Excitonic perfect absorbers in the visible region

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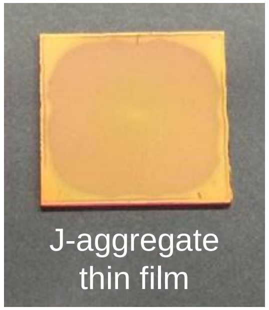
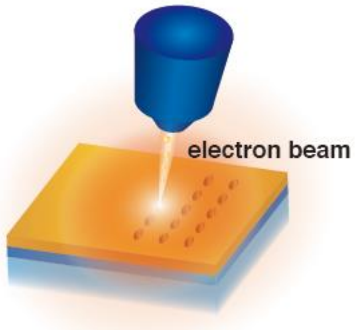
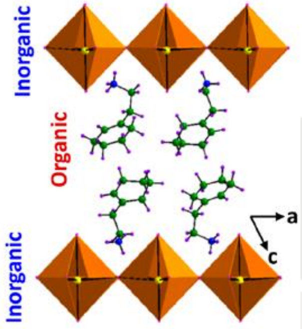


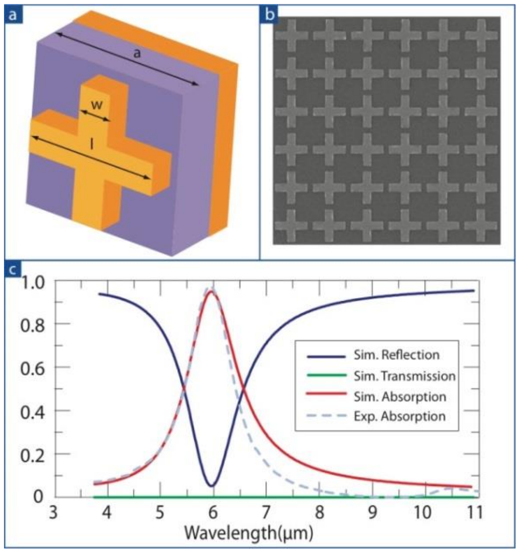




Perfect absorption of light in metal nanostructures

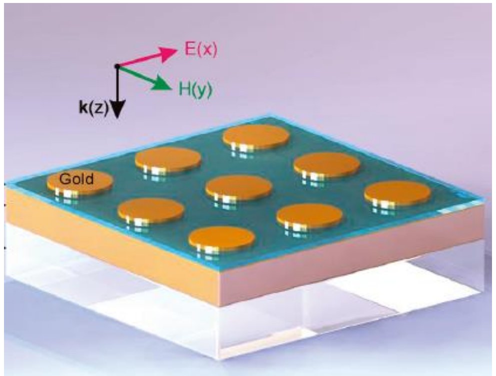
- Fundamental study: **Total absorption of light in a deep subwavelength volume**
- Device application: **Filters, Sensors, Detectors, Highly efficient energy conversion**
- Perfect absorber is a hot topic now
  - mainly based on *metal* nanostructures
  - the absorbed light energy is mostly lost due to metal losses ..

Impedance matching ($\mu/\varepsilon \sim 1$)



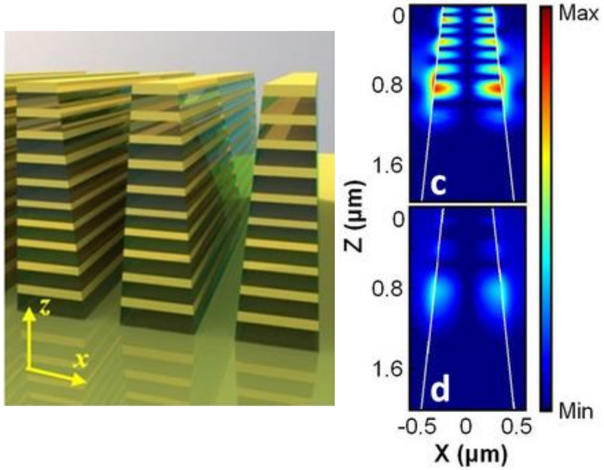
W. Padilla, PRL (2008)

Destructive interference
(R = 0, T = 0 → A = 1)



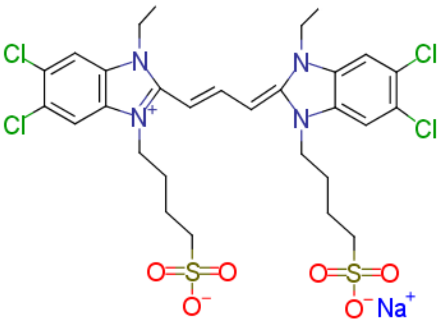
H. Giessen, Nano Lett (2010)

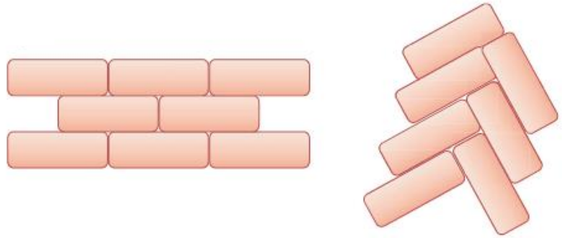
Broadband light trapping (or slow light) structures



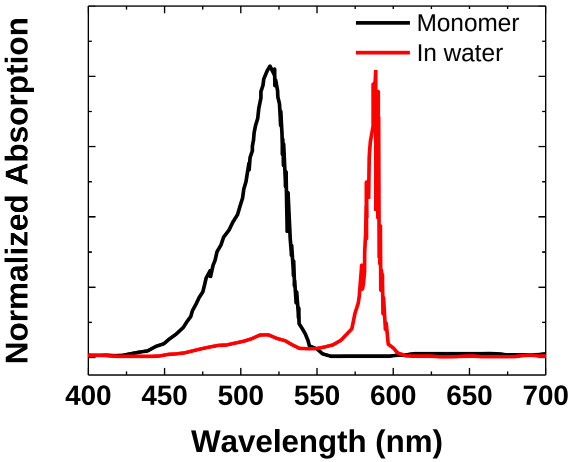
Q. Gan, Sci. Rep. (2014)

Organic dyes as tunable metals in visible



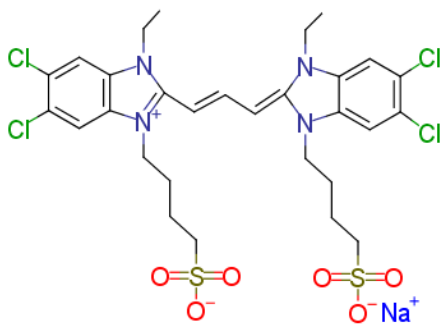


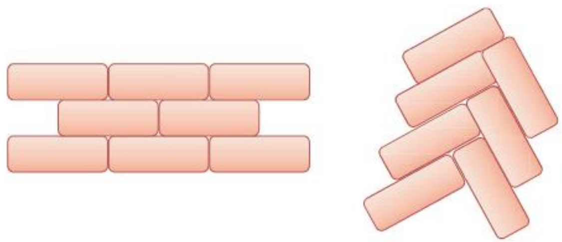
J-aggregates of TDBC dye
(***J*-aggregates** exhibit high/sharp absorption)



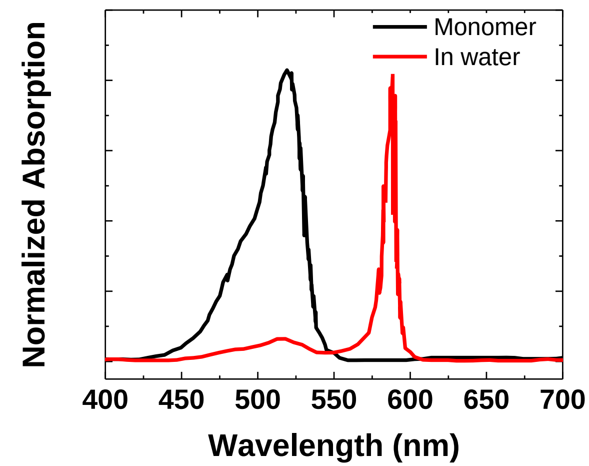
- Some organic molecules can have very large oscillator strengths
  → Large absorption at a certain wavelength due to Frenkel **exciton** formation
  → **Metallic response (Re[ε] < 0) from the Kramers-Kronig relation**
- Support Surface exciton polaritons (SEPs), similar to SPPs or SPhPs
- SEPs have been known since 1980's, but regained attention recently
- Recent reports: APL 103, 021104 (2013), Nano Lett 14, 2339 (2014)

Organic semiconductor as tunable metals in visible





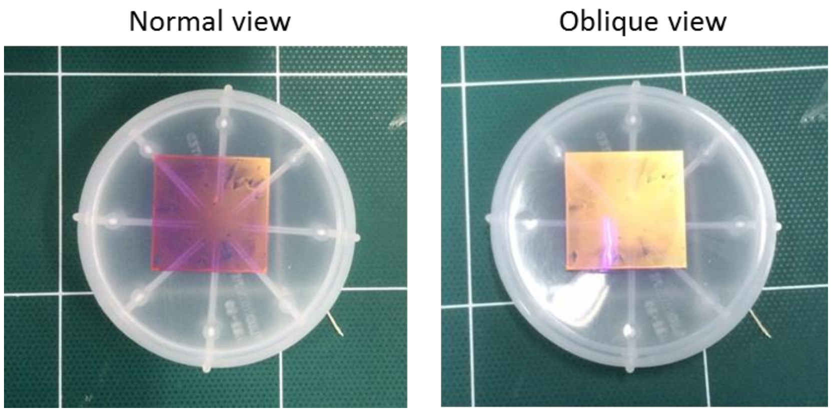
J-aggregates of TDBC dye
(***J*-aggregates** exhibit high/sharp absorption)



- For example, cyanine dyes form **J-aggregates** in water, and exhibit very high & sharp absorption peaks
- **TDBC** is one of such cyanine dyes (absorption peak in water ~ 590 nm)
- Support coherent exciton transport (over a sub-micrometer scale)
- TDBC has been used for strong coupling experiments

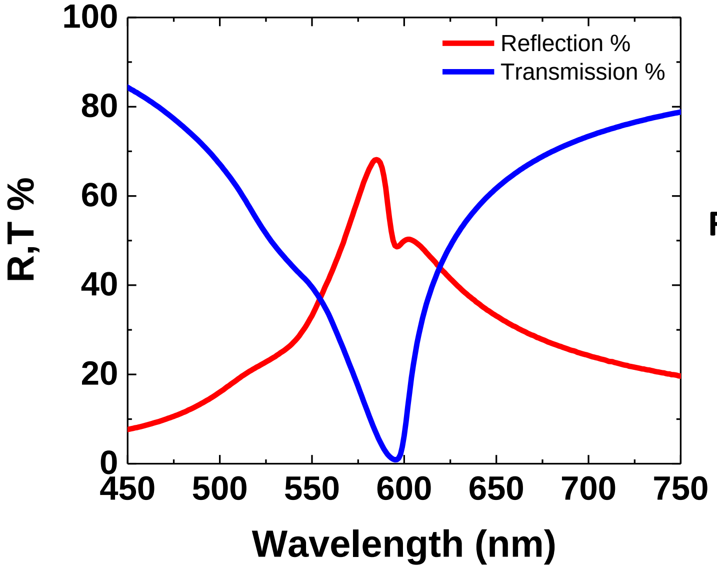
  (strong coupling b/w a cavity mode and excitons, b/w SPPs and excitons)
- Absorption peak positions are tunable over the whole visible region

Metallic response from Excitonic films

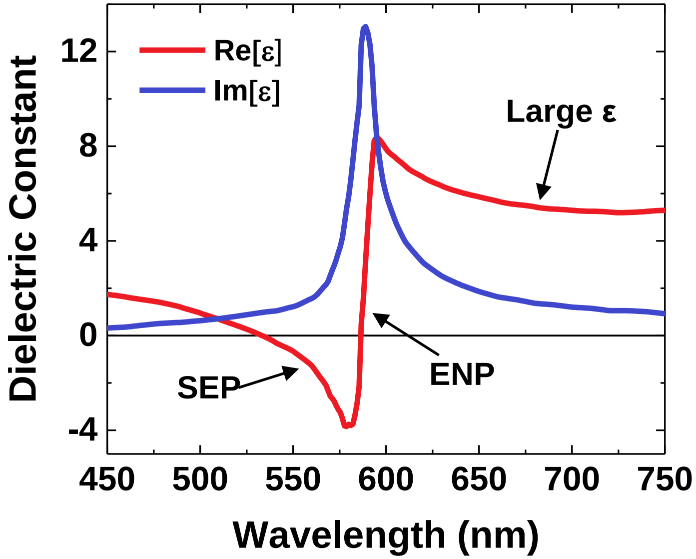


Spin-coated TDBC/PVA mixture
(sample prepared in our lab)

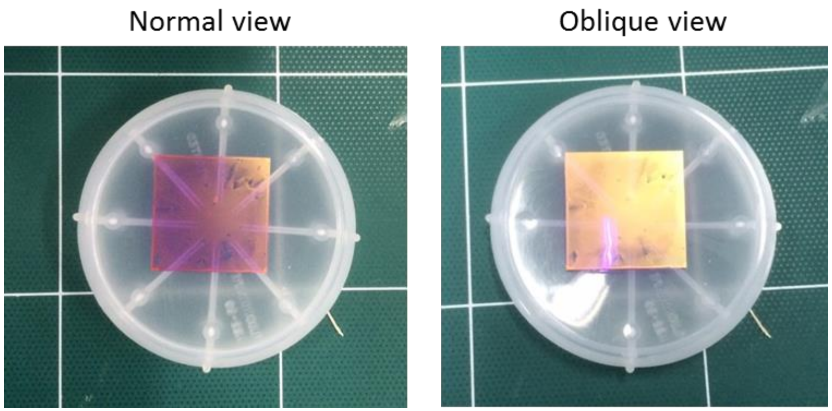
- Spin-coated TDBC/PVA mixture on quartz
  - 2.5 wt% TDBC + 2 wt% PVA
  - Spin speed ~ 4000 rpm
  - Thickness ~ 40 nm
- Possible to control the film thickness by varying composition or spin speed



Fitting to Fresnel equations →

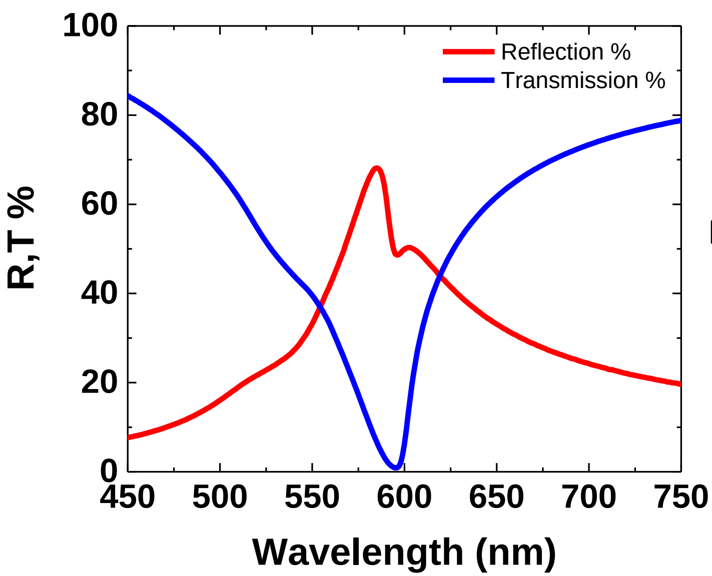


Metallic response from Excitonic films



Spin-coated TDBC/PVA mixture
(sample prepared in our lab)

- Various features appear in the visible region (SPP/SEP, ENP, Large-ε, etc)
- Possible to fine tune the dielectric constants with the film composition
- Potentially, mixing with gain materials can help reducing loss further



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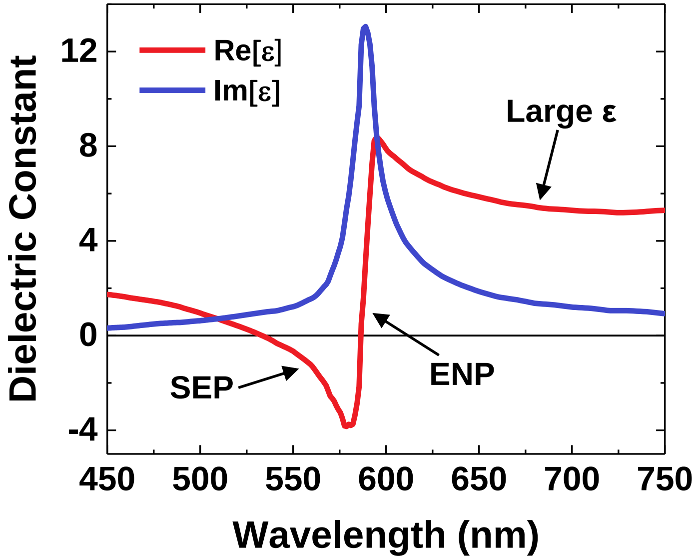
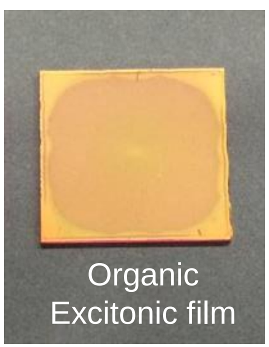
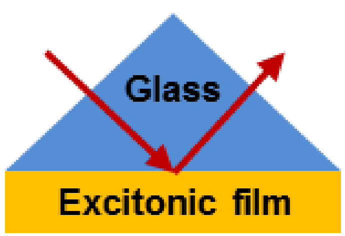


This talk: Excitonic perfect absorbers

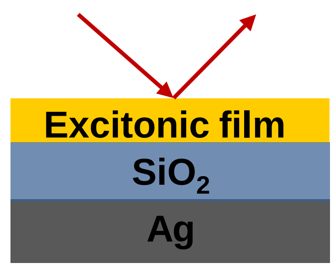
- Diverse absorption spectra in planar J-aggregate thin films
- Strong absorption even away from the excitonic pole
- Perfecta absorption & Enhanced photoluminescence
- Electron-beam-induced nanoscale patterning
- Extension to 2D-layered Perovskites: absorption below E_g

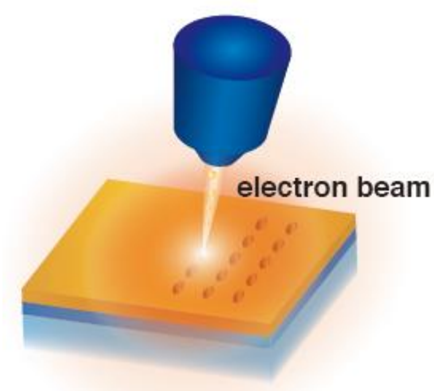


(i) ATR condition



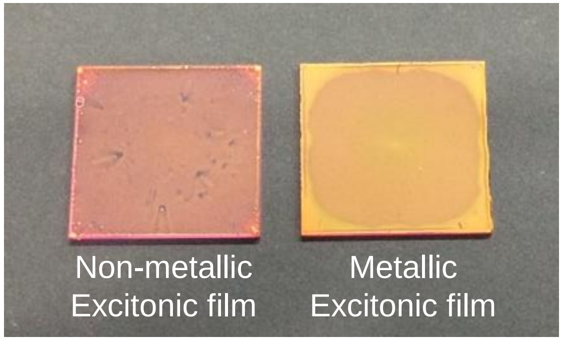
(ii) Silver substrate with a phase controller



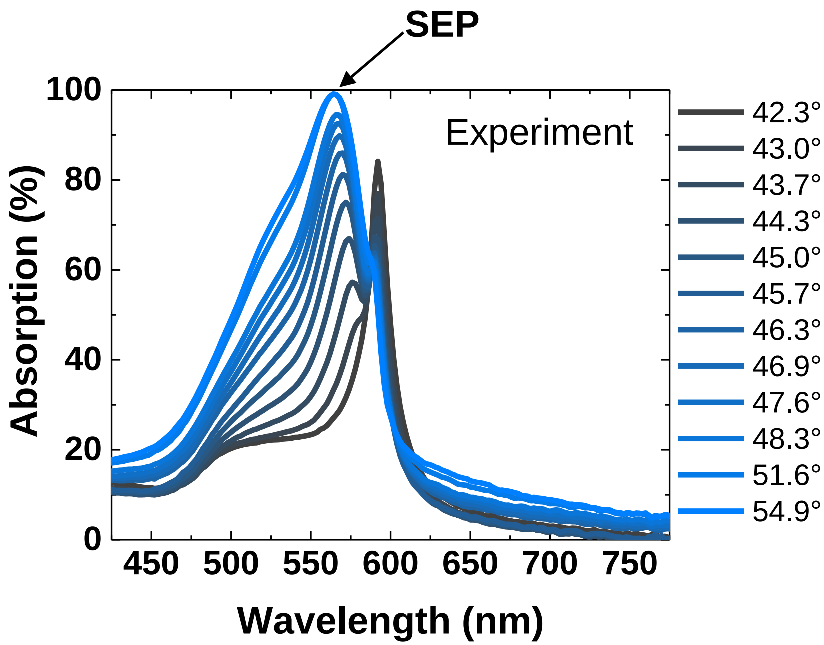


Perfect absorption in excitonic films: (i) ATR condition

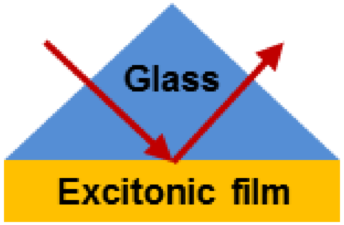
ACS Photonics 4, 1138 (2017)



- Absorption spectra in the ATR condition
  (reveal mode coupling features)
- Perfect absorption observed at SEP and ENP wavelengths
- Compare *metallic* and *non-metallic* films

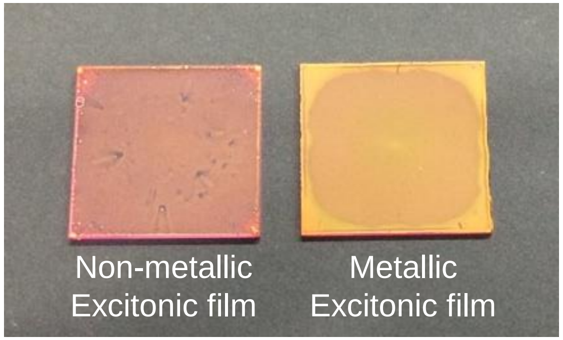


p-polarized spectra for the *metallic* excitonic film



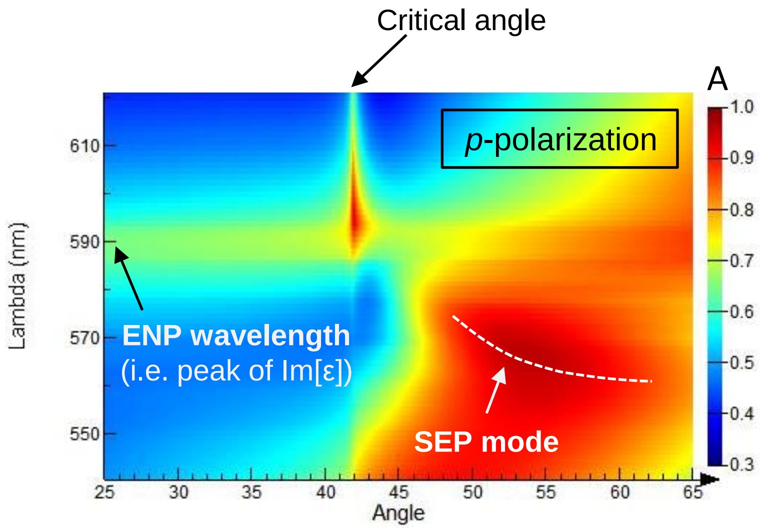
Perfect absorption in excitonic films: (i) ATR condition

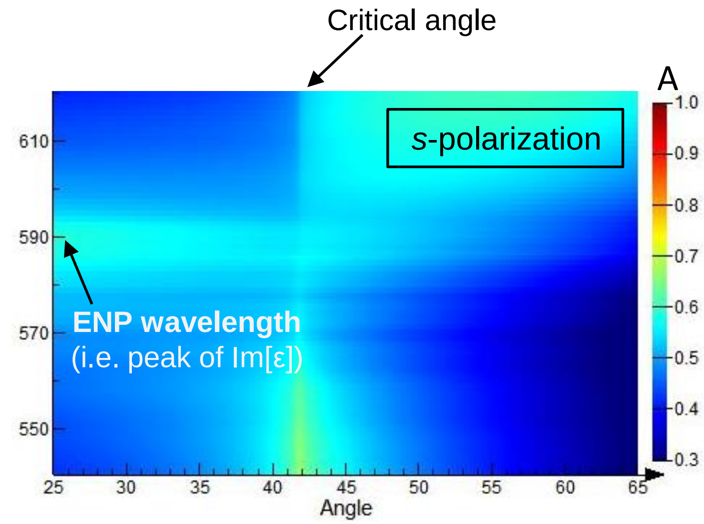
ACS Photonics 4, 1138 (2017)



- Absorption spectra in the ATR condition (reveal mode coupling features)
- Perfect absorption observed at SEP and ENP wavelengths
- Compare *metallic* and *non-metallic* films

TMM calculation





Perfect absorption in excitonic films: (i) ATR condition

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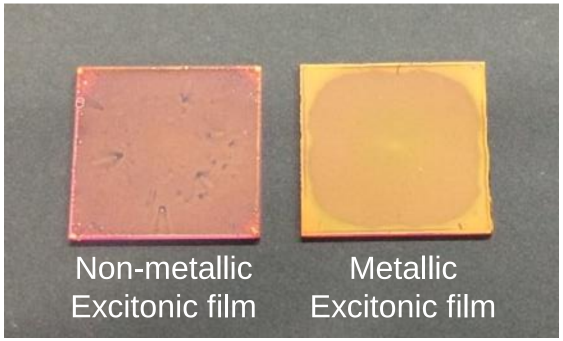
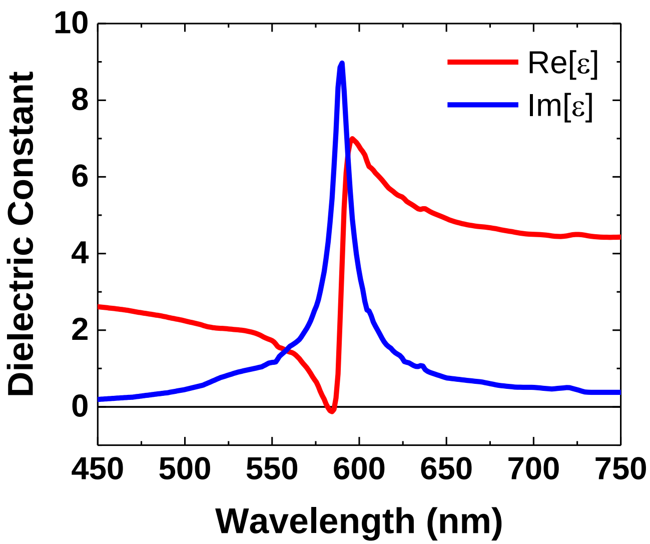
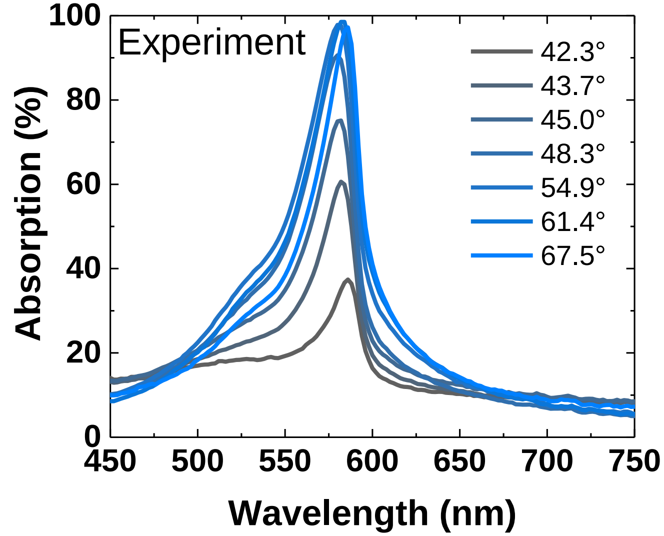
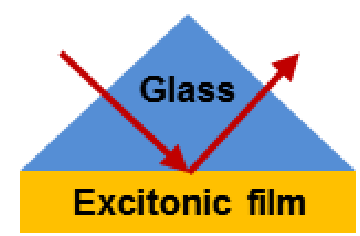


- Absorption spectra in the ATR condition (reveal mode coupling features)
- Perfect absorption observed at SEP and ENP wavelengths
- Compare *metallic* and *non-metallic* films



p-polarized spectra for the *non-metallic* excitonic film





Perfect absorption in excitonic films: (i) ATR condition

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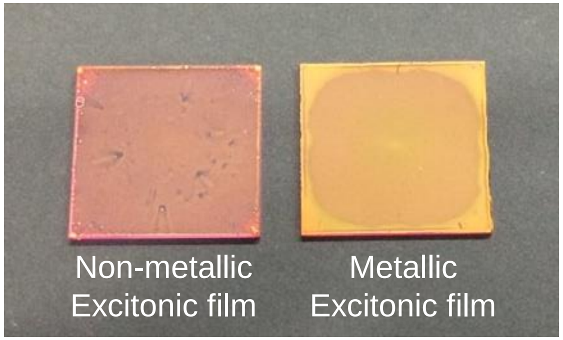
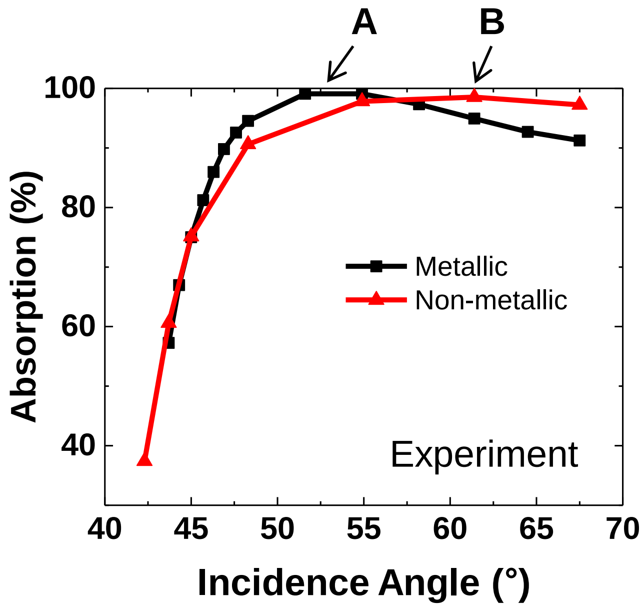
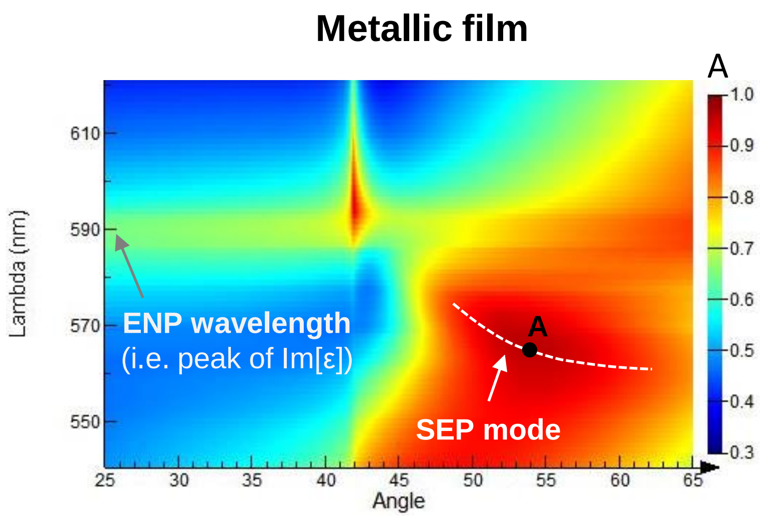
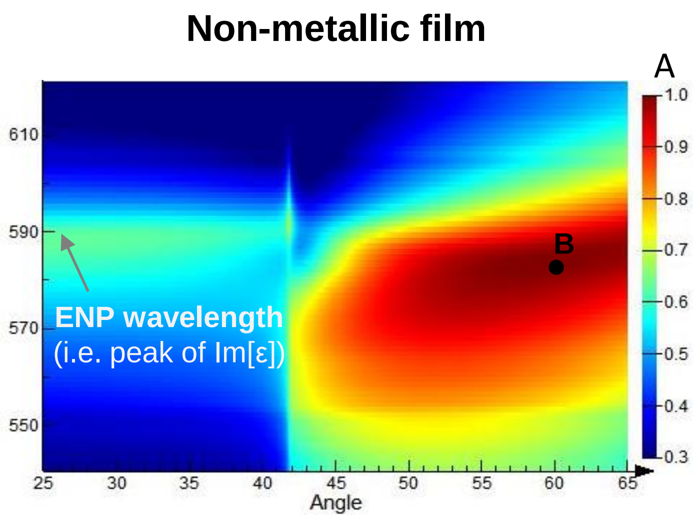




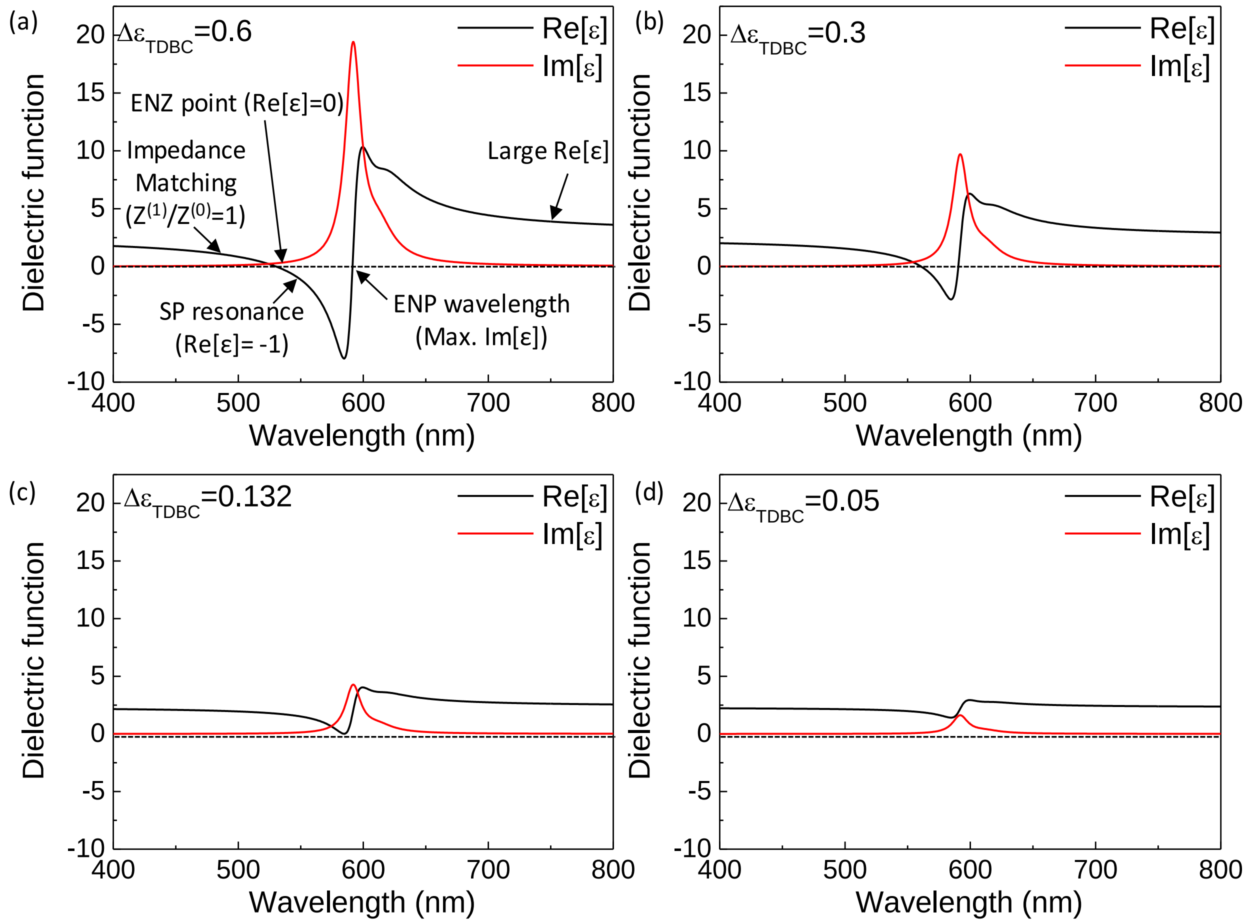
p-polarized ATR measurements

TMM calculation





Optical modes in excitonic thin films



$$\varepsilon(\omega) = \varepsilon_{PVA} + \Delta\varepsilon_{TDBC} \sum_i A_i \frac{\omega_i^2}{\omega_i^2 - \omega^2 - i\omega\gamma_i}$$

* From ellipsometer measurements:

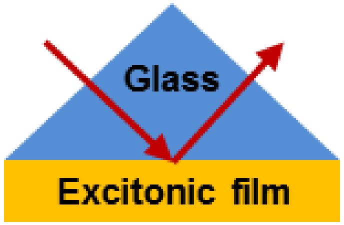
ε_{PVA} = 2.26

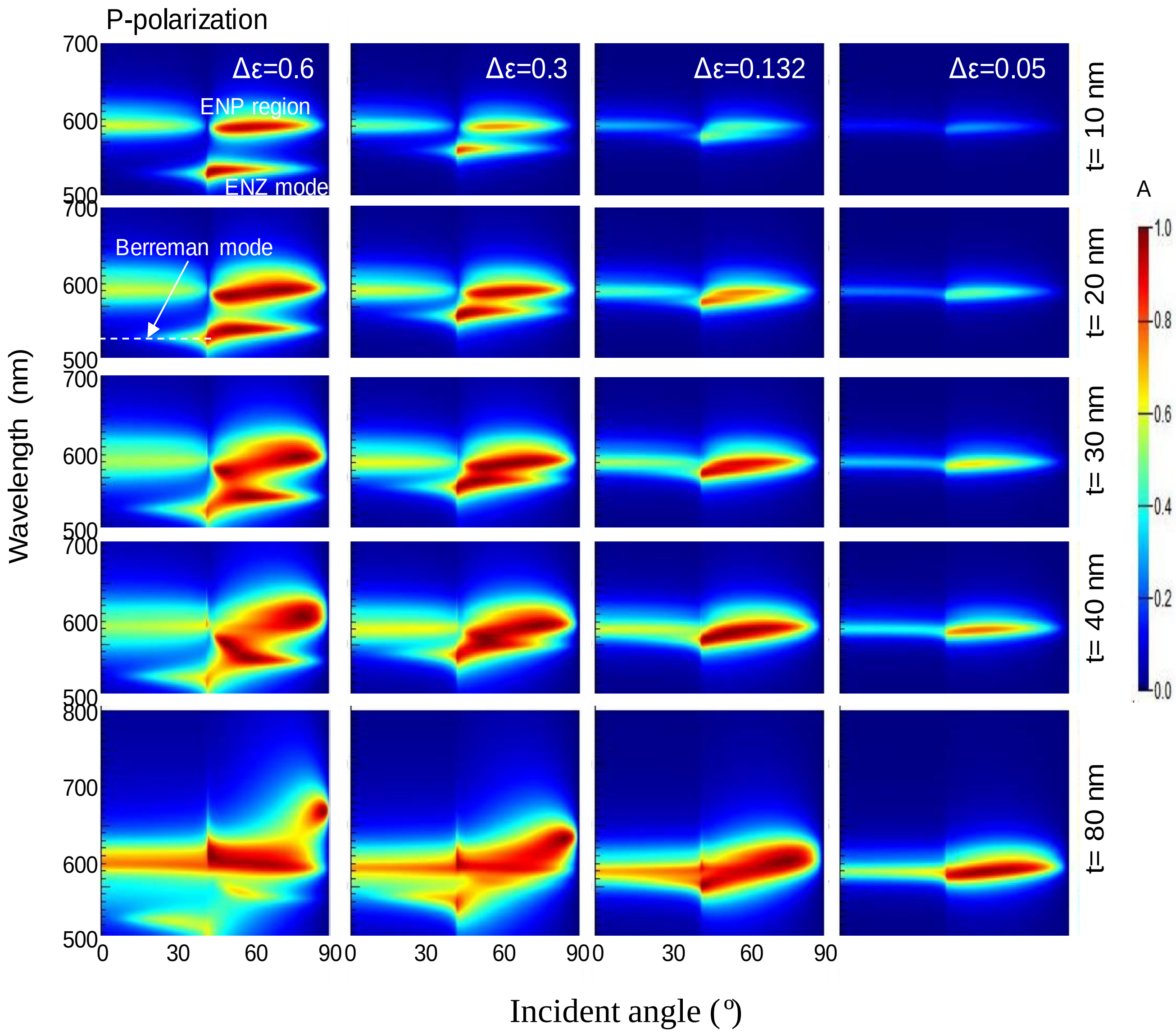
ω_1=2.10 eV, γ_1=0.053 eV

ω_2=2.03 eV, γ_2=0.0988 eV

Optical modes in excitonic thin films

TMM calculation





Optical modes in excitonic thin films

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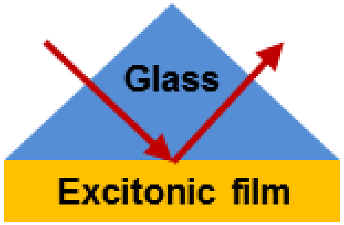
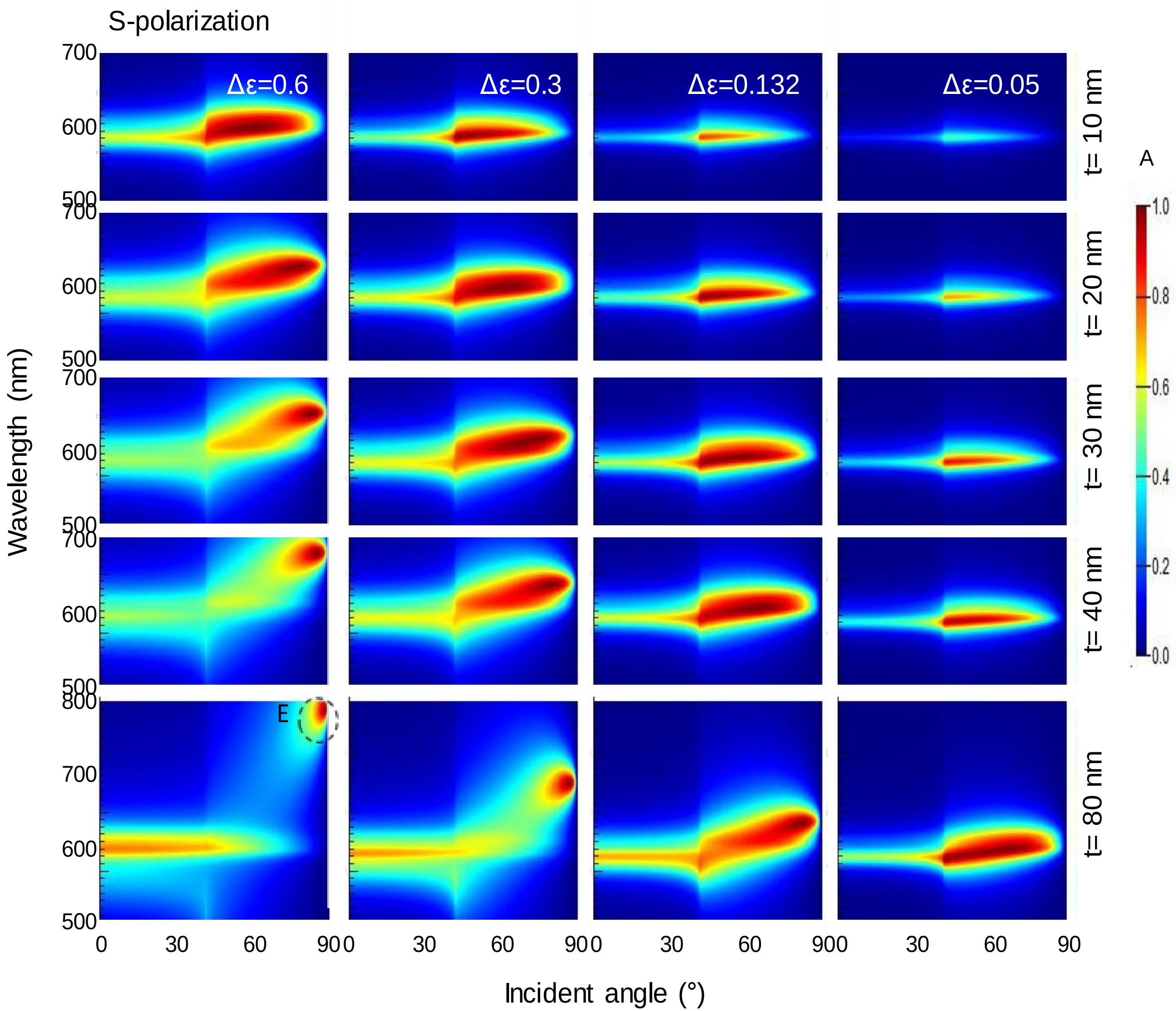
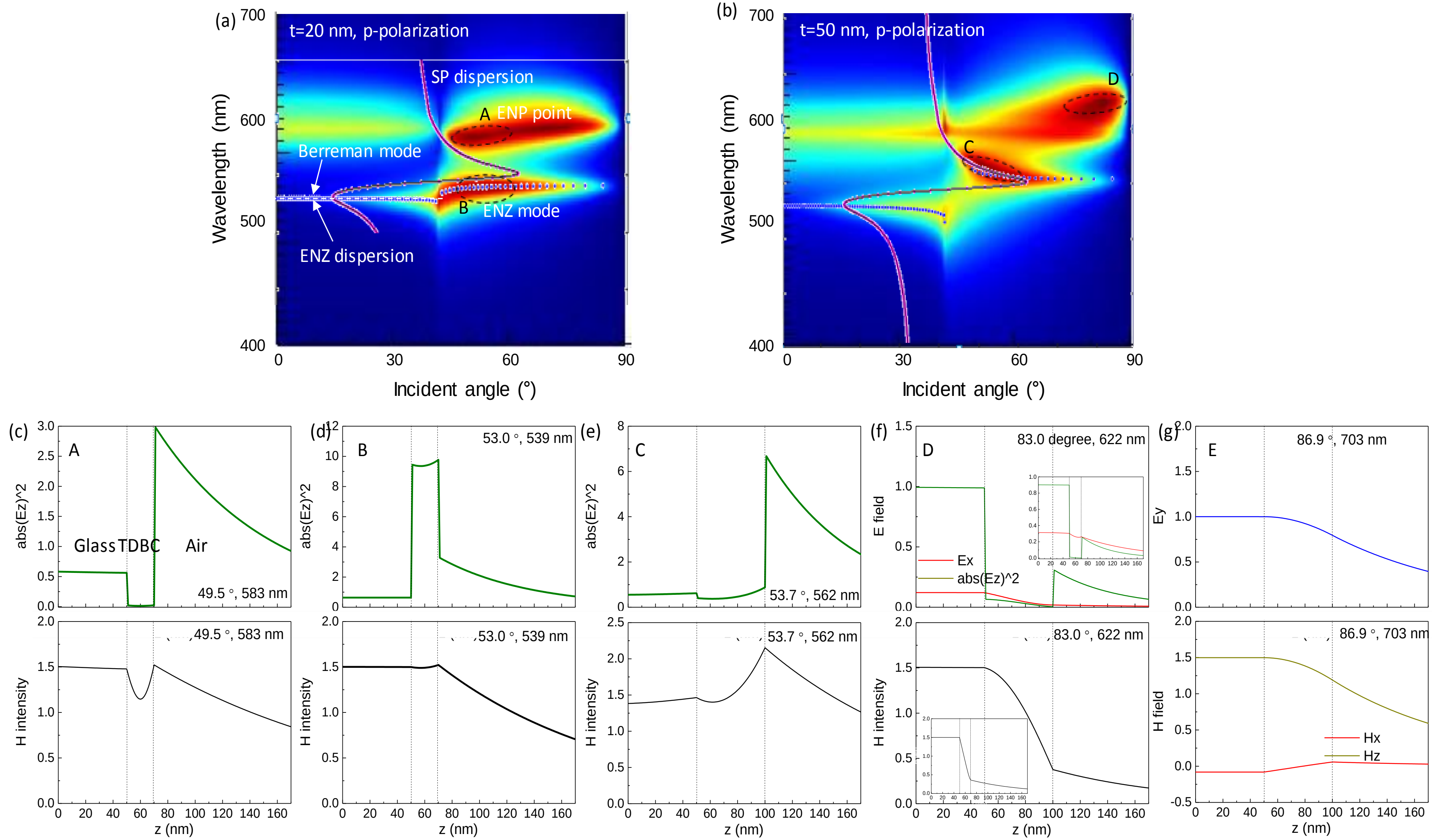
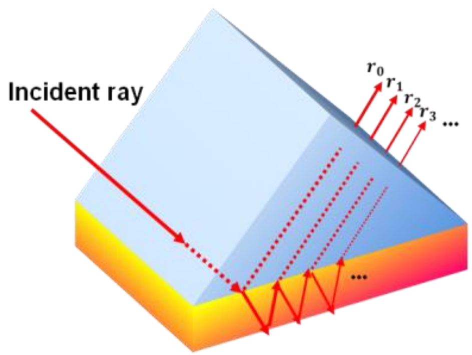


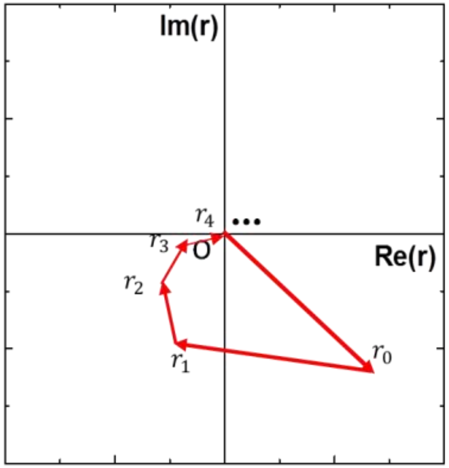


Optical modes in excitonic thin films



Perfect absorption in *non-metallic* films



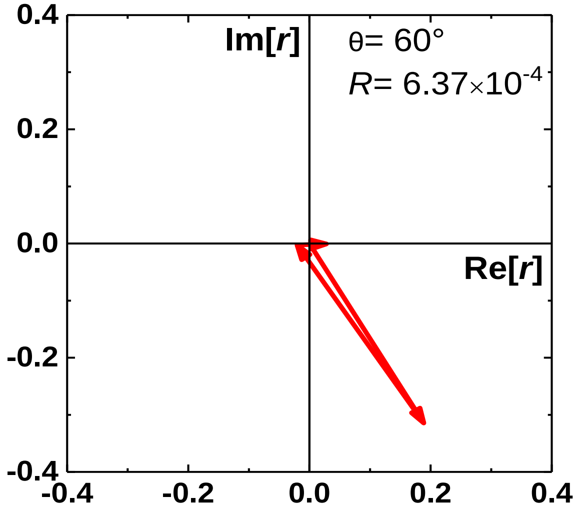


Partial reflected waves

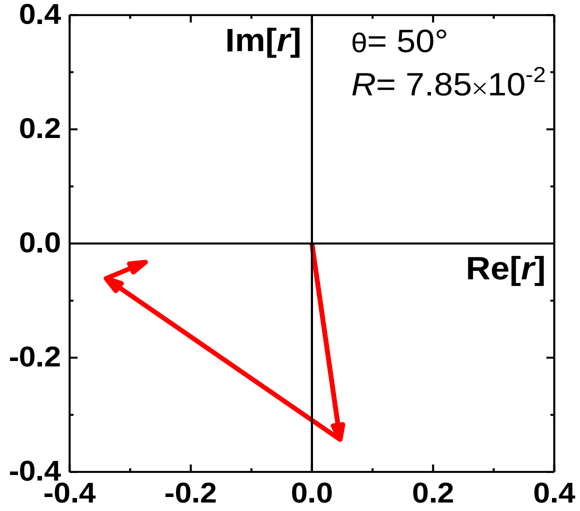
$$r = \sum_{m=0}^{\infty} r_m = r_{12} + \sum_{m=1}^{\infty} t_{12} r_{23}^{m} r_{21}^{m-1} t_{21} e^{2mi\beta}$$

Lossy film → Thin film interference occurs even in *ultra-thin* films

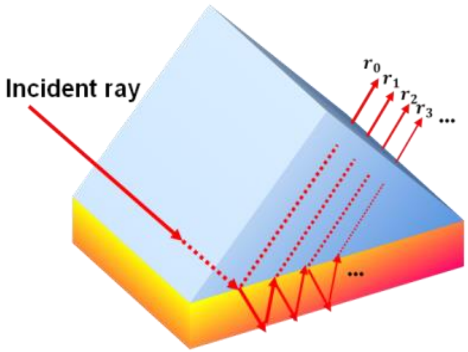
On-resonance



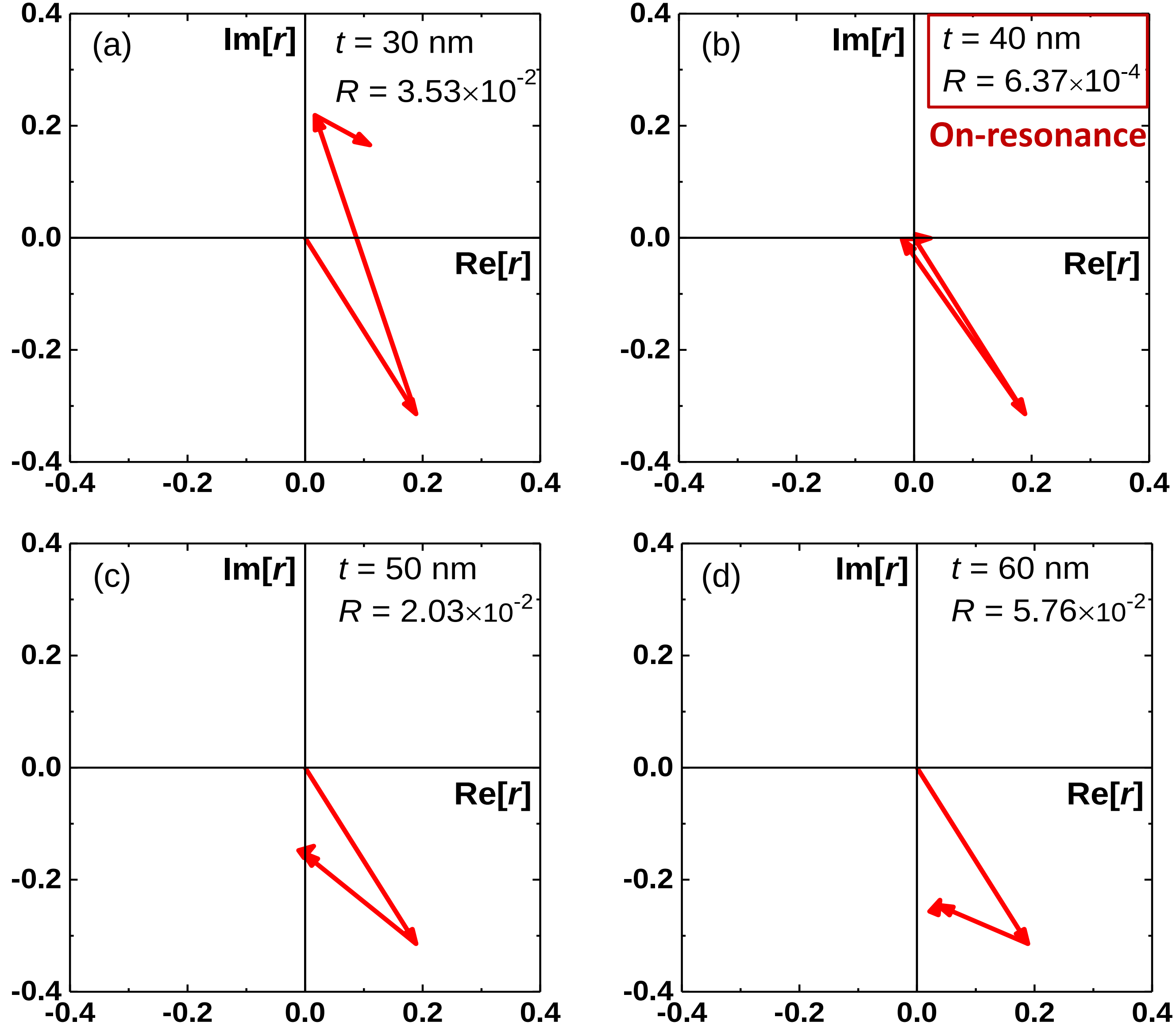
Off-resonance



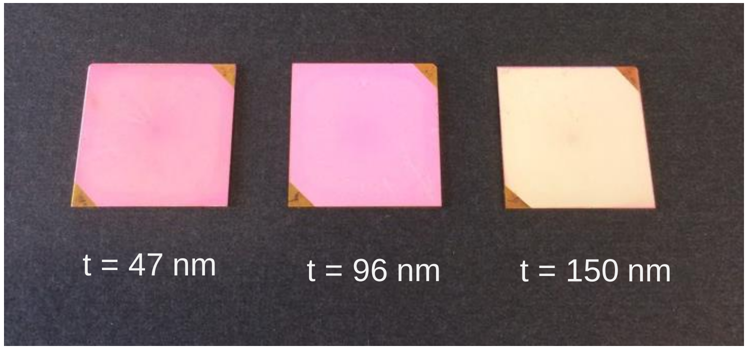
Perfect absorption in *non-metallic* films



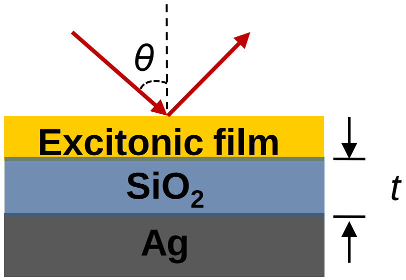
- Incidence Angle: 60º
- Vary film thickness t



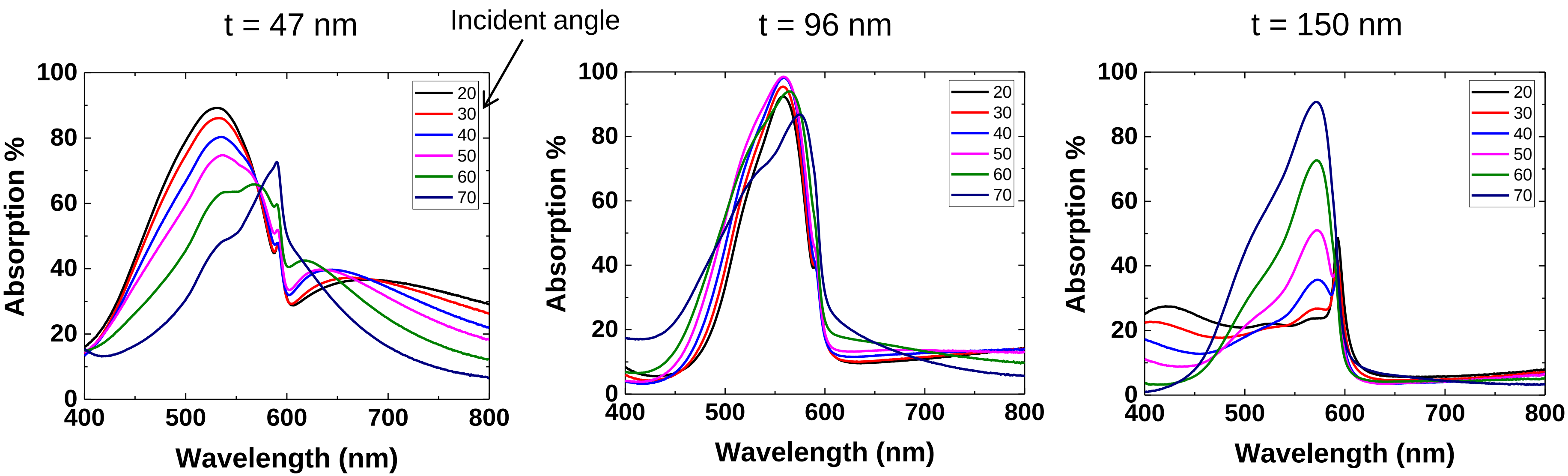
Perfect absorption in excitonic films: (ii) SiO_2/Ag



Metallic **excitonic film on Ag substrate with a *phase controller***



p-polarization (Air incidence)



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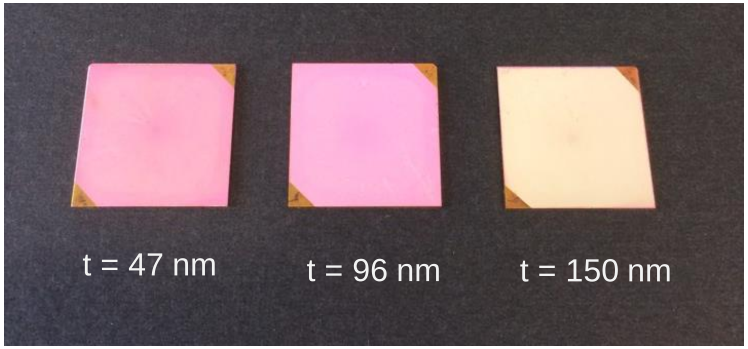

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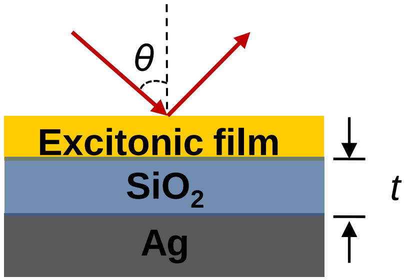
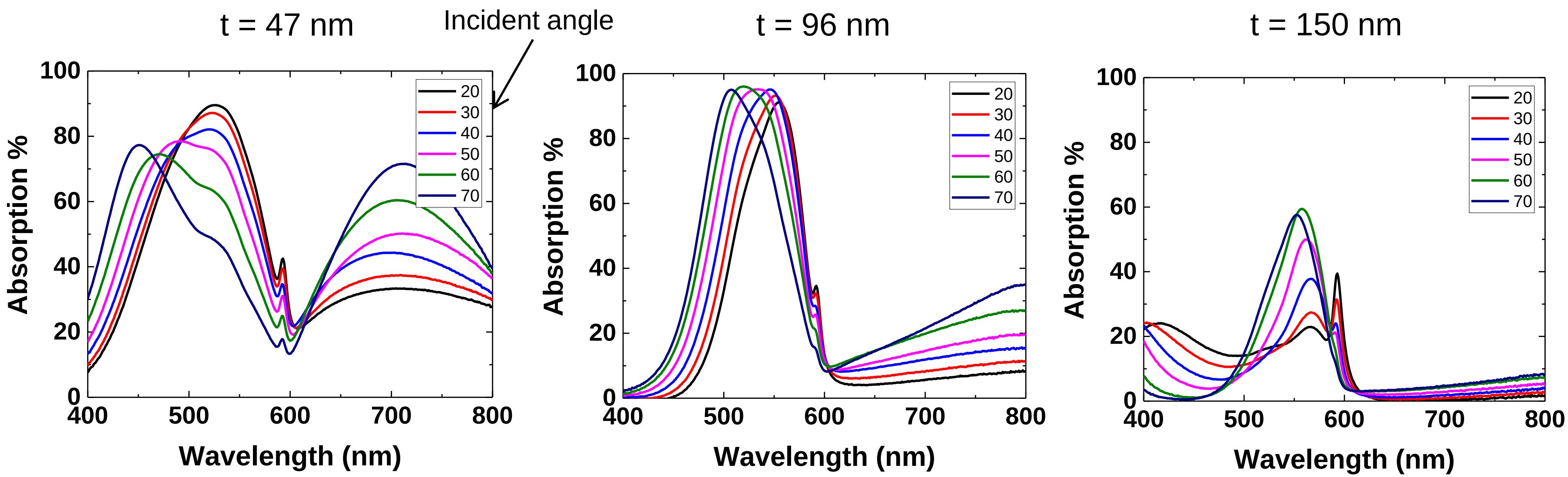


s-polarization (Air incidence)



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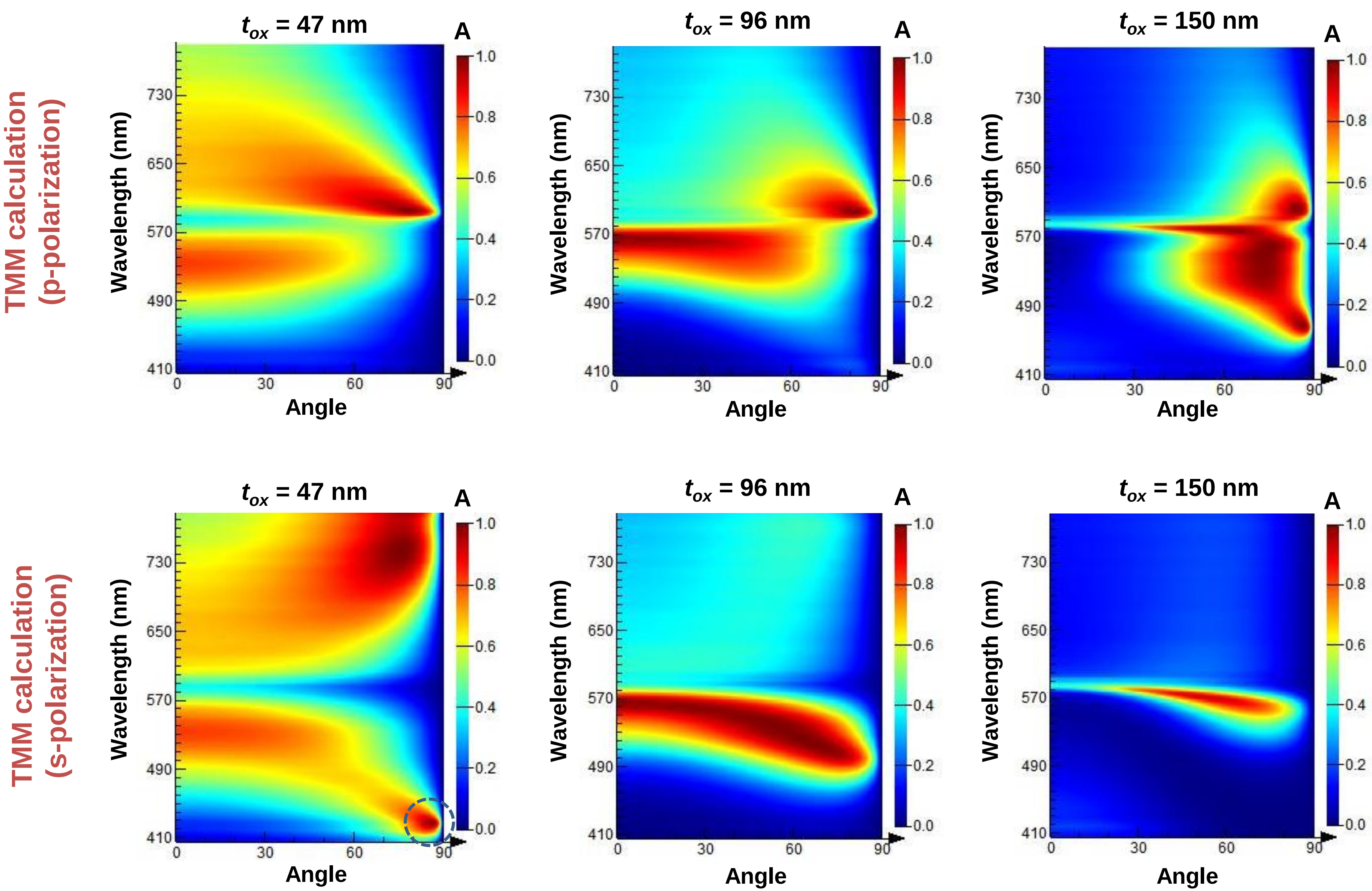

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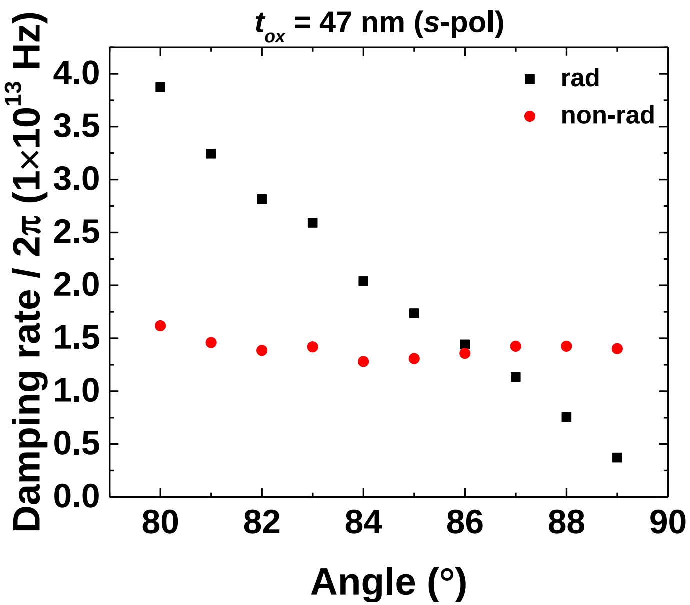


$$A(\omega) = \frac{4\gamma_{rad}\gamma_{nonrad}}{(\omega-\omega_0)^2 + (\gamma_{rad}+\gamma_{nonrad})^2}$$

From temporal coupled mode theory

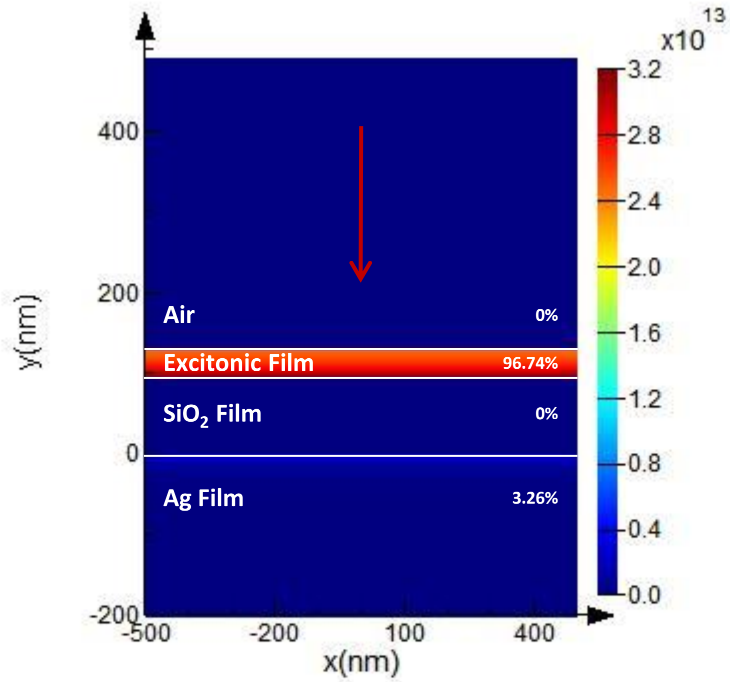
$\omega = \omega_0$

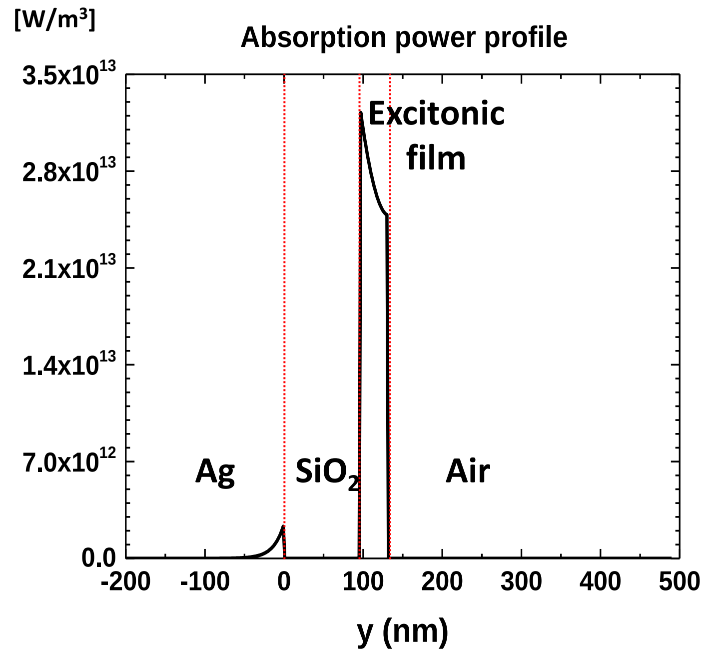
$$A_m = \frac{4\gamma_{rad}\gamma_{nonrad}}{(\gamma_{rad}+\gamma_{nonrad})^2}$$

$\gamma_{rad} = \gamma_{nonrad}$ → $A_m = 1$

"Critical coupling"

Perfect absorption in excitonic films: (ii) SiO_2/Ag





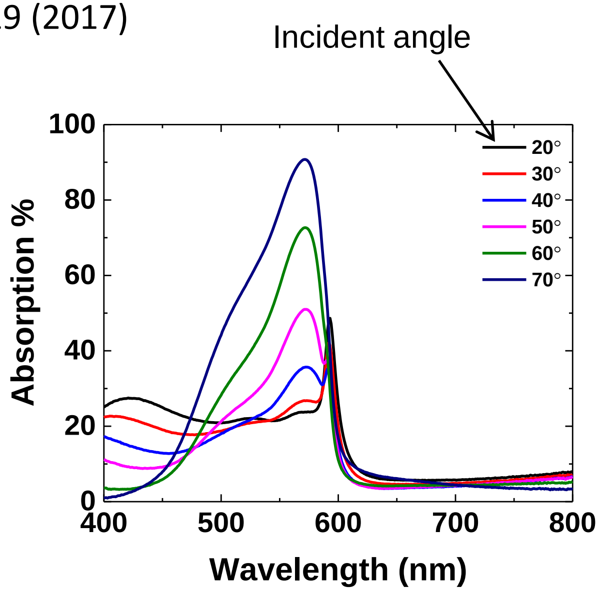
- Incident light is strongly absorbed in the excitonic film
- This absorbed light energy can be potentially extracted out
- However, in more conventional perfect absorbers made of '*metal*' nanostructures, the absorbed light energy is mostly lost due to metal losses in most cases..

Photoluminescence enhancement at strong absorption conditions

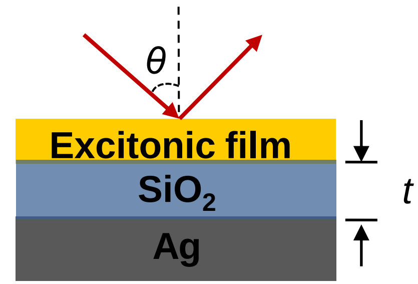
Optics Express 25, 28619 (2017)

t = 150 nm

Absorption spectra
(incident light: p-pol)

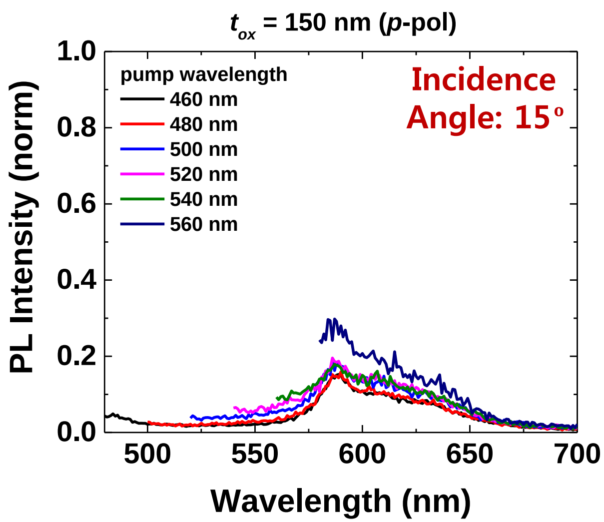


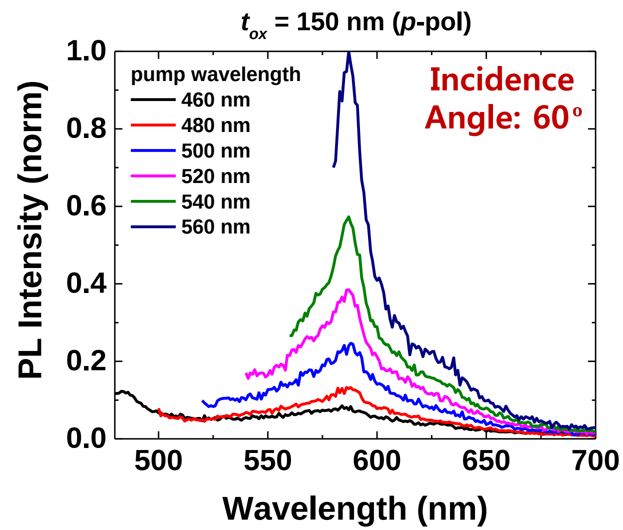
Metallic **excitonic film on Ag substrate with a phase controller**



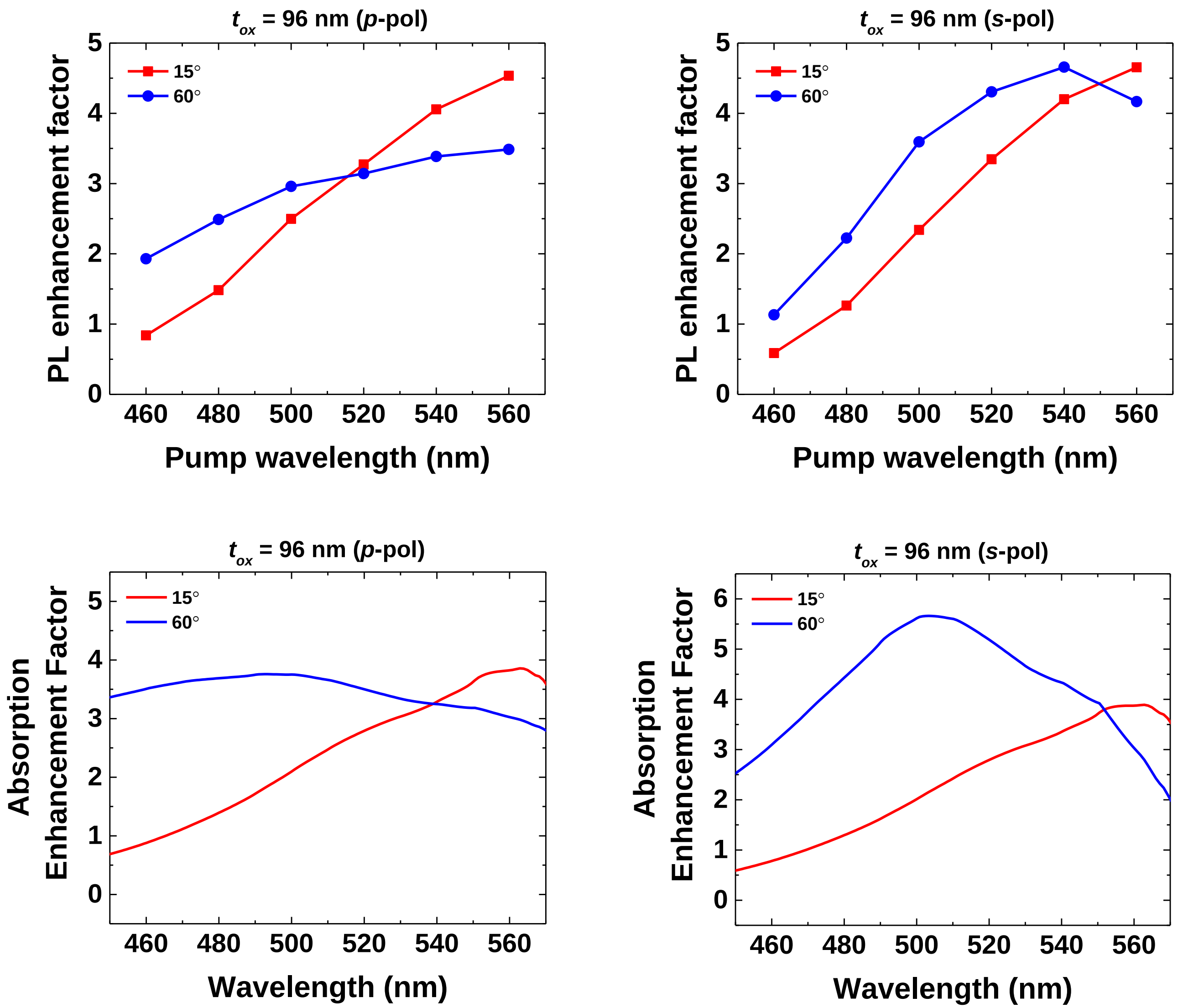
Photoluminescence
(incident light: p-pol)

Vary pump wavelength,
Measure PL spectrum





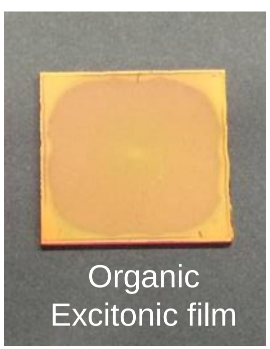
Perfect absorption in excitonic films: (ii) SiO_2/Ag



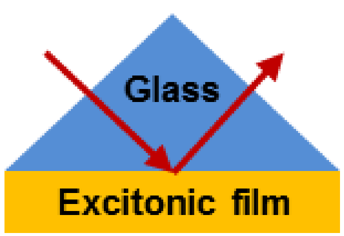
Summary: Excitonic perfect absorbers

- **Demonstrated diverse absorption spectra in planar thin films**
- **Strong absorption occurs even away from the excitonic pole**
- **Studied Perfect absorption and Enhanced photoluminescence**
- **Demonstrated direct nanopatterning of J-aggregate thin films**
- **2D-layered Perovskites: absorption enhancement below E_g**
- **Excitonic films could be a promising platform for many interesting optical studies in the visible region**

Thanks for your attention ..



(i) ATR condition



(ii) Silver substrate with a phase controller