

# Study of surface plasmon polaritons (SPPs) from periodic metallic arrays by coupled mode theory

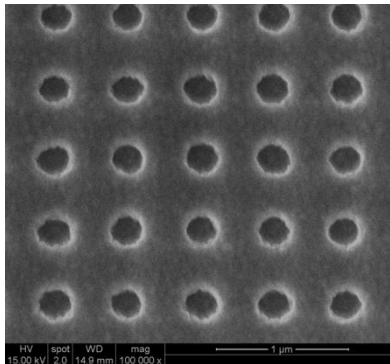
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Acknowledgement

Hong Kong Research Grant Council and Innovative Technology Fund



# Outline

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- How we fabricate and characterize periodic arrays
- Coupled mode theory (CMT)
- Determination of the absorption and radiative decay rates of SPPs
- Maximize the field strength by matching the absorption and radiative decay rates
- Control the phase difference between p- and s-polarizations for SPR sensing



# Outline

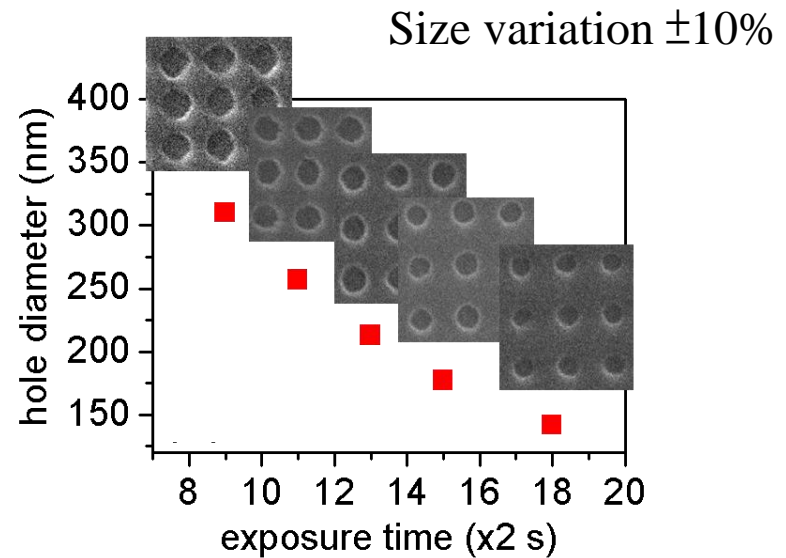
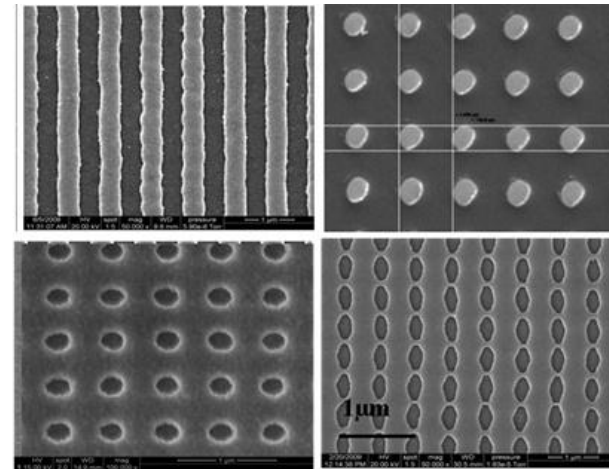
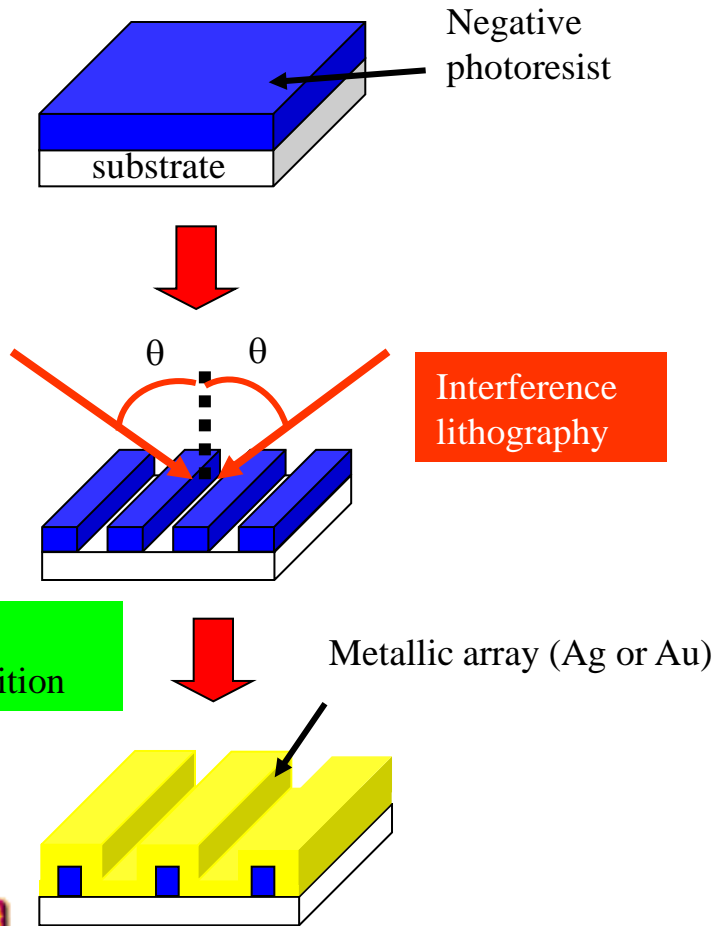
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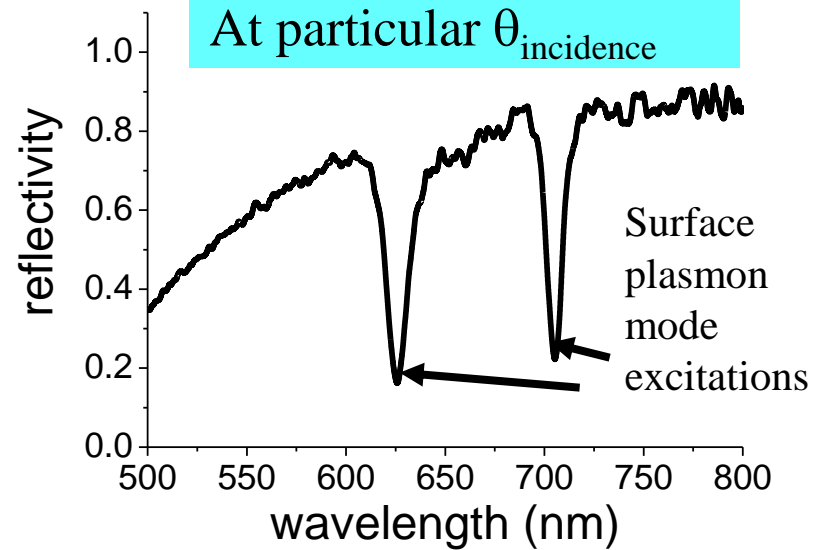
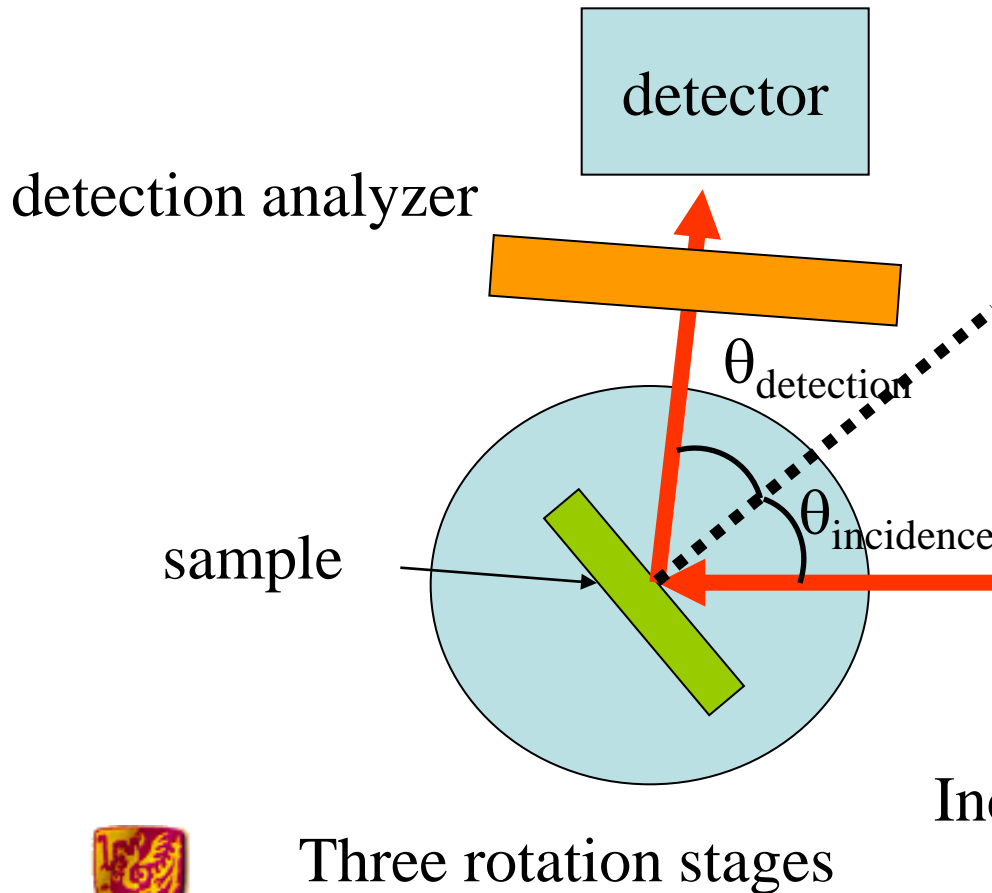
# Fabrication of periodic metallic hole arrays

Combine interference lithography and thin film deposition

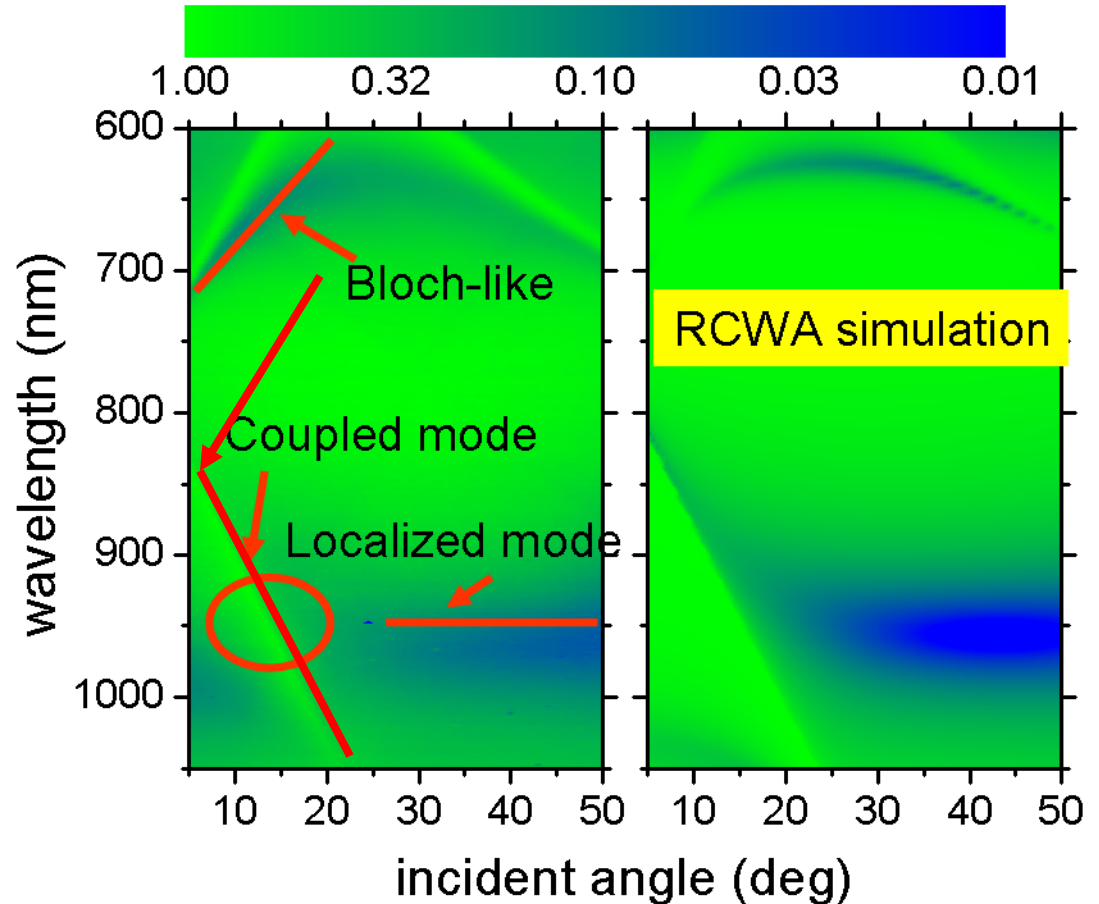
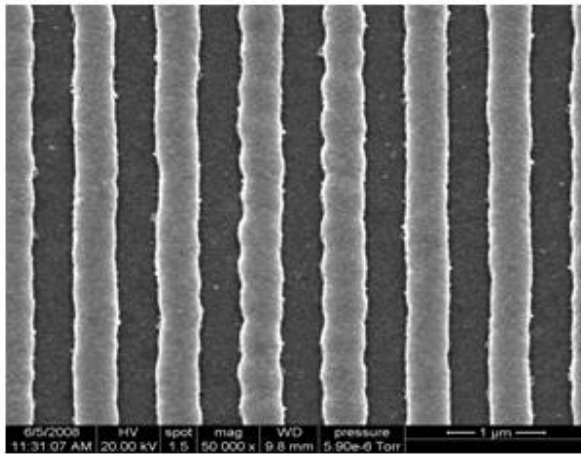


# Optical characterization

Measure specular or total reflectivity



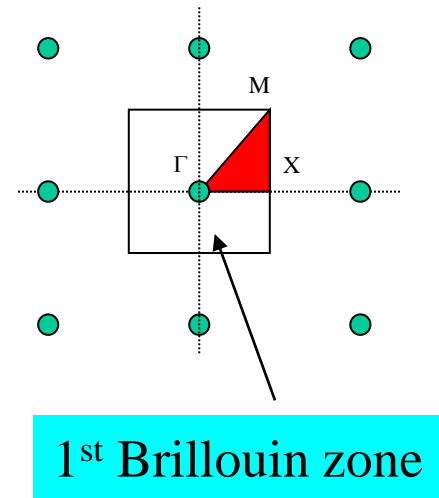
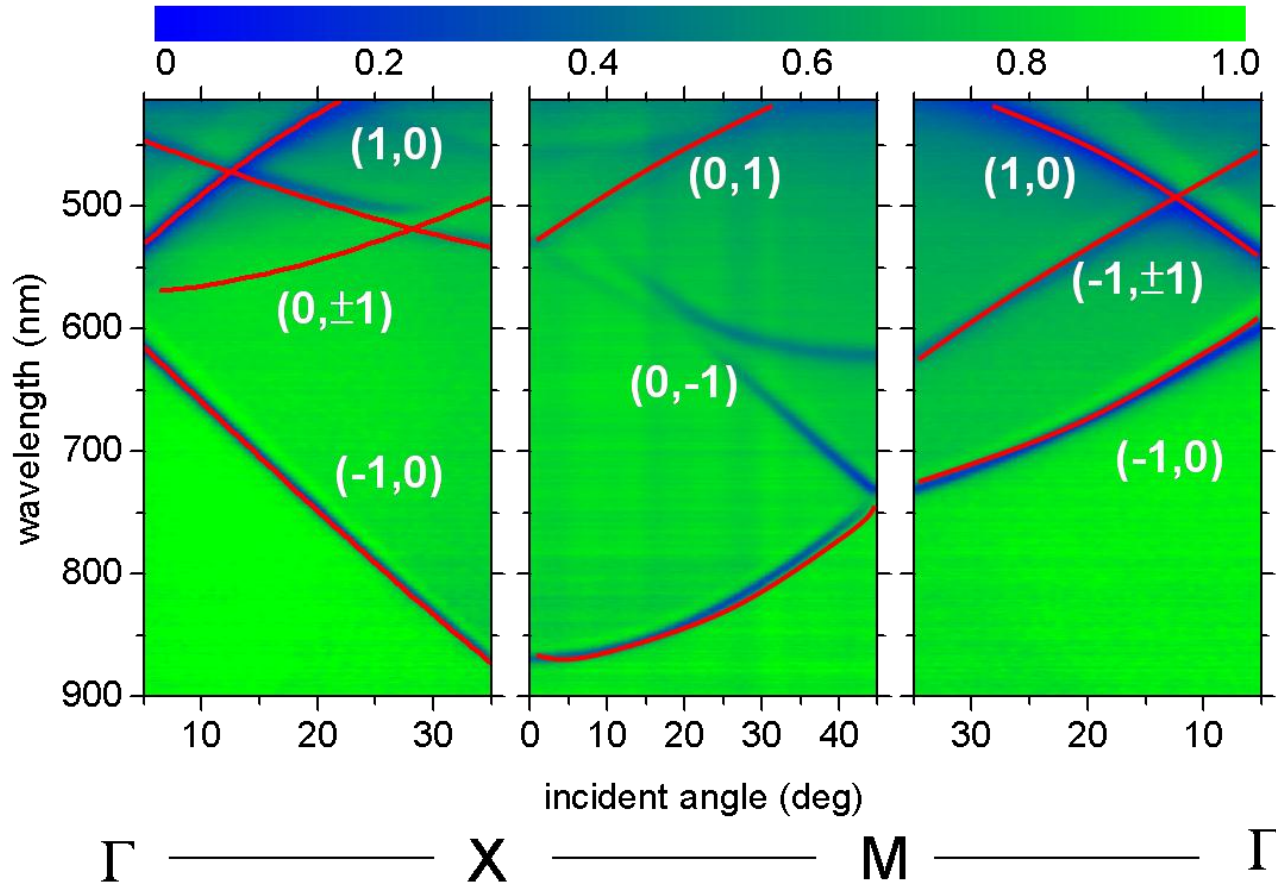
# P-reflectivity mapping from 1D Au grating



Blue region : low reflectivity dips  
Green region: high reflectivity



# From 2D Ag hole array



$$k_{sp} = \sqrt{\left(\frac{\omega}{c} \sin \theta \cos \varphi + \frac{2m\pi}{a}\right)^2 + \left(\frac{\omega}{c} \sin \varphi \cos \theta + \frac{2n\pi}{a}\right)^2}$$



# Numerical vs analytical

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- Many parameters are involved. Period, hole depth and radius, wavelength, incident angle, type of resonance modes, etc.

Difficult to find the right condition.

- Experimental: databank.
- Numerical methods: time consuming, resource demanding, lack of overall picture, etc.
- Analytical: may be qualitative, broad picture and physical insight.





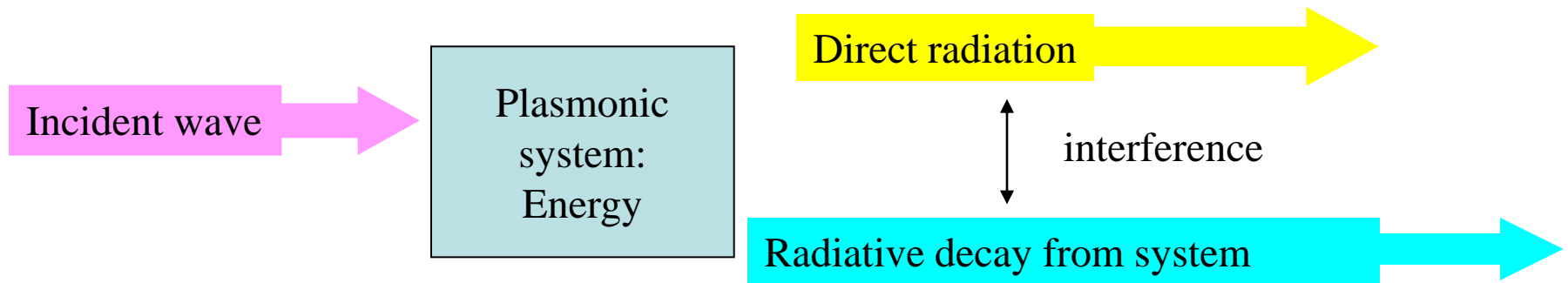
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# Coupled mode theory (CMT)



H.A. Haus, "Waves and Fields in Optoelectronics," 1984; S. Fan, "Photonic crystal theory: temporal coupled-mode formalism," *Optical Fiber Telecommunications V A: Components and Subsystems*, 2008; S. Fan et al, *JOSA A* 20, 569 (2003); L. Verslegers et al, *JOSA B*, 27, 1947 (2010); T.J. Seok et al, *Nano Lett.* 11, 2606 (2011); S.A. Maier, *Opt. Exp.* 14, 1957 (2006); J.B. Khurgin et al, *APL* 94, 1911106 (2009), *APL* 94, 101103 (2009), *APL* 95, 171103 (2009))



For single port, the mode amplitude,  $a$ , is given as:

$$\frac{da}{dt} = i\omega_o a - \underbrace{\frac{1}{2}(\Gamma_{abs} + \Gamma_{rad})}_{\text{Decay of SPP}} a + \underbrace{\kappa \sqrt{\frac{\Gamma_{rad}}{2}}}_{\text{Coupling from incident wave}} s_+$$

Decay rates are very important!!!

$\omega_o$  = resonance frequency

$|a|^2$  = energy in the resonator

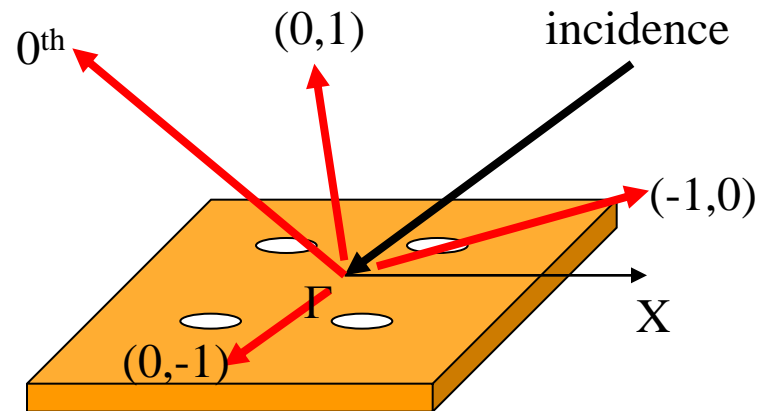
$\kappa$  = coupling constant

$|s_+|^2$  = power carried by incident wave

$\Gamma_{abs}$  = absorption rate

$\Gamma_{rad}$  = radiative decay rate

$\Gamma_{tot} = \Gamma_{abs} + \Gamma_{rad}$



- Can be extended to multiple ports
- Can be applied to different dimensionalities and systems. For example, thin films, nanoparticles, etc.



- Solve for a

$$a = \frac{\kappa \sqrt{\frac{\Gamma_{rad}}{2}} s_+}{i(\omega - \omega_o) + \Gamma_{tot} / 2}$$

- For (-1,0) SPP, single port CMT yields Fano-like reflectivity

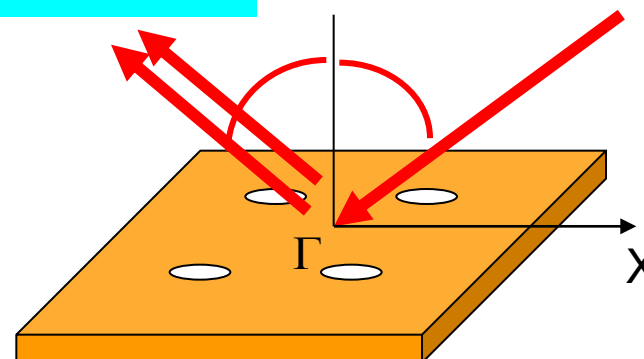
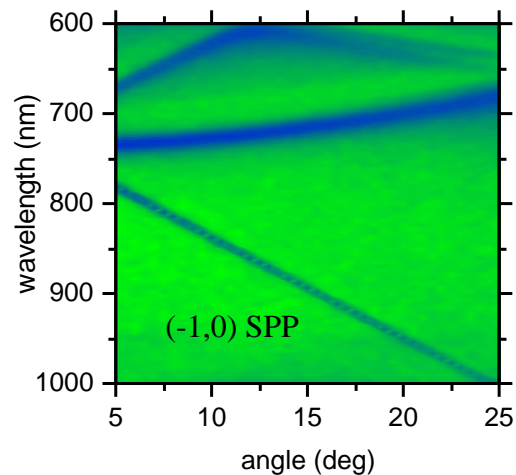
$$R = \frac{|s_-|^2}{|s_+|^2} = \left| \alpha + \kappa \sqrt{2} \frac{a}{s_+} \right|^2 = \left| \alpha + \underbrace{\frac{\kappa^2 \frac{\Gamma_{rad}}{2} e^{i\phi}}{i(\omega - \omega_o) + \Gamma_{tot} / 2}} \right|^2$$

outgoing
incoming

Direct reflection

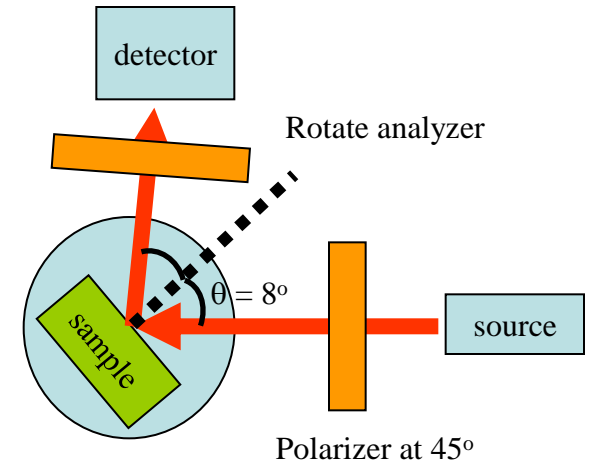
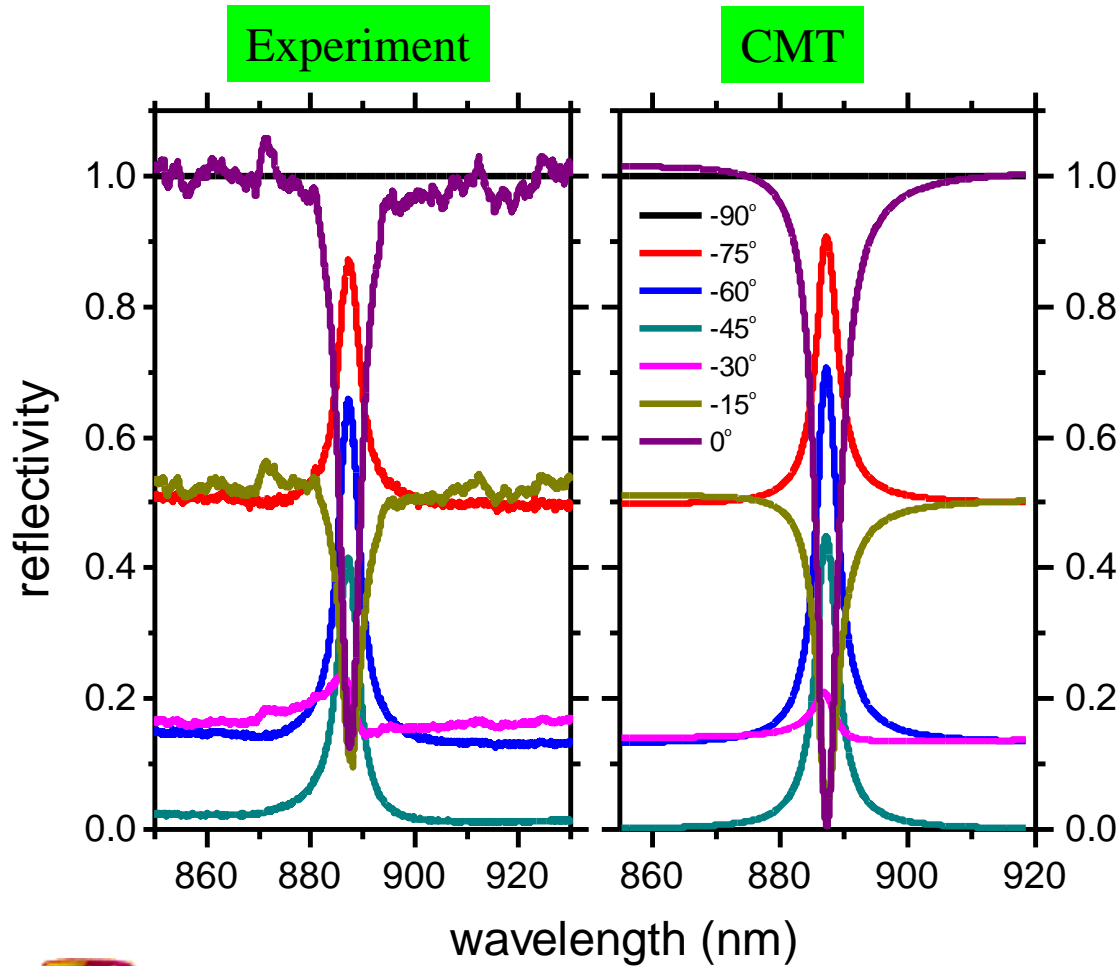
Radiative decay of SPP

Specular reflection



# Polarization-dependent spectroscopy to verify CMT

Au 2D hole array, period = 760 nm, diameter = 210 nm



Best fit 0° spectrum and calculate, -15° → -90° spectra by CMT and Jones calculus



Normalized w.r.t. -90° spectrum

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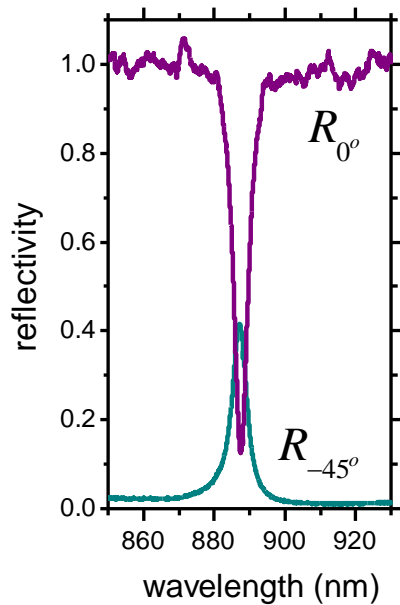


By conservation of energy

$$|s_+|^2 - |s_-|^2 = \Gamma_{abs} |a|^2 + \Omega_{Au} |s_+|^2$$

SPP absorption

Flat Au absorption



0° reflectivity

From fitting

From fitting

$$\Gamma_{abs} = \frac{(1 - \Omega_{Au} - R_{0^\circ}) |\kappa^2 \Gamma_{rad}|}{8R_{-45^\circ}}$$

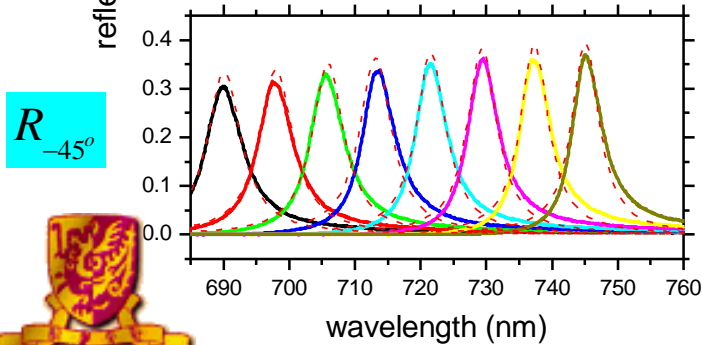
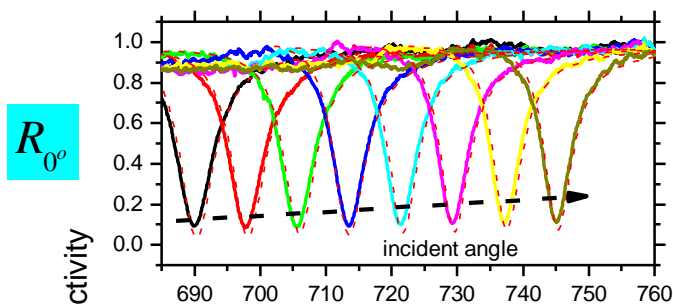
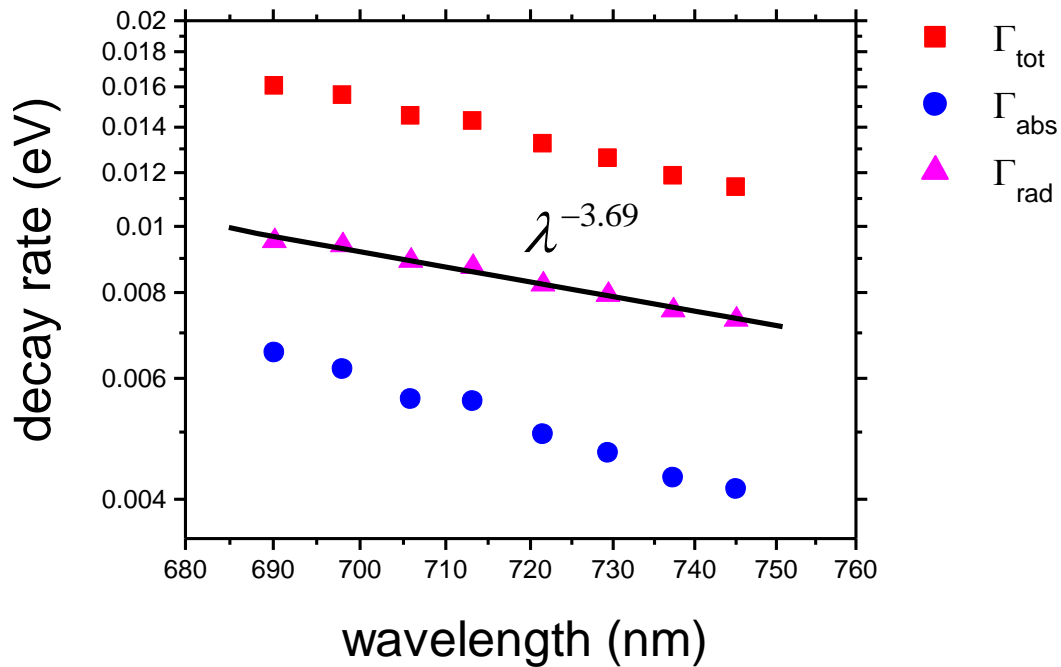
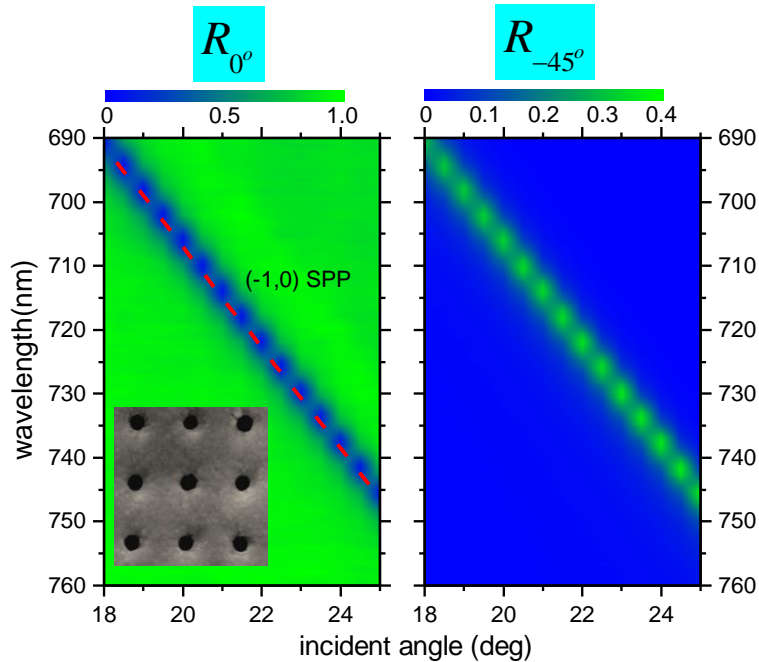
-45° reflectivity

$$\Gamma_{rad} = \Gamma_{tot} - \Gamma_{abs}$$

From fitting



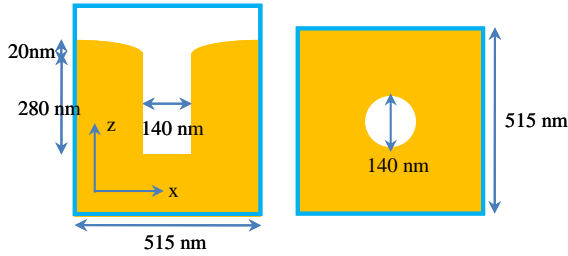
P = 515 nm, hole depth H and diameter D = 280 and 140 nm





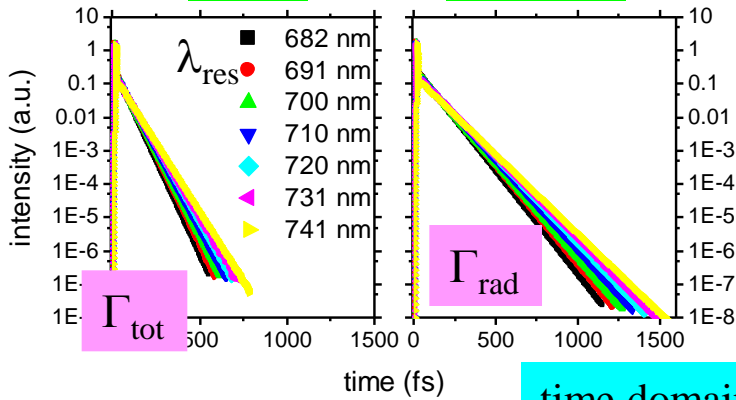
# FDTD simulation

Unit cell

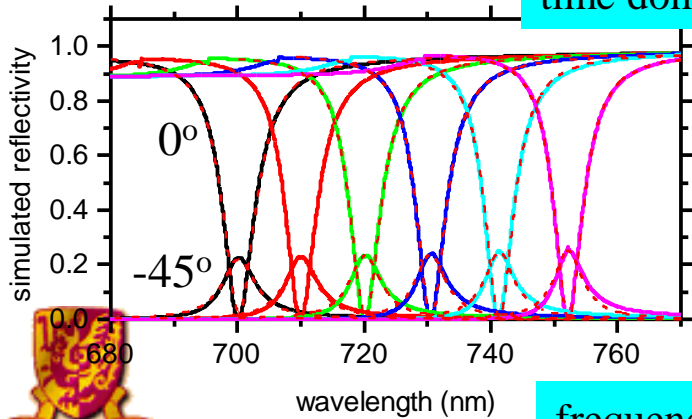


$\epsilon''_{Au}$

$0.1\epsilon''_{Au}$



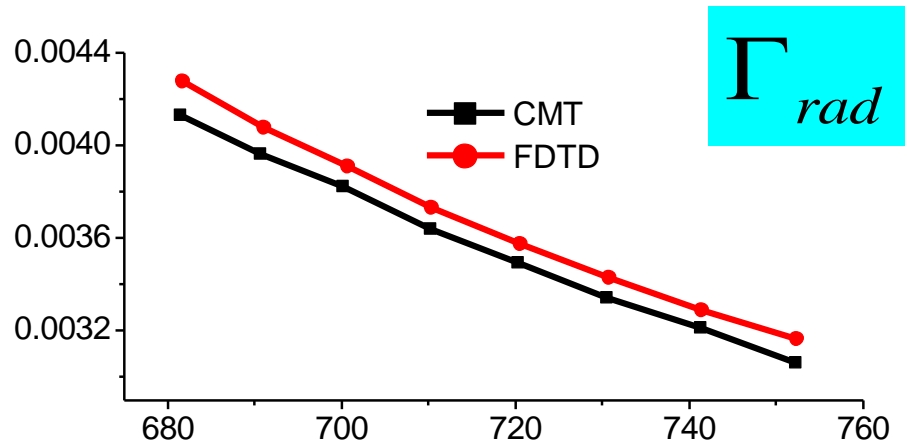
time domain



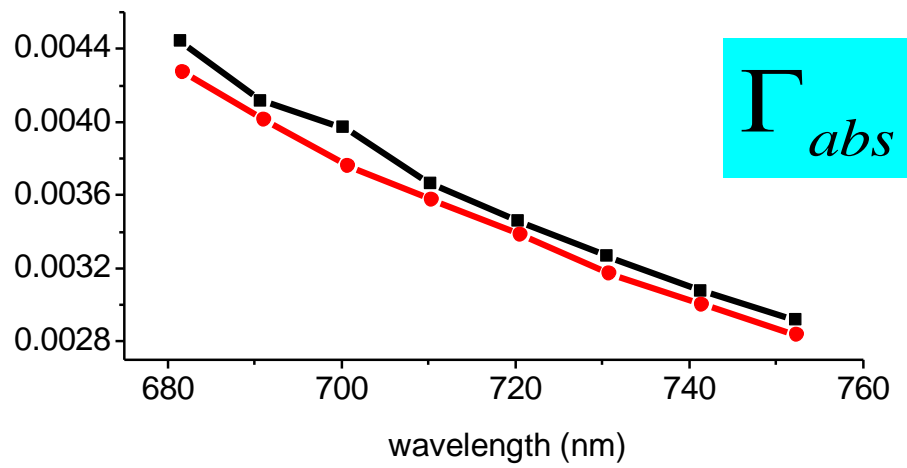
frequency domain

radiative decay rate (eV)

absorption rate (eV)



$\Gamma_{rad}$



$\Gamma_{abs}$

(-1,0) SPP



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# Maximize the field strength?

Energy of SPP,  $|a|^2$ , is given as:

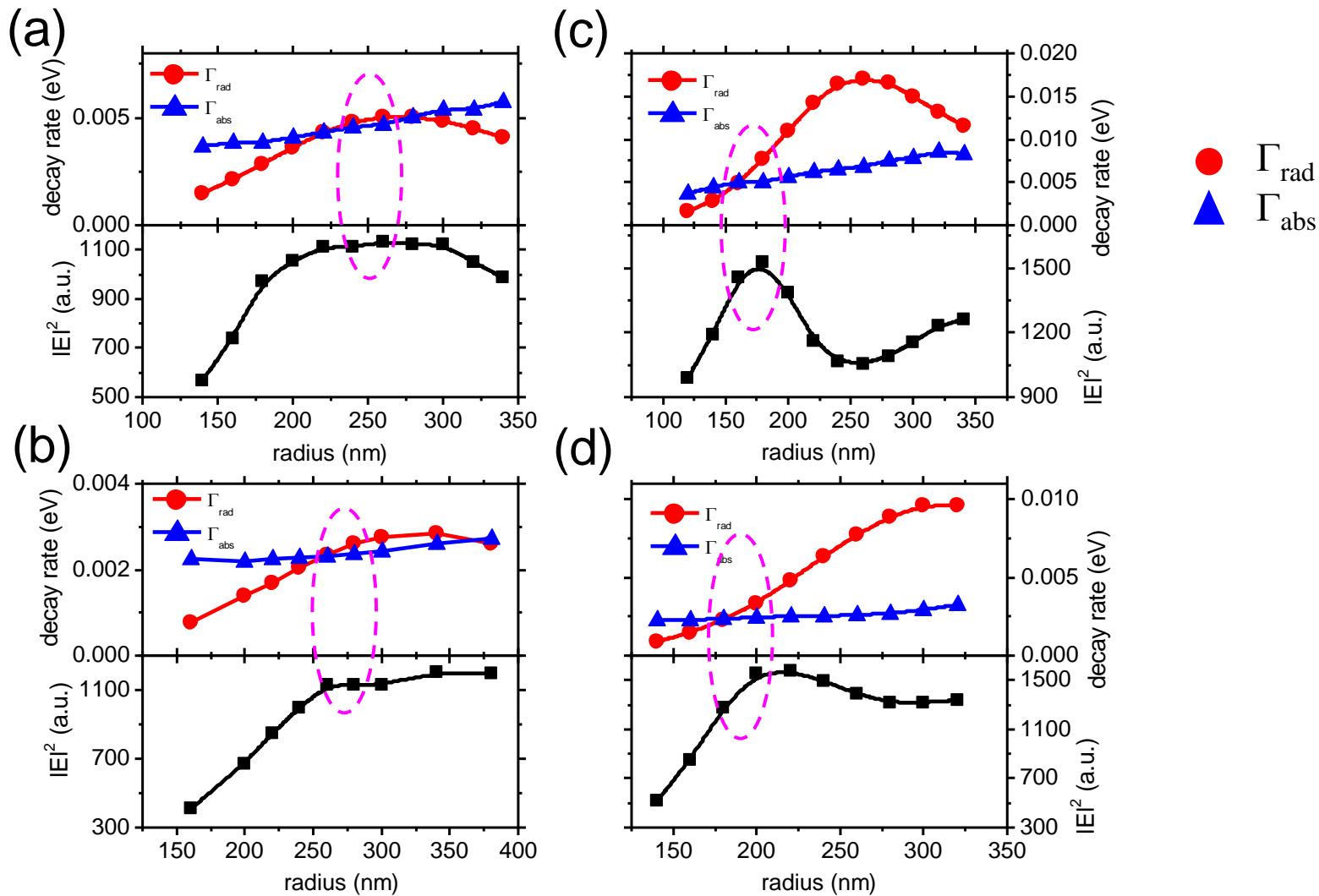
$$|a|^2 = \frac{\kappa^2 \frac{\Gamma_{rad}}{2} |s_+|^2}{(\omega - \omega_o)^2 + \left(\frac{\Gamma_{tot}}{2}\right)^2} \quad \longrightarrow \quad |E|^2 \propto \frac{|a|^2}{V_{eff}}$$

One special case, if  $\Gamma_{abs}$  and  $\kappa$  do not vary much,  $|a|^2$  is maximal when  $\Gamma_{abs} = \Gamma_{rad}$ .

$$\frac{d|a|^2}{d\Gamma_{rad}} = 0$$



# 2D Au periodic arrays (-1,0) SPP: FDTD



(a)  $P = 760$  nm and depth = 60 nm, (b)  $P = 900$  nm and depth = 60 nm, (c)  $P = 760$  nm and depth = 120 nm, and (d)  $P = 900$  nm and depth = 120 nm.

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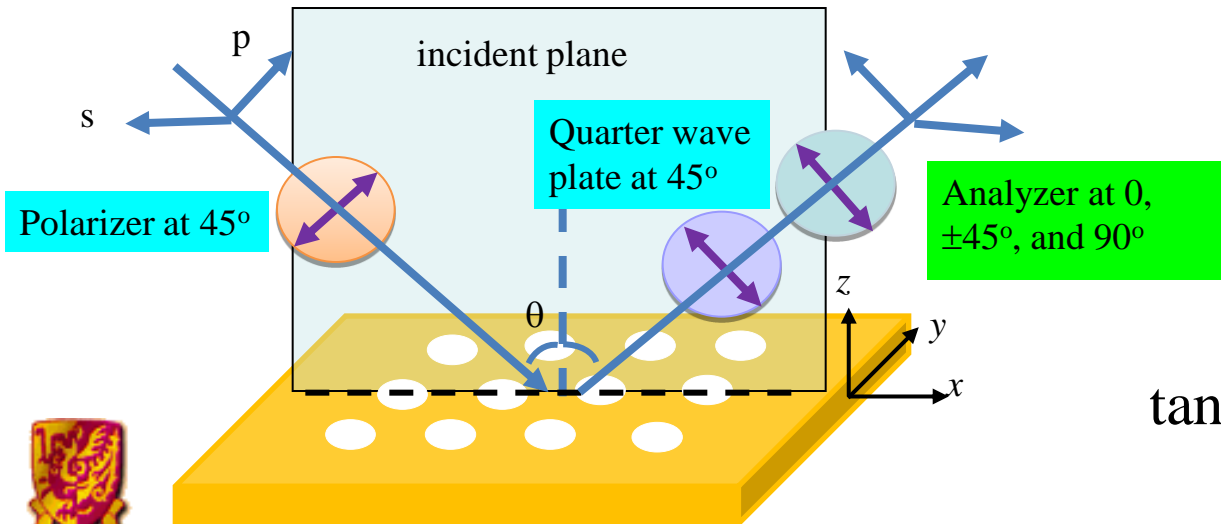
# P- and s-reflectivity

- P- and s-reflections and p-s phase difference

$$\begin{aligned}
 \text{p} \quad r_p &= r_p' e^{i\phi_p} = \alpha + \frac{\kappa^2 \frac{\Gamma_{rad}}{2} e^{i\phi}}{i(\omega - \omega_o) + \frac{\Gamma_{tot}}{2}} && \text{by CMT} \\
 \text{s} \quad r_s &= r_s' e^{i\phi_s} = \beta && \text{radiative decay} \\
 &&& \text{direct reflection}
 \end{aligned}$$

$\phi = \phi_p - \phi_s$   
P-s phase difference

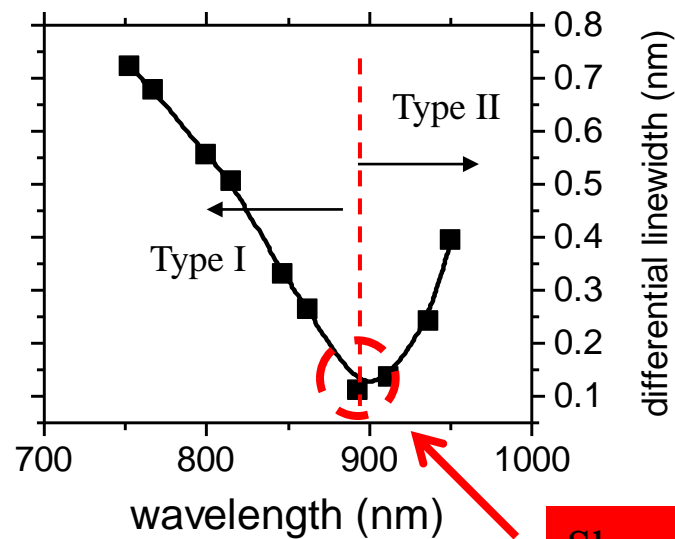
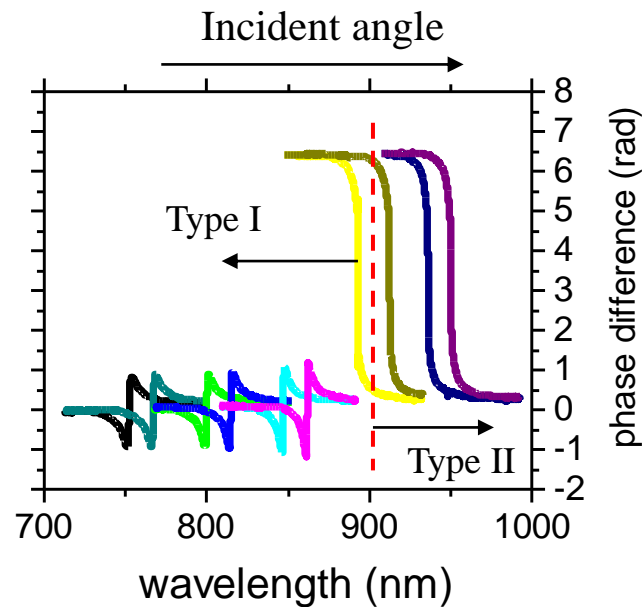
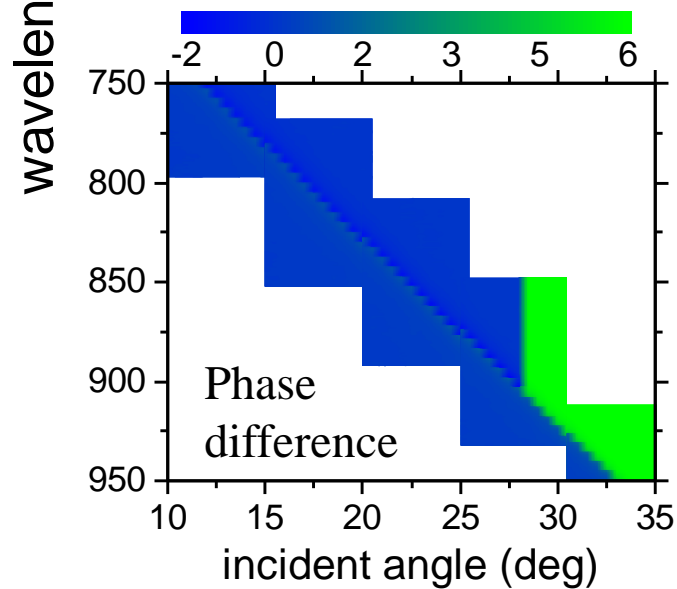
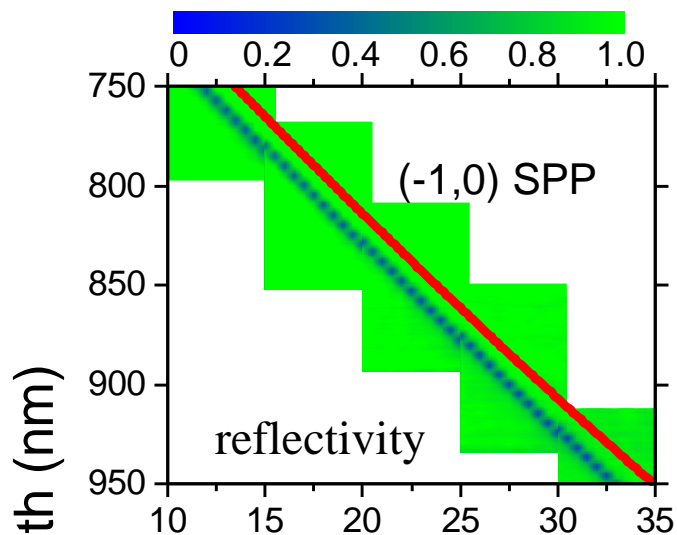
- Phase difference measurement by angle-resolved phase quadrature common-path interferometry



By Jones matrix

$$\tan \phi(\lambda) = \frac{I_0(\lambda) - I_{90}(\lambda)}{I_{+45}(\lambda) - I_{-45}(\lambda)}$$

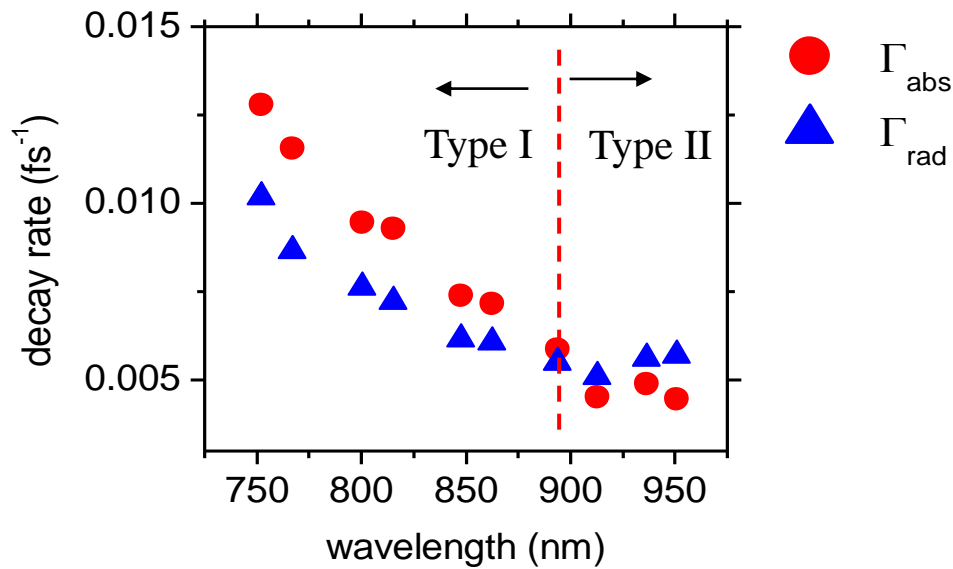




Sharpest phase jump  
Good for SPR sensing

Period = 600 nm, hole depth and radius = 80 and 60 nm





By Jones matrices to calculate the phase difference

$$\begin{bmatrix} r_p \\ r_s \end{bmatrix} = \begin{bmatrix} \alpha + \frac{\kappa^2 \Gamma_{rad} e^{i\varphi}}{2} \\ i(\omega - \omega_o) + \frac{\Gamma_{tot}}{2} \\ \beta \end{bmatrix}$$

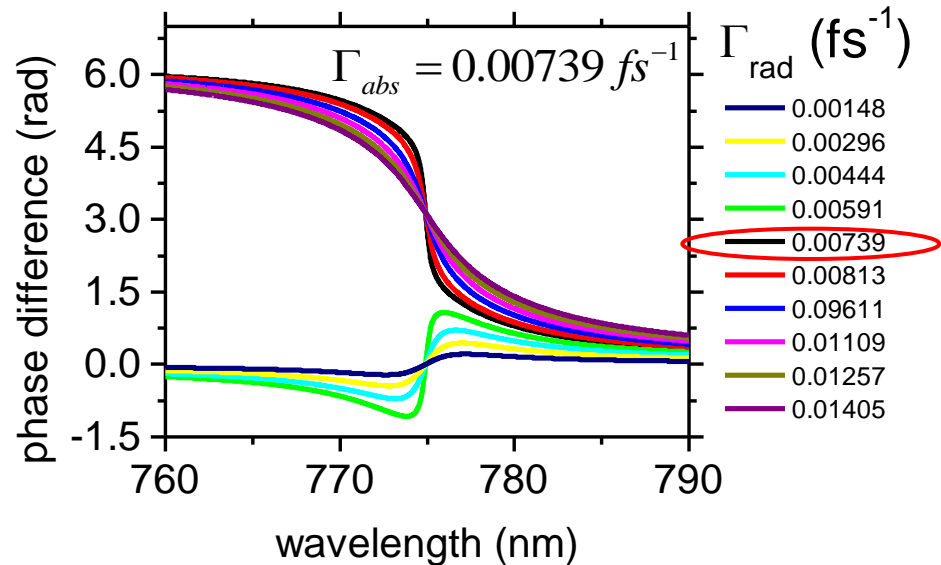
$$\alpha = \beta = 0.95$$

$$\kappa = \sqrt{2}$$

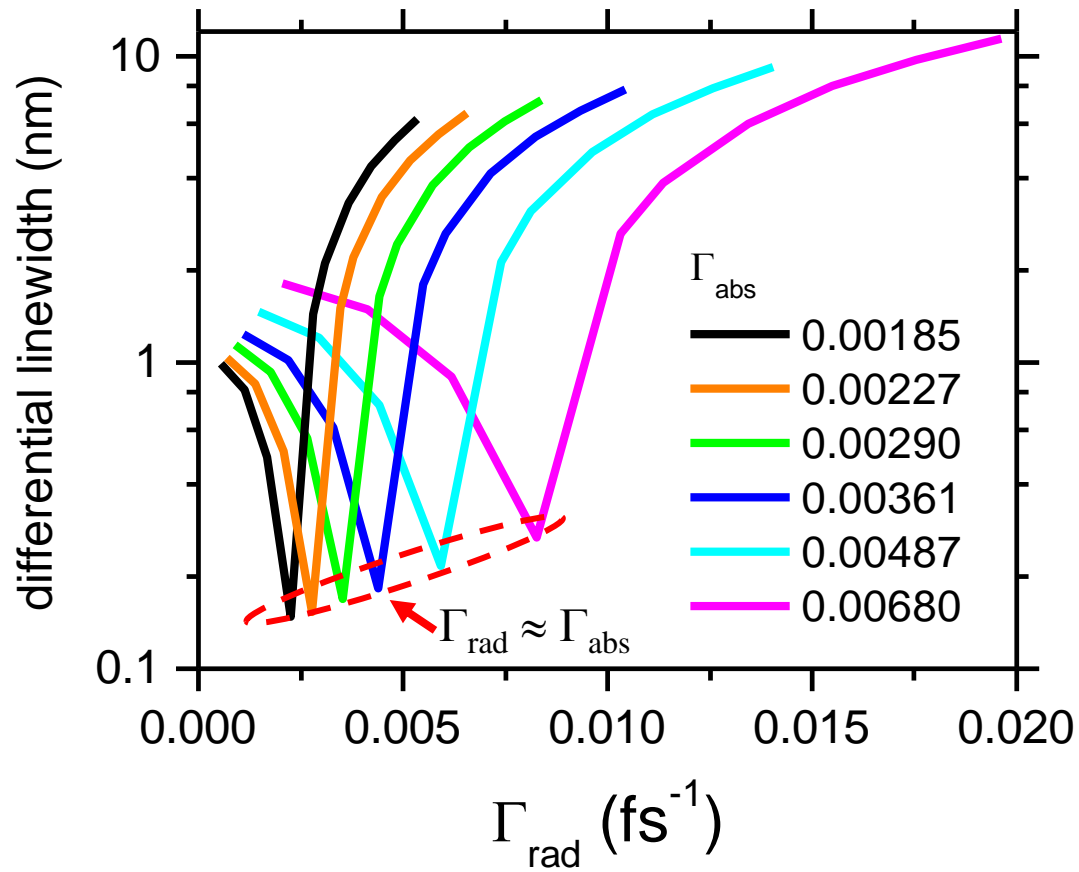
$$\varphi = \pi$$

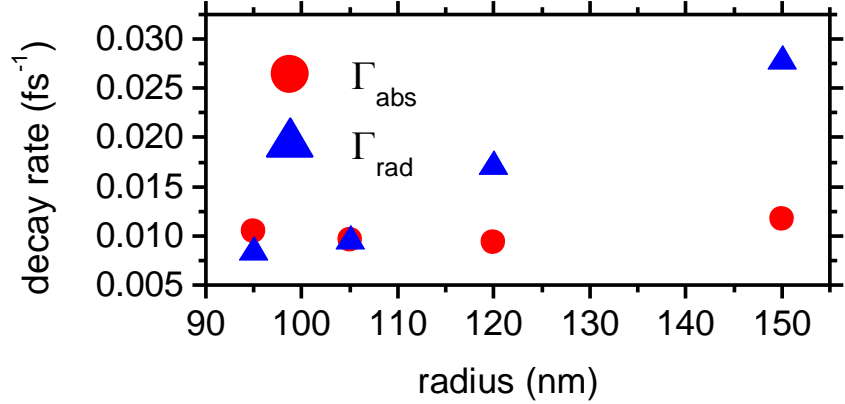
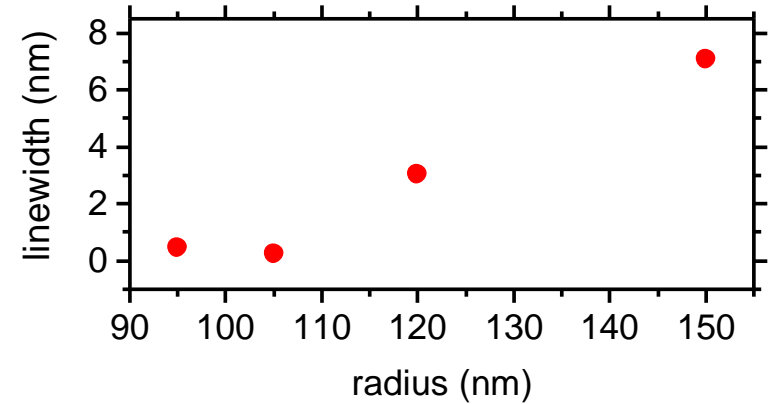
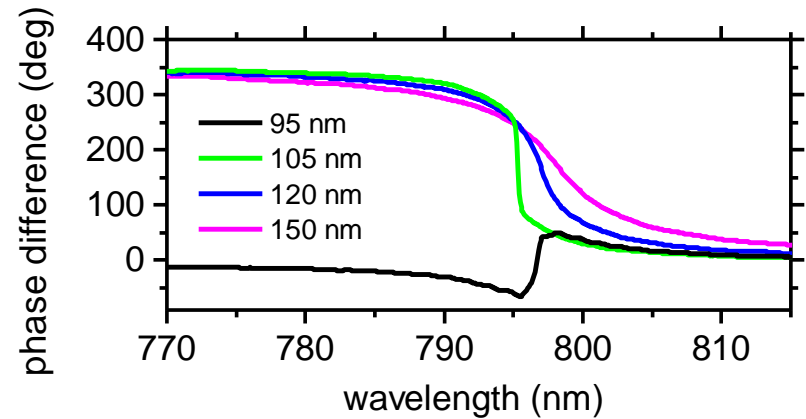
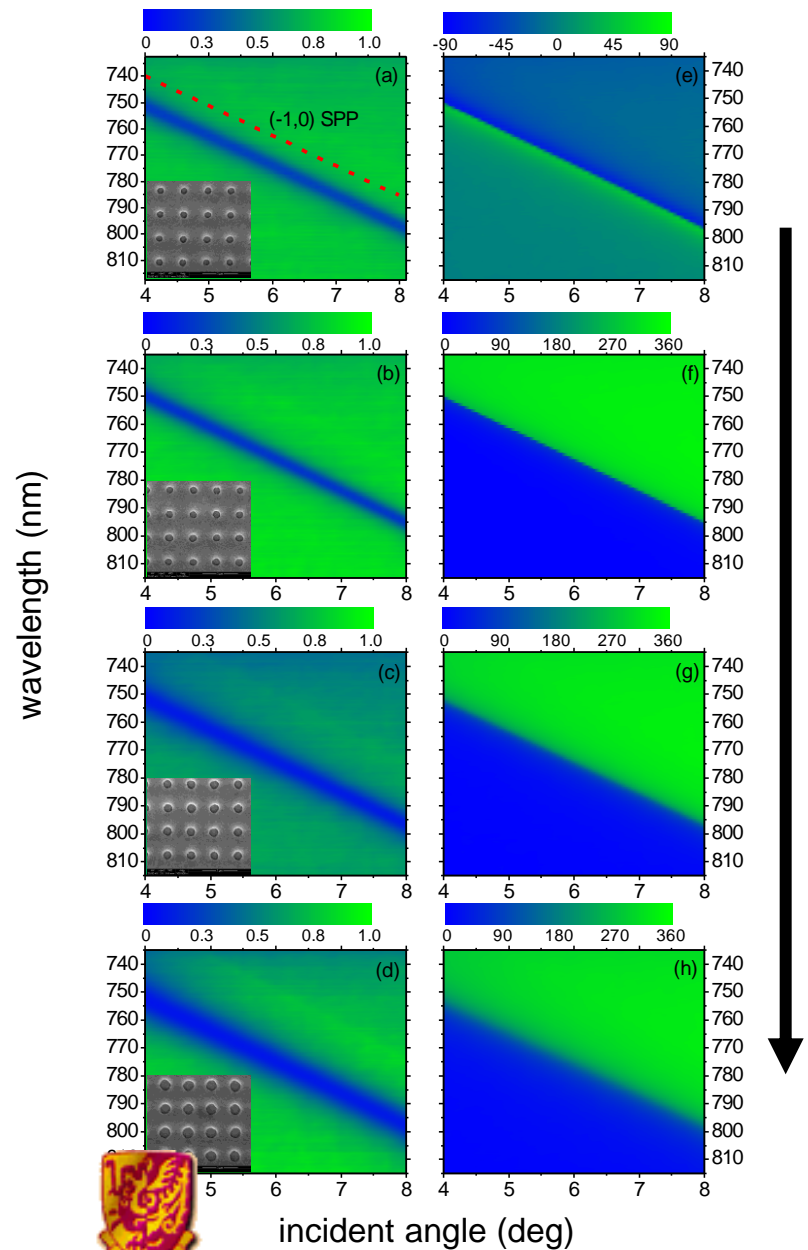
$$\Gamma_{abs} = 0.00739 \text{ fs}^{-1}$$

Transition at  $\Gamma_{rad} \approx \Gamma_{abs}$



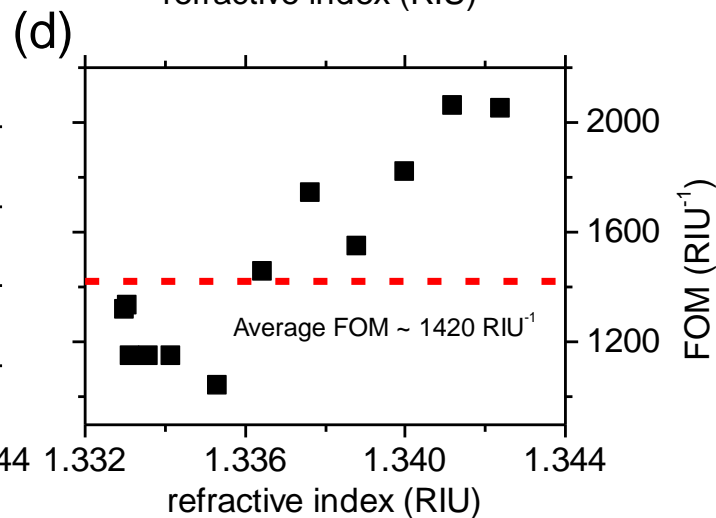
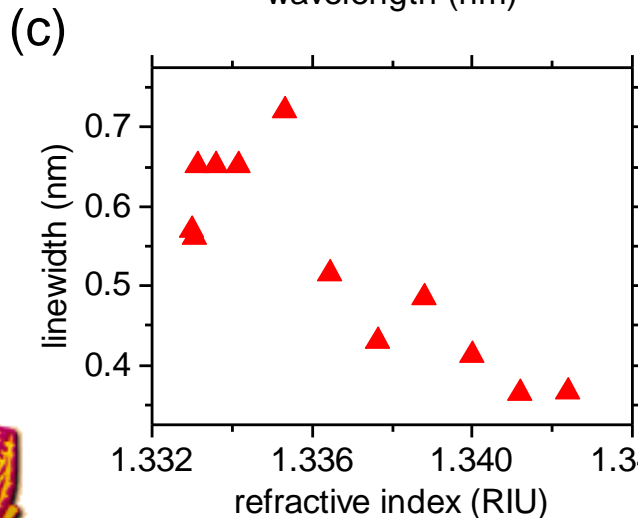
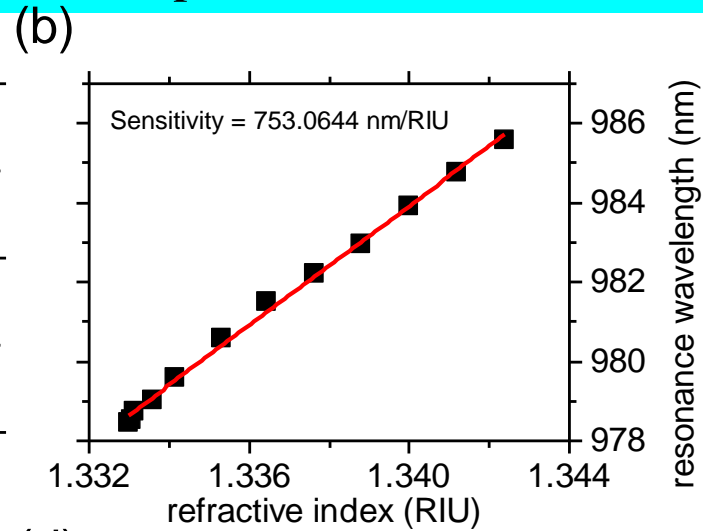
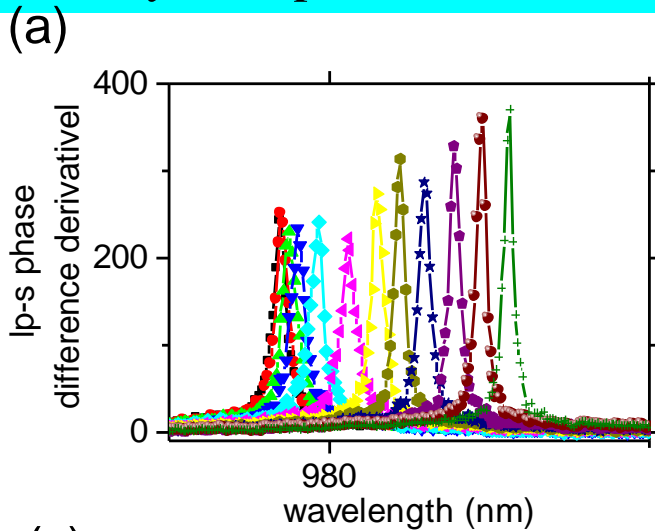






# Phase-based SPR

2-D Au array with period = 750 nm, hole depth = 100 nm and radius = 80 nm



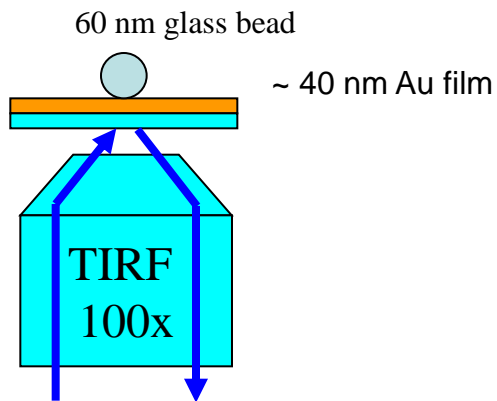
15 times  
higher than  
commercial  
SPR

$$FOM = \frac{\text{sensitivity}}{\text{linewidth}}$$

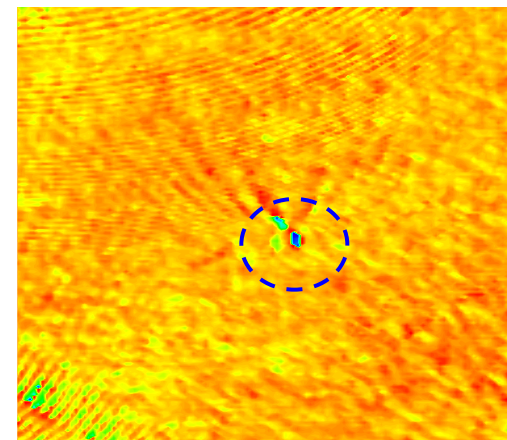
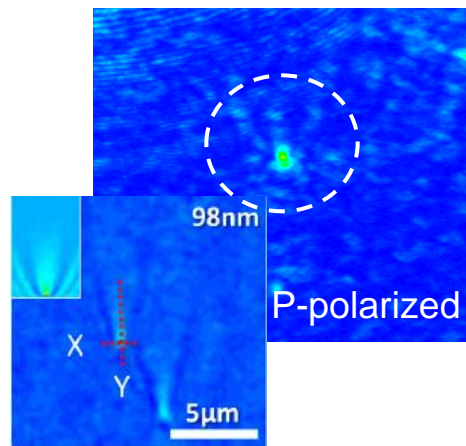


# Something extra: SPR imaging

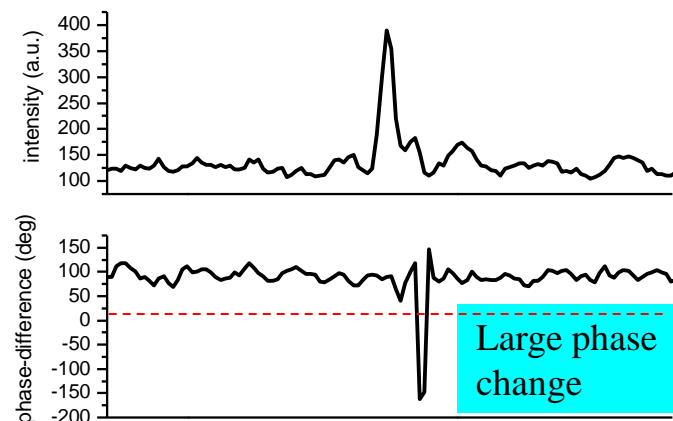
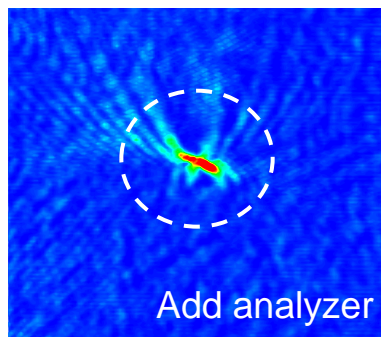
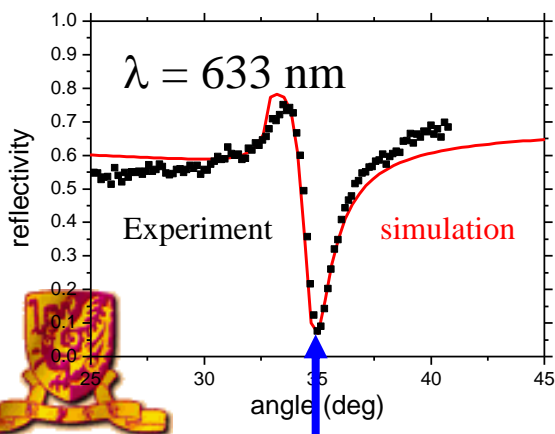
- CMT can be applied to attenuated total reflection thin film as well
- Change Au film thickness to tune  $\Gamma_{\text{rad}} \approx \Gamma_{\text{abs}}$  (Raether, Springer)



angle-resolved phase quadrature  
common-path interferometry



Wang et al, PNAS, 107, 16028 (2010)



# Conclusions

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- Coupled mode theory (CMT) is a useful analytical tool for studying the optical properties of period metallic arrays.
- Determine the absorption and radiative decay rates of SPPs.
- Under some special condition, the field strength is strongest when absorption rate = radiative decay rate.
- Sharpest p-s phase difference phase jump when absorption rate = radiative decay rate. Possible to have very high FOM.

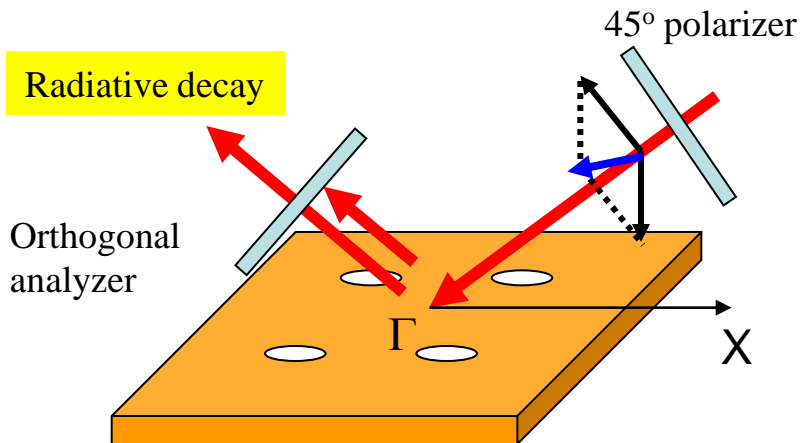


# Radiative decay of SPPs

For (-1,0) SPP, the Fano-like reflectivity contains radiative decay

$$R = \frac{|s_-|^2}{|s_+|^2} = \left| \alpha + \kappa\sqrt{2} \frac{a}{s_+} \right|^2 = \left| \underbrace{\alpha}_{\text{Direct reflection}} + \underbrace{\frac{\kappa^2 \Gamma_{rad} e^{i\varphi}}{2}}_{\text{Radiative decay of (-1,0) SPP}} \right|^2$$

Radiative decay of (-1,0) SPP



By Jones matrix

Lorentzian

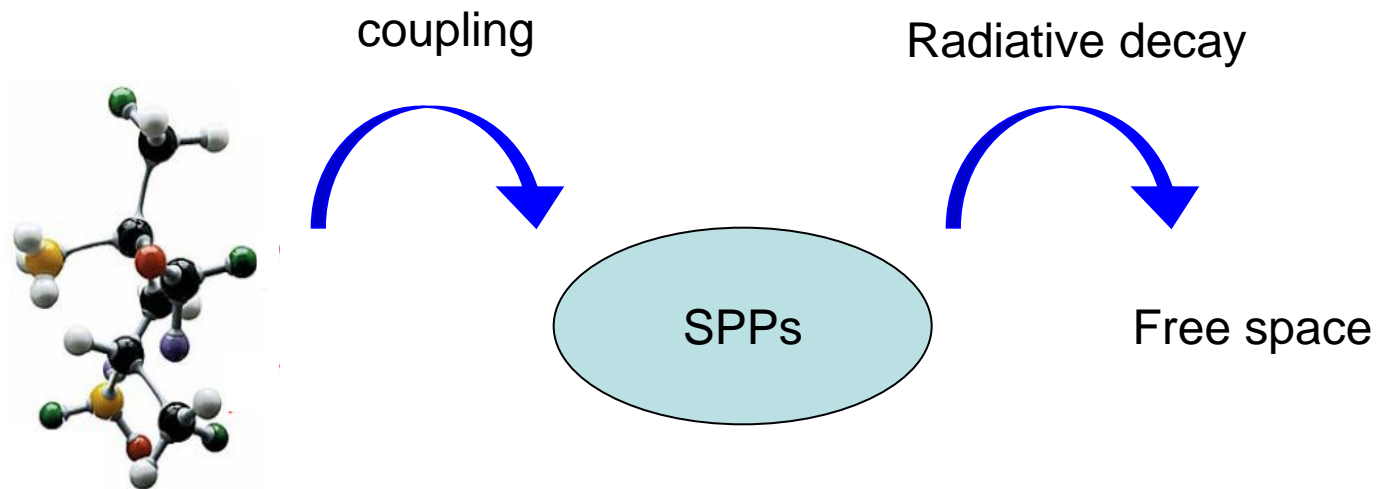
$$\frac{1}{4} \frac{(\kappa^2 \Gamma_{rad} / 2)^2}{(\omega - \omega_o)^2 + (\Gamma_{tot} / 2)^2}$$

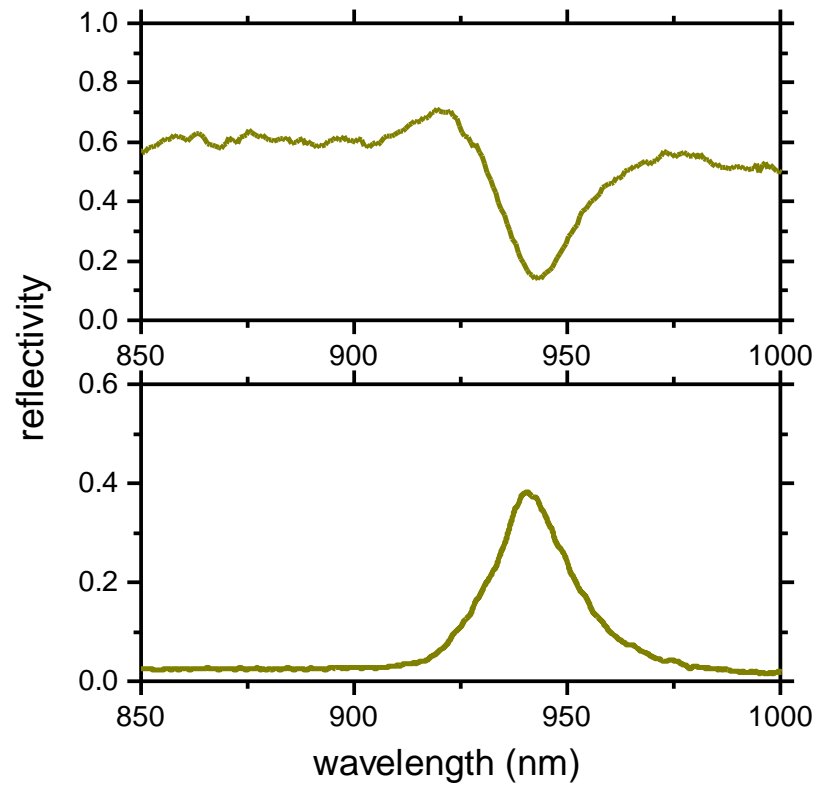
Power ratio between radiative scattering and incident field



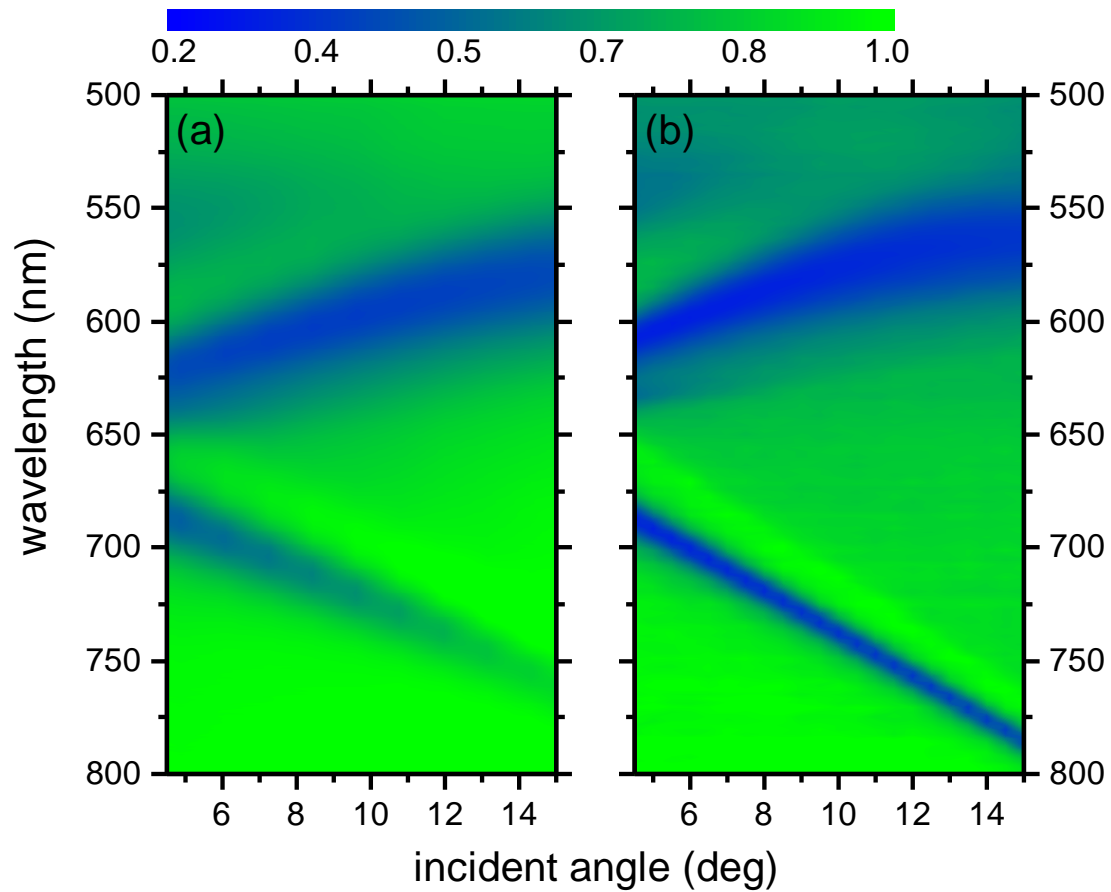
# Importance of radiative scattering

For Raman and fluorescence



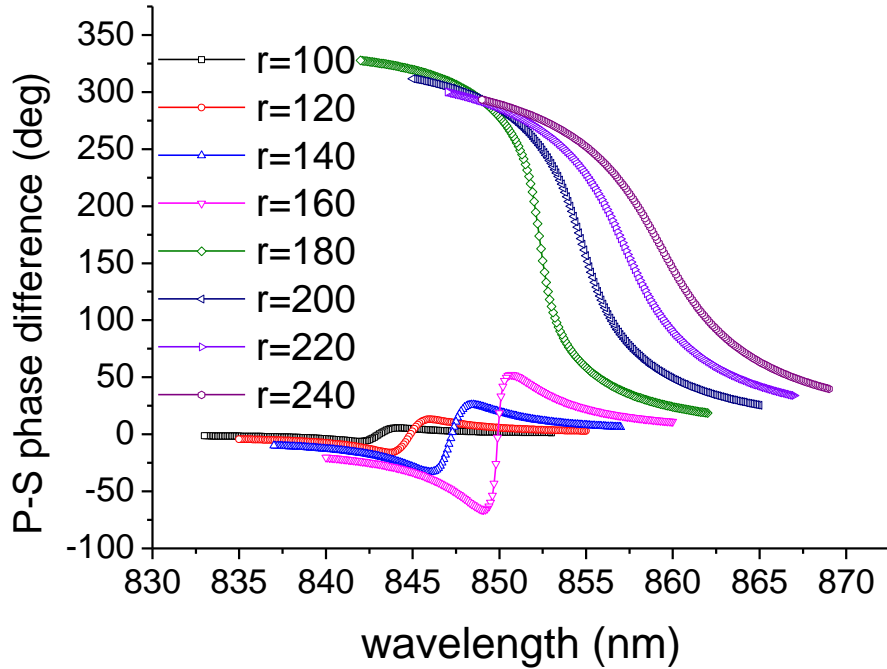




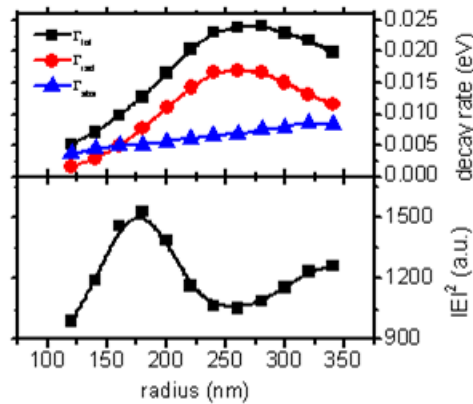
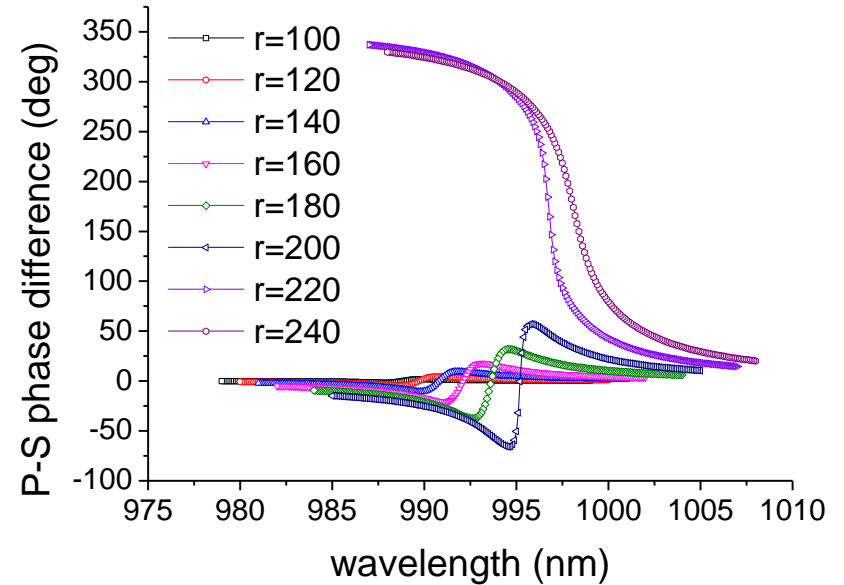


# Phase change

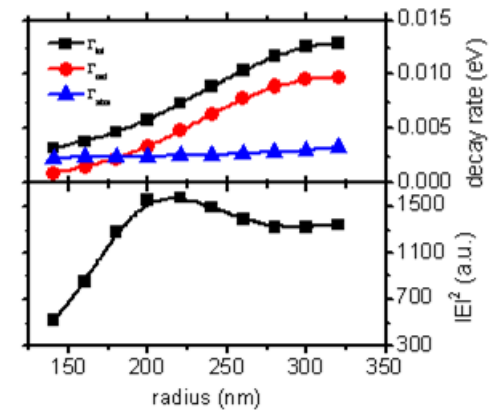
Period = 760nm,  $\lambda \sim 860$ nm



Period = 900nm,  $\lambda \sim 1000$ nm



Hole depth = 120 nm



Can we relate  $\Gamma_{\text{abs}}$  and  $\Gamma_{\text{rad}}$   
wavelength and geometry?

# We have for 2D hole arrays

- Combined quasi-static model, FDTD, and experiment to show

$$\Gamma_{tot} = \Gamma_{abs} + \Gamma_{rad}$$

$$\Gamma_{abs} = \frac{1.3 \times 10^7}{\lambda^{3.3}}$$

$$\Gamma_{rad} \approx \frac{6\pi^6 \lambda_p c}{P} \left\{ \frac{R^3 H^2}{\lambda^6} + 5\pi^2 \frac{R^{4.3} H^{2.7}}{\lambda^8} \right\}$$

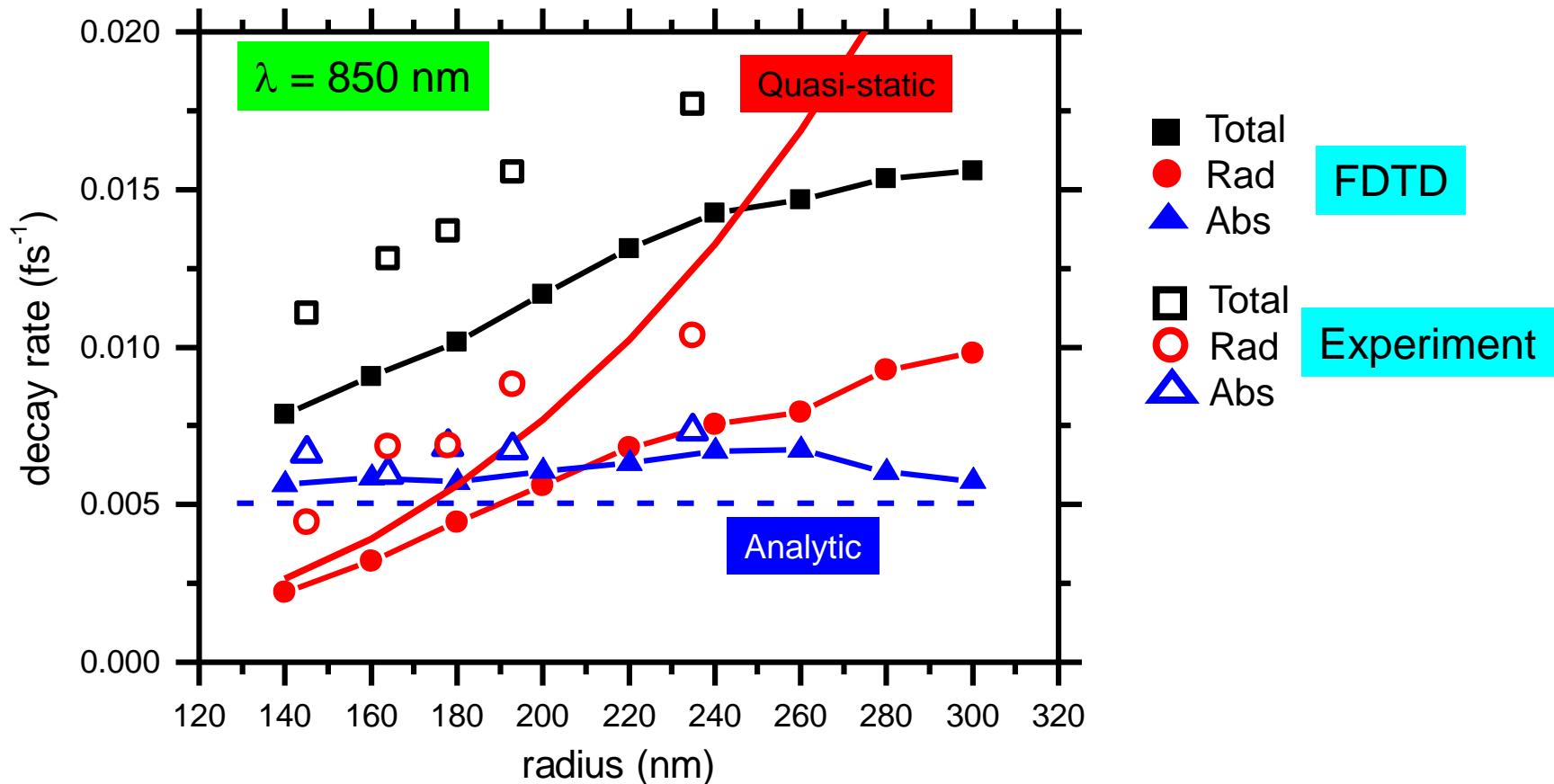
Both are functions of period (P), hole radius (R), and hole depth (H)

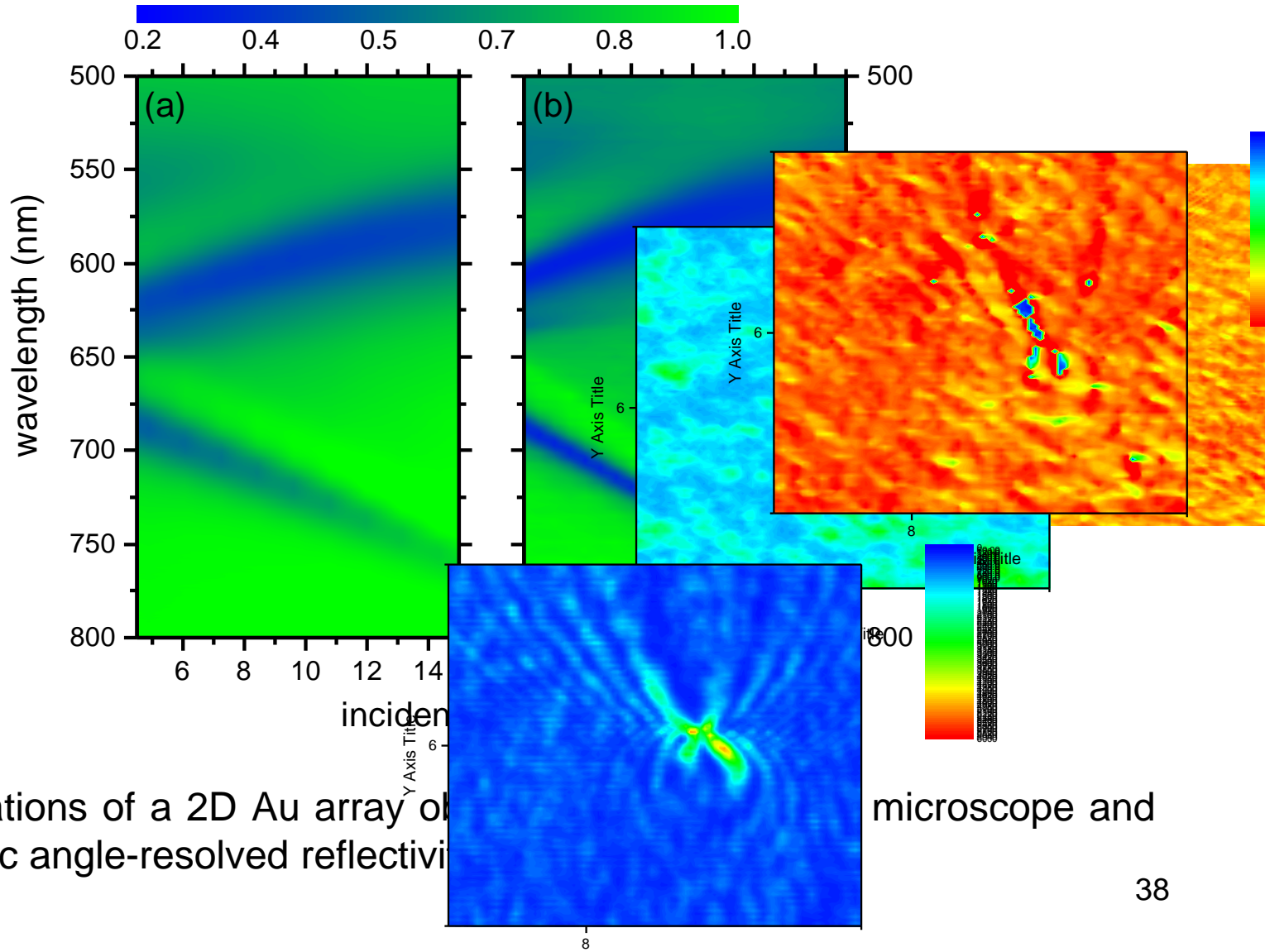
Give us guideline to control the decay process by using geometry



# One result

2D Au periodic arrays, period = 760 nm and hole depth = 60 nm





Dispersion relations of a 2D Au array of  $100 \text{ nm}$  period and  $50 \text{ nm}$  height (a) macroscopic angle-resolved reflectivity and (b) microscopic angle-resolved reflectivity.

microscope and