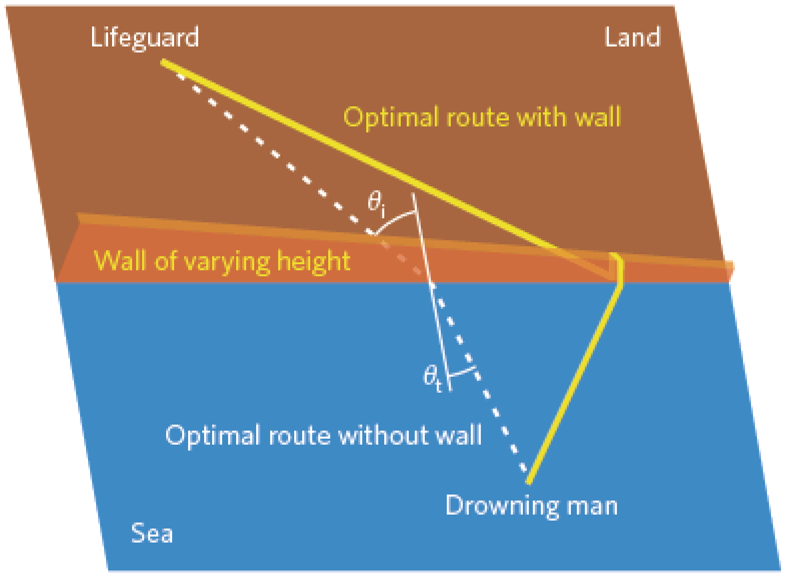

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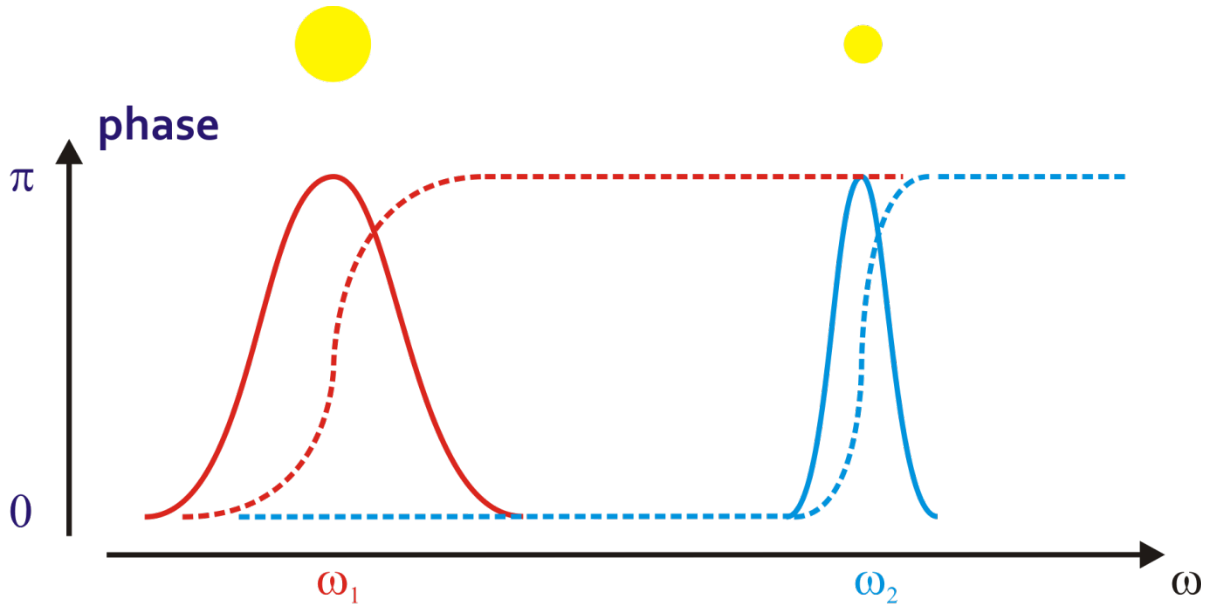
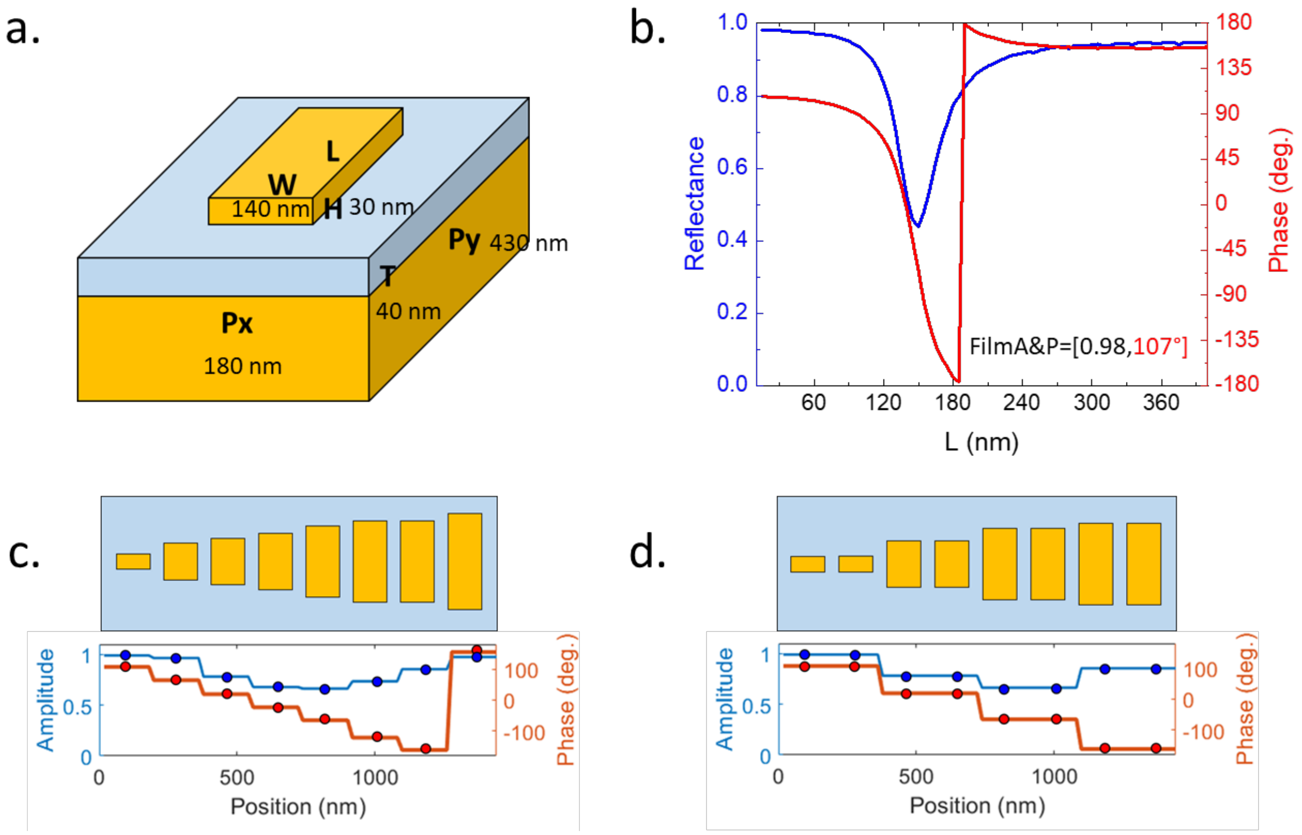
Plasmon resonance – Phase control through the size

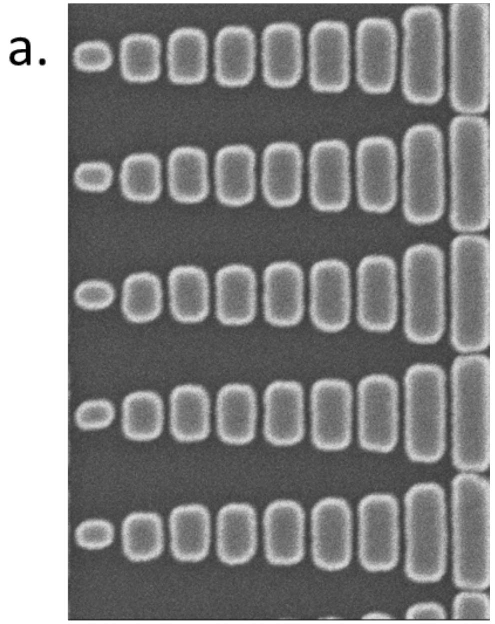
- Since particles with different sizes have plasmon resonances at different frequencies, the phase shift can be engineered by using different particle sizes



Plasmon resonance – Phase control through the size

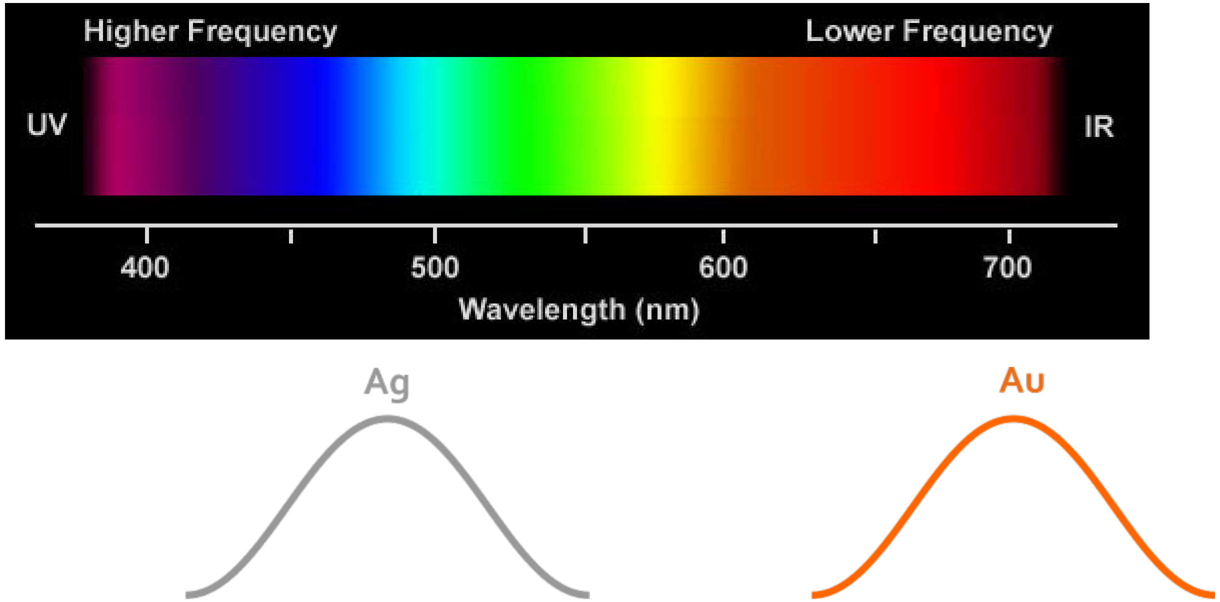
- Since particles with different sizes have plasmon resonances at different frequencies, the phase shift can be engineered by using different dimensions
- The amplitude should however remain as constant as possible





Plasmon resonance – Phase control through the composition

- In the previous example, we controlled the phase through a change of shape
- It is also possible to control the phase through a change of material – here a change of stoichiometry in and Au-Ag alloy
- Only a few plasmonic metals exist and each works at a specific wavelength



D. Ray, Advanced Materials vol. 2022, p. 2108225 (2022)

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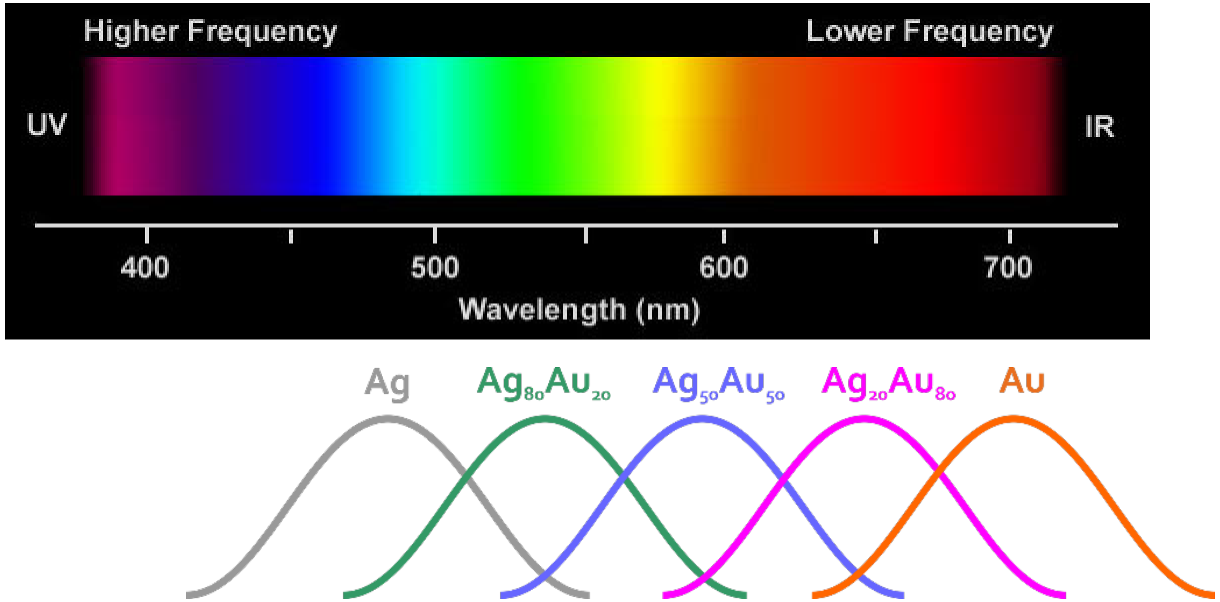
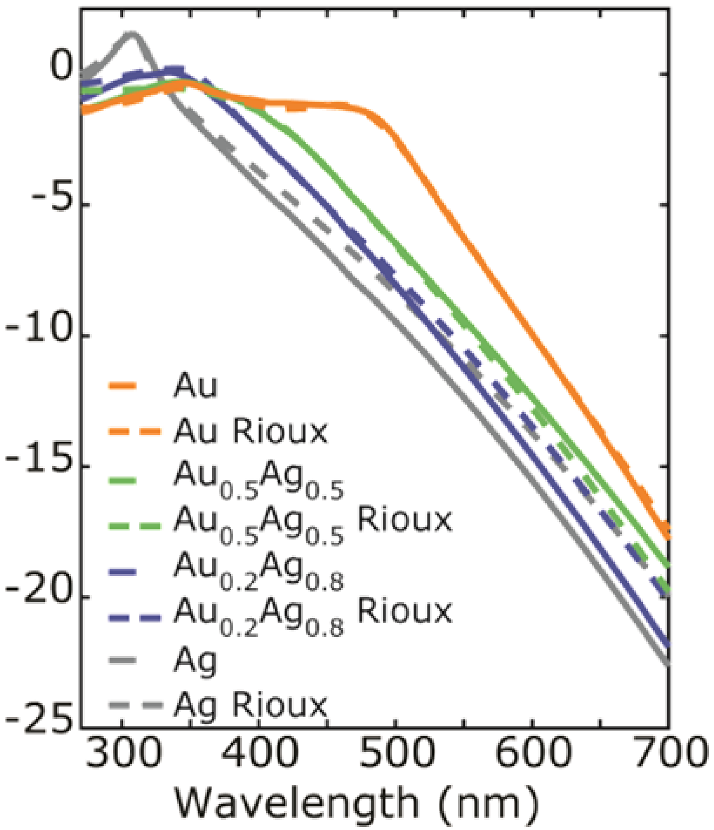


D. Ray, Advanced Materials vol. 2022, p. 2108225 (2022)

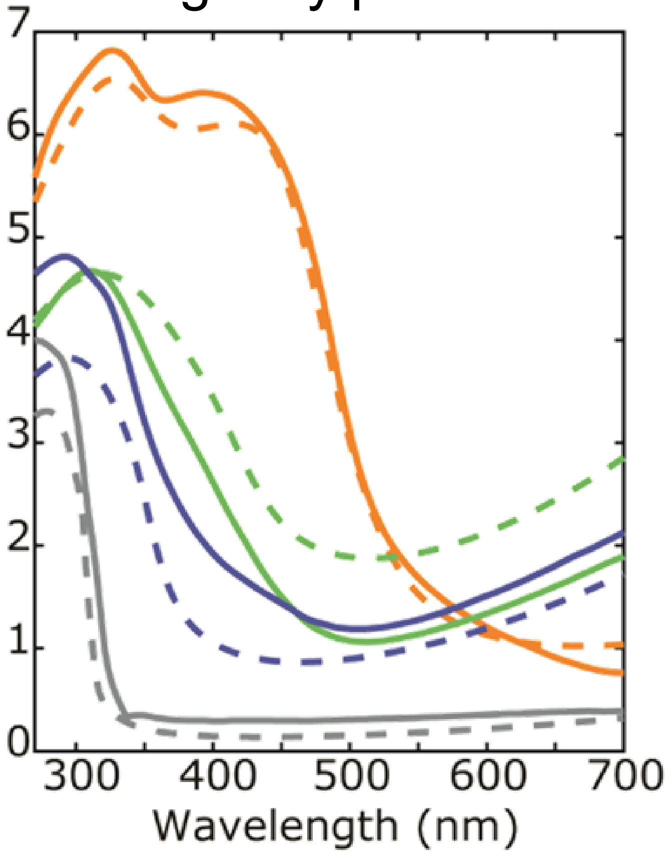
Plasmonic alloys

- Continuous variation of the dielectric function vs. stoichiometry

Real part of ε



Imaginary part of ε

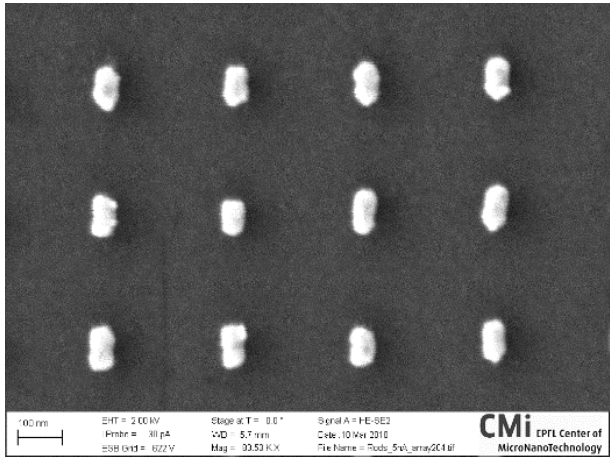


D. Ray, Advanced Materials vol. 2022, p. 2108225 (2022)

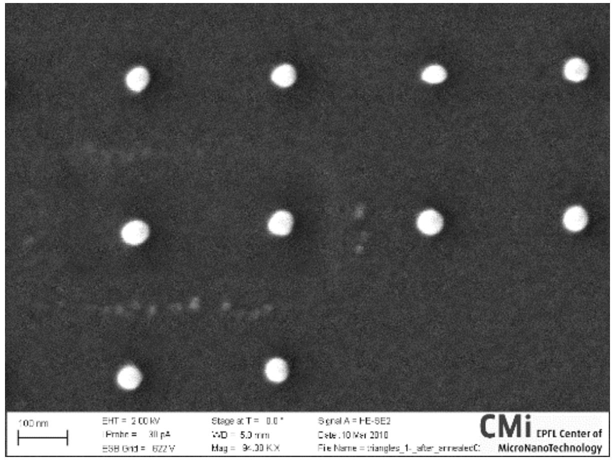
Alloyed plasmonic nanostructures

- Normally, Au_xAg_{1-x} alloying is performed at high temperature
- Can we maintain the shape of nanostructures during the alloying process…?

Before annealing



After annealing



- … no! the shape is lost during the high temperature annealing

D. Ray, Advanced Materials vol. 2022, p. 2108225 (2022)

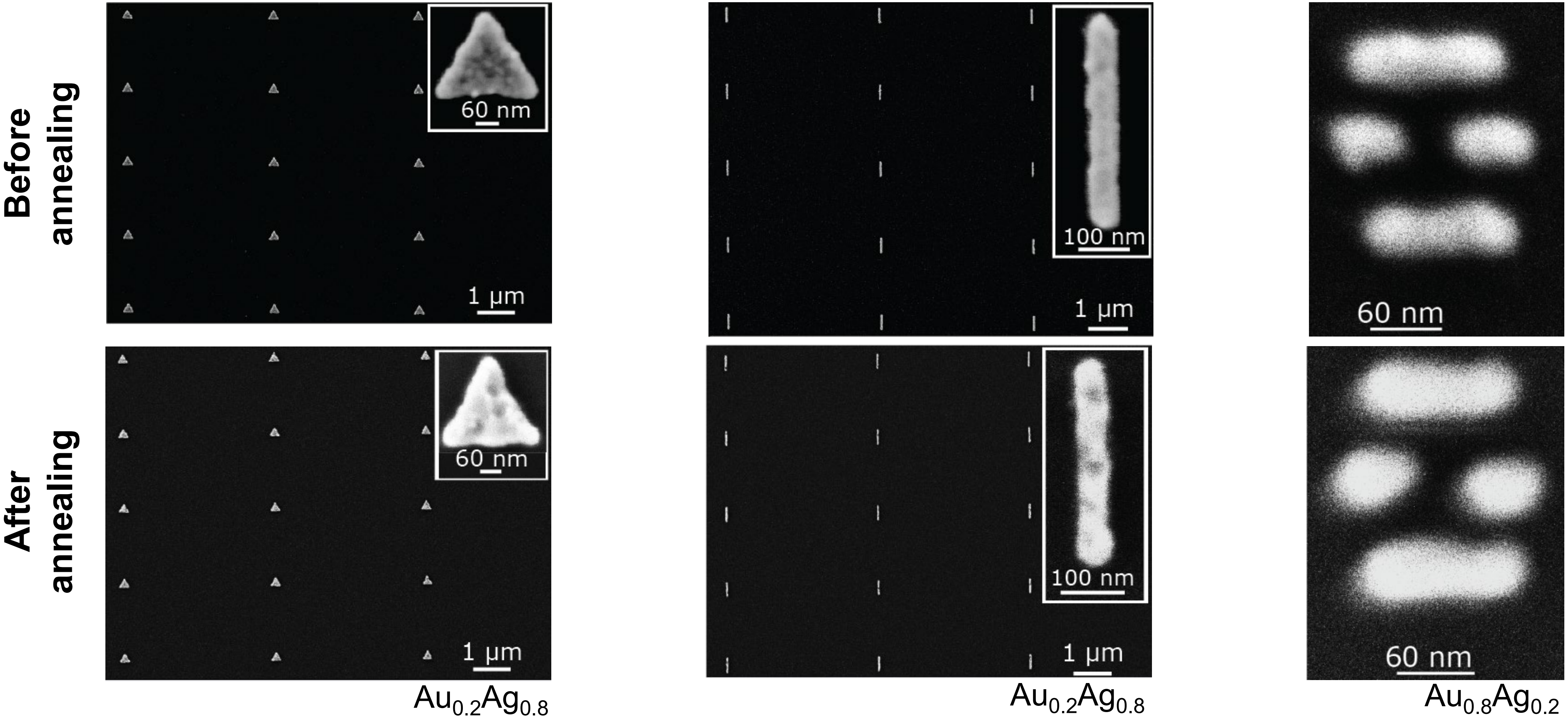
Alloyed plasmonic nanostructures

- Challenge:
  - Find a low temperature alloying procedure…
  - … that enables any alloy composition
- Solution:
  - Define the composition by the Au and Ag thicknesses
  - Alloy at 300°C for 8 hours and 450°C for 30 mins

D. Ray, Advanced Materials vol. 2022, p. 2108225 (2022)

Alloyed plasmonic nanostructures

- A low temperature annealing process can maintain the particle shape, whilst producing well alloyed materials



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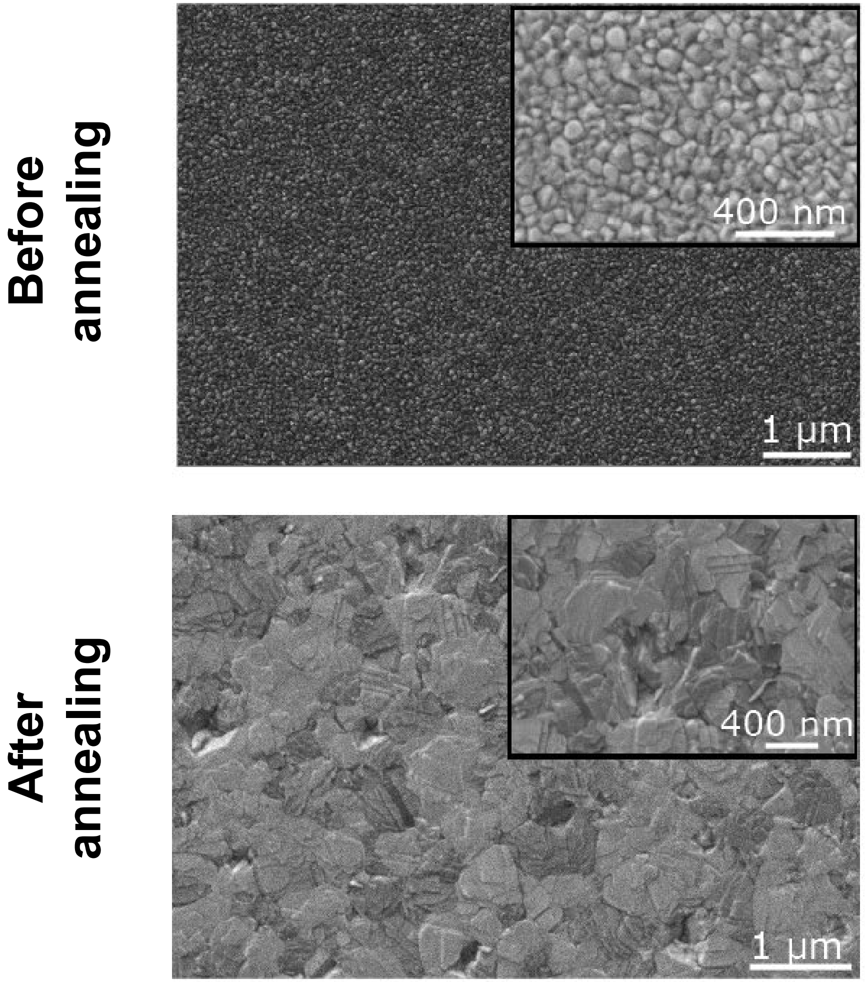
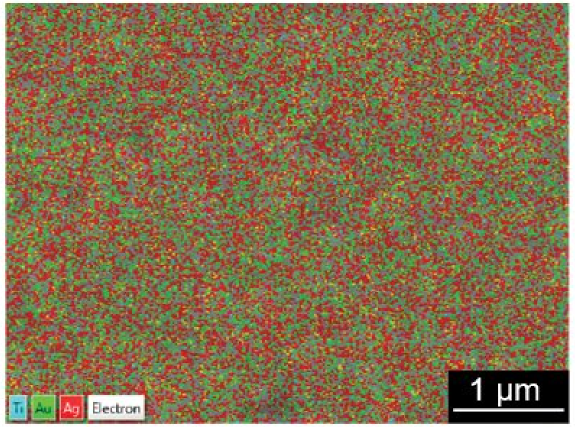
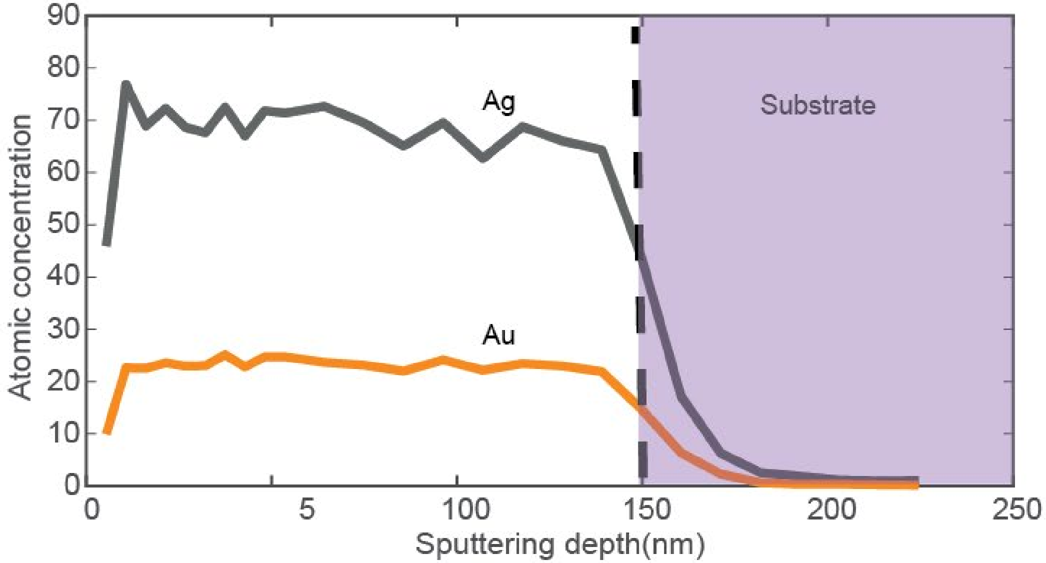
Alloyed plasmonic nanostructures

- Well alloyed materials (EDX and XPS)



$Au_{0.2}Ag_{0.8}$

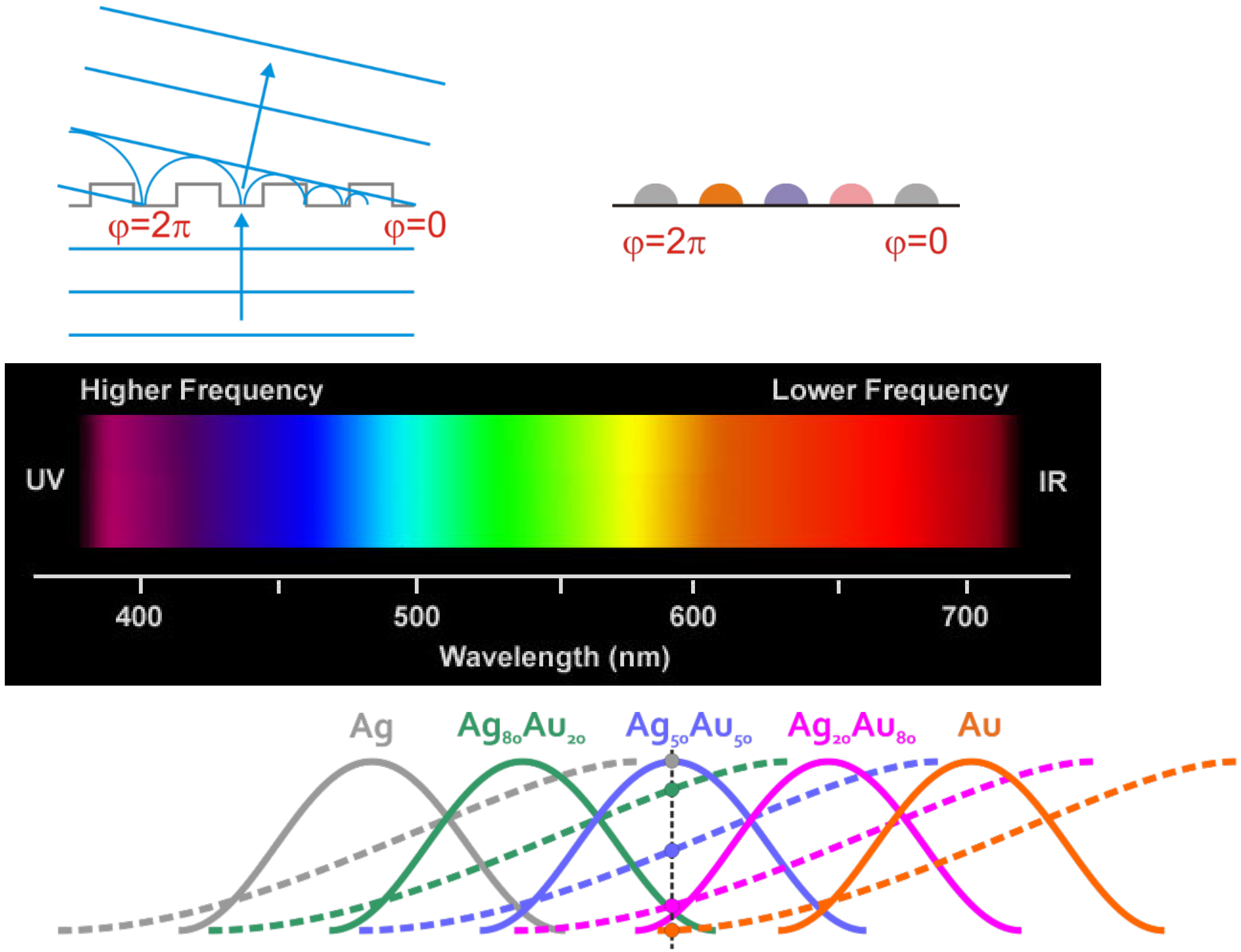




D. Ray, Advanced Materials vol. 2022, p. 2108225 (2022)

Plasmonic metasurfaces

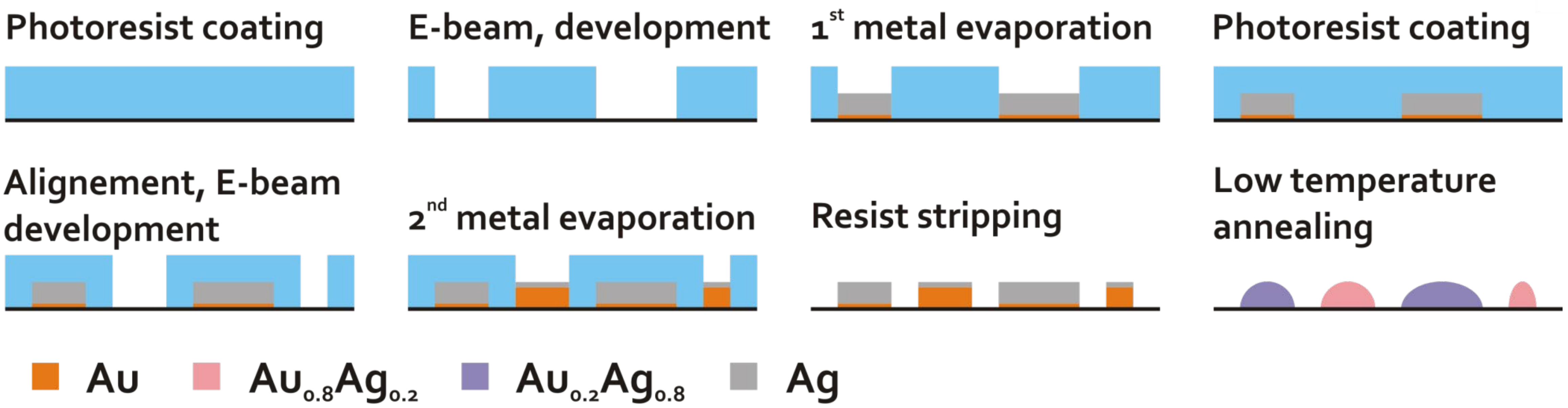
- Instead of using different geometries, we can use the same geometry and change the stoichiometry to obtain different phases on the metasurface:



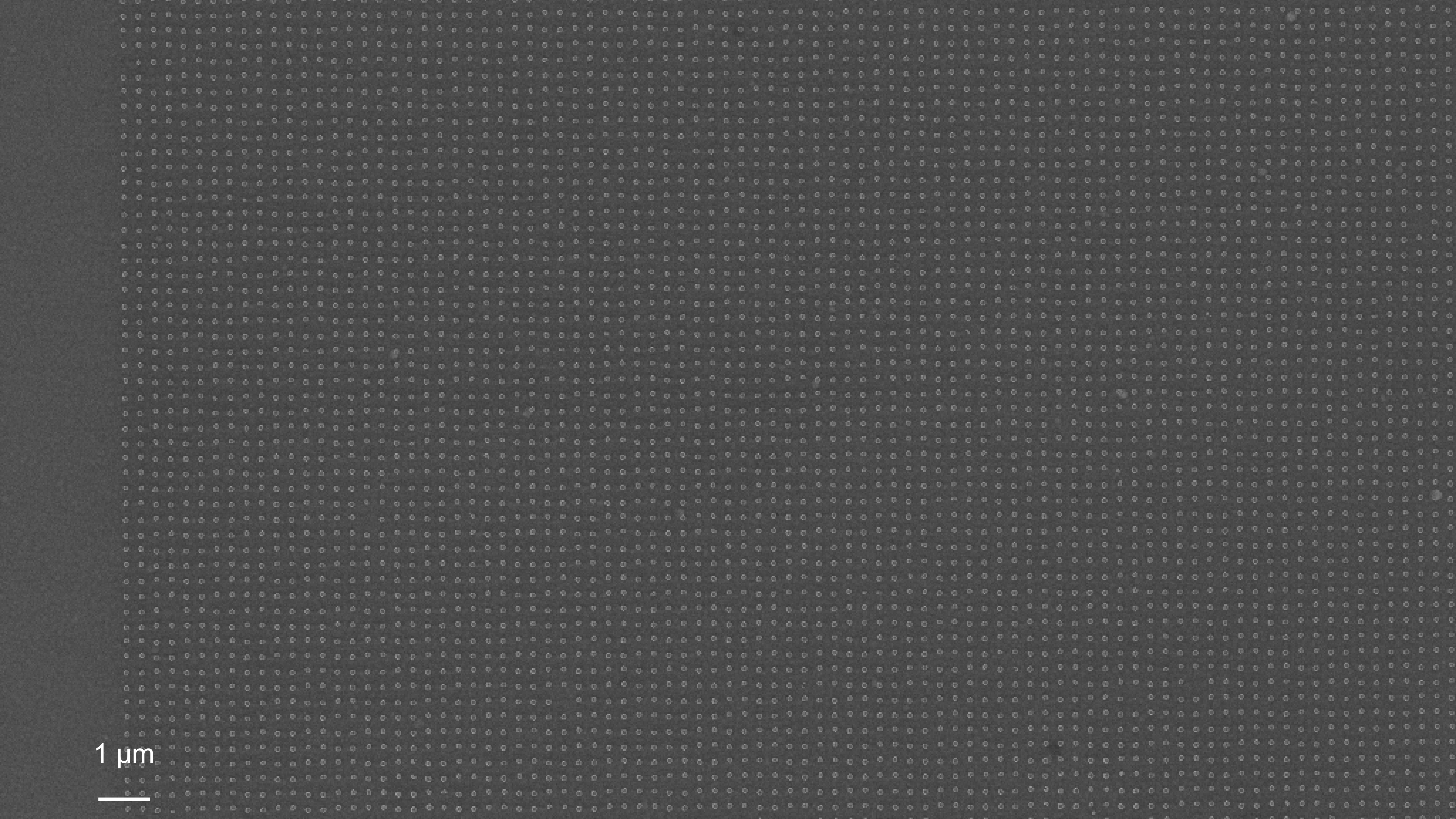
D. Ray, Advanced Materials vol. 2022, p. 2108225 (2022)

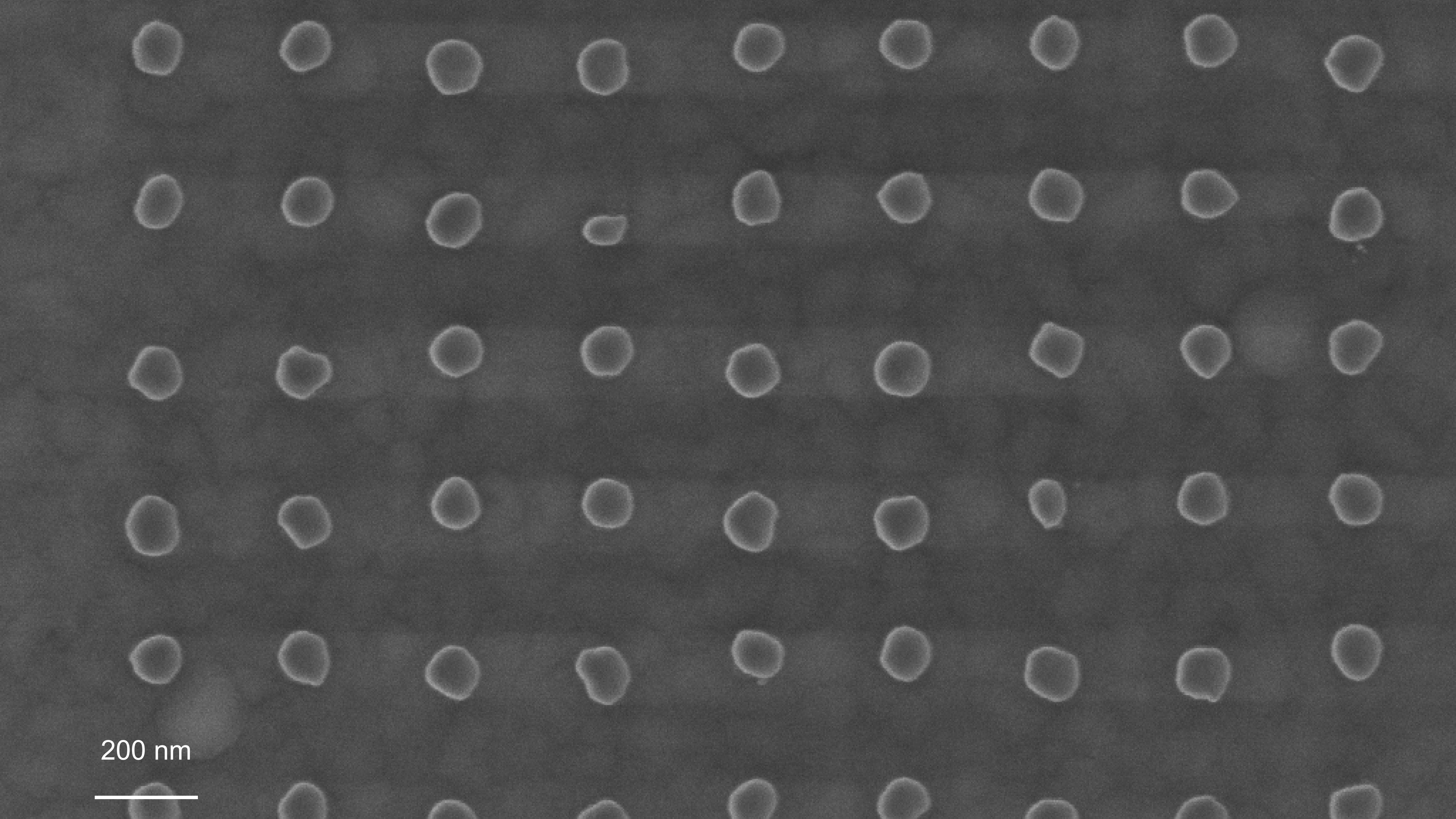
Alloyed plasmonic metasurfaces

- Combinations of different alloy compositions on the same sample to produce more complex optical effects

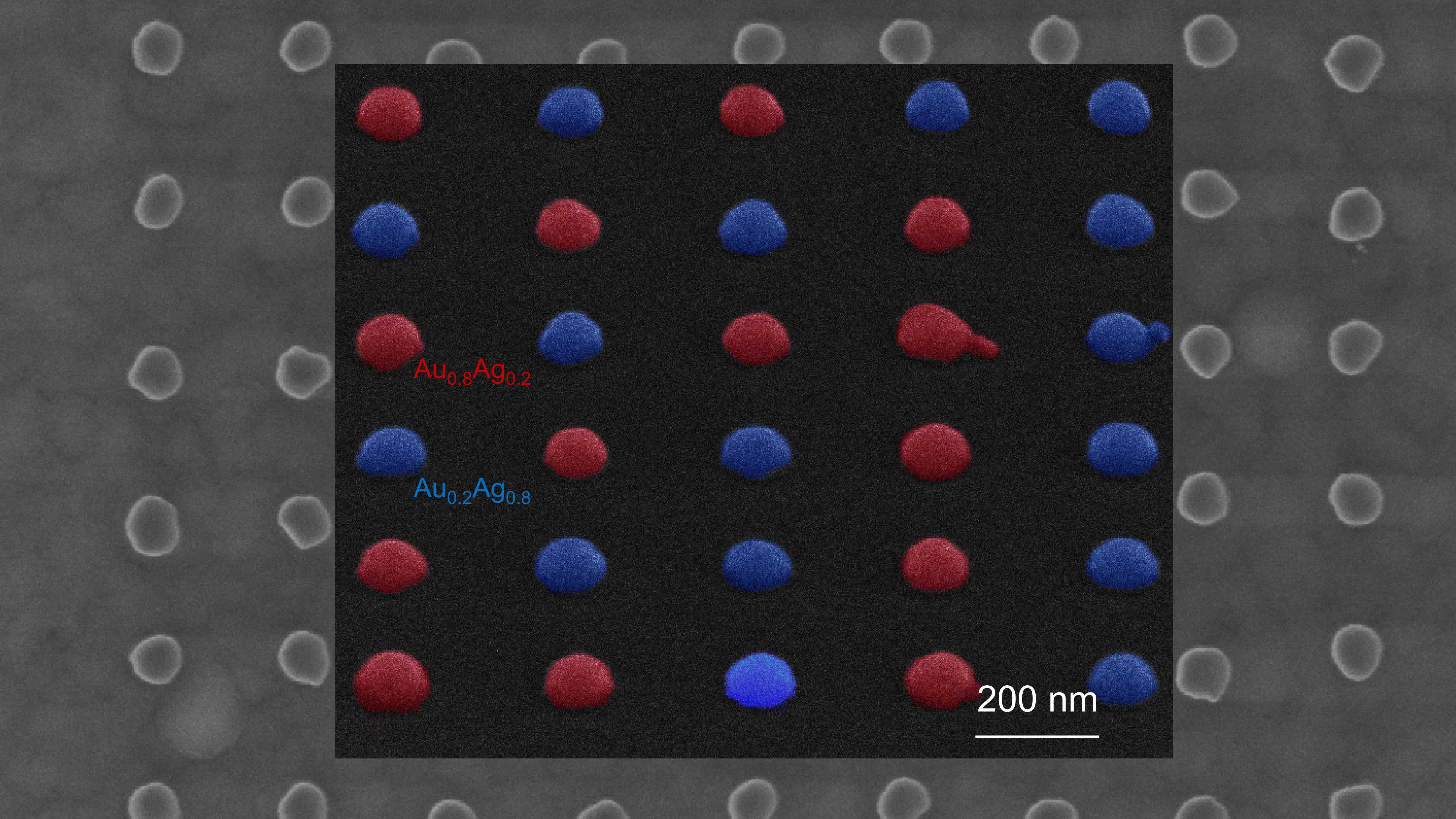


D. Ray, Advanced Materials vol. 2022, p. 2108225 (2022)


1 µm



200 nm

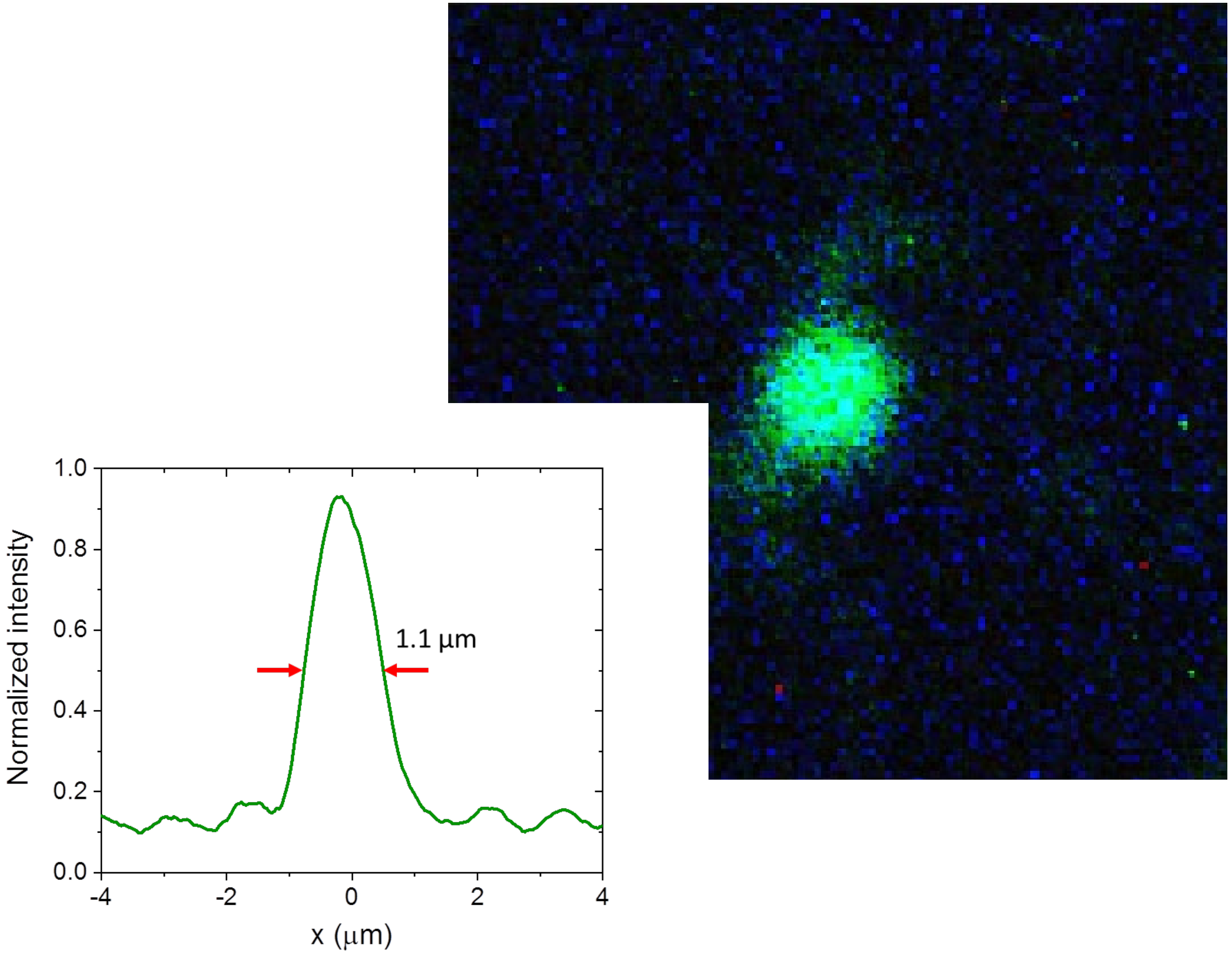

$Au_{0.8}Ag_{0.2}$
$Au_{0.2}Ag_{0.8}$
200 nm

Complex metasurfaces

- Hologram:

λ=532 nm

- Lens:



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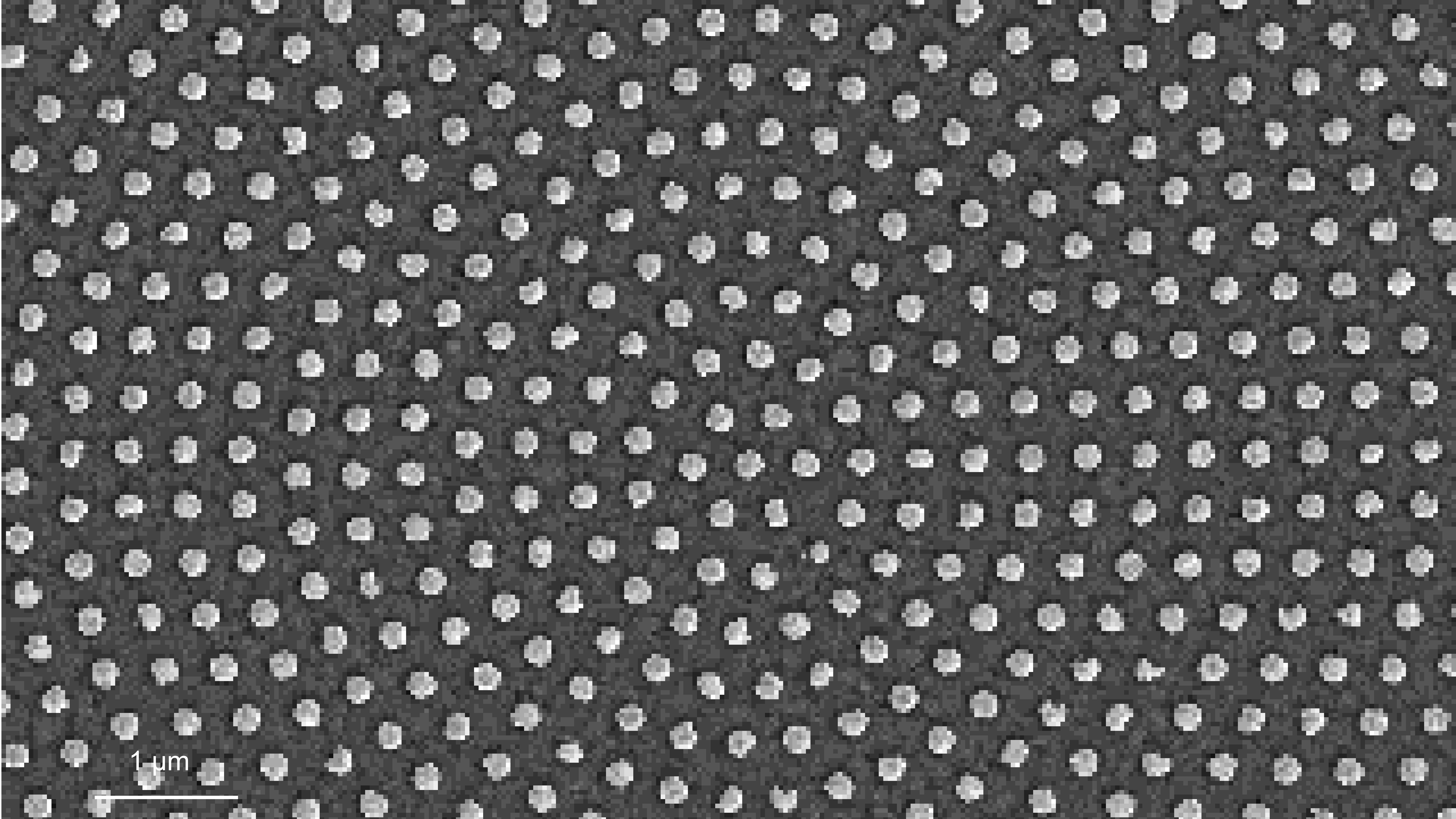
1 µm

Selected Topics in Advanced Optics

Week 12 – part 2

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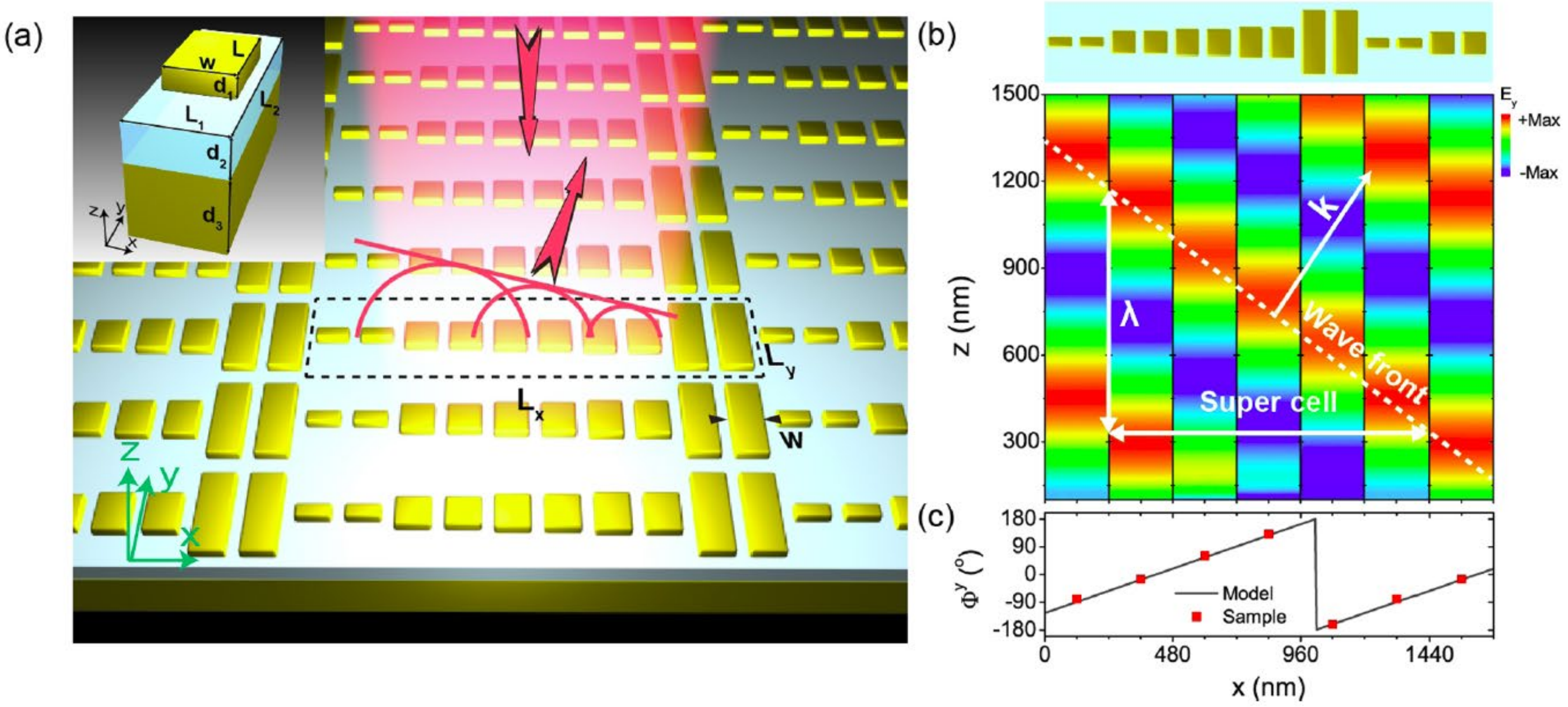
Generalized Snell's law – Anomalous reflection

- The engineering of the phase is arbitrary in those systems
- Each structure produces a different phase shift



S. Sun, Nano Letters vol. 12, p. 6223 (2012)

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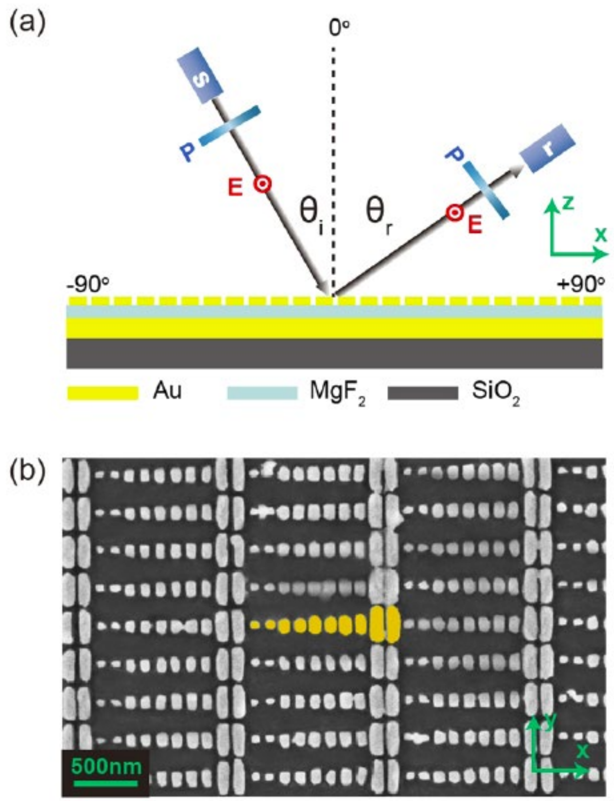

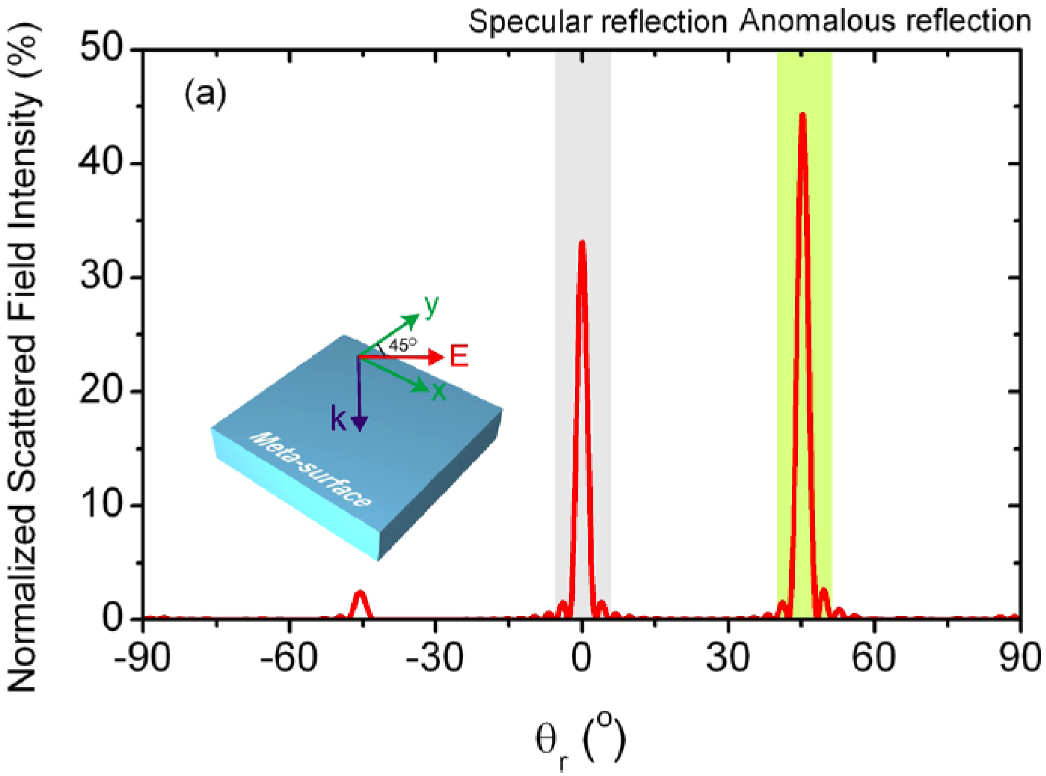

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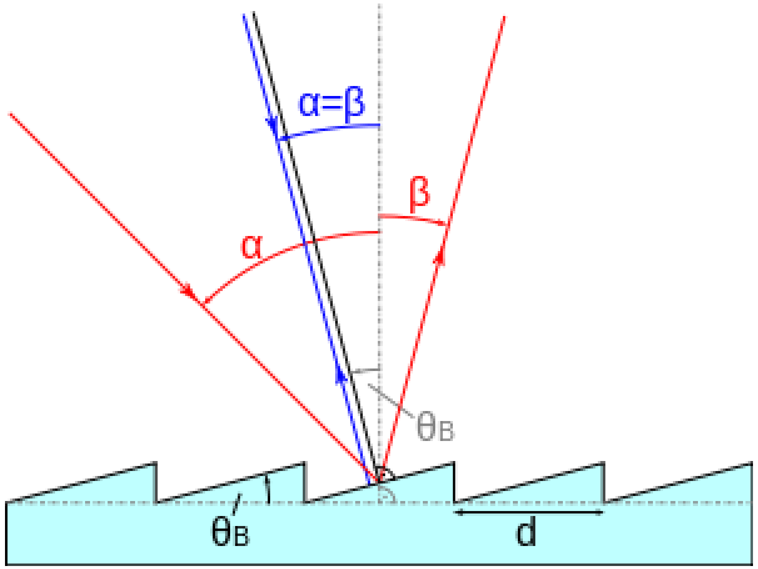
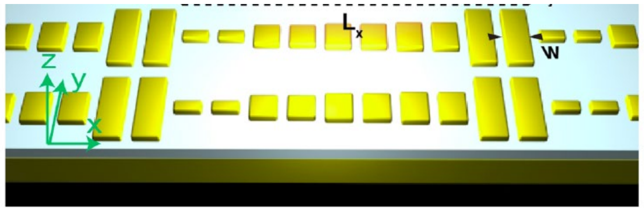
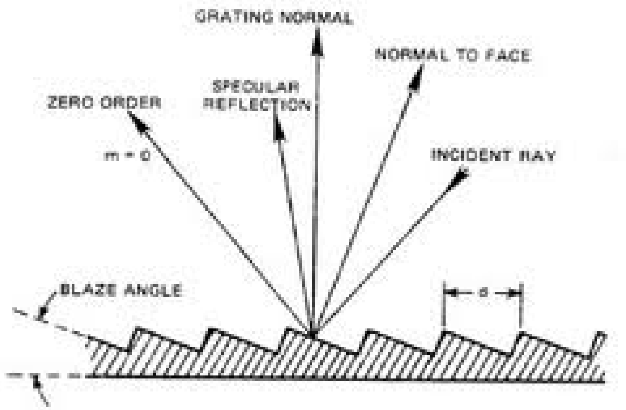
Blazed grating

- One should not forget classical optics, which remains a fantastic source of inspiration!

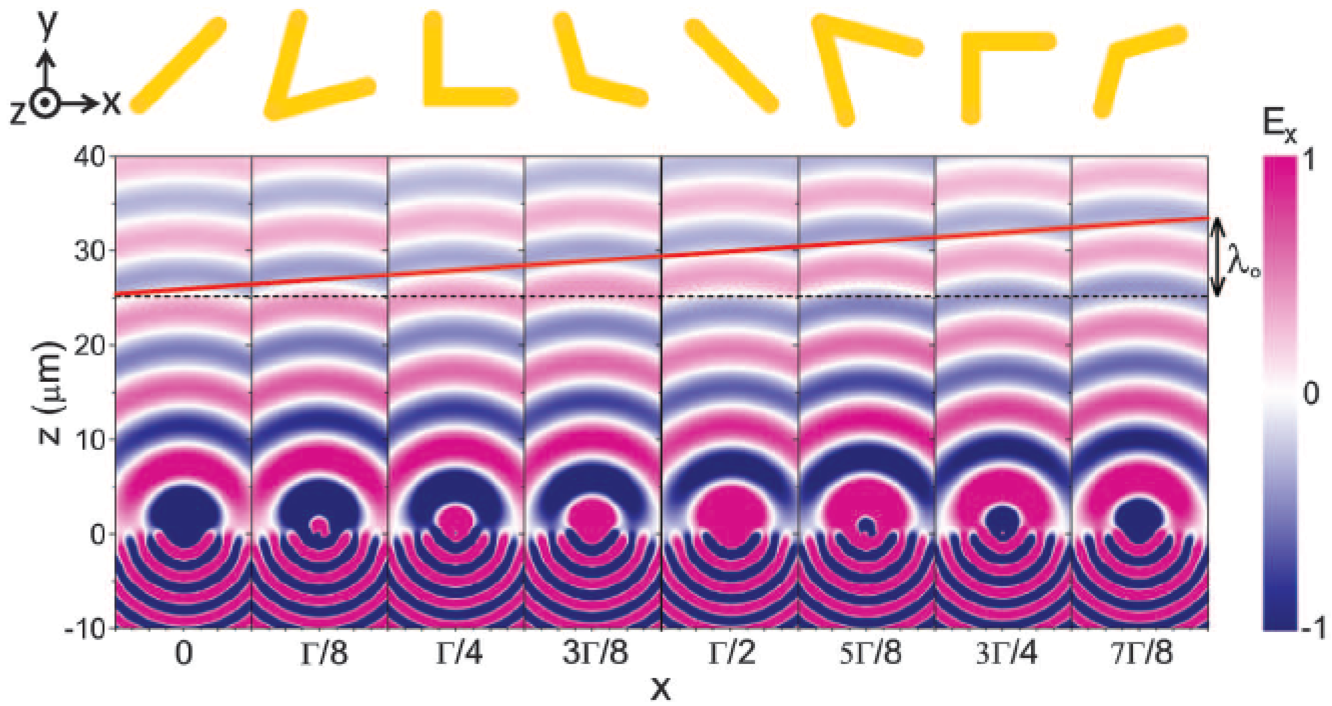


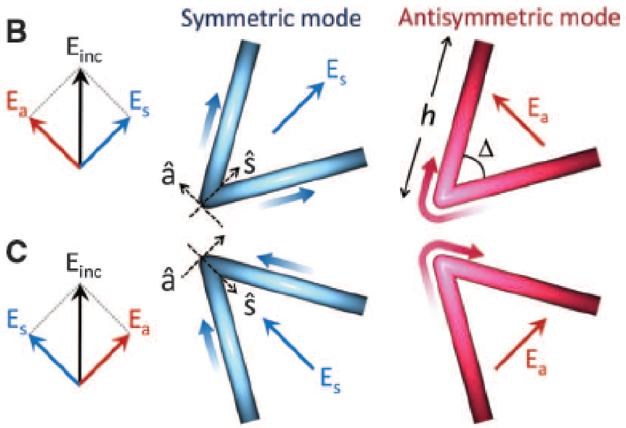




Generalized Snell's law

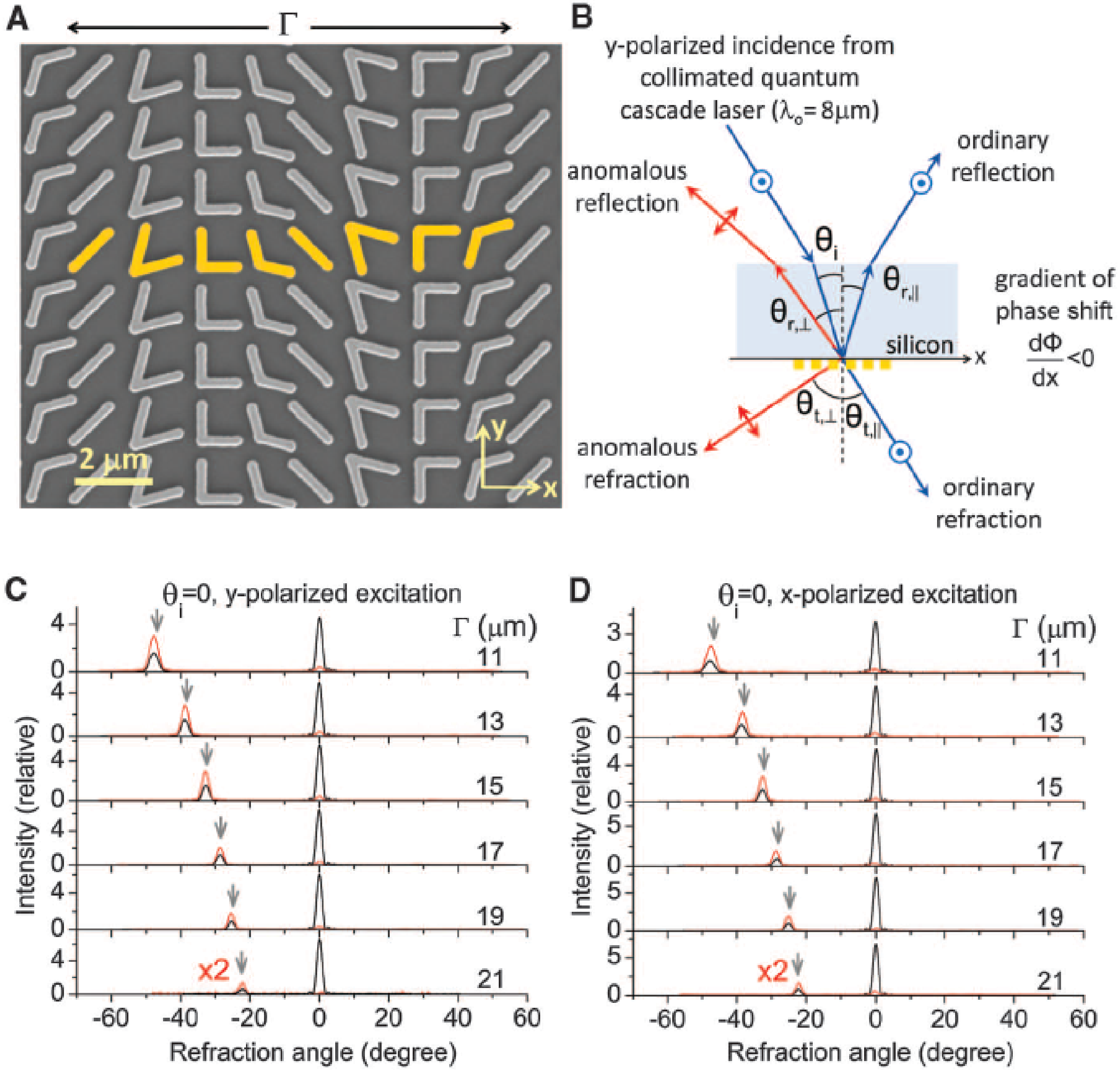
- A plasmonic V-shaped nanostructure can introduce a phase in the scattered wave, which originates from the interplay between the two modes that can be excited in the structure





Science vol. 334, p. 333 (2011)

Generalized Snell's law – Negative refraction can be produced this way

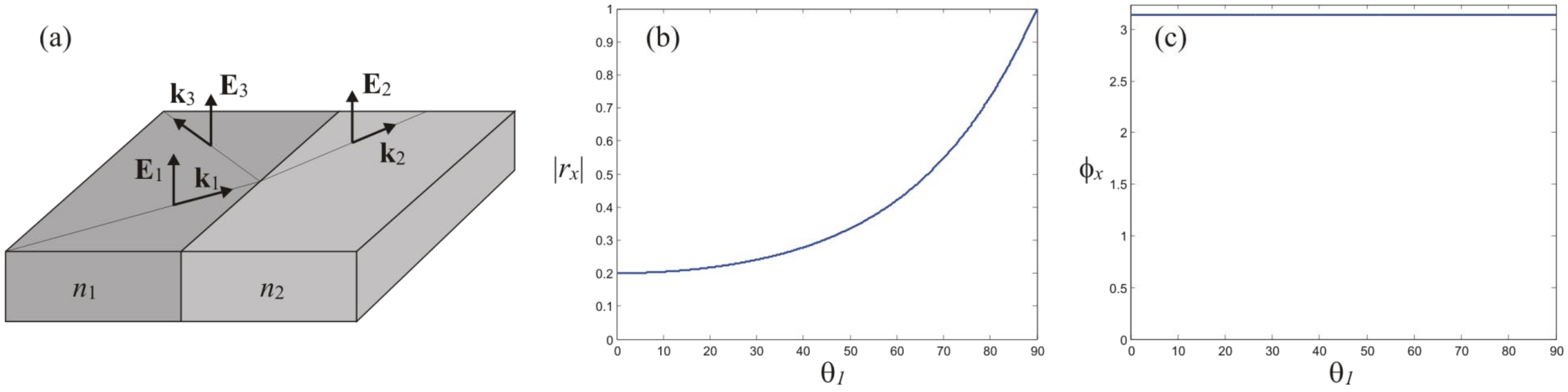


Science vol. 334, p. 333 (2011)

Extending the phase beyond π

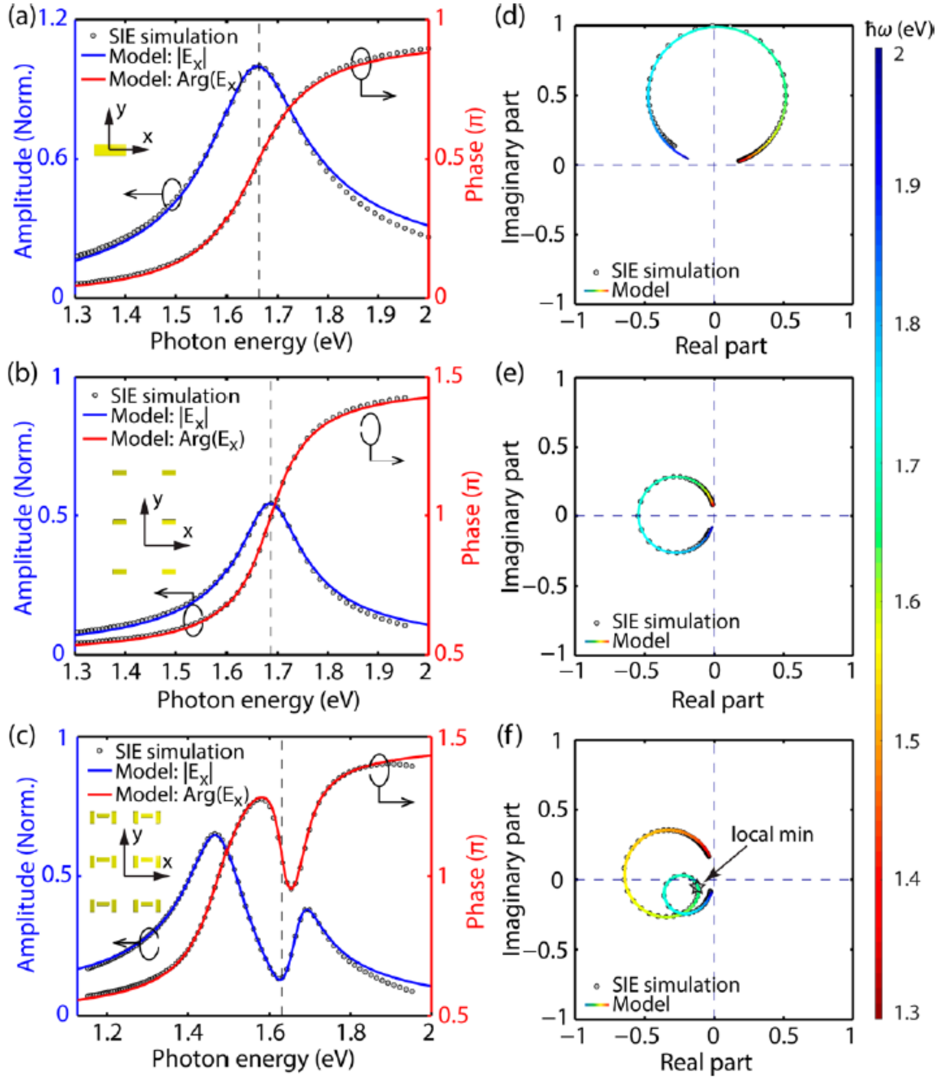
- Normally, any optical resonance is associated with a phase shift of π. To produce a metasurface, we need a phase shift of 2π
- This can be achieved with a mirror under the nanostructures

Fresnel coefficients for external reflection ($n_1<n_2$, TE):

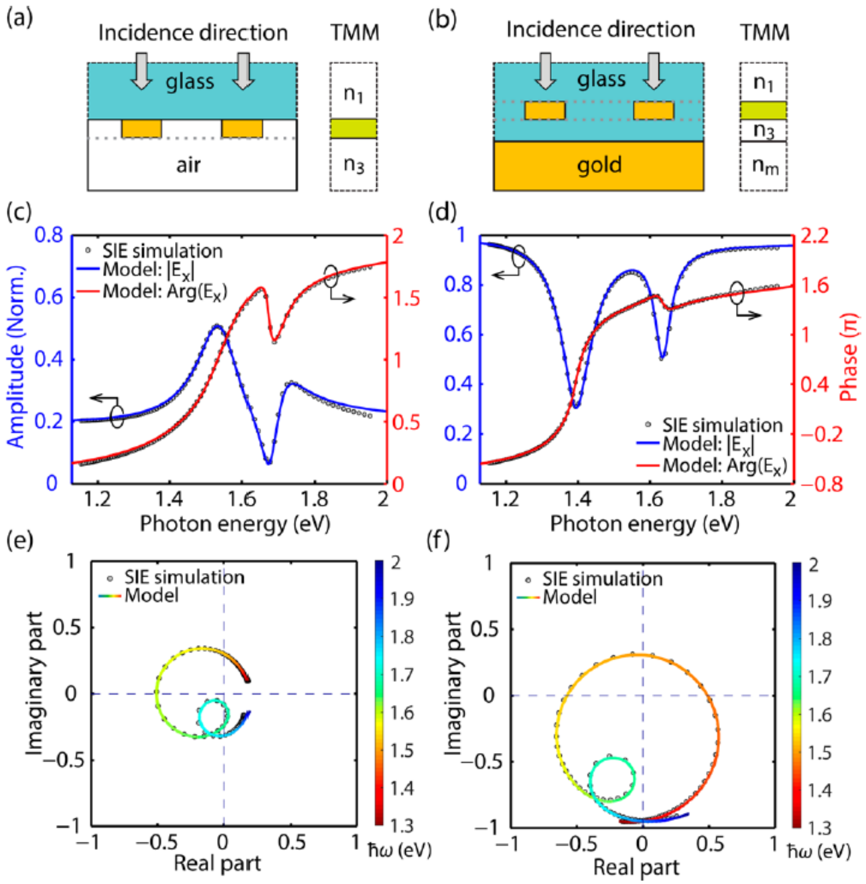


Extending the phase beyond π

Without a surface:



With a surface/mirror:



ACS Photonics vol. 4, p. 852-860 (2017)

Selected Topics in Advanced Optics

Week 12 – part 3

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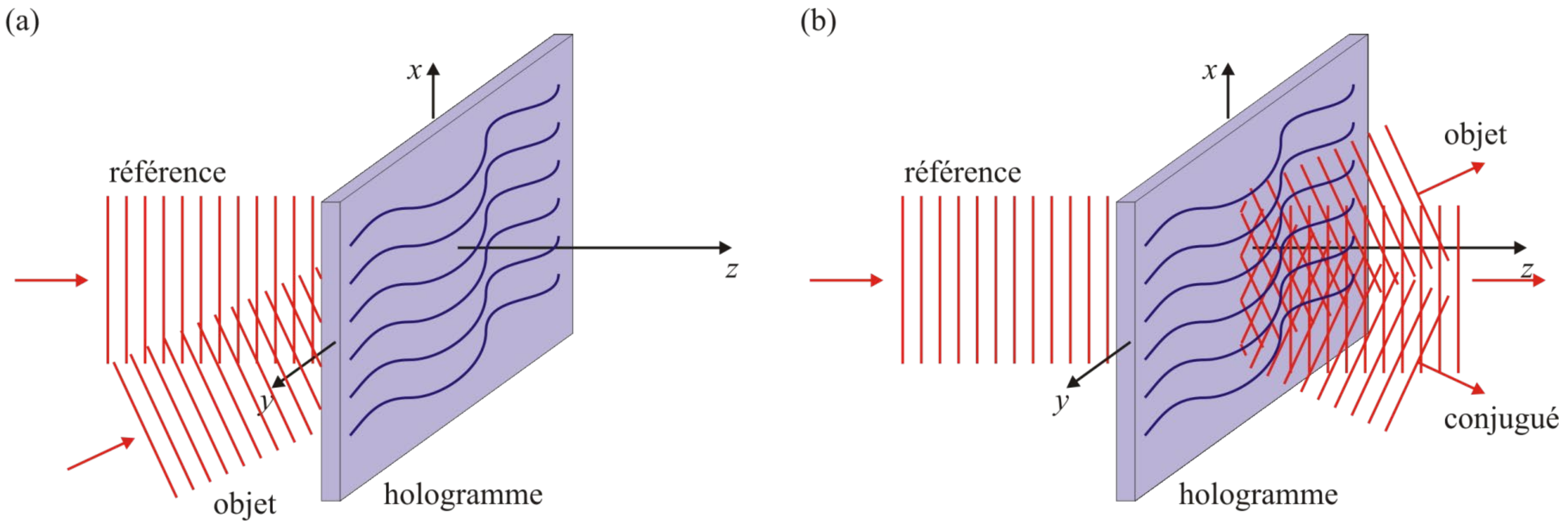
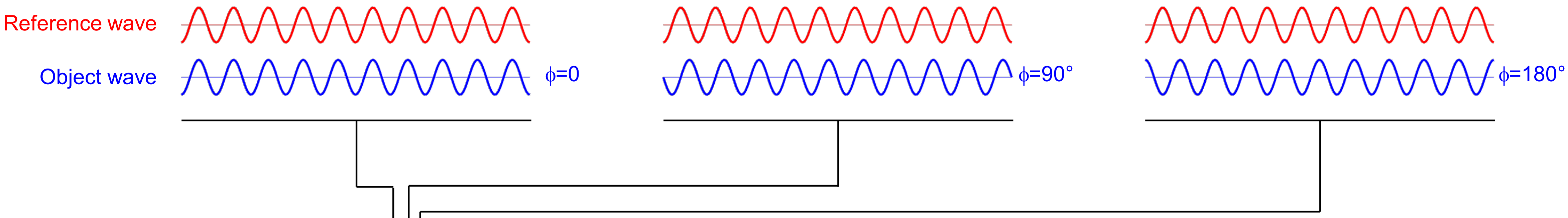
Designing a hologram with meta-atoms

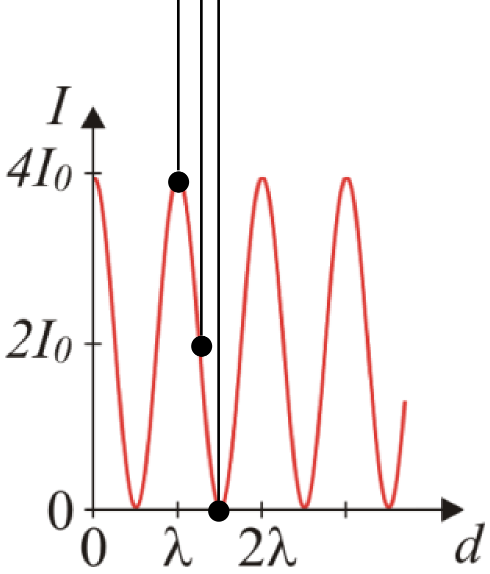
- Monochromatic object wave $U_o(x, y)$
- Its complex amplitude is recorder by performing an interference with a reference wave:
$$t \propto |U_o + U_r|^2 = |U_r|^2 + |U_o|^2 + U_r^* U_o + U_r U_o^* = I_r + I_o + U_r^* U_o + U_r U_o^* \, .$$
- The hologram is reconstructed with the same reference wave:
$$U = U_r t \propto U_r I_r + U_r I_o + I_r U_o + U_r^2 U_o^* \, .$$



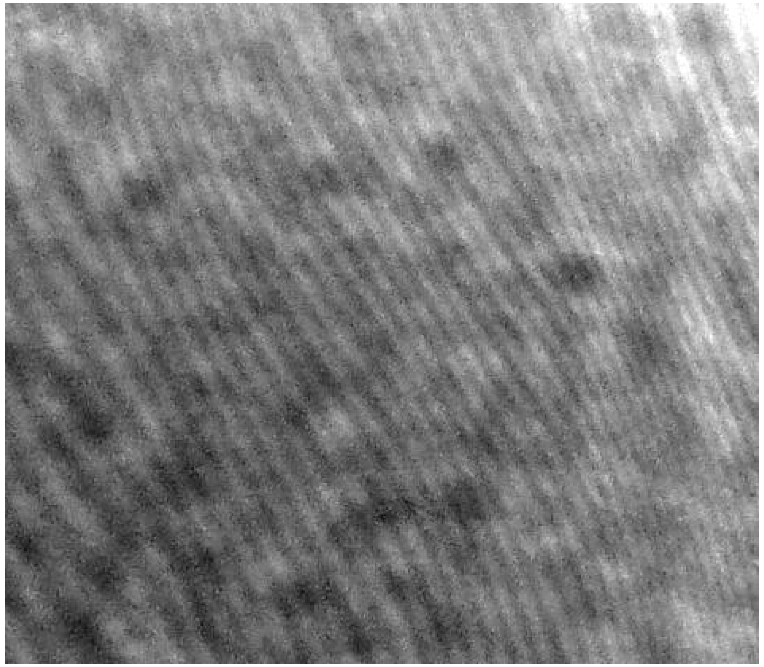
Designing a hologram with meta-atoms

- A hologram encrypts the amplitude and phase of an object by recording its interference with a reference wave





Hologram surface



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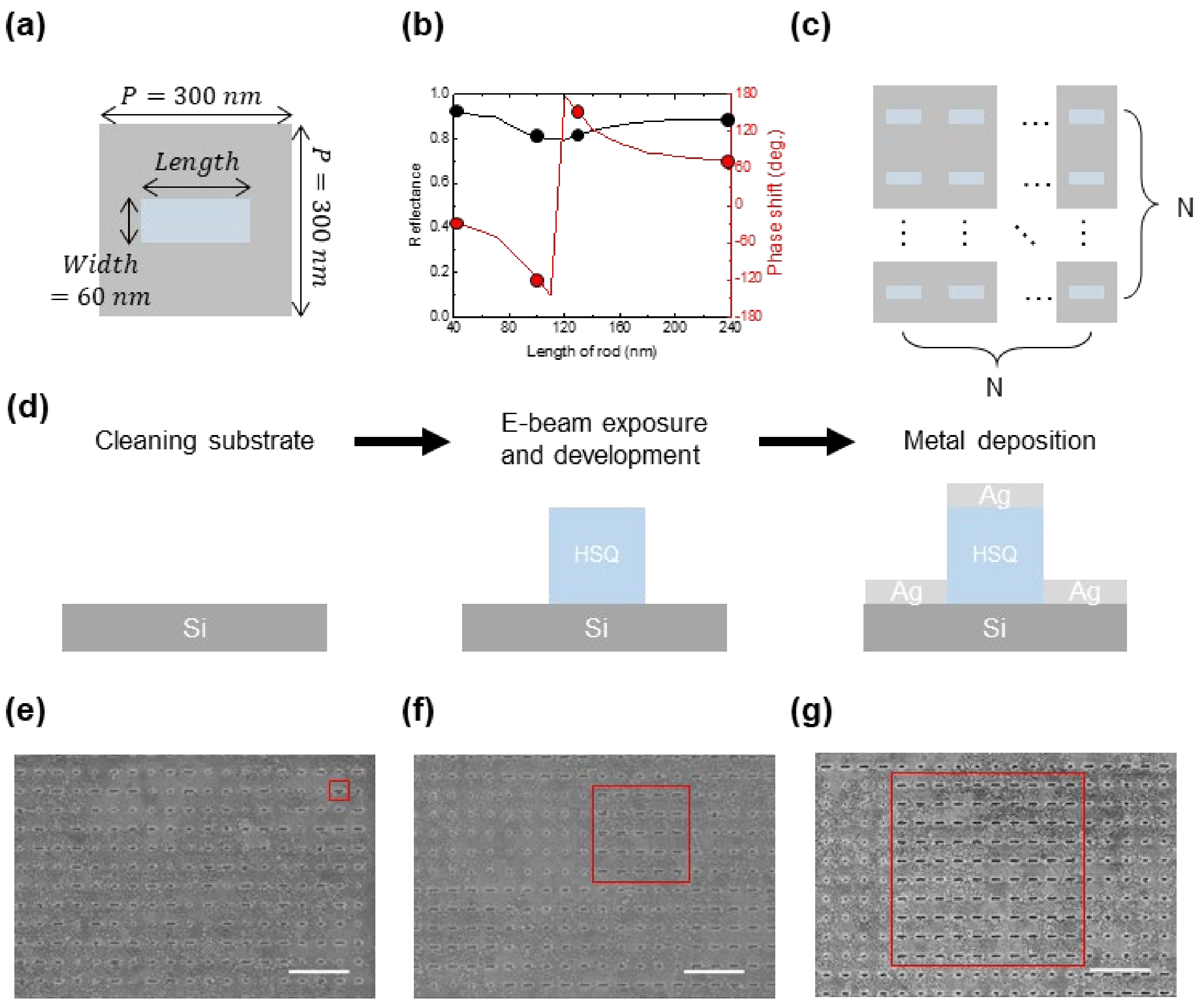
Meta-hologram design

- The phase distribution on the hologram surface is discretized with nanostructures that produce different phases
- Only the phase is usually discretized (although one could also discretize the amplitude)
- For example nanorods with different lengths produce different phases with an almost constant reflectance



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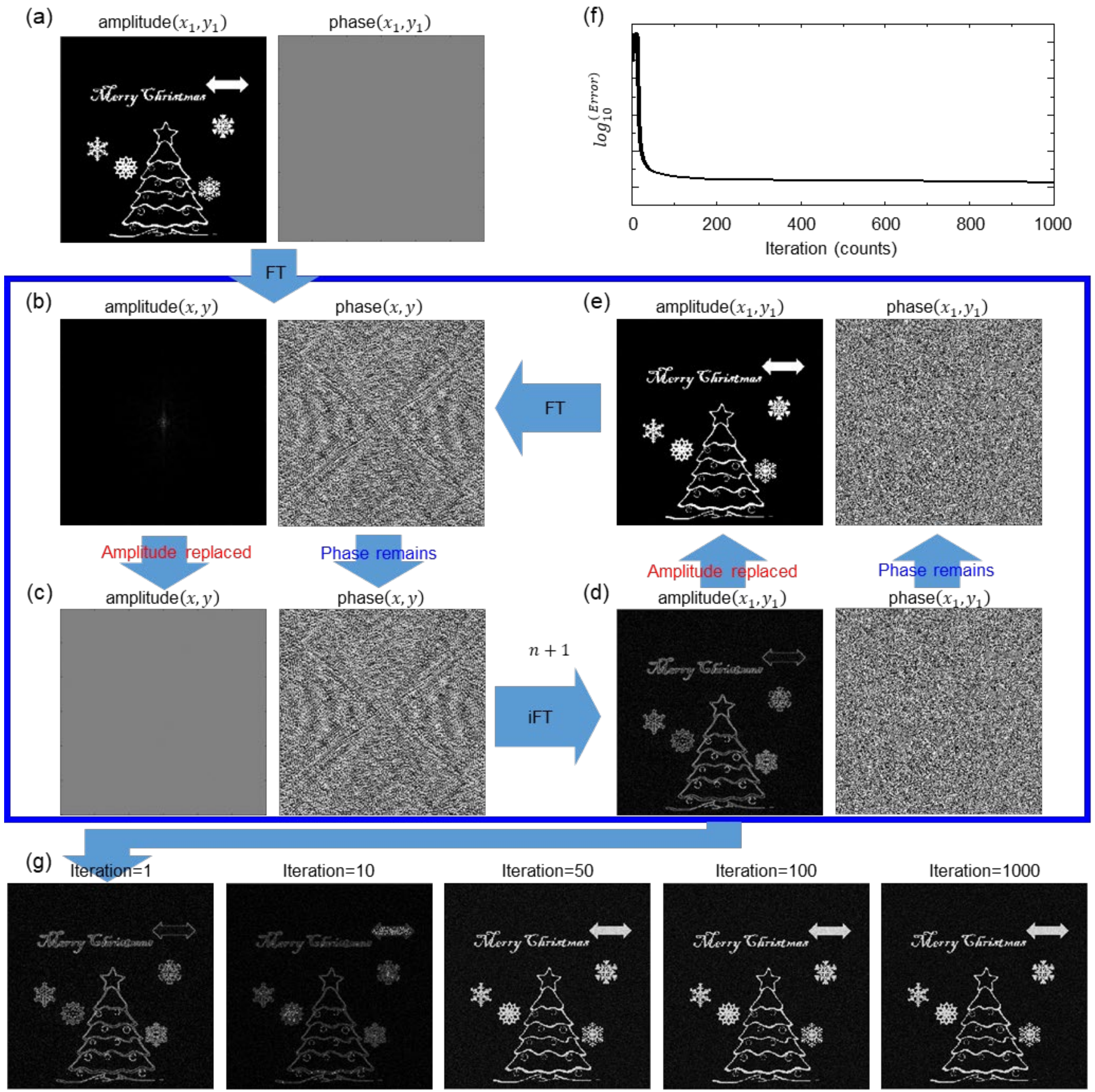
Meta-hologram design

- An iterative procedure is used to obtain the target image by only adjusting the phase
- As the iterations progress, the hologram image appears more clearly



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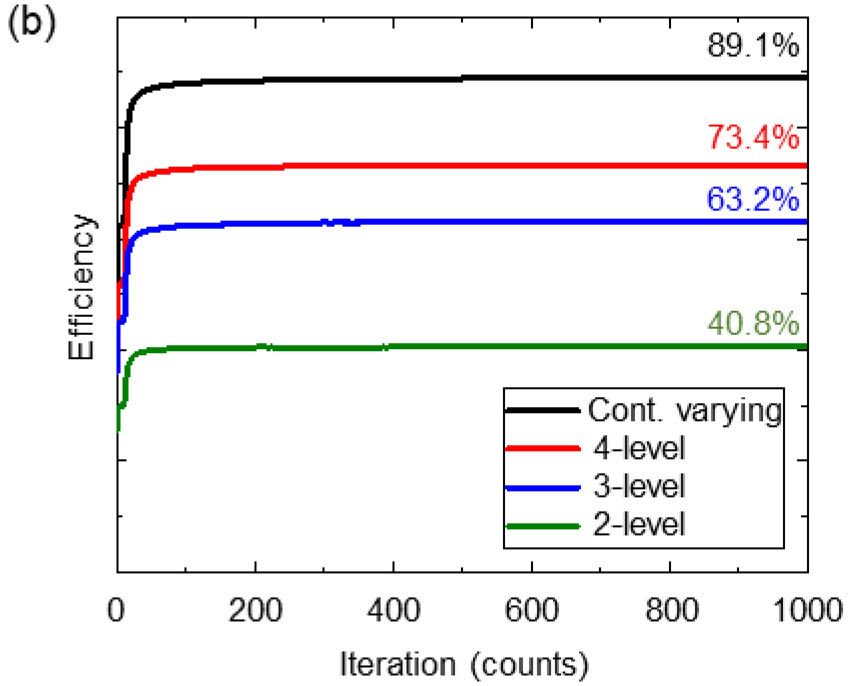
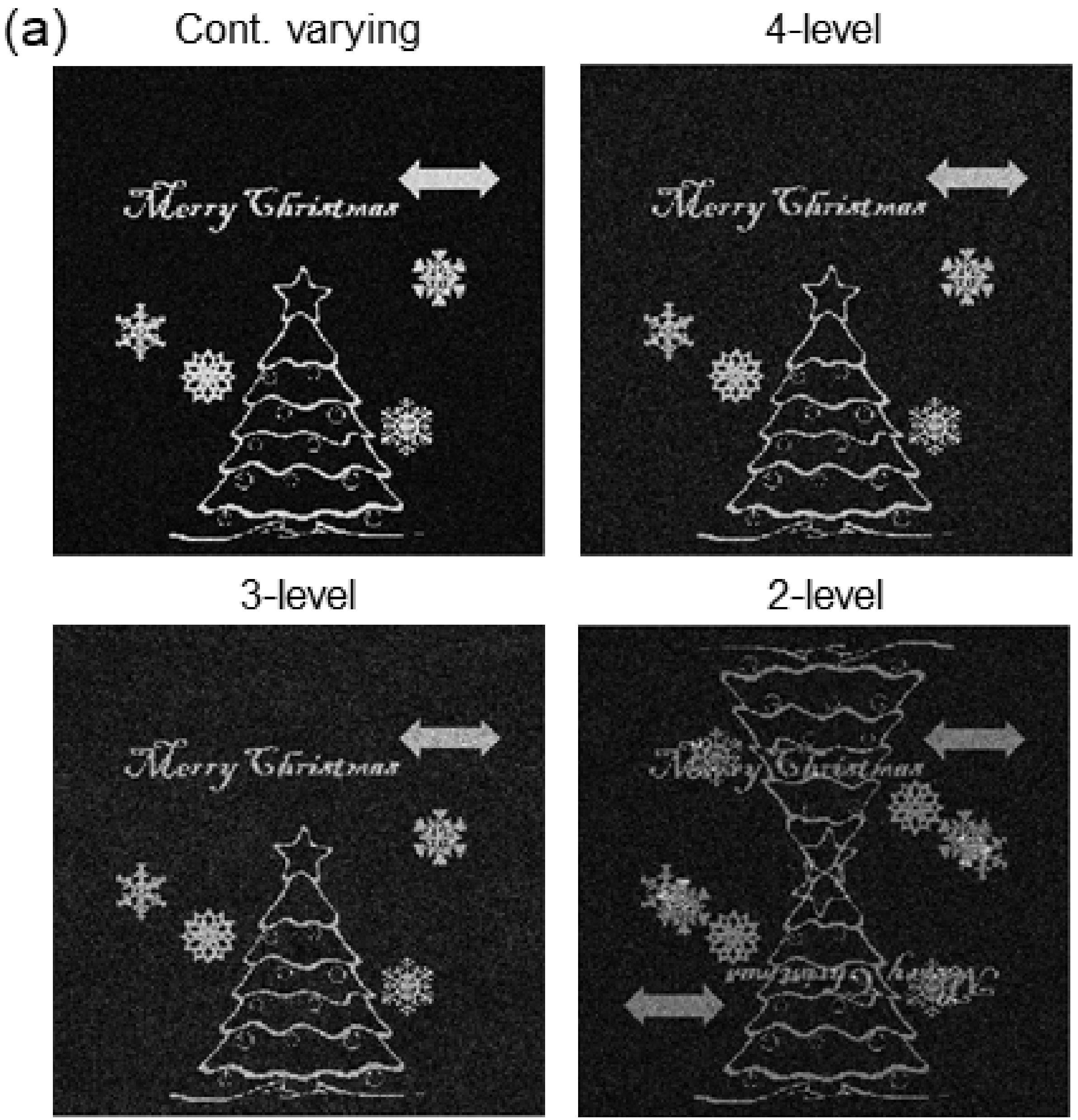
Meta-hologram design

- The hologram efficiency depends on the number of discrete phases (levels) used to reproduce the image
- Some spurious effects can occur when only 2-levels are used





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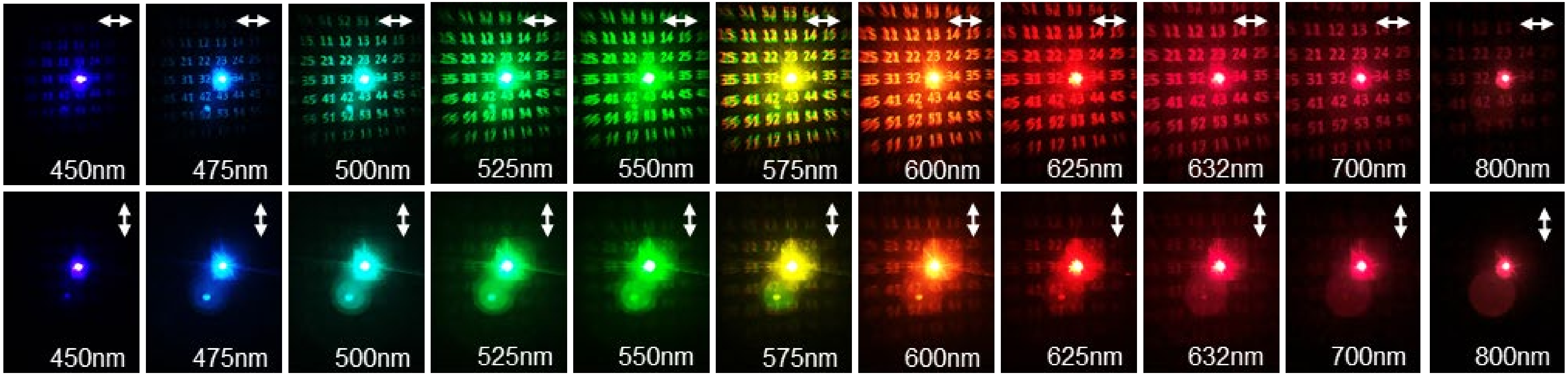
Meta-hologram design

- Depending on the nanostructures used, the rendering is polarization-dependent
- Since the nanostructure resonance depends on the wavelength, the hologram will also change with the illumination wavelength



Selected Topics in Advanced Optics

Week 12 – part 4

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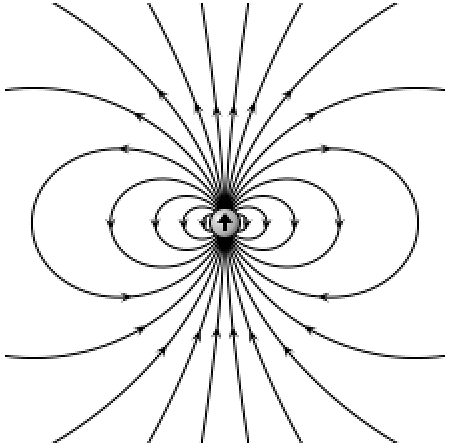
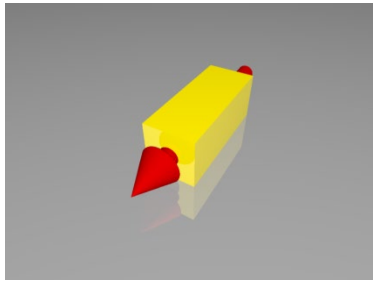
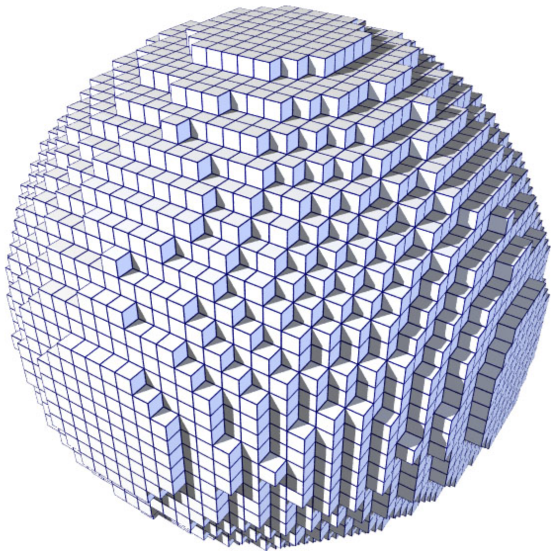
Plasmonic bright mode

- A bright mode is a mode that can radiate into the far-field
- The simplest and most useful bright mode is an electric dipole mode
- More complex modes can be obtained by the superposition of electric dipoles
- Numerical technique:
  coupled-dipole approximation
  (or discrete-dipole approximation)

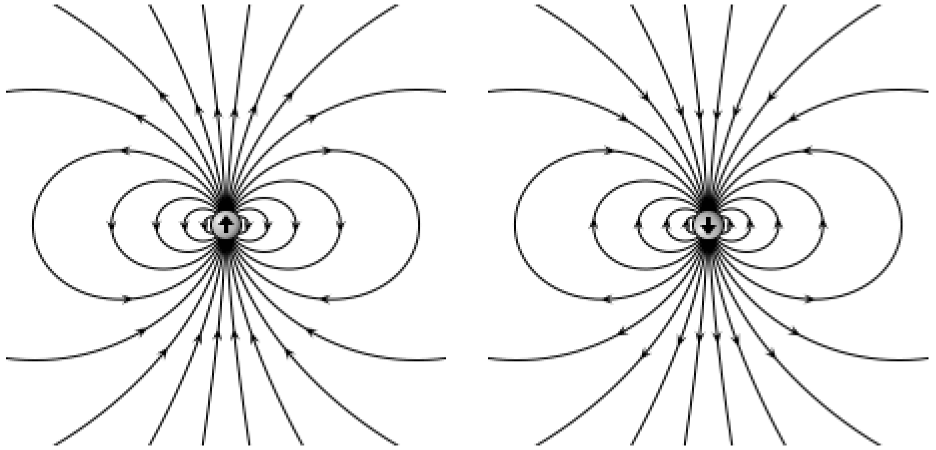


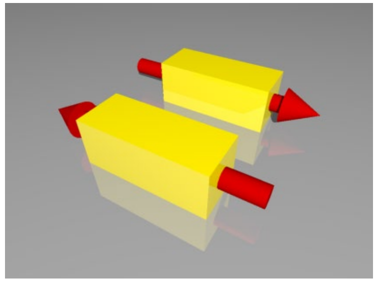




Plasmonic dark mode

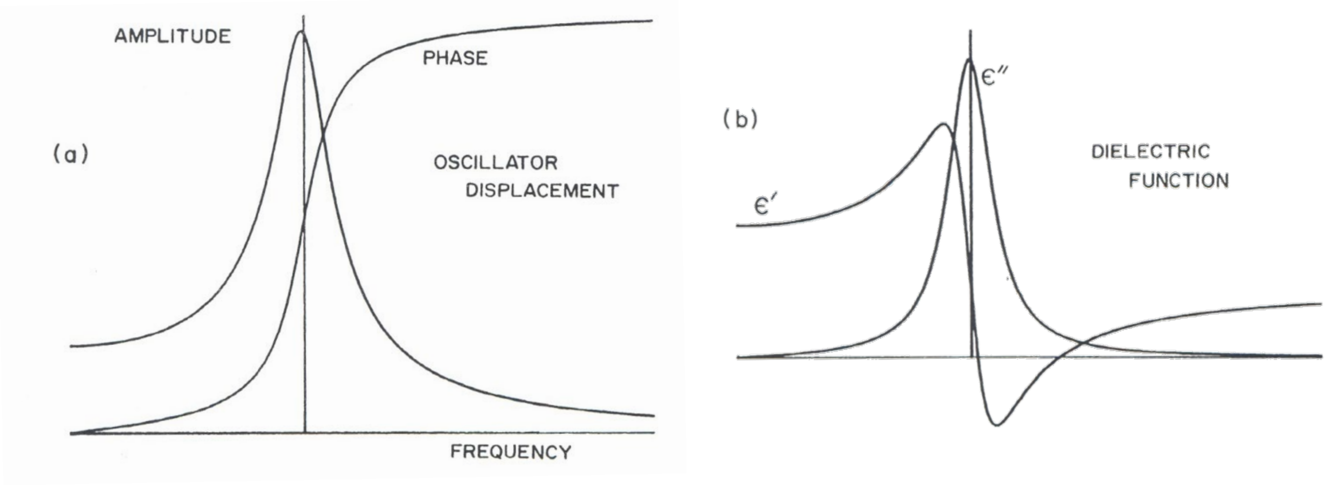
- A dark mode does not radiate into the far-field…
  … but it can have a strong near-field
- The simplest dark mode is an electric quadrupole:





- An electric quadrupole corresponds to a small current loop (magnetic resonance)

Lorentz model

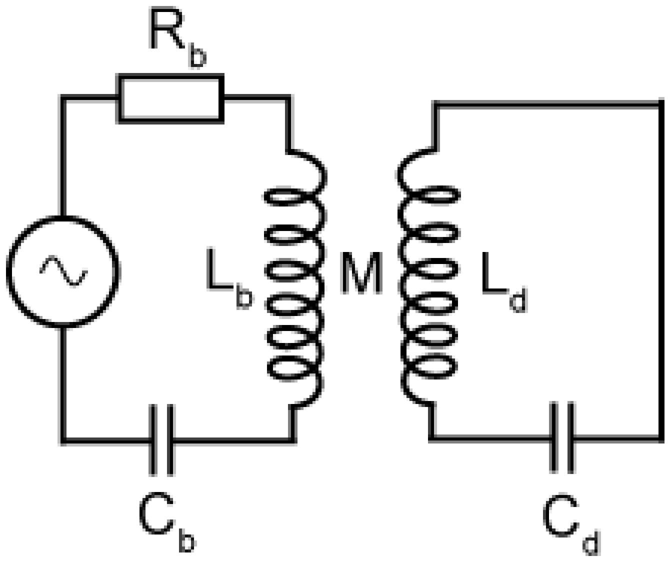


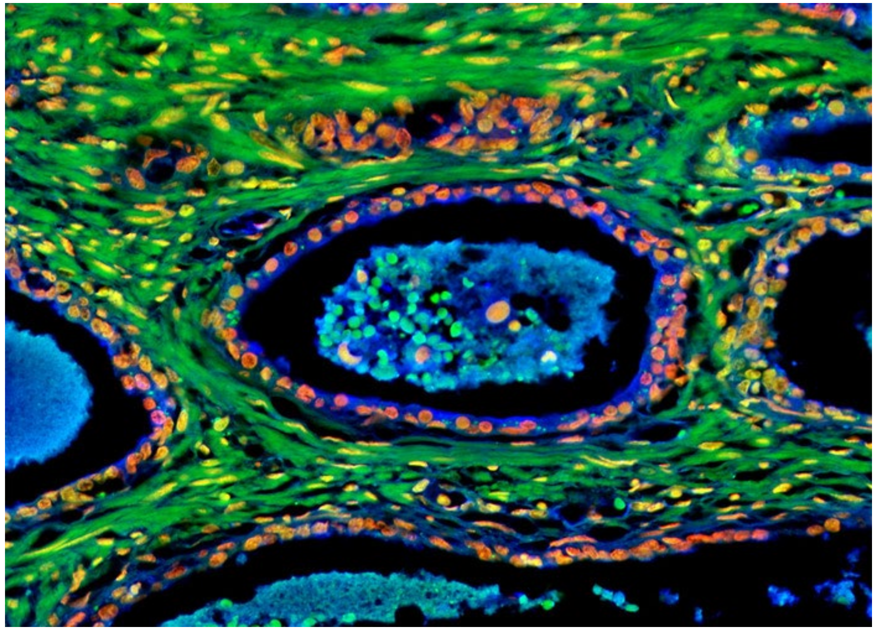
- The amplitude is maximum for $\omega = \omega_0$ and the width inversely proportional to γ
- At low frequency the oscillator is in-phase $(\Theta = 0)$ and at high frequency it is out of phase by 180°. The change occurs at $\omega \simeq \omega_0$
- The induced dipole moment of a single oscillator is $\mathbf{p} = e\mathbf{x}$
- For a collection of n oscillators per volume unit, the polarization is $\mathbf{P} = ne\mathbf{x}$

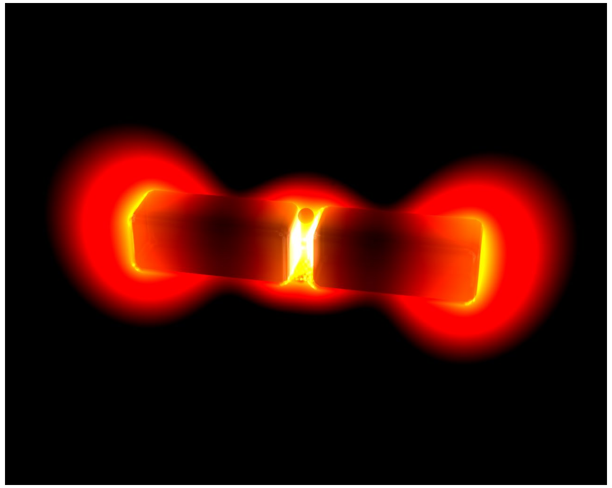
$$\mathbf{P} = \frac{\omega_p^2}{\omega_0^2 - \omega^2 - j\gamma\omega}\varepsilon_0\mathbf{E} \quad \text{plasma frequency} : \omega_p^2 = ne^2 / m\varepsilon_0$$

- Since $\mathbf{P} = \varepsilon_0 \chi \mathbf{E} \rightarrow \boxed{\varepsilon_r = 1 + \chi = 1 + \frac{\omega_p^2}{\omega_0^2 - \omega^2 - j\gamma\omega}}$

Resonant electromagnetic systems



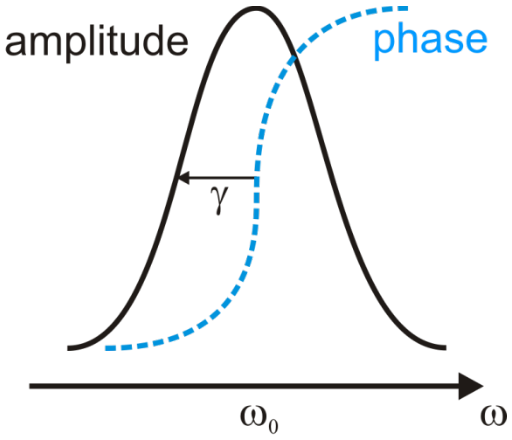




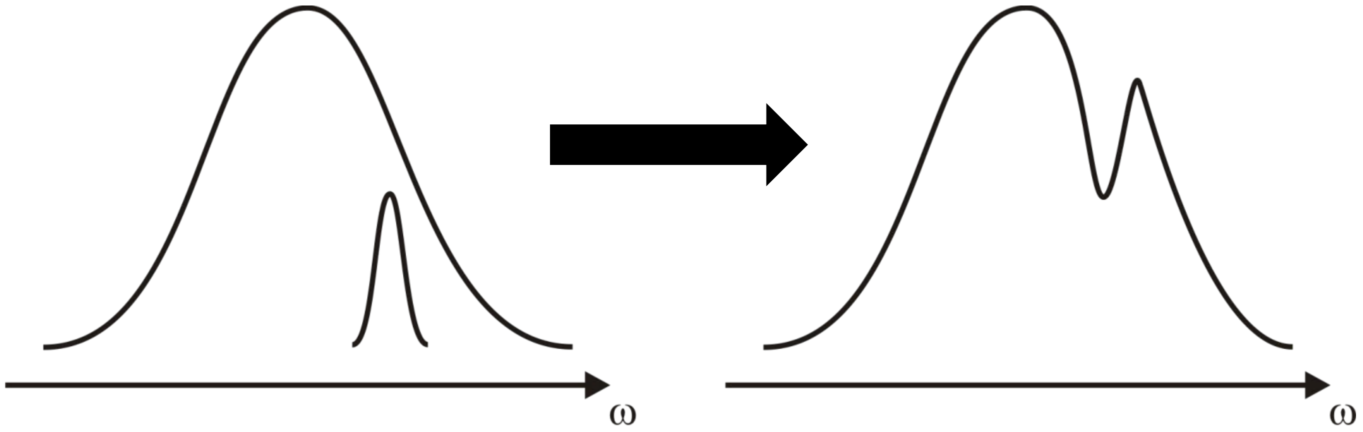
Lorentzian lineshape

- These different systems have a Lorentzian response
  (the signal amplitude as a function of energy/frequency follows a Lorentzian curve)

$$\varphi(\omega)=\frac{\gamma}{(\omega-\omega_0)^2+\gamma^2}$$

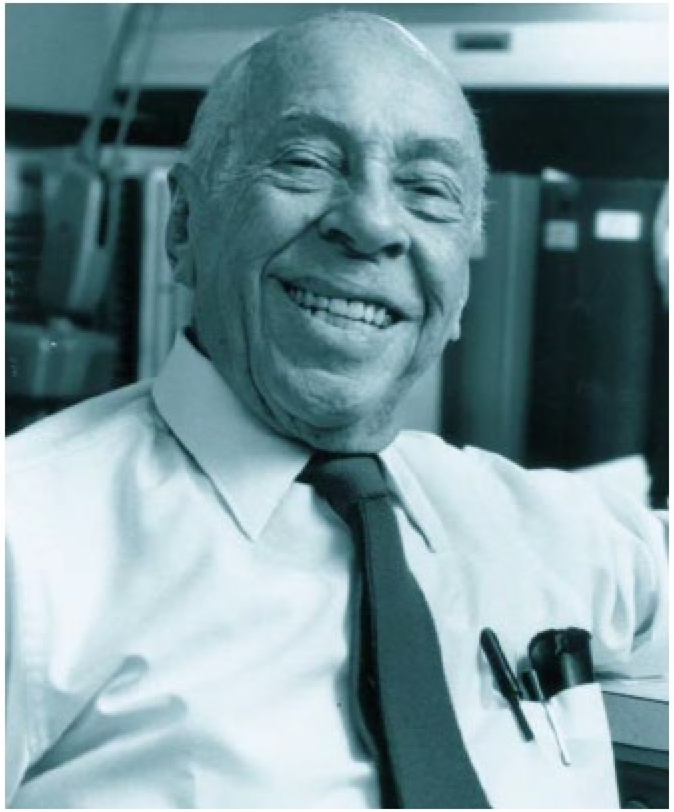


- When two oscillators are interacting, their responses can be combined, producing a Fano lineshape:



The original work of Ugo Fano in atom spectroscopy

- The asymetric lineshape originates from the inteference of two ionization processes



Ugo Fano
(1912-2001)

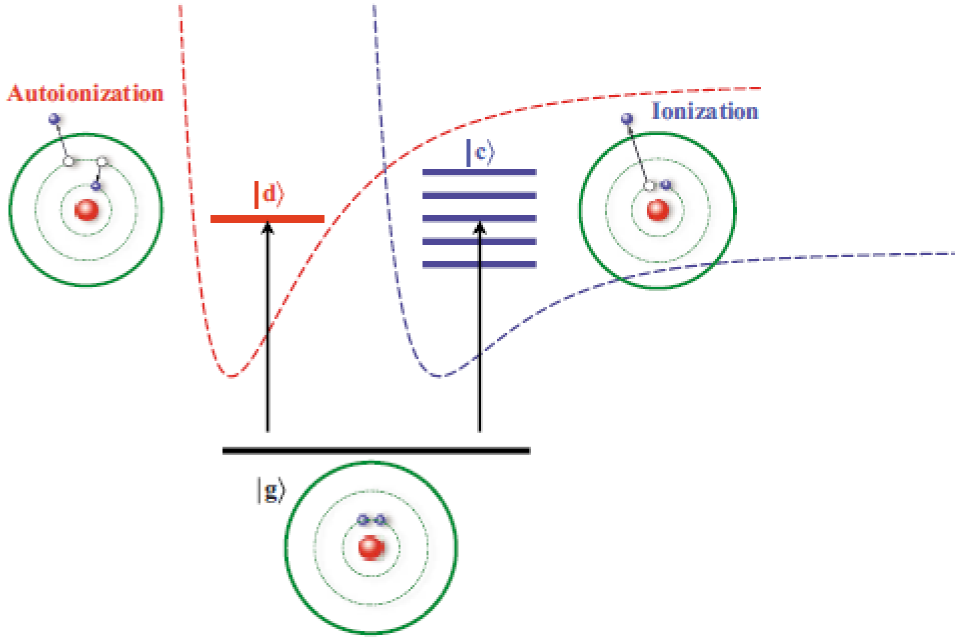
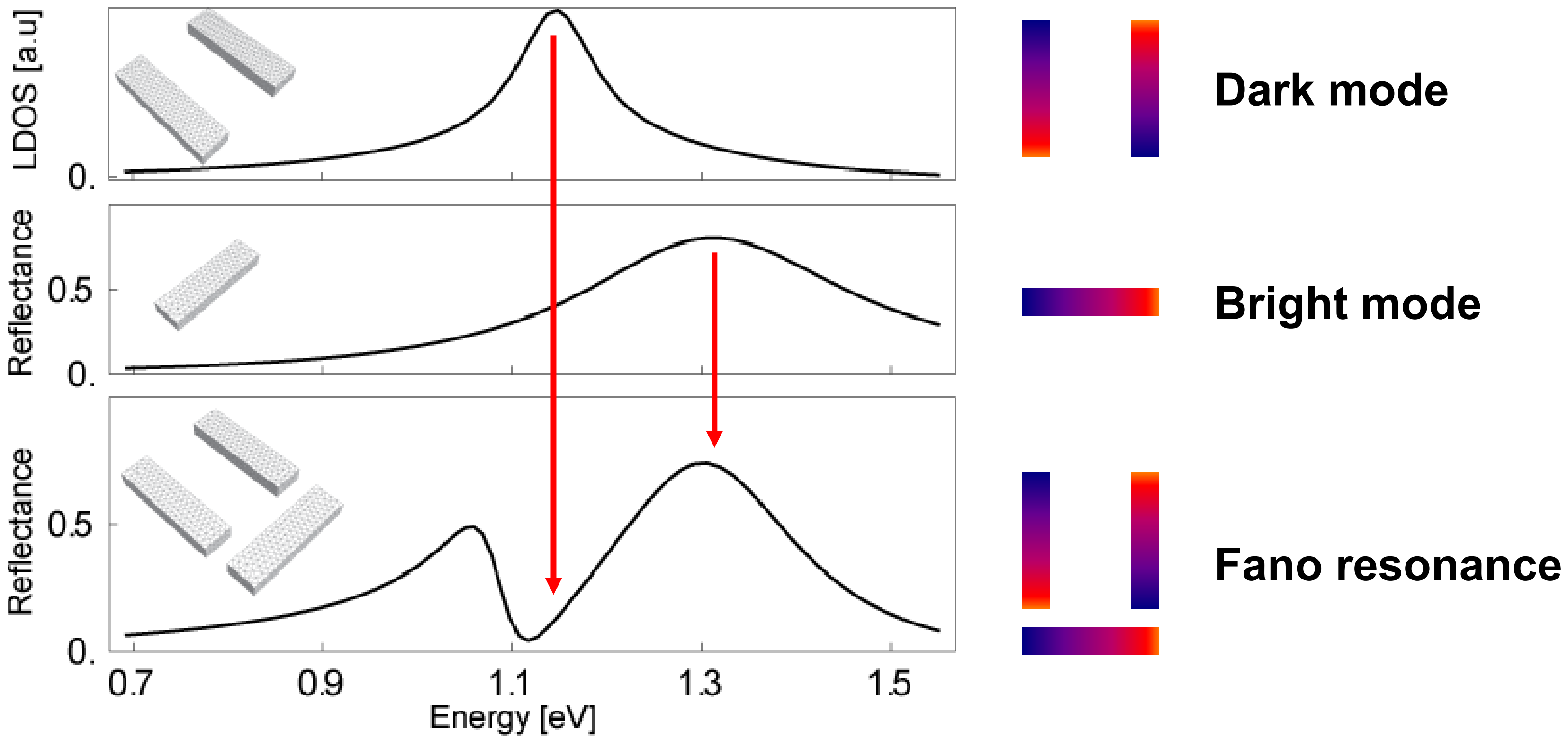


FIG. 3. (Color online) Fano resonance as a quantum interference of two processes: direct ionization of a deep inner-shell electron and autoionization of two excited electrons followed by the Auger effect. This process can be represented as a transition from the ground state of an atom $|g\rangle$ either to a discrete excited autoionizing state $|d\rangle$ or to a continuum $|c\rangle$. Dashed lines indicate double excitations and ionization potentials.

U. Fano, Phys. Rev. vol. 124, p. 1866 (1961)

Fano resonances in plasmonic systems

- Constructive and destructive interference of a bright mode with a dark mode



- Fano resonances are best studied in energy scale
- The interference can be constructive or destructive, depending on the portion of the spectrum

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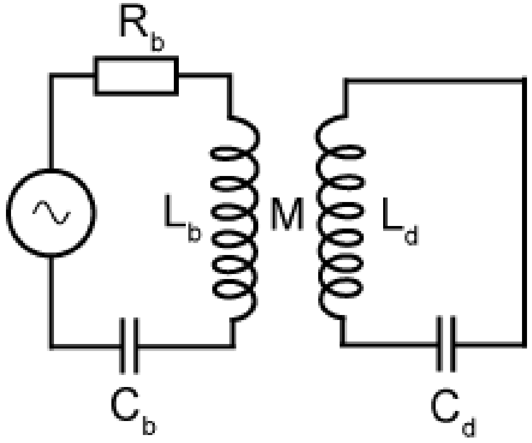
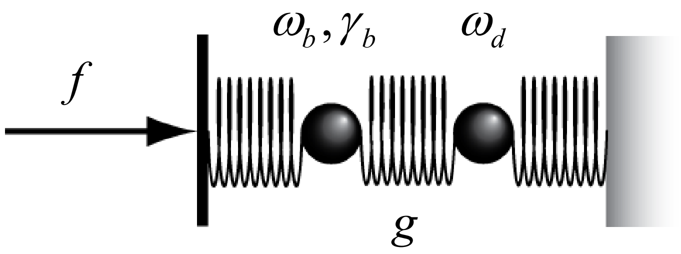
Coupled oscillator models

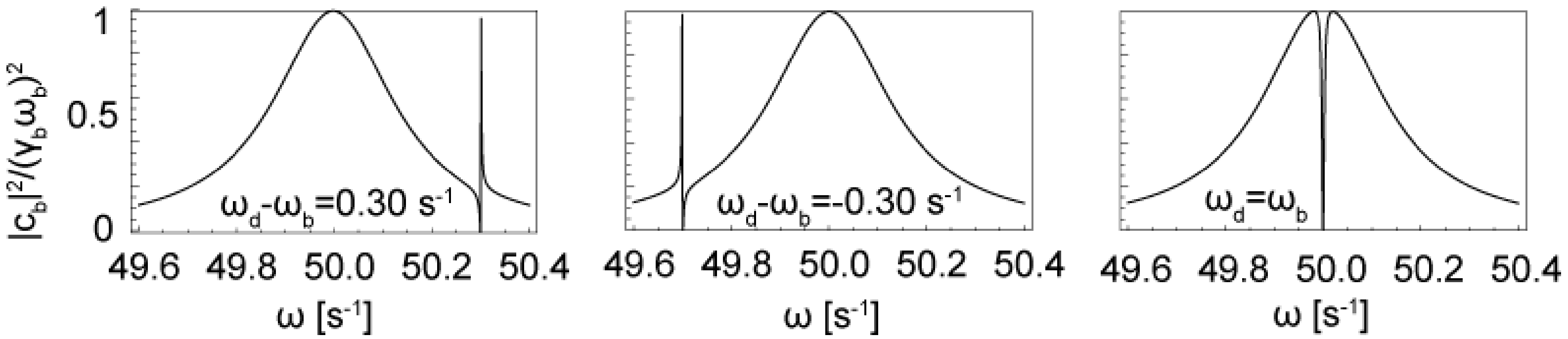
Bright mode Dark mode



Bright mode Dark mode



- Bright mode response: $$c_b = \frac{\omega_d^2 - \omega^2}{\left(\omega_b^2 + i\gamma_b\omega - \omega^2\right)\left(\omega_d^2 - \omega^2\right) - g^2} f$$

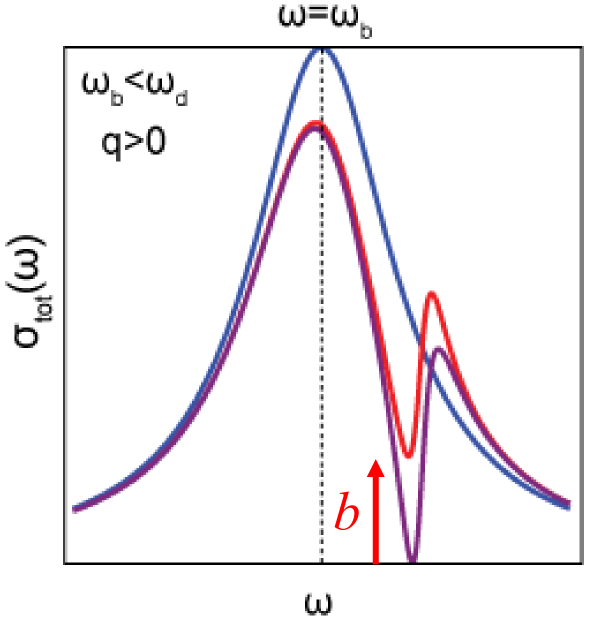


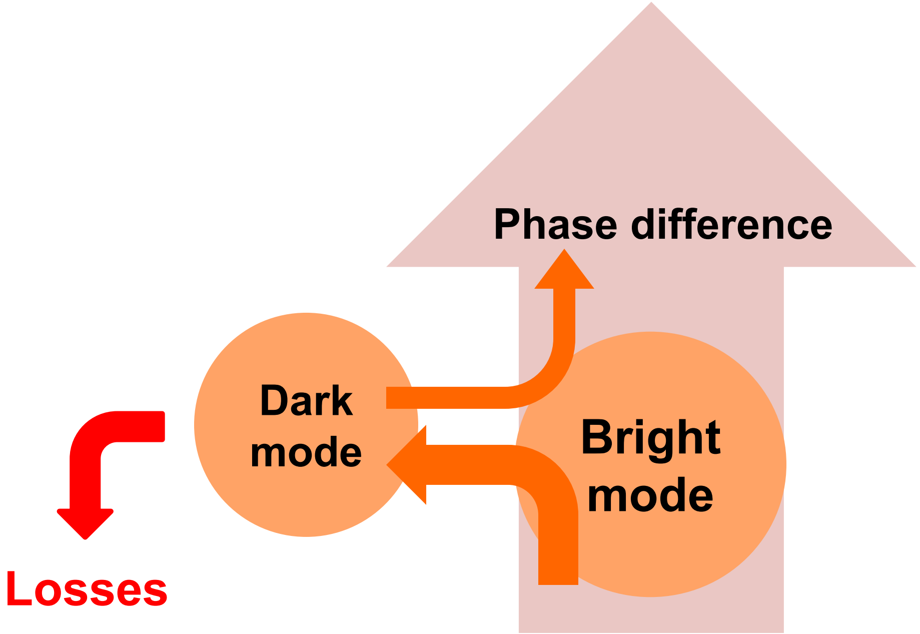
W.E. Lamb, Phys. Rev. ***81****, 222 (1951)*

C.L.G. Alzar, Am. J. Phys. ***70****, 37 (2002)*

Y.S. Joe, Phys. Scr. ***74****, 259 (2006)*

Coupled oscillators models





- Modified Fano formula:

$$|c_b|^2 = \frac{|f|^2}{|C|^2}\frac{(\kappa+q)^2+b}{\kappa^2+1}$$

$$\Gamma = \Gamma_i + \Gamma_c$$

Intrinsic losses — Coupling losses

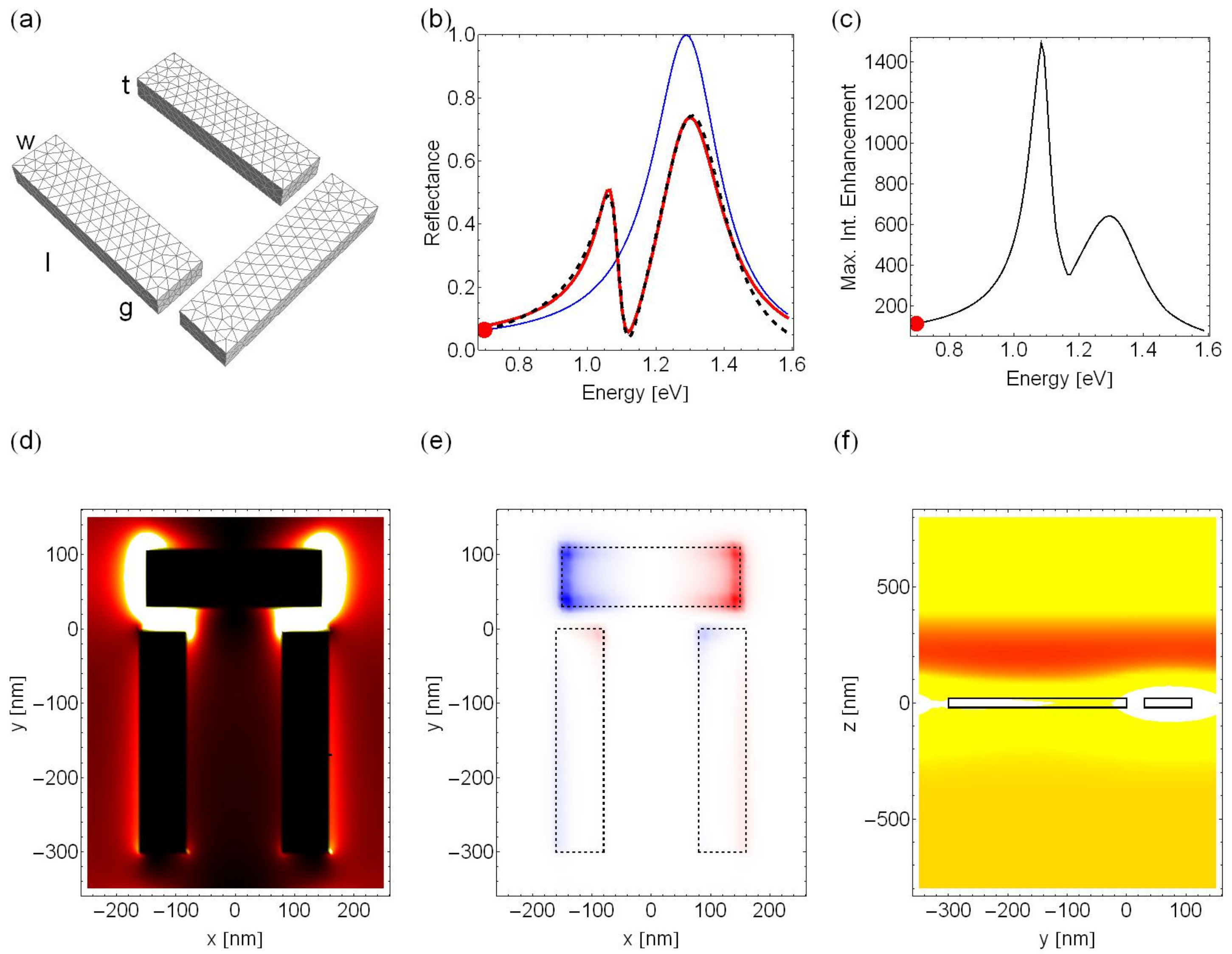
- Modulation damping by losses:

$$b = \frac{\Gamma_i^2}{(\Gamma_i+\Gamma_c)^2}$$

$$q = \frac{\omega_d^2-\omega_b^2}{\gamma_b\omega_d\left(1+\Gamma_i/\Gamma_c\right)}$$

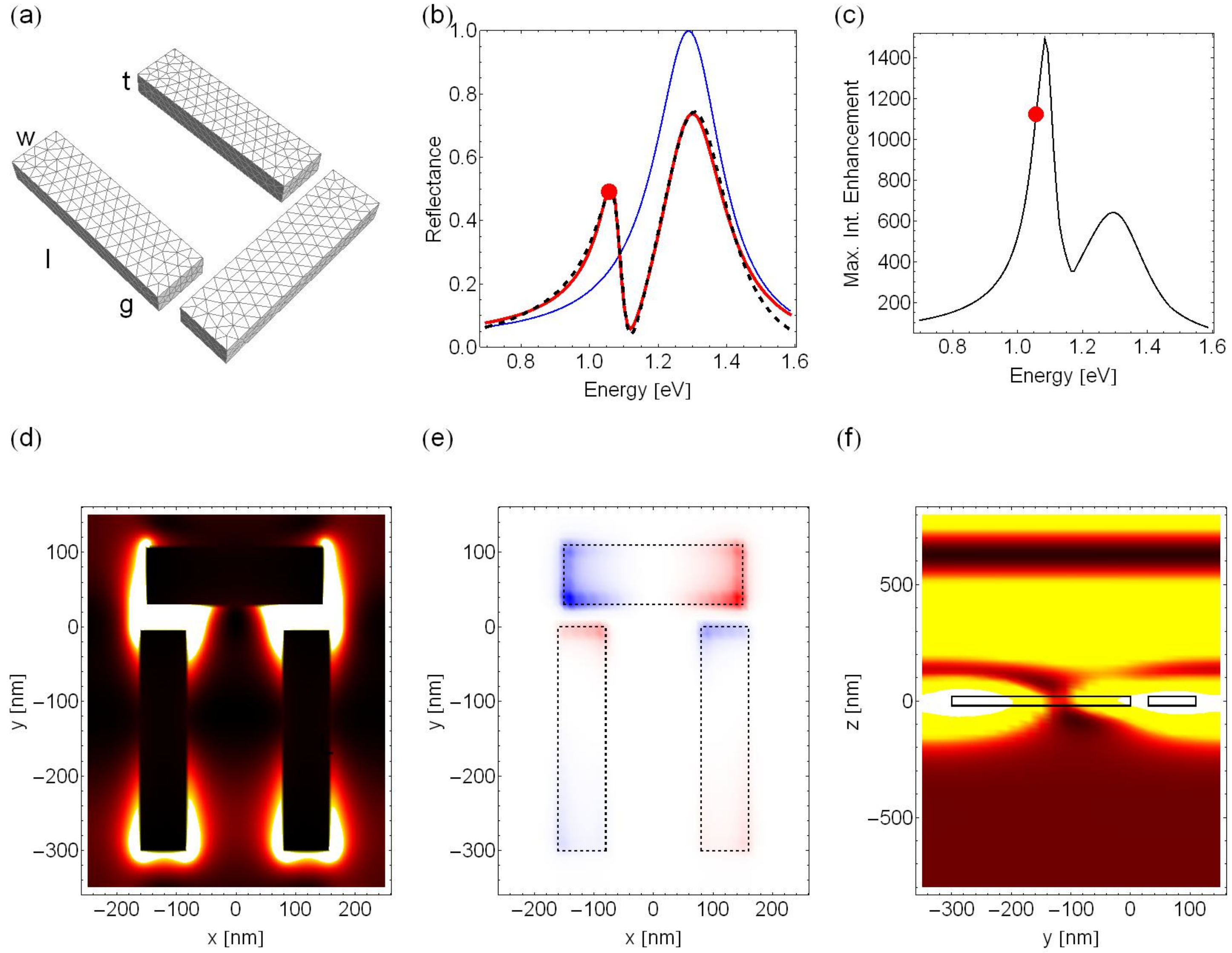
B. Gallinet, Phys. Rev. B vol. 83, p. 235427 (2011)

Combining bright and dark modes



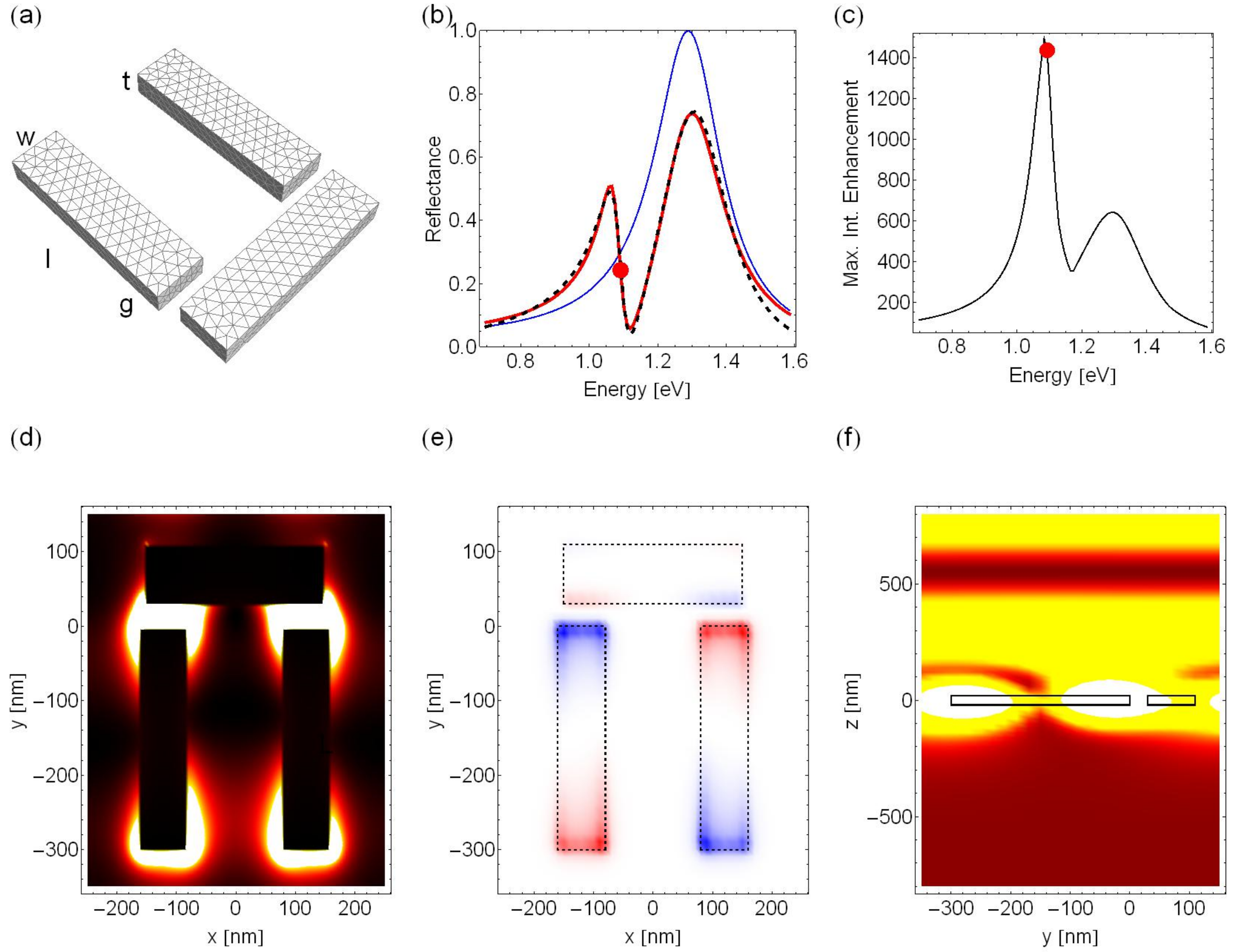
B. Gallinet, Optics Express vol. 19, p. 22167 (2011)

Combining bright and dark modes



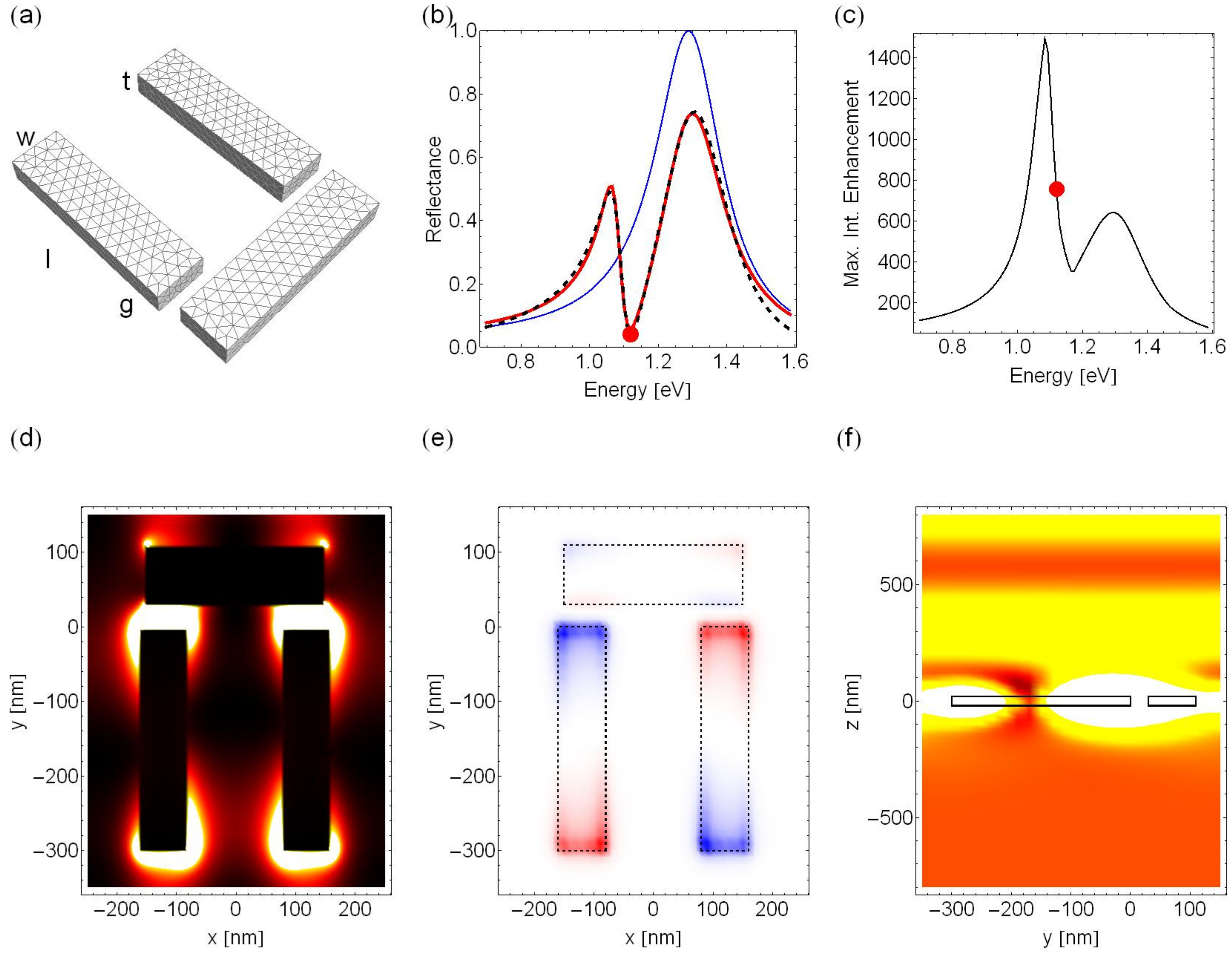
B. Gallinet, Optics Express vol. 19, p. 22167 (2011) Olivier J.F. Martin

Combining bright and dark modes



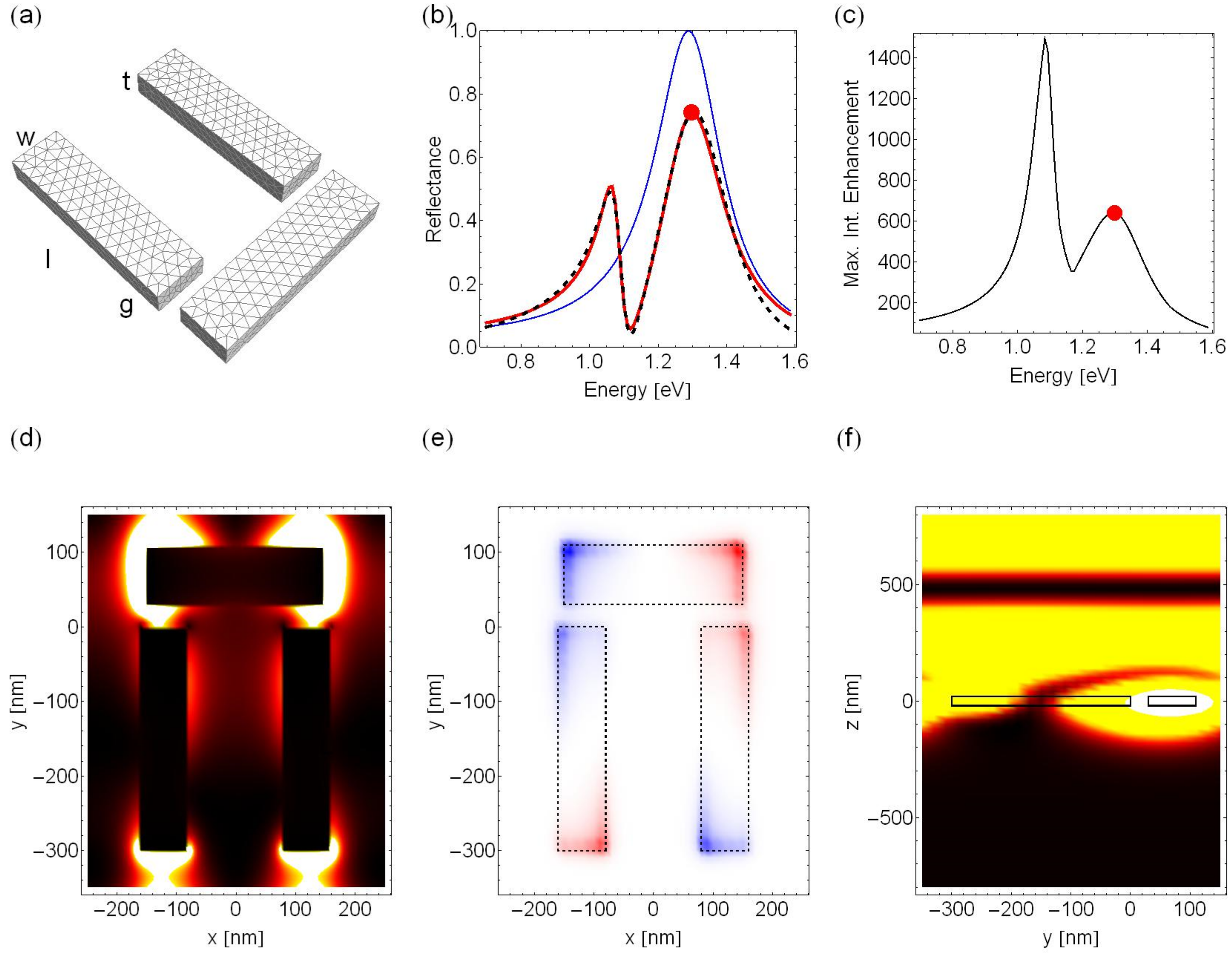
B. Gallinet, Optics Express vol. 19, p. 22167 (2011) Olivier J.F. Martin

Combining bright and dark modes



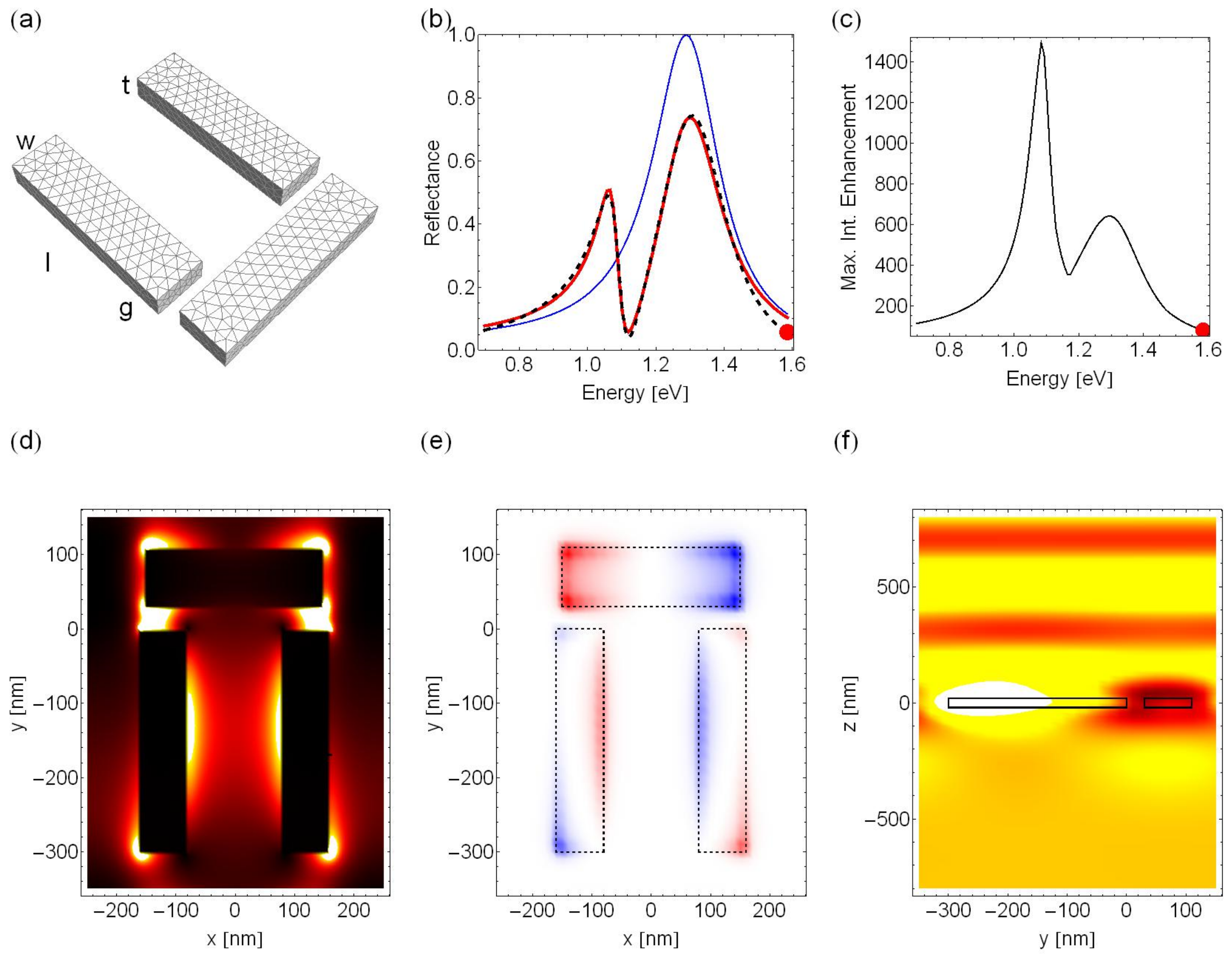
B. Gallinet, Optics Express vol. 19, p. 22167 (2011) Olivier J.F. Martin

Combining bright and dark modes



B. Gallinet, Optics Express vol. 19, p. 22167 (2011) Olivier J.F. Martin

Combining bright and dark modes

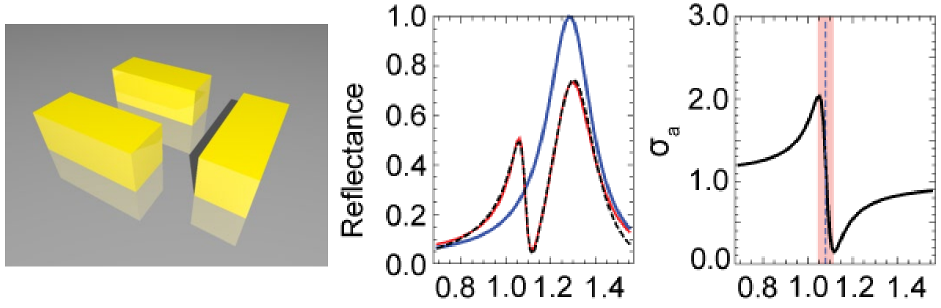


B. Gallinet, Optics Express vol. 19, p. 22167 (2011)

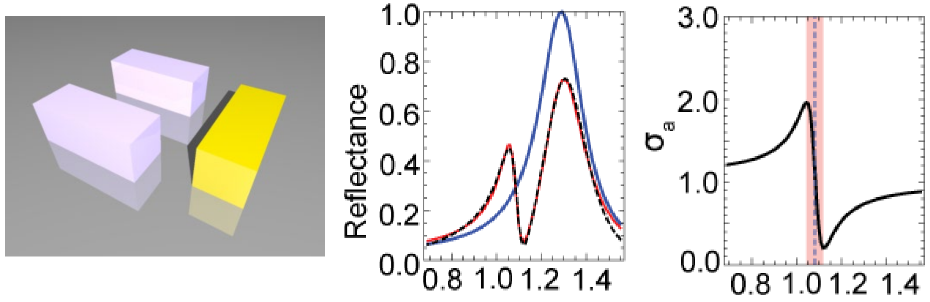
Fano resonance engineering

- Decompose the experimental signal by fitting the formula

Numerical/experimental (black dashed) **Bright mode** (blue)
Generalized formula (red) **Dark mode** (black)

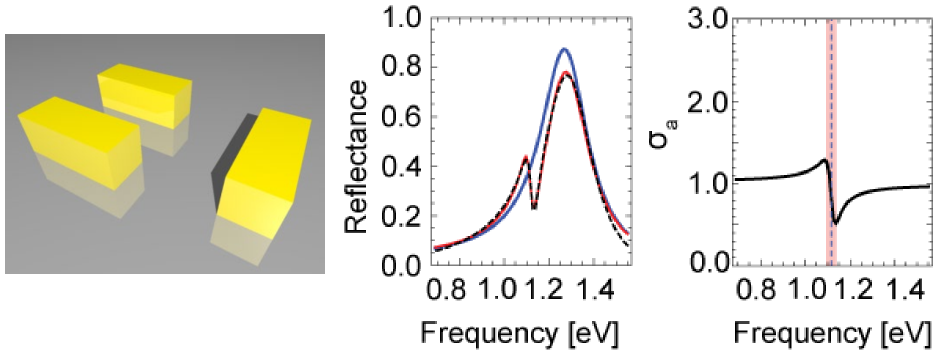


Increasing losses

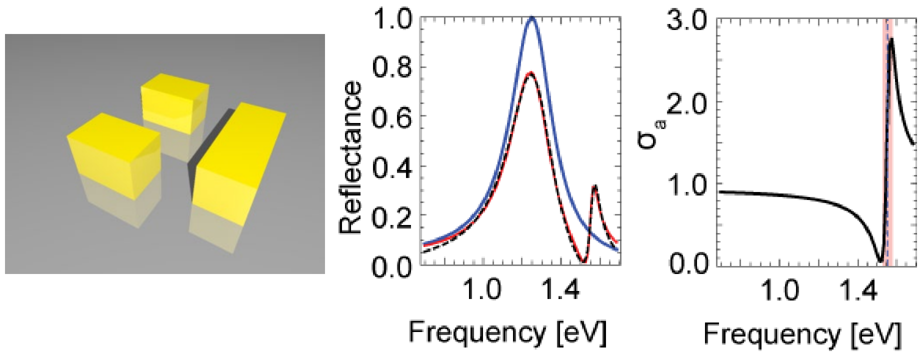


➢**Less modulation, wider resonance**

Decreasing coupling



Reversing modes detuning



➢**Less modulation, narrower resonance** ➢**Asymmetry reversal**

B. Gallinet, ACS Nano vol. 11, p. 8999 (2011)

Selected Topics in Advanced Optics

Week 12 – part 5

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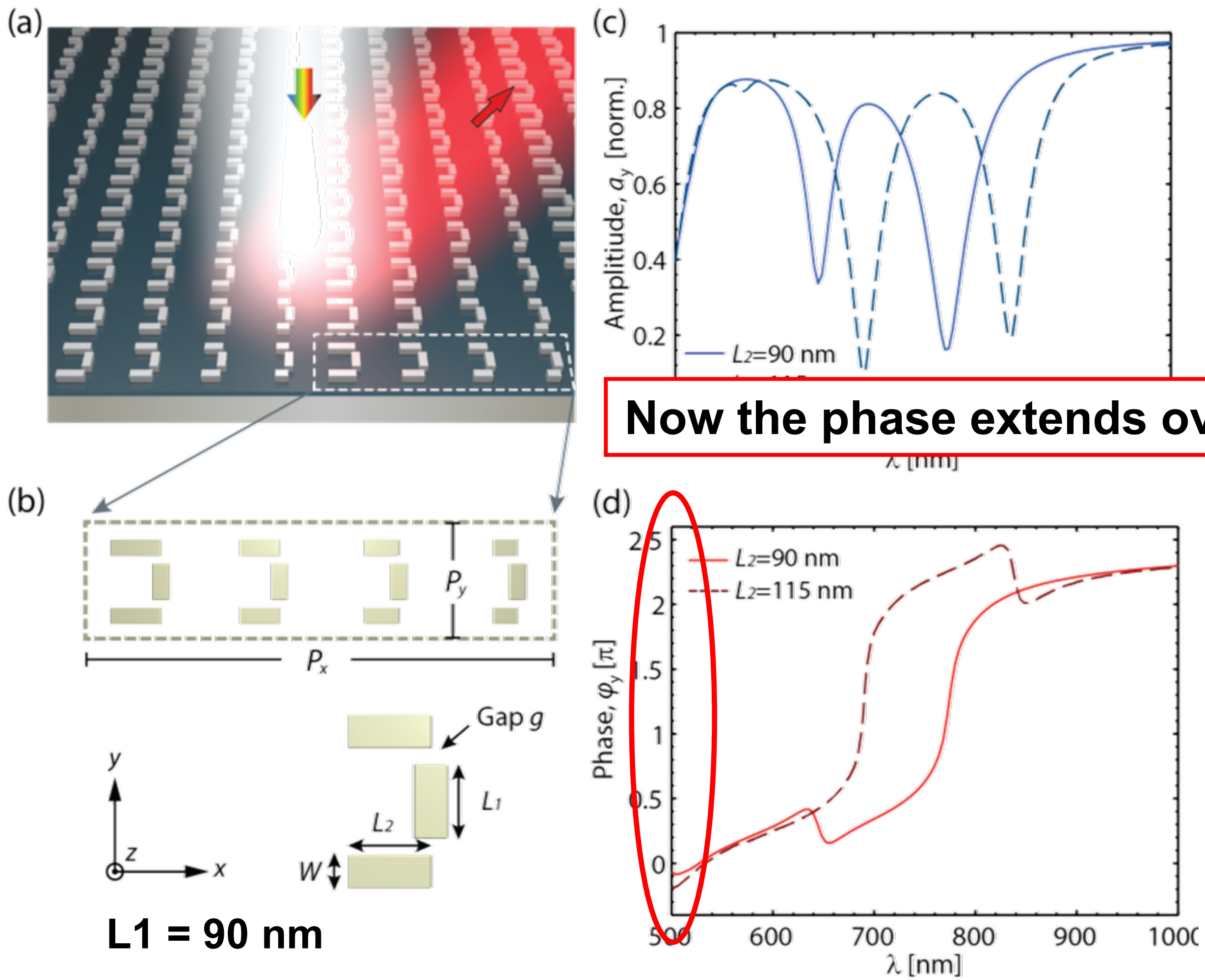
Narrow band Fano-resonant metasurface

- A cell contains 4 Fano-resonant nanostructures that each provide a different phase to the reflected wave



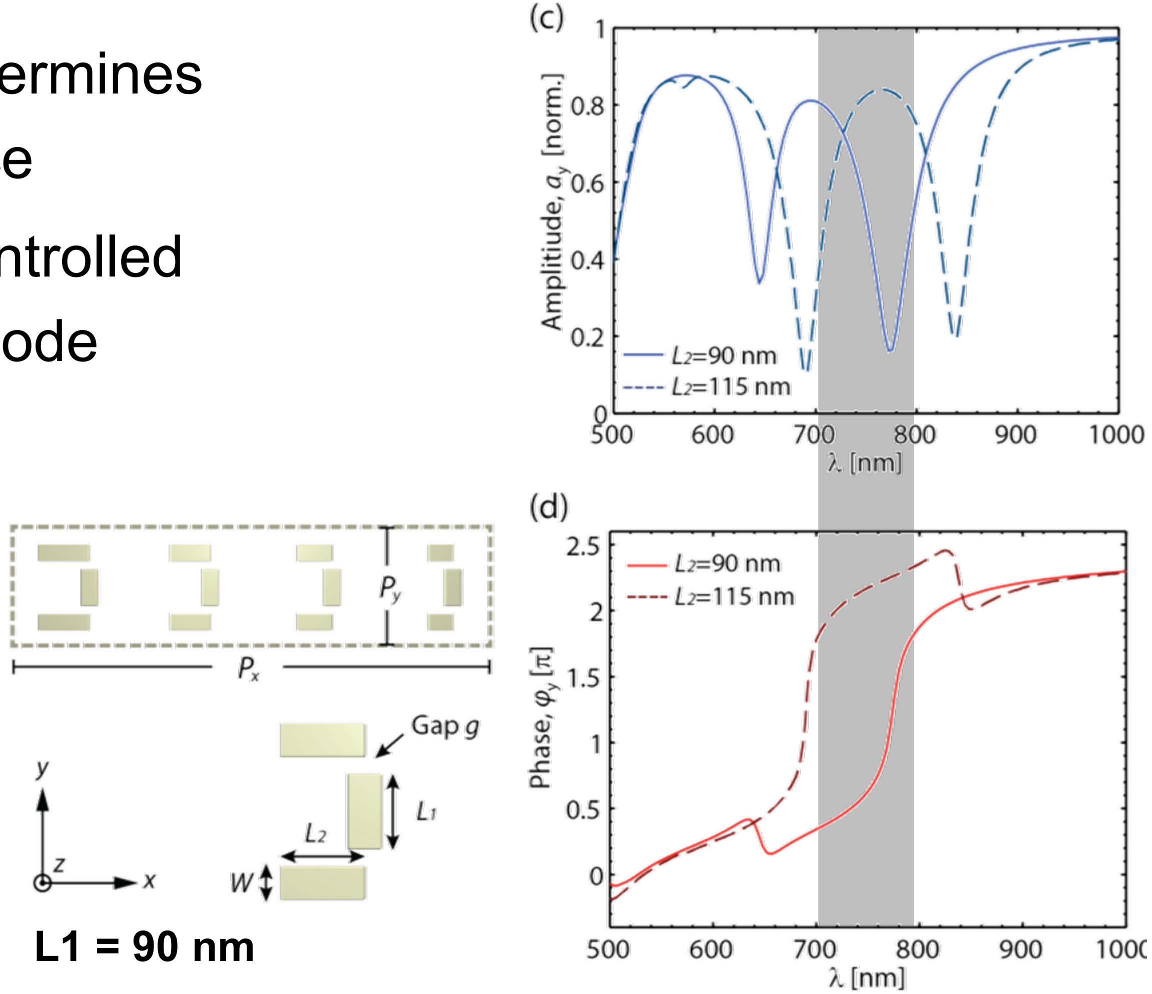
Now the phase extends over 2π

L1 = 90 nm

Y. Chen, Nature Light: Science and Applications vol. 6, p. e17017 (2017)

Narrow band Fano-resonant metasurface

- A cell contains 4 Fano-resonant nanostructures that each provide a different phase to the reflected wave
- The bright mode determines the central resonance
- The bandwidth is controlled by tuning the dark mode

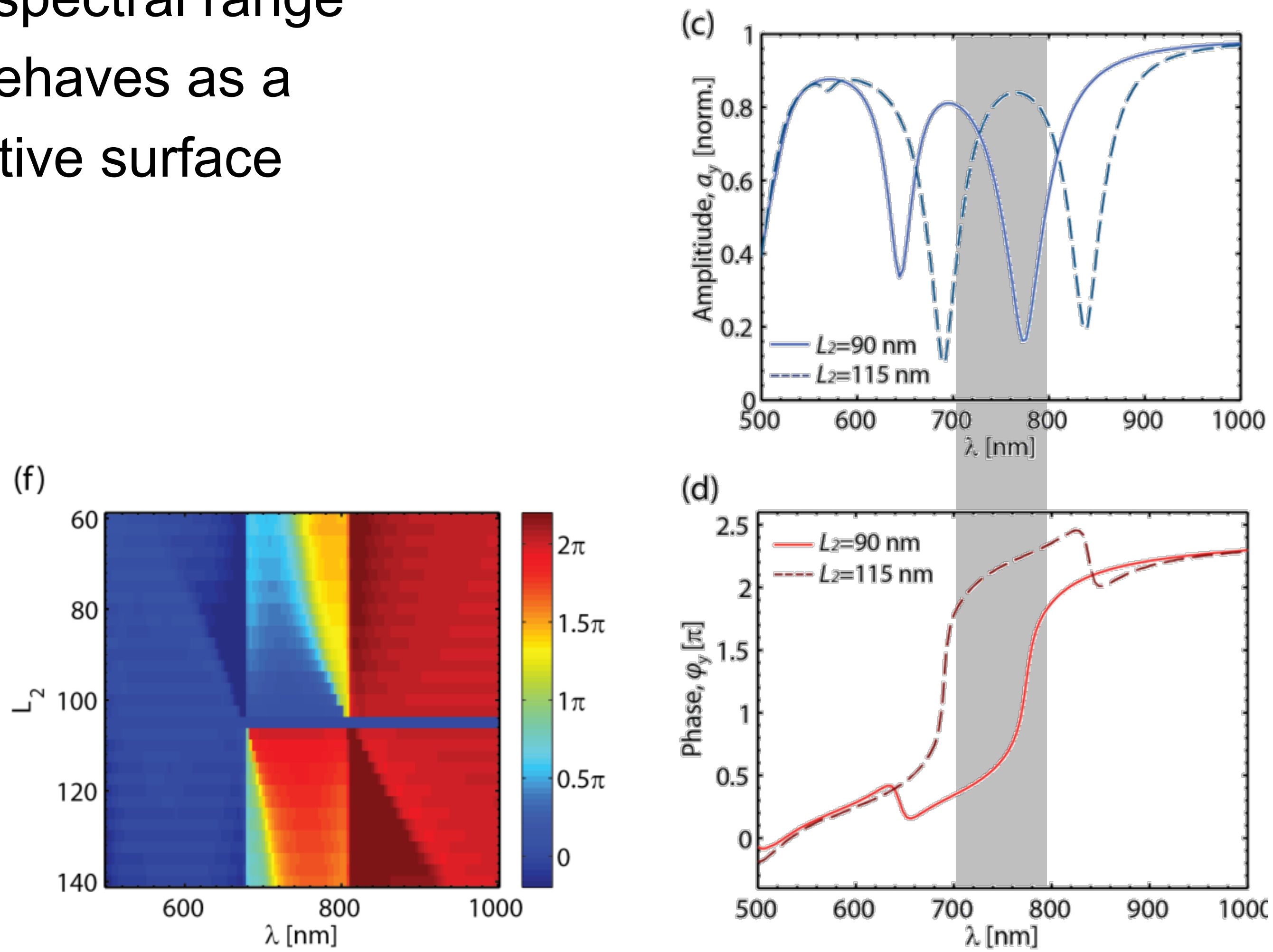


L1 = 90 nm

Y. Chen, Nature Light: Science and Applications vol. 6, p. e17017 (2017)

Narrow band Fano-resonant metasurface

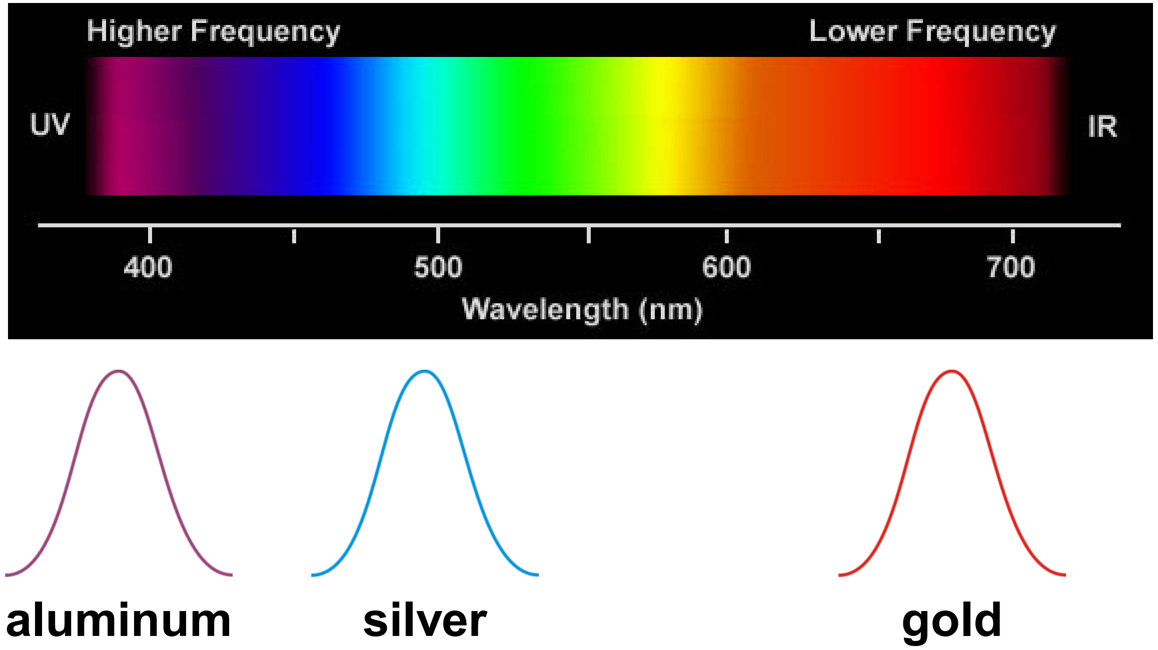
- Spectral range of ~ 100 nm with a highly tunable phase shift between 0 and 1.9 π
- Outside that spectral range the system behaves as a normal reflective surface



Y. Chen, Nature Light: Science and Applications vol. 6, p. e17017 (2017)

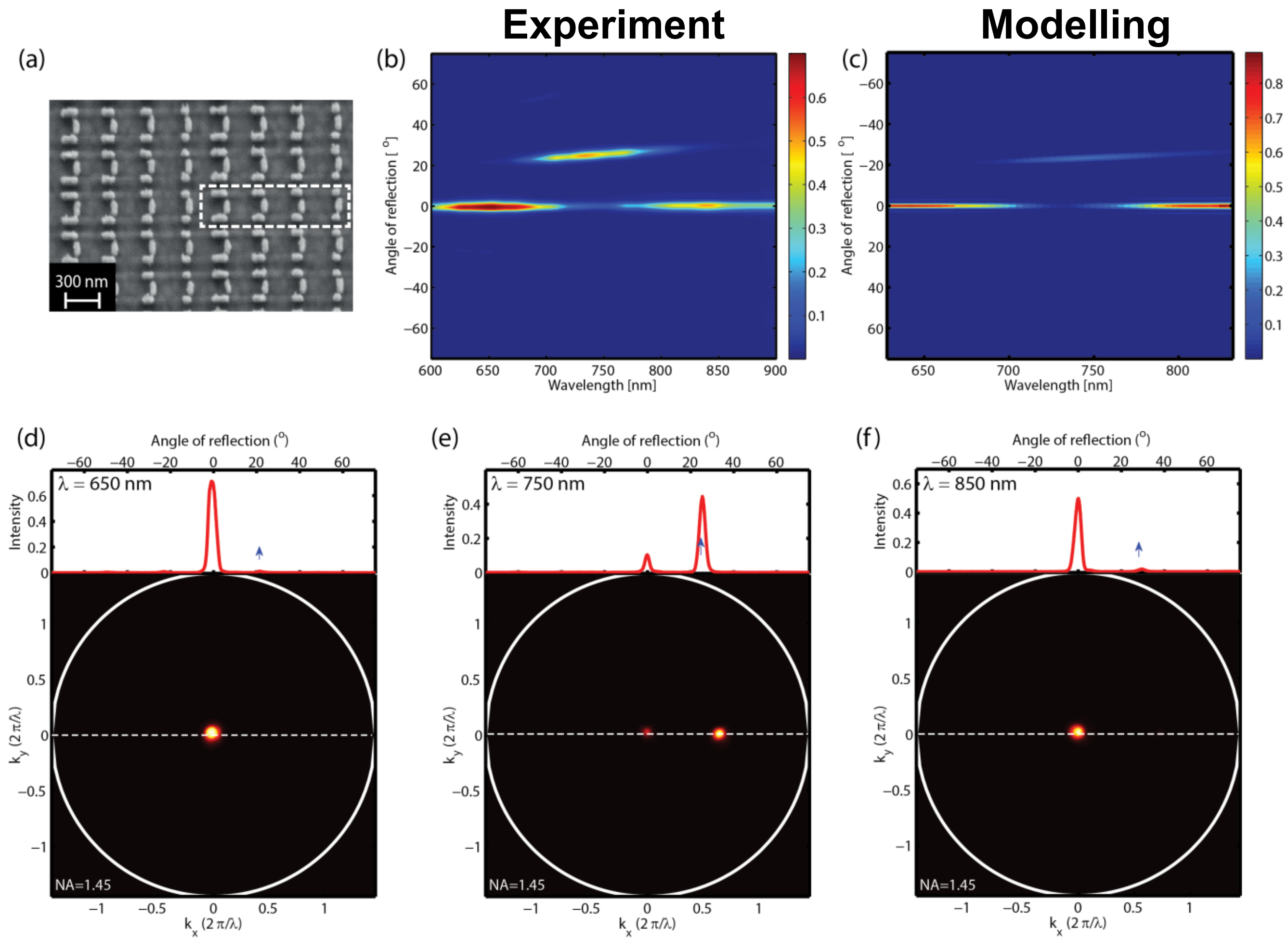
Narrow band Fano-resonant metasurface

- We want to work in the visible range, which imposes additional constrains on the possible coinage metal to be used (silver and aluminum are especially well suited)



Narrow band Fano-resonant metasurface

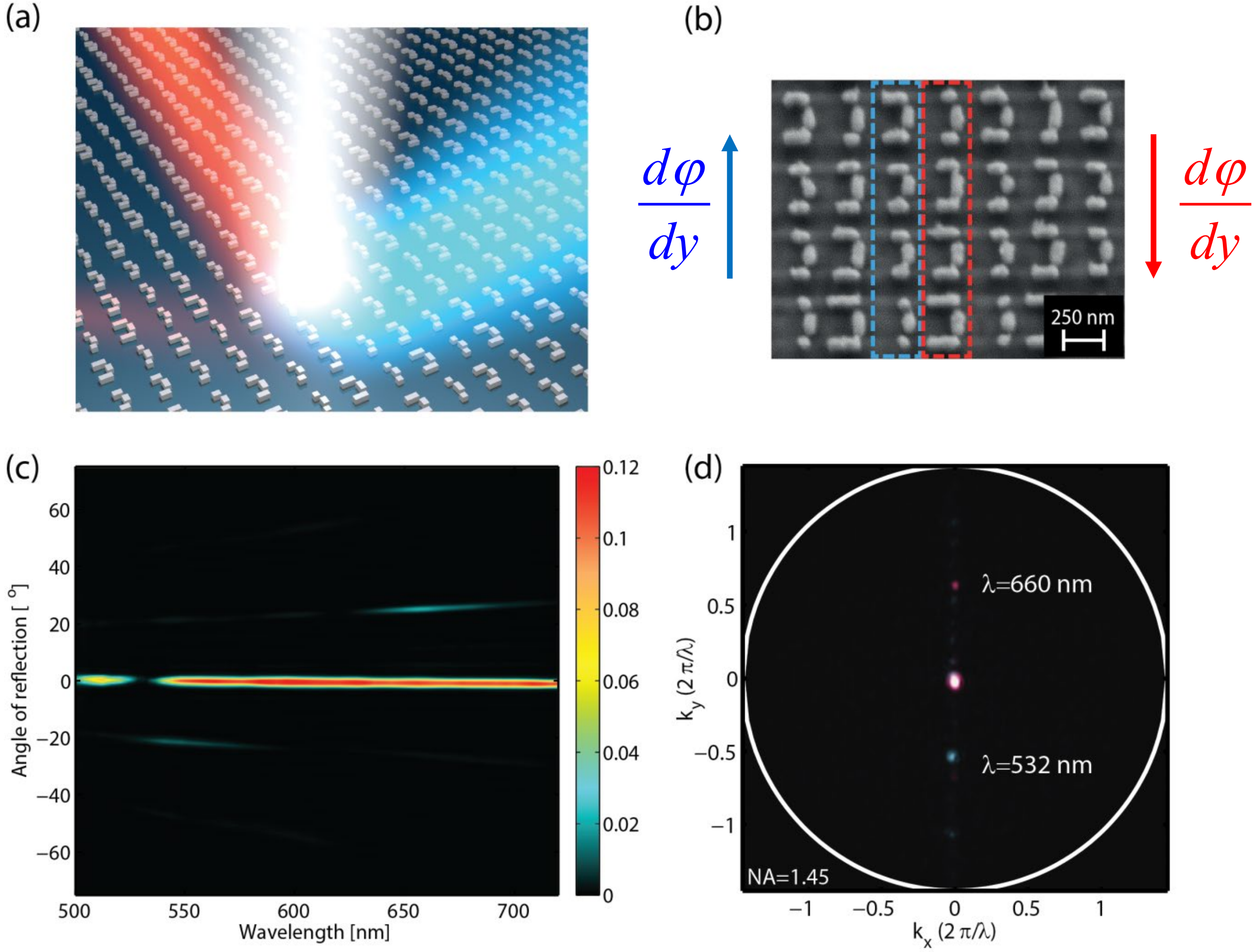
- At λ = 750 nm, anomalous reflection with high efficiency
- Good agreement with the generalized Snell law



Y. Chen, Nature Light: Science and Applications vol. 6, p. e17017 (2017)

Colour routing with a Fano-resonant metasurface

- Two different unit cells centered at λ_1 = 532 and λ_2 = 660 nm



Y. Chen, Nature Light: Science and Applications vol. 6, p. e17017 (2017)