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Diffractive coupling in gold nanorod arrays

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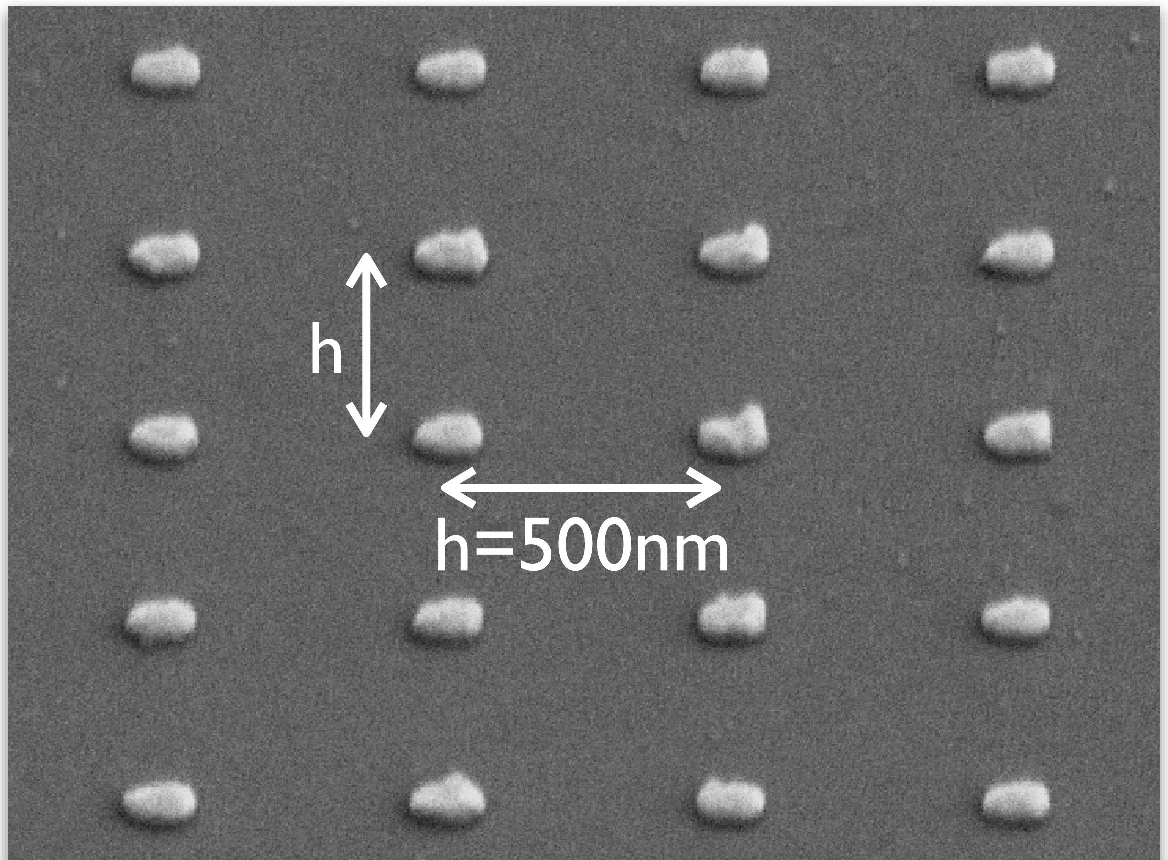
Outline

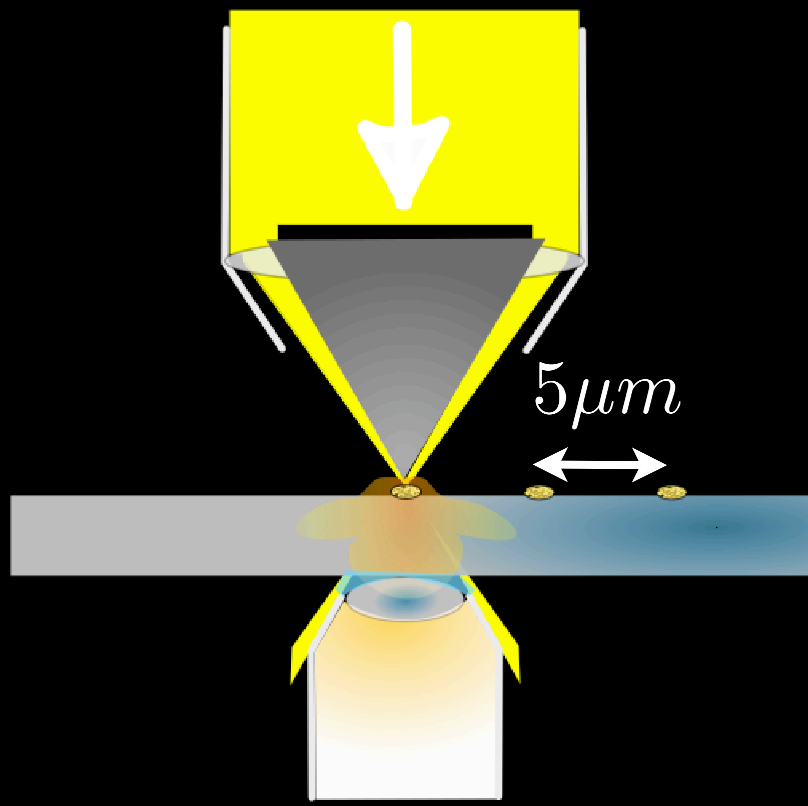
- Scattering of light by gold nanorods, localised plasmons
- Coupled dipole model: prediction of narrow spectral features
- Regular arrays: influence of the periodicity
- Varying particle size and aspect ratio

Gold nanorods

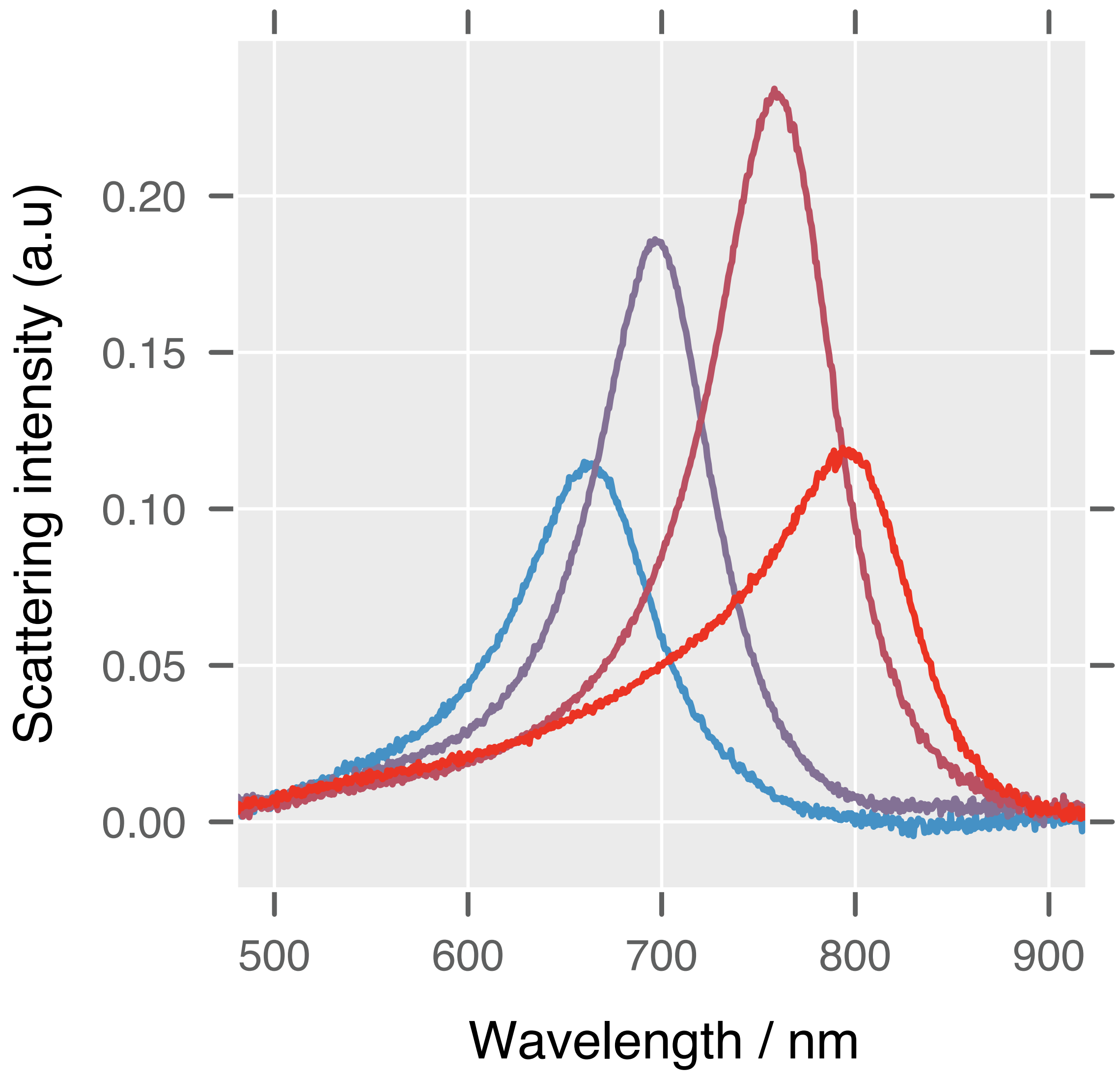


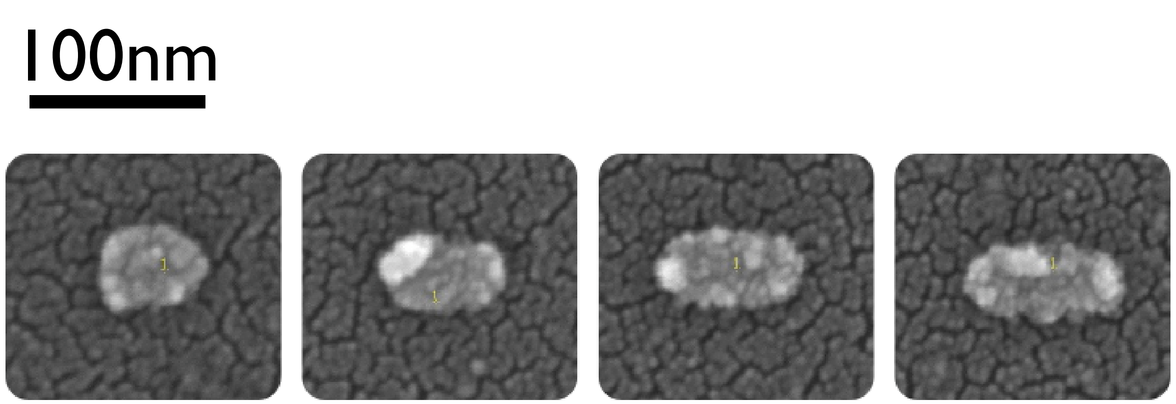
- excitation of Localised Surface Plasmon Resonances (LSPR)
  ↳ resonant scattering and absorption in the visible
- high field confinement

Isolated nanorods: dark field microscopy

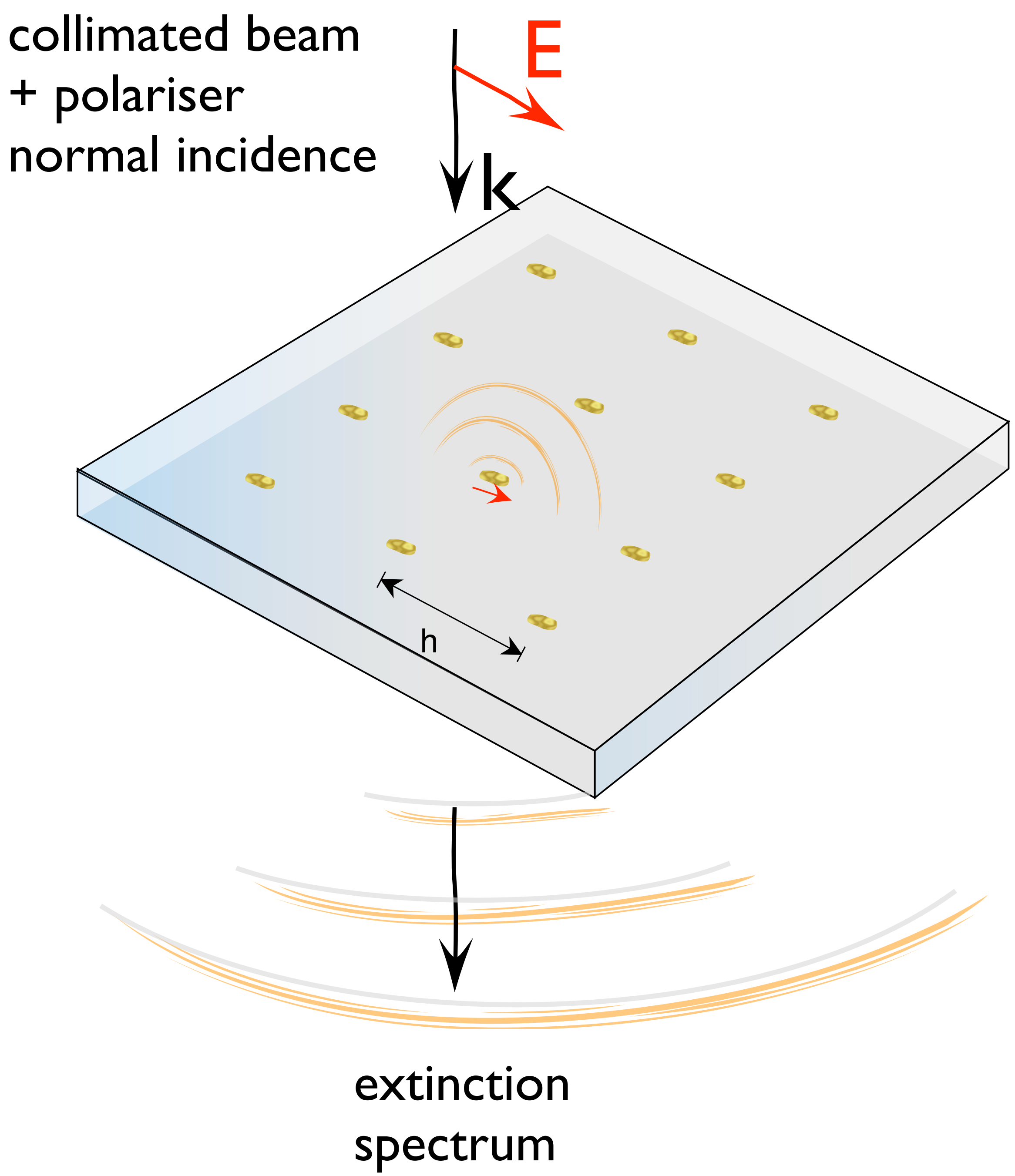


- measure forward-scattered light
- shape and size dictate the spectral lineshape





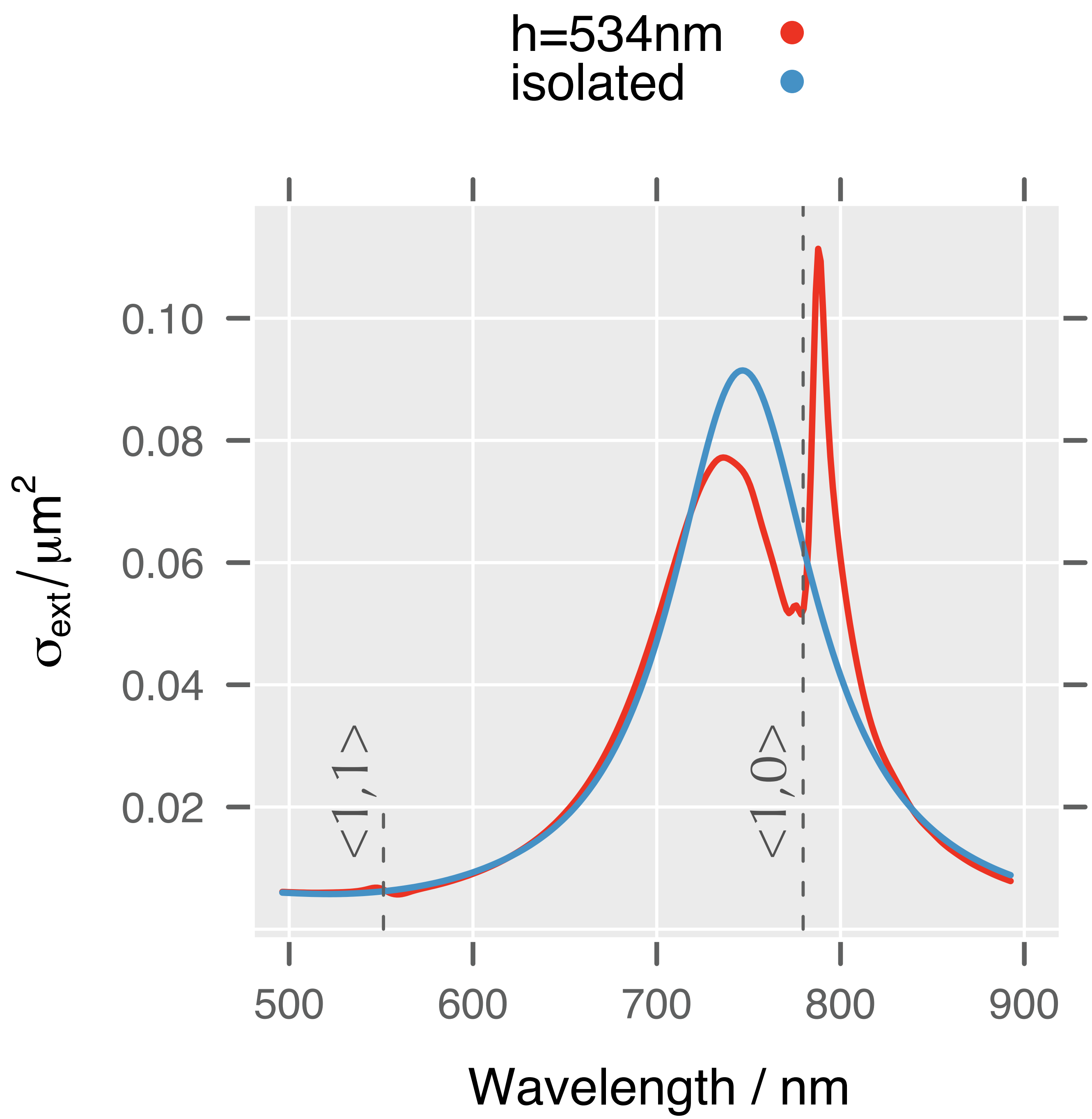
Transmission measurements in 2D arrays



- excitation of LSPR
- multiple scattering in the plane
- phase-matching condition
  ↳ geometrical resonance
- transmission measurement,
  Fano-type interference

Very narrow spectral peaks were predicted

Coupled dipole approximation



Extinction:

$$\sigma_{\text{ext}} \propto k\Im(\alpha)$$

Polarizability :
(isolated dipole)

$$\alpha = V\frac{\varepsilon_m - \varepsilon_d}{3\varepsilon_d + 3\chi(\varepsilon_m - \varepsilon_d)}$$

Effective polarizability:

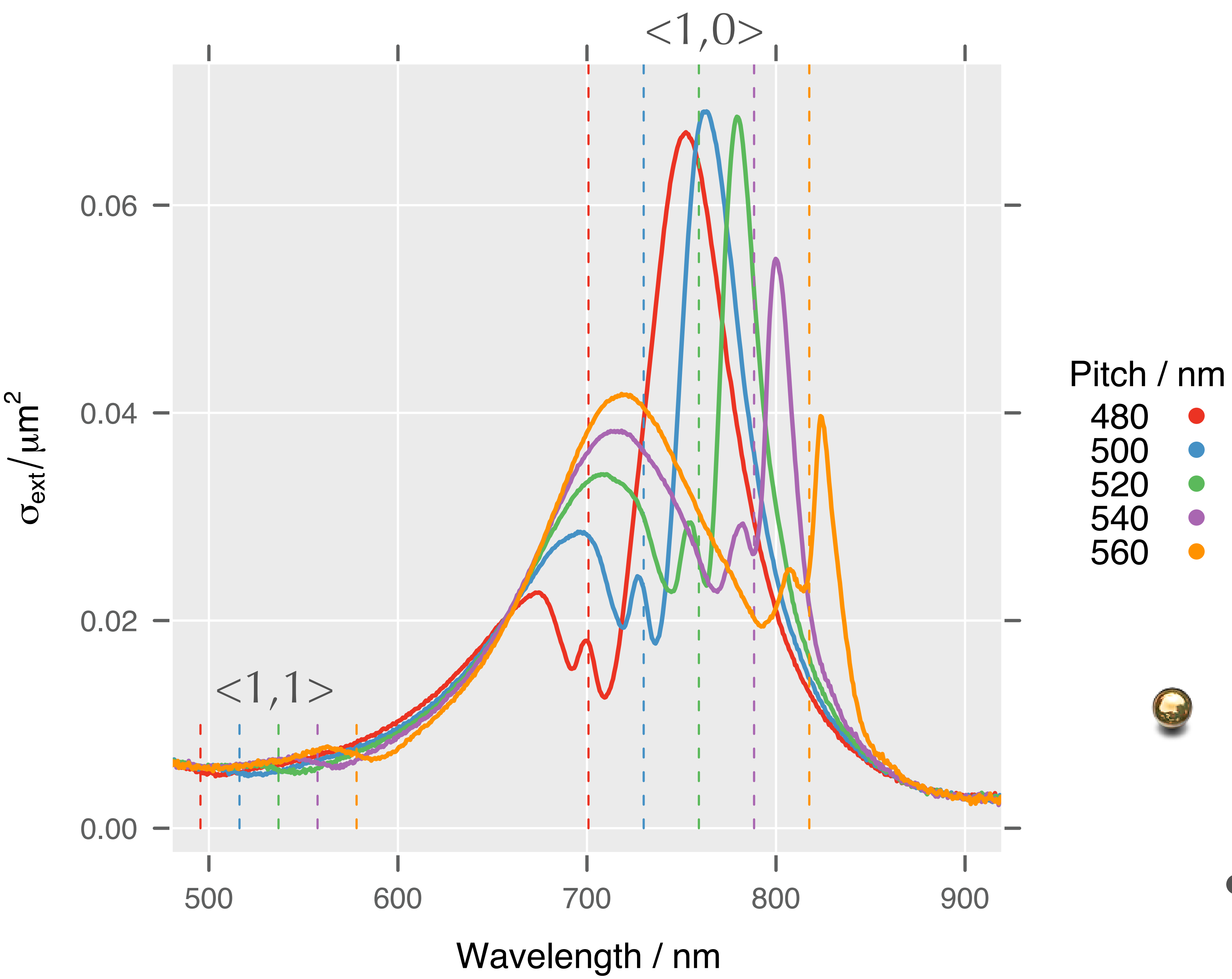
$$\alpha^* = \frac{\alpha}{1 - \alpha S}$$

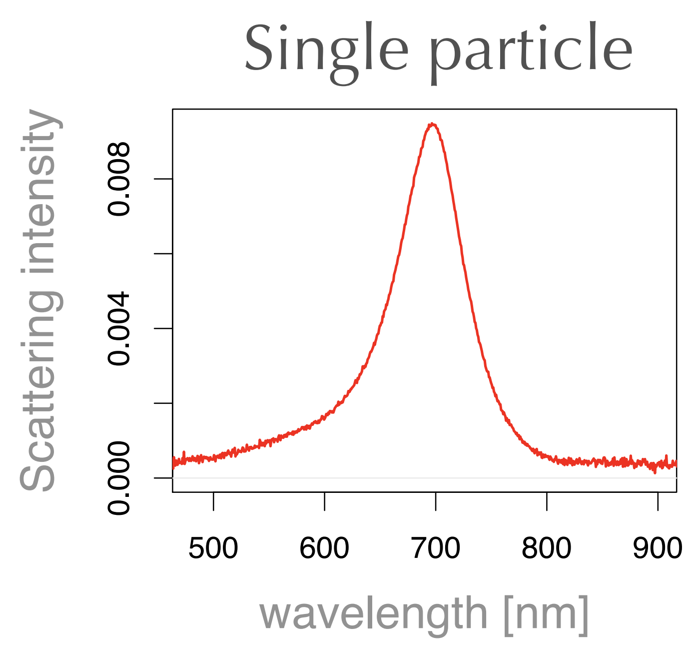
Array factor:

$$S = \sum_{\text{other dipoles}} \left[\frac{(1 - ikr)(3\cos^2\theta - 1)\exp(ikr)}{r^3} + \frac{k^2 \sin^2\theta \exp(ikr)}{r} \right]$$

Zhao, Schatz, Kelly. Journal of Physical Chemistry B (2003) vol. 107
Markel. J Phys B-At Mol Opt (2005) vol. 38
García de Abajo. Rev Mod Phys (2007)

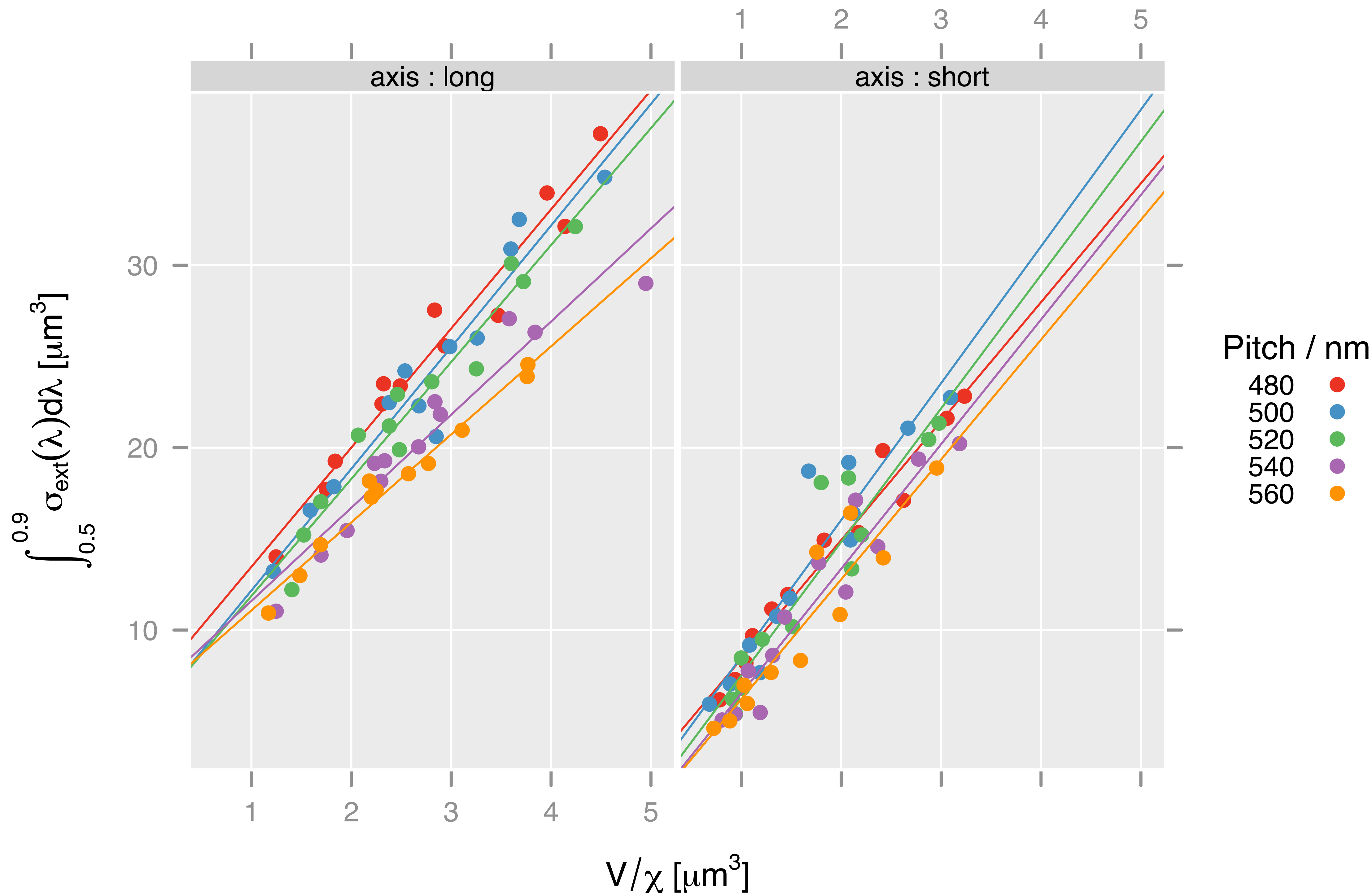
Diffractive coupling in 2D arrays: varying particle separation



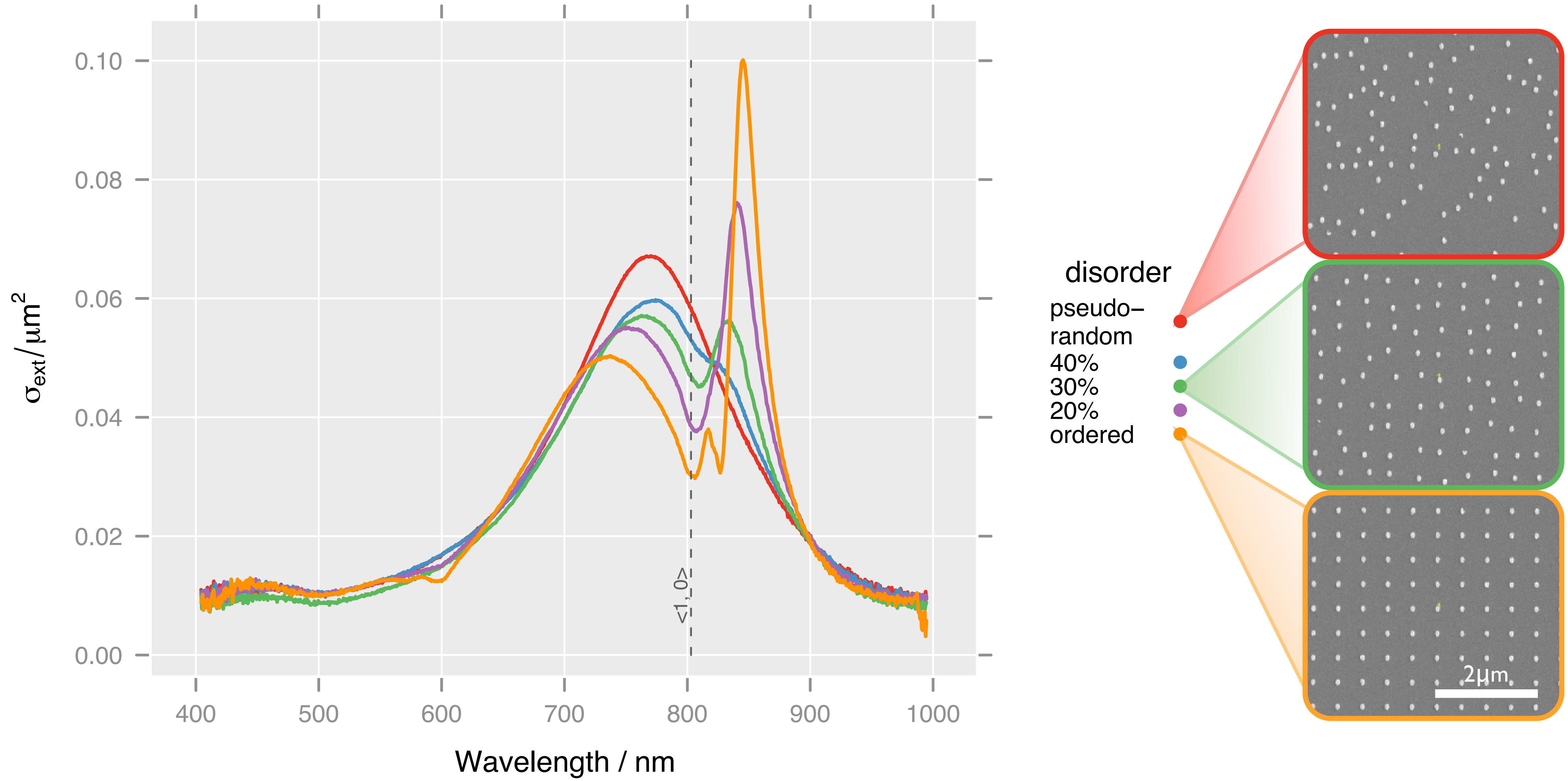


- Lineshape shows an interplay between :
  - localised resonance (LSPR)
  - geometrical resonance (phase matching)
- Sum rule : constant area

Sum rule for extinction $\int_0^\infty \sigma(\lambda)\mathrm{d}\lambda \propto \frac{V}{\chi}$



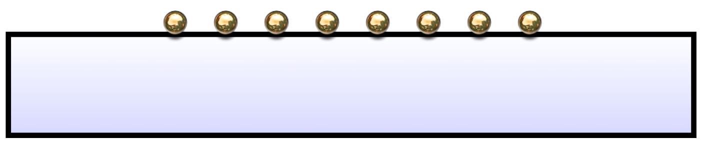
Influence of disorder



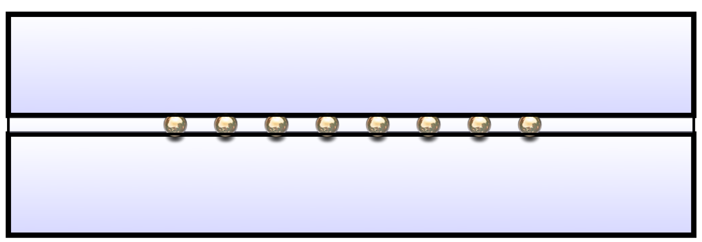
- the sharp spectral features disappear with a decrease of spatial correlation between scattering centers
- the inhomogeneously broadened LSPR spectrum is retrieved for spatially uncorrelated positions

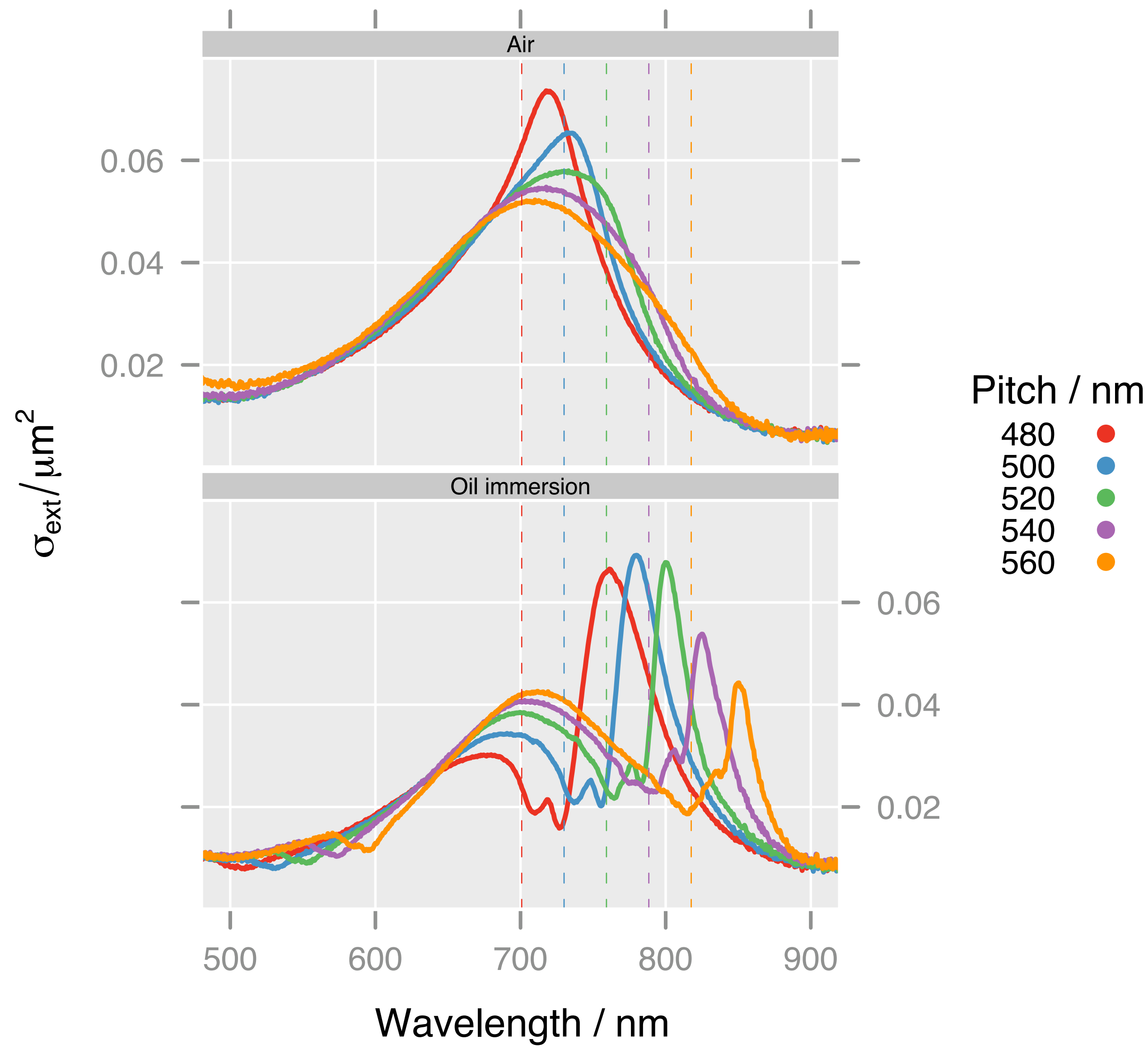
Diffractive coupling in 2D arrays

air superstrate



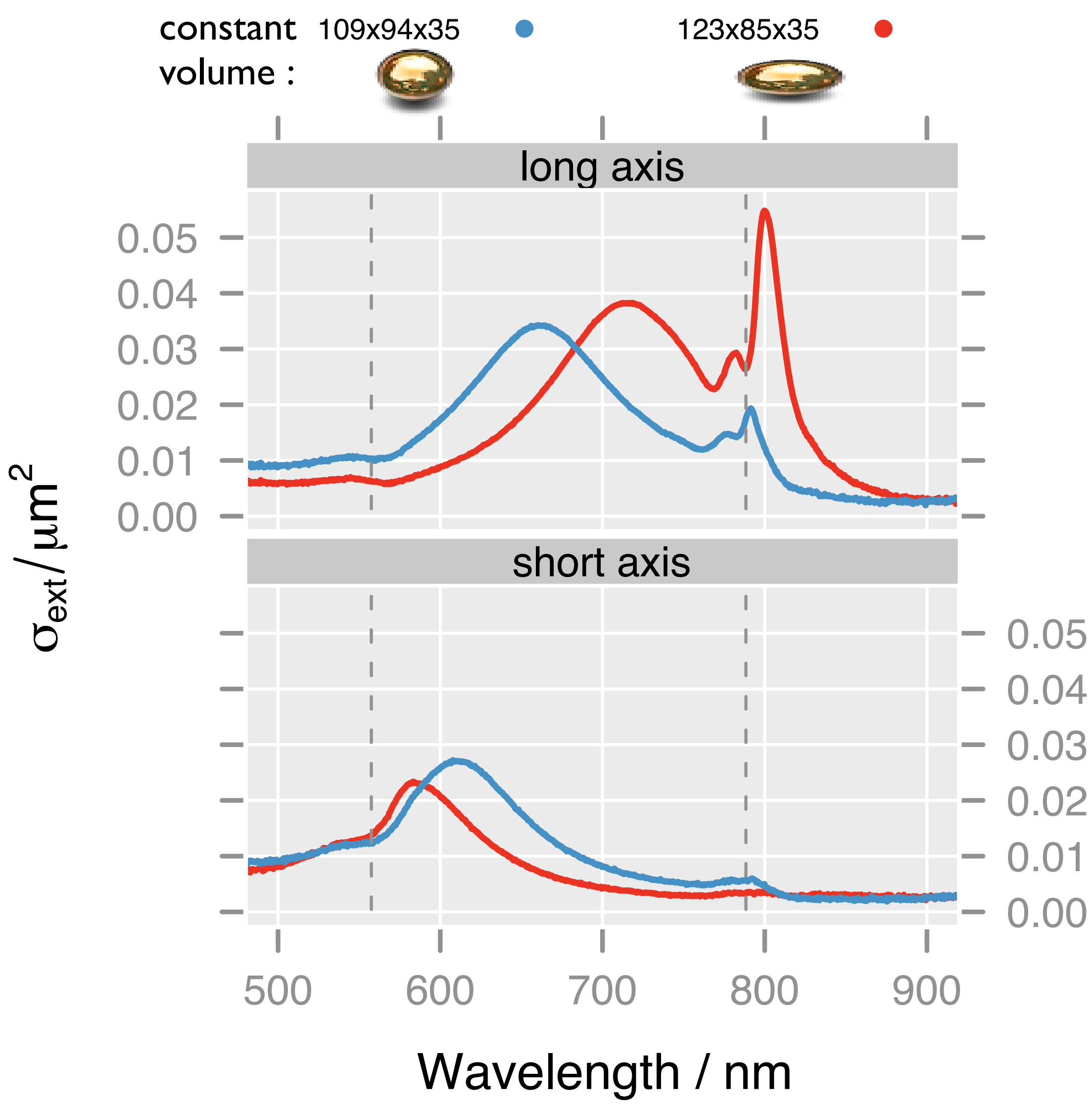
index-matched

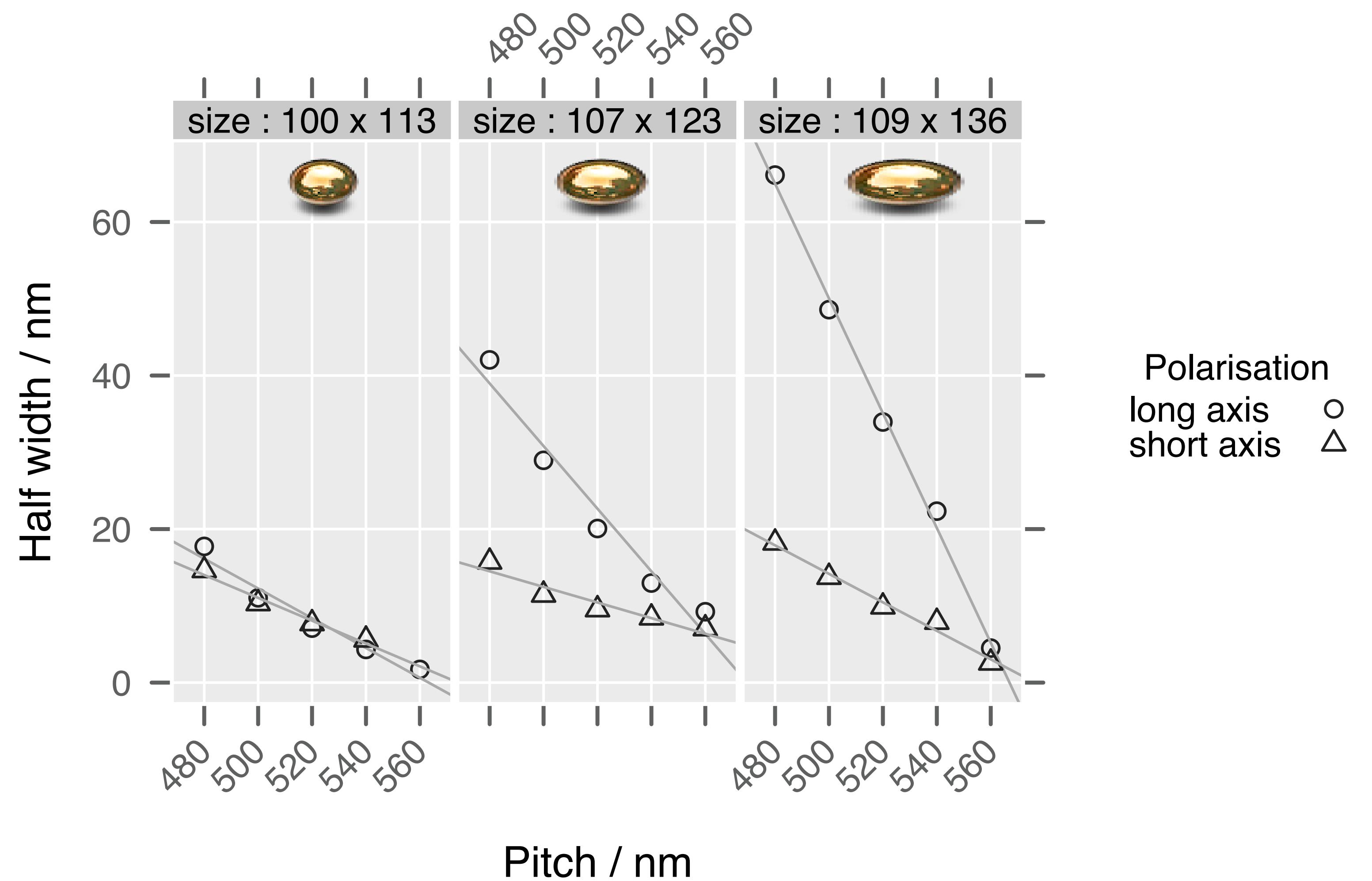




- an homogeneous surrounding index is needed
- reflection off the surface inhibits the radiative coupling

Size and aspect ratio: effect on the diffractive peak





- intensity and width strongly depend on the volume and aspect ratio of the particles
- width is a non-linear, decreasing function of the particle separation

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Conclusions

- a rich interplay between resonant scattering (excitation of LSPR) and a geometrical resonance is observed
- requires an homogeneous surrounding medium (in transmission measurements)
- the spectral lineshape depends on the periodicity and size and aspect ratio of the particles

Ref.: B. Auguié, W.L. Barnes: "Collective resonances in gold nanoparticle arrays" (accepted for publication in PRL, Sept 2008)

Additional material

- sum rule for extinction
- diagonal disorder
- off-normal incidence
- LSPR linewidth

+

Sum rule for extinction

KK: $$\Im\left[S(\omega)/\omega^2\right] = \frac{-2\omega}{\pi}\mathcal{P}\int_0^\infty \mathrm{d}\Omega \frac{\Re\left[S(\Omega)/\Omega^2\right]}{\Omega^2-\omega^2}$$

Optical th.: $$\sigma_{\mathrm{ext}}(\omega) = \frac{4\pi}{k^2}\Re\left[S(0,\omega)\right]$$

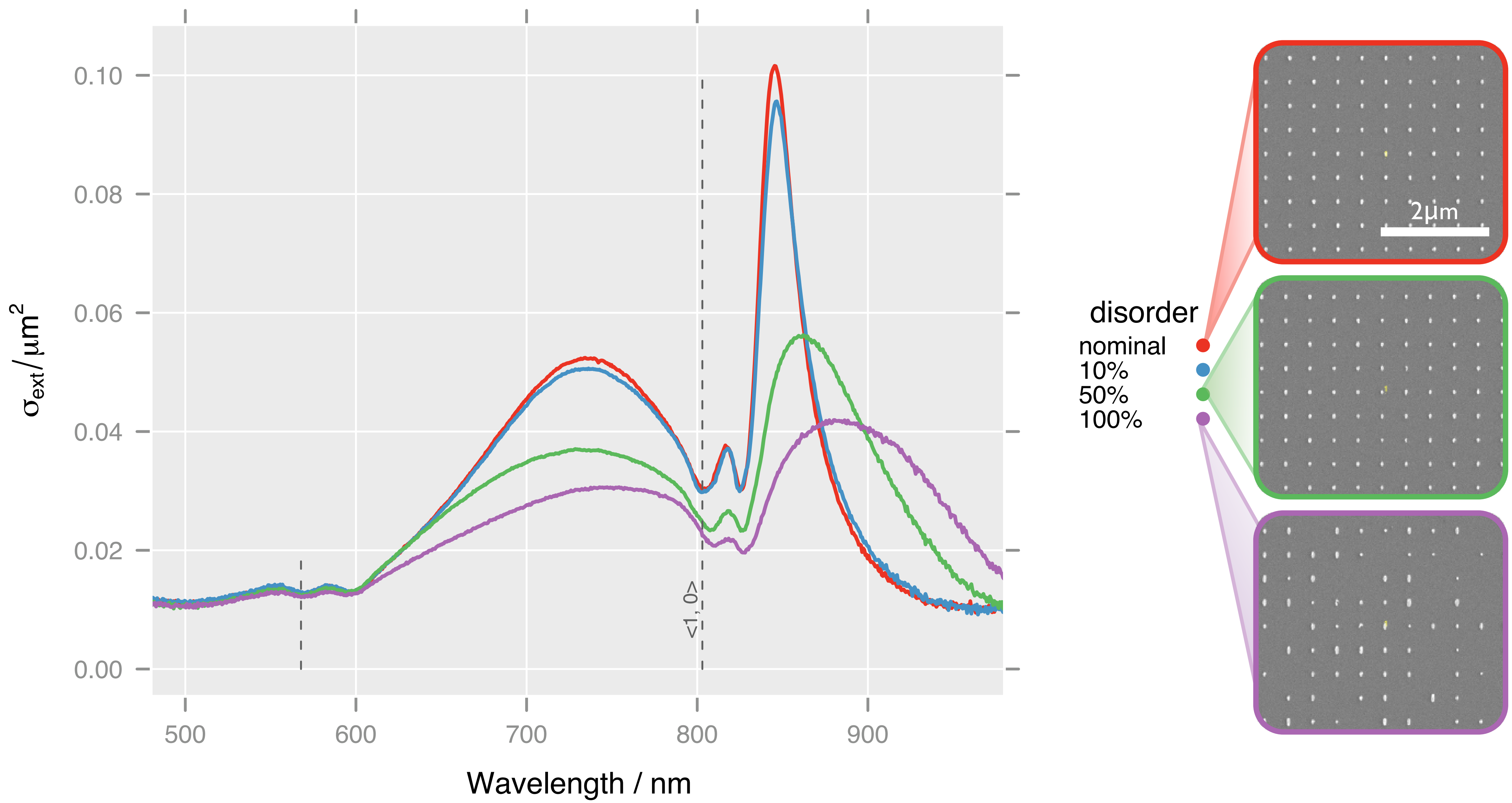
Electrostatic limit: $$S(0,\omega) = \frac{-\imath\omega^3}{c^3}\alpha_{\mathrm{static}}$$

$$\alpha_{\mathrm{static}} = V\frac{\varepsilon_m - \varepsilon_d}{\varepsilon_d + \chi(\varepsilon_m - \varepsilon_d)}$$

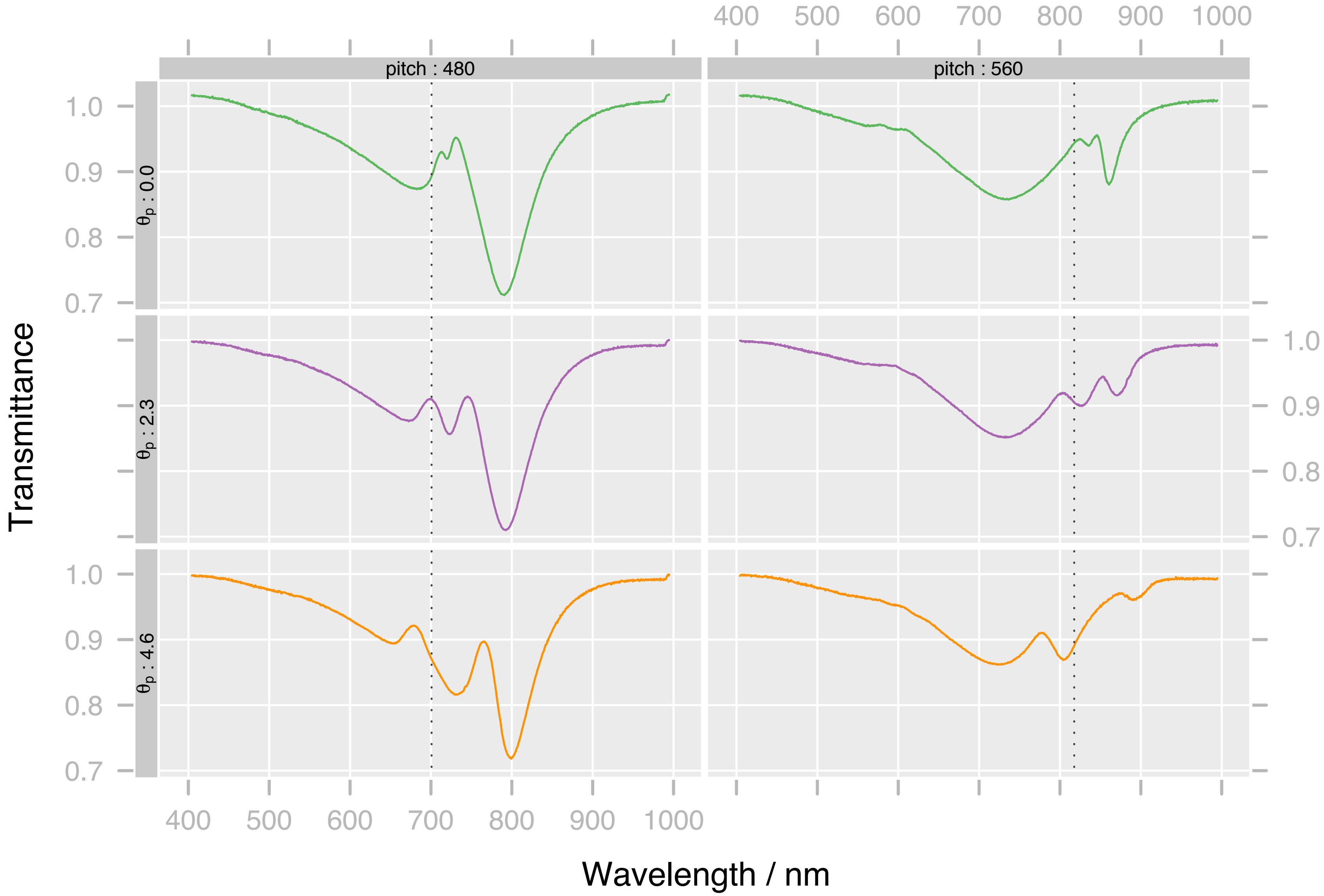
Metals: $\varepsilon_m \longrightarrow \infty$

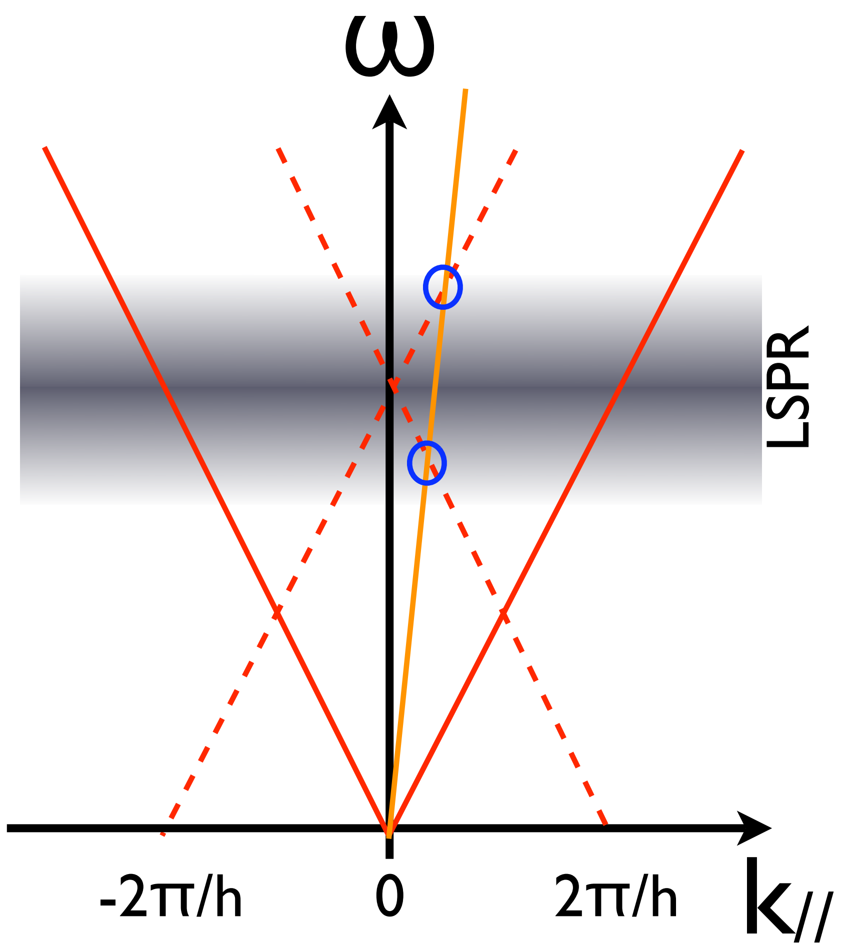
$$\int_0^\infty \sigma(\lambda)\mathrm{d}\lambda \propto \frac{V}{\chi}$$

Influence of diagonal disorder



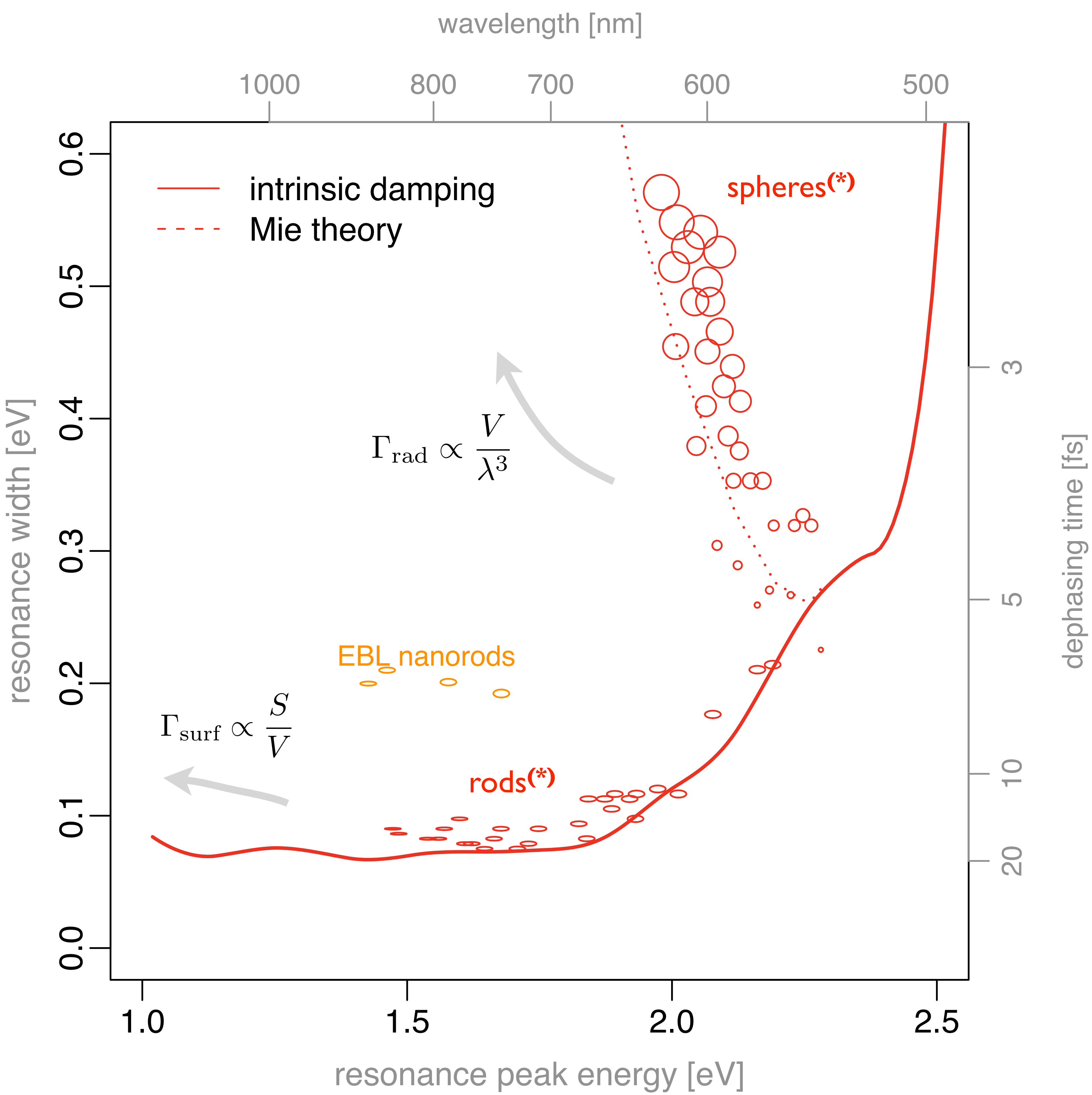
Deviation from normal incidence





- secondary dip grows with off-axis tilt
- the general picture is similar

What dictates the resonance width?



energy density in a dispersive medium:

$$u = \frac{d(\omega\varepsilon)}{d\omega}\mathbf{E}^2$$

rate of **energy loss** (Joule)

$$\omega\varepsilon_i\mathbf{E}^2$$

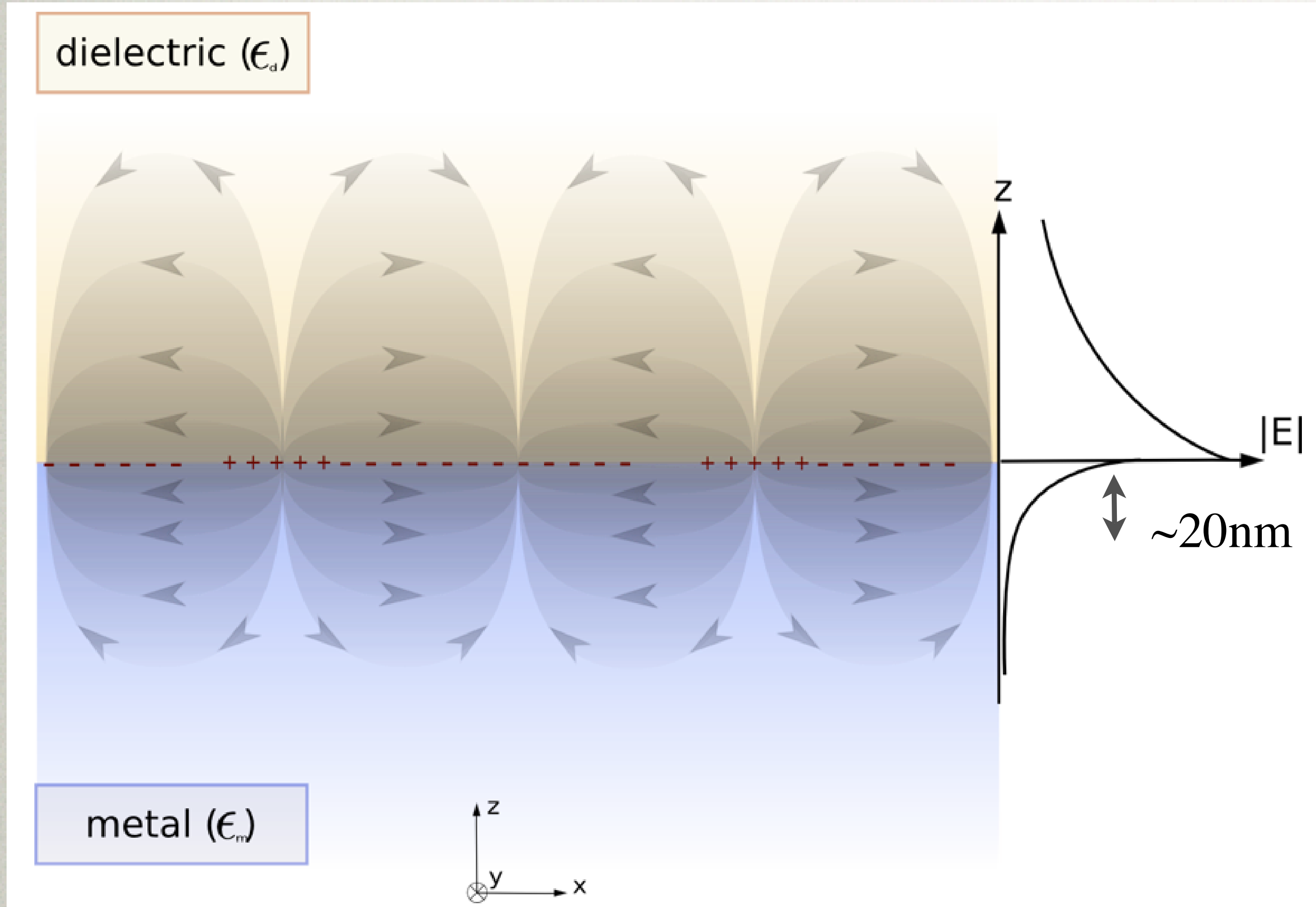
⇩

“intrinsic damping”

$$\Gamma = \frac{2\varepsilon_i}{\sqrt{\left(\frac{d(\varepsilon_r)}{d\omega}\right)^2 + \left(\frac{d(\varepsilon_i)}{d\omega}\right)^2}}$$

[*]: Sonnichsen et al. Phys. Rev. Lett., 88(7):077402, Jan 2002

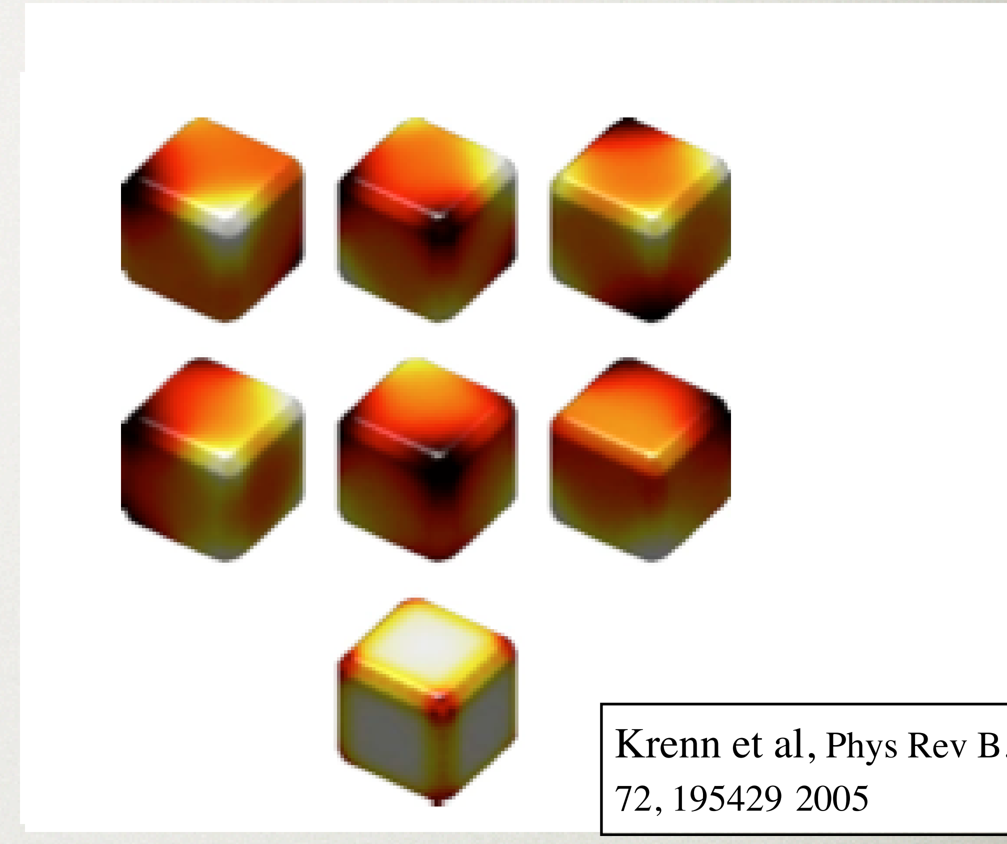
Confinement of surface plasmons using nanostructures



Surface plasmon-polaritons

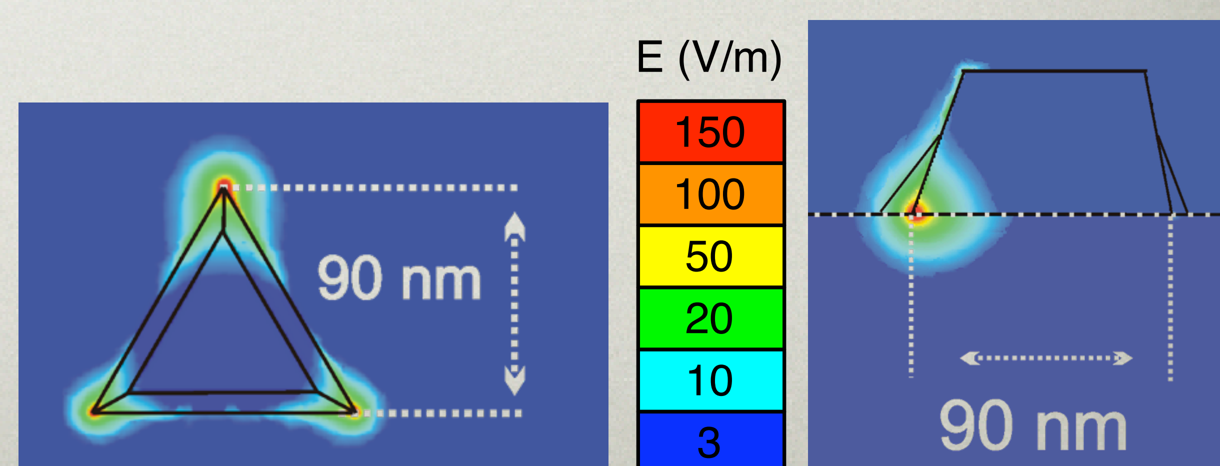
Skin depth in the metal is roughly 20 nm
propagation : typically microns

What happens if we constrain the dimensions to a sub-wavelength volume?

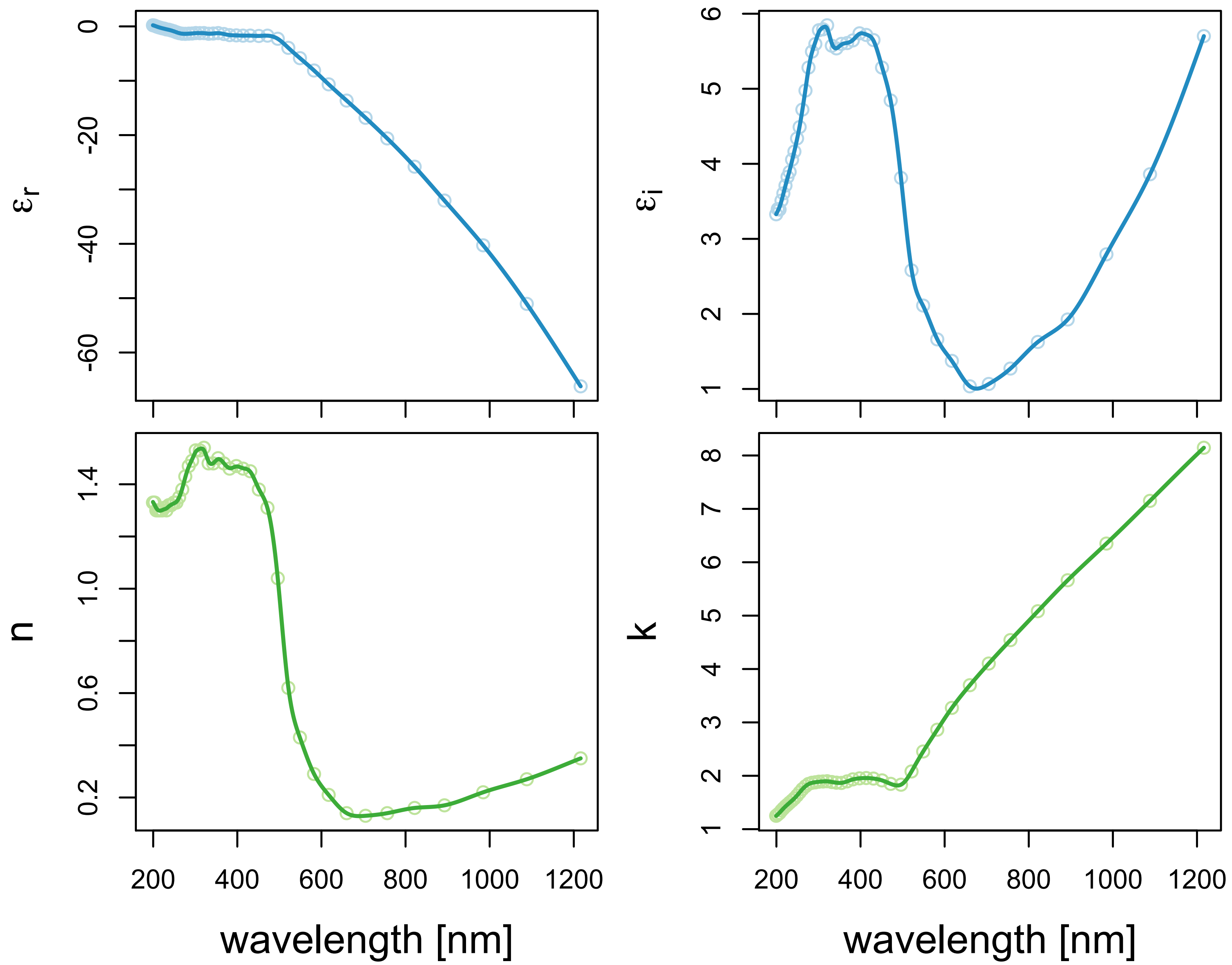


localised plasmons

eigenmodes of the surface charge density excitations

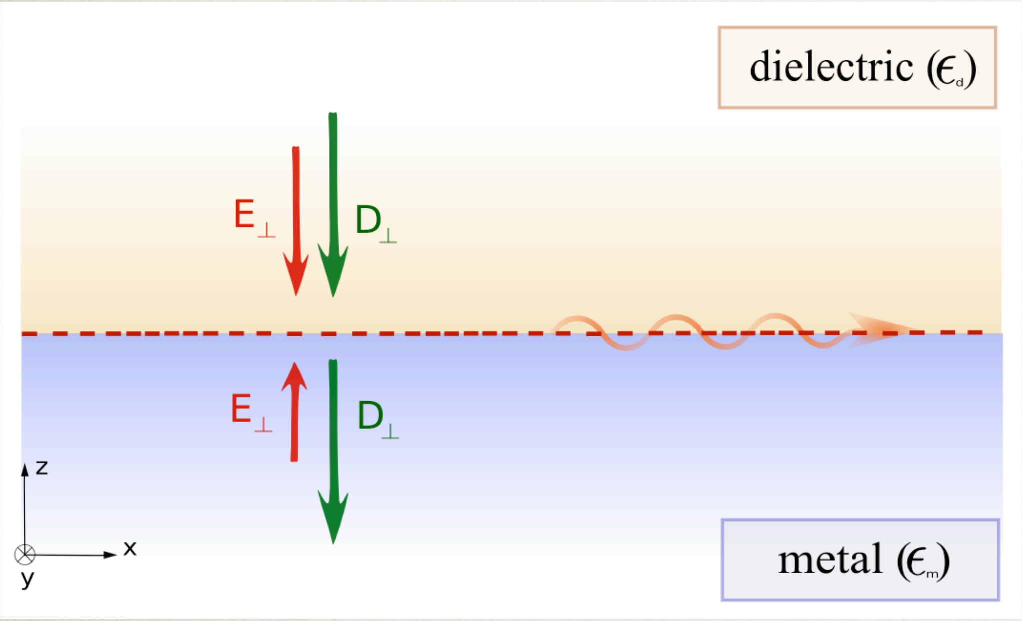


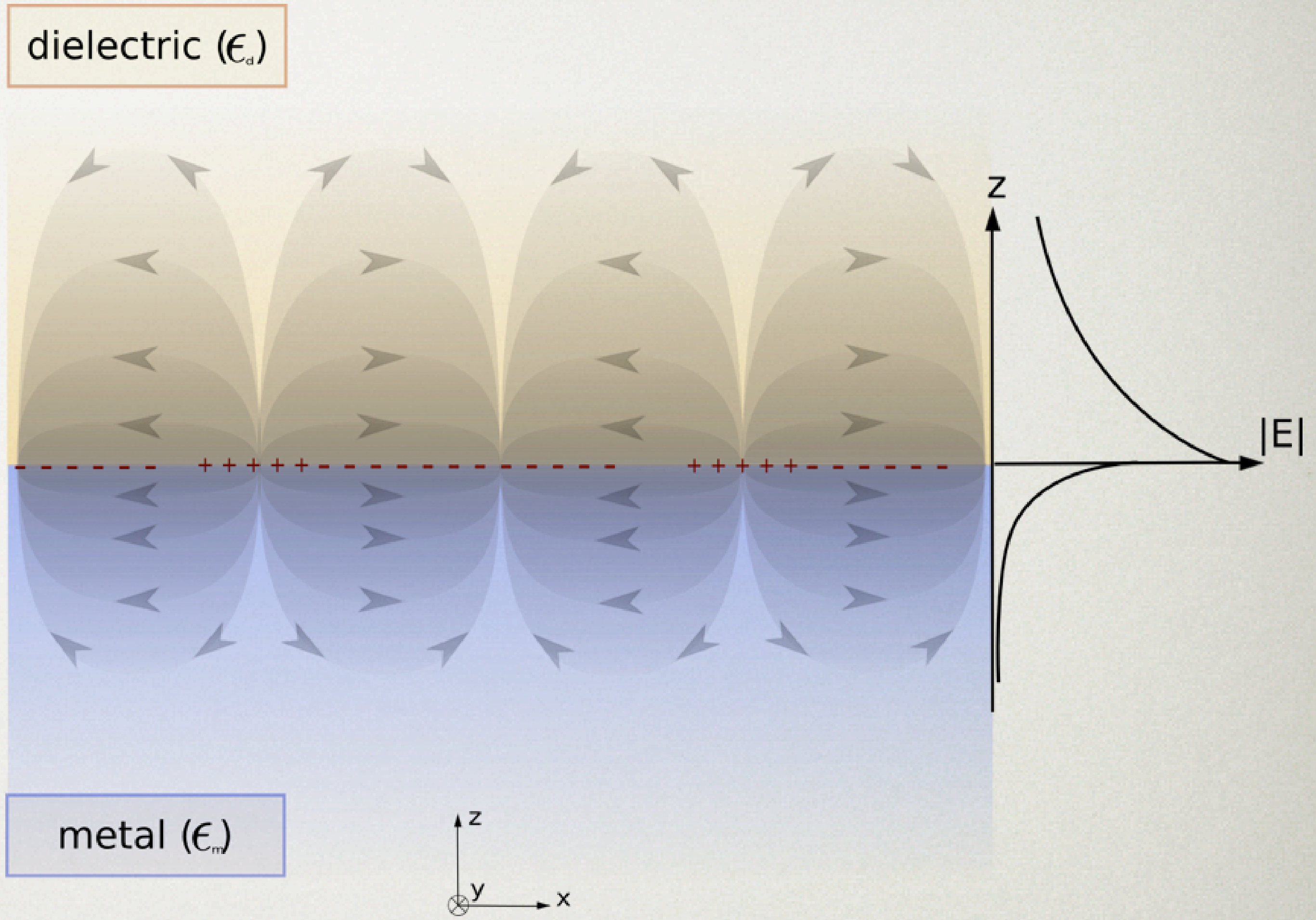
Gold nanorods

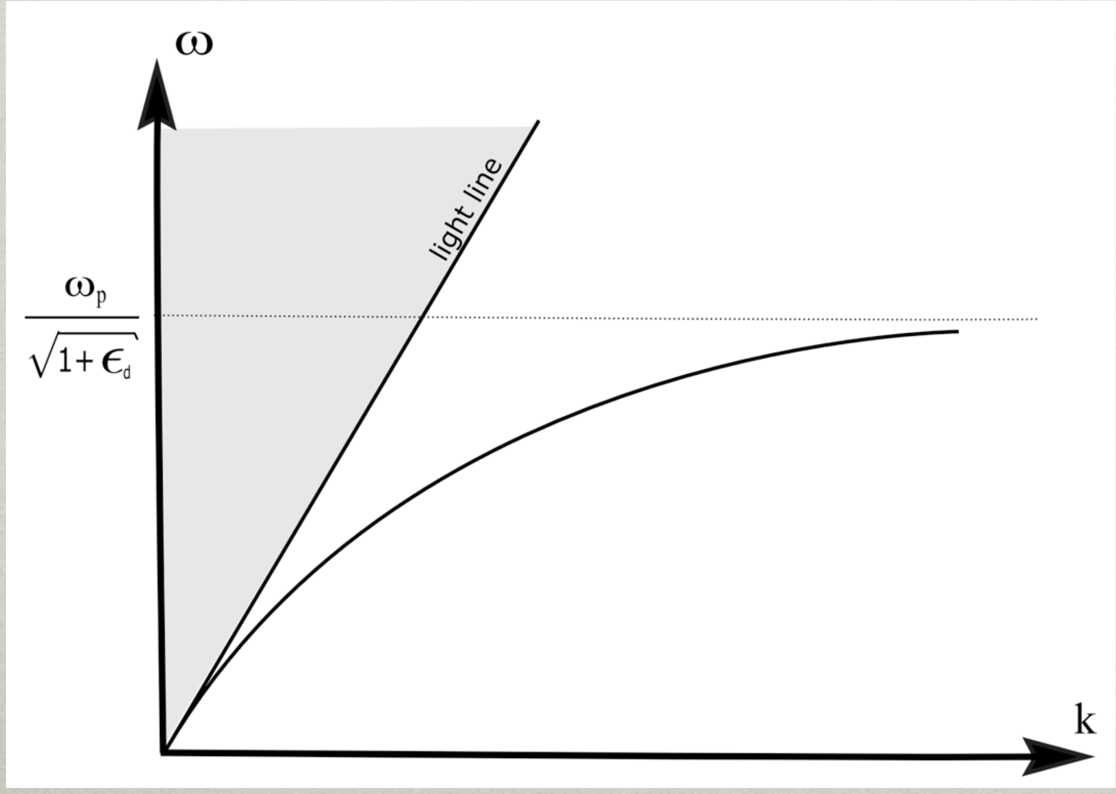


Optical properties of gold

Surface plasmon-polaritons







Dispersion relation

$$k_{\rm SPP} = k_0 \sqrt{\frac{\varepsilon_d \varepsilon_m}{\varepsilon_d + \varepsilon_m}}$$