PIER Graduate Week 2017

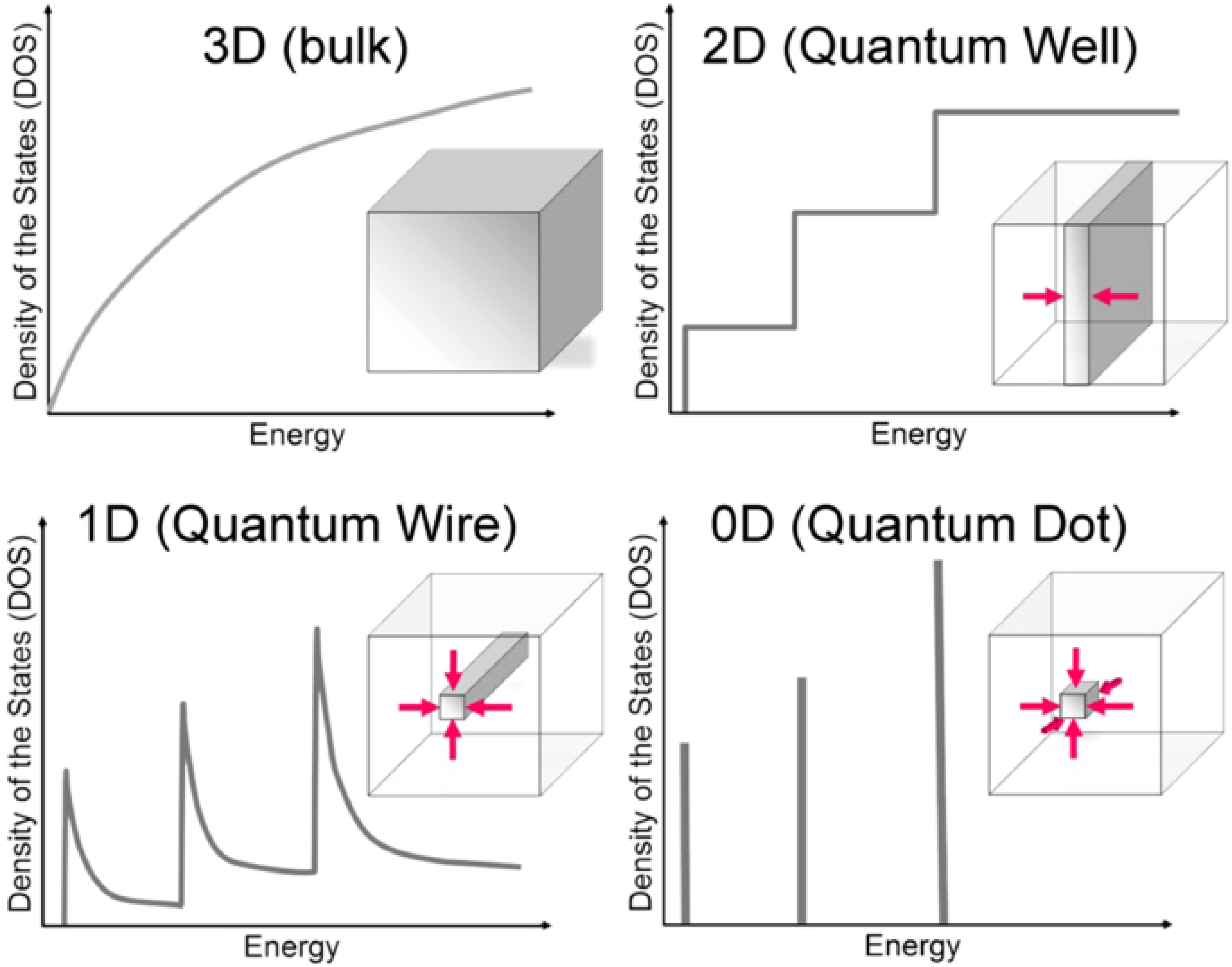
Nano-Optics of low dimensional semiconductor structures

Lecture 2:
Nano-Optics of 2D Semiconductors

Martin Kroner

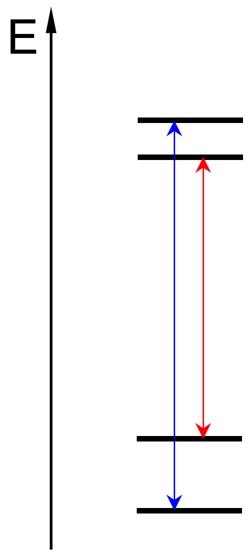
Quantum Photonics Group, ETH Zürich, Switzerland

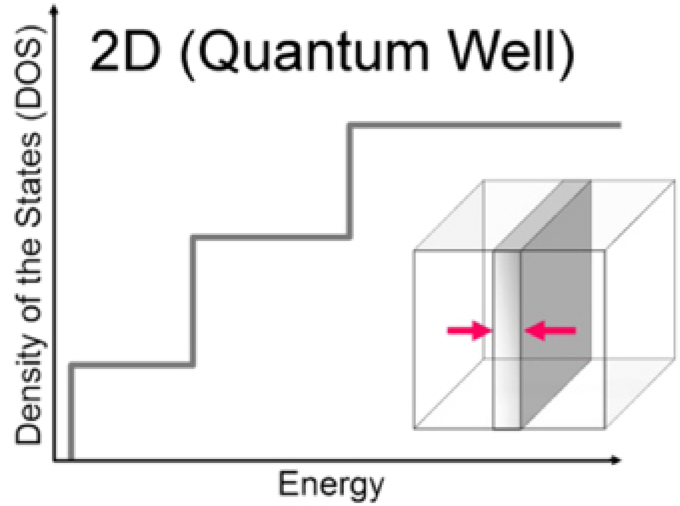
Dimensionality of optical systems

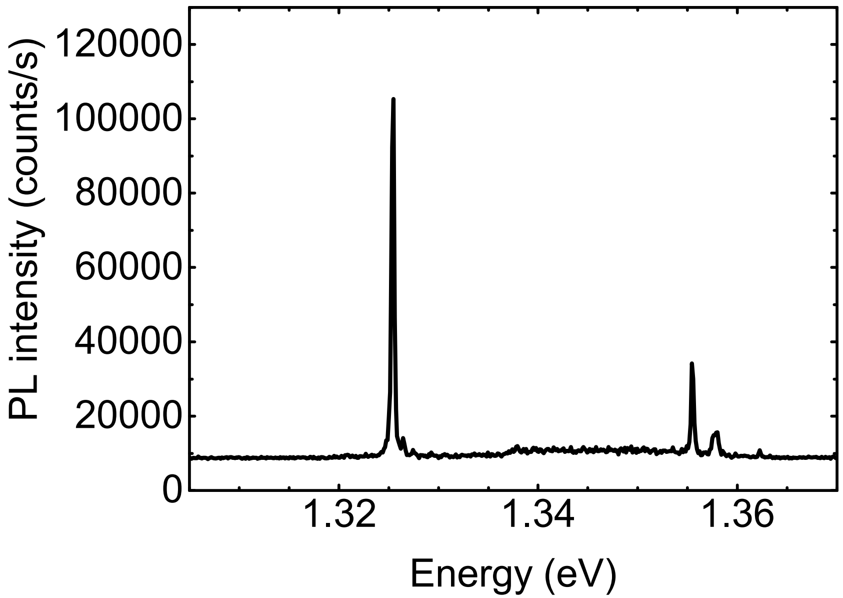


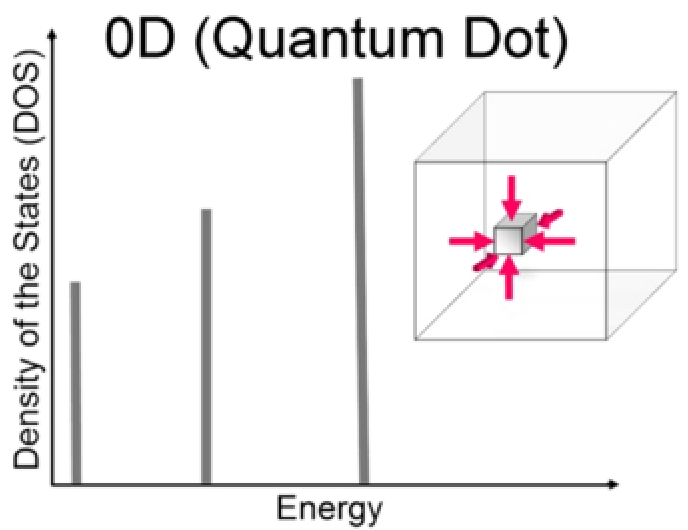
L. Mino et al, J. Phys. D: Appl. Phys. 46, 423001 (2013)

Optical Properties of a 0D System

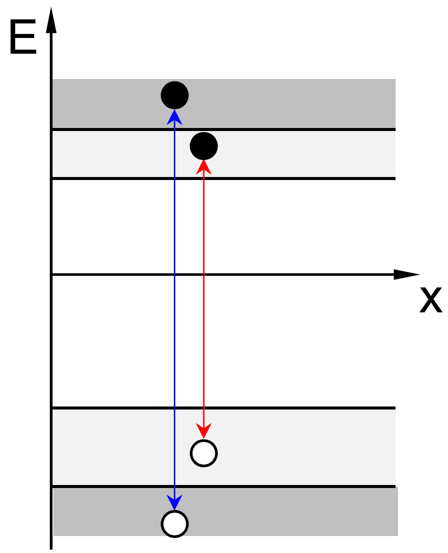


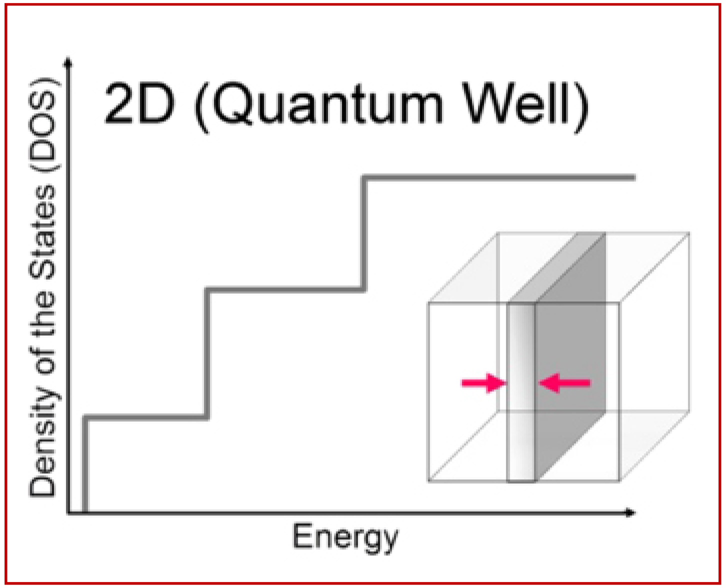


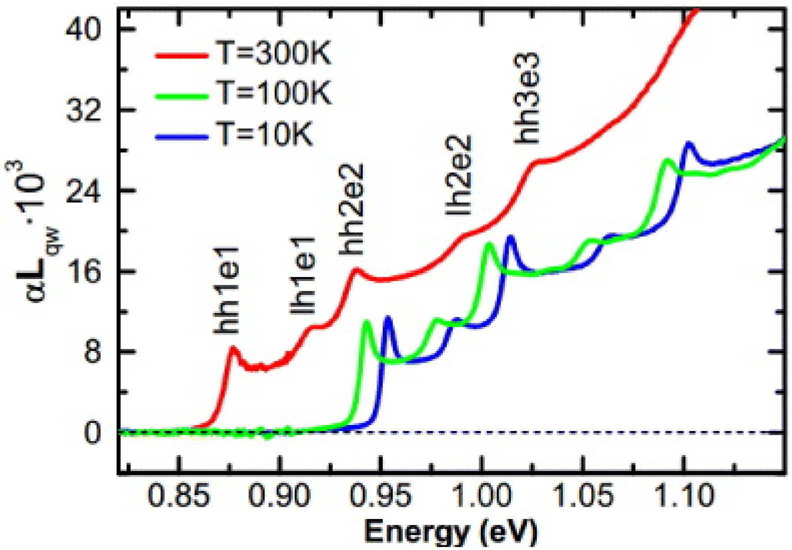


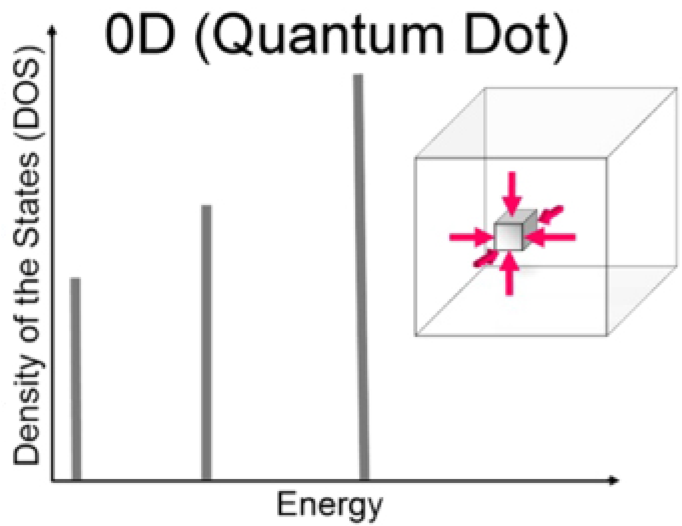


Optical Properties of a 2D System









N.S. Köster, et al, NJoP 15, 075004 (2013)

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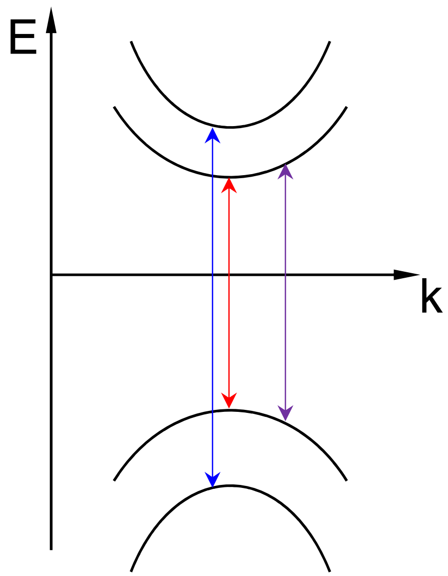

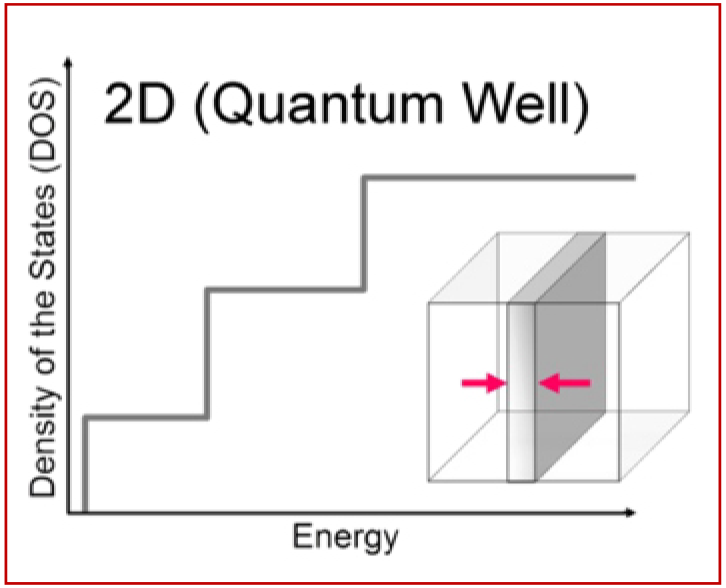

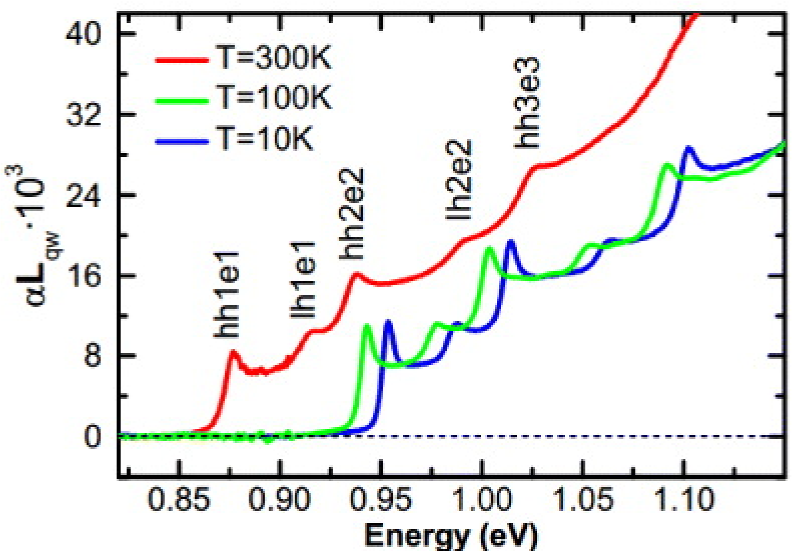

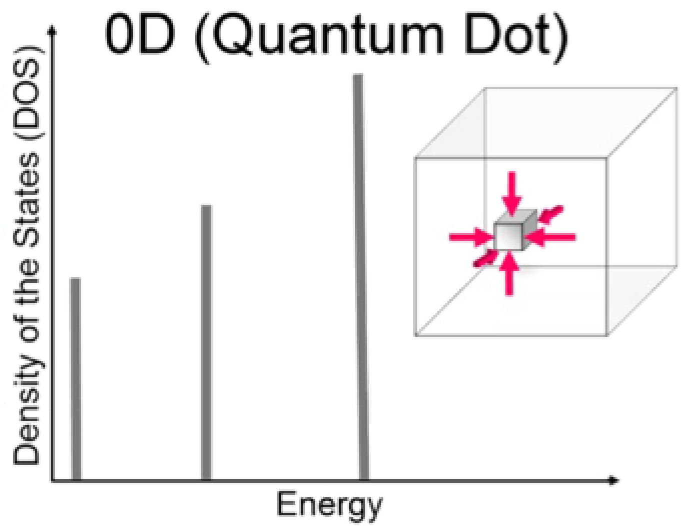

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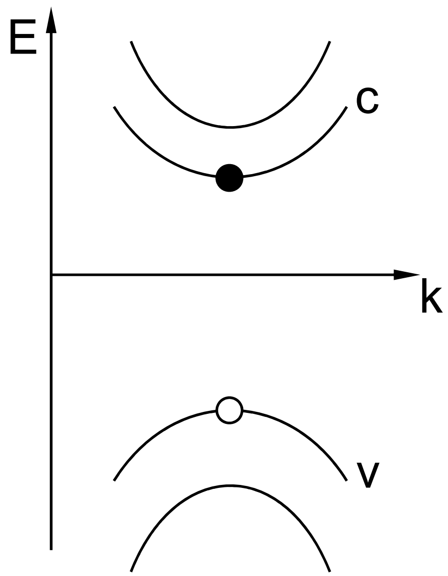
Excitons in 2D

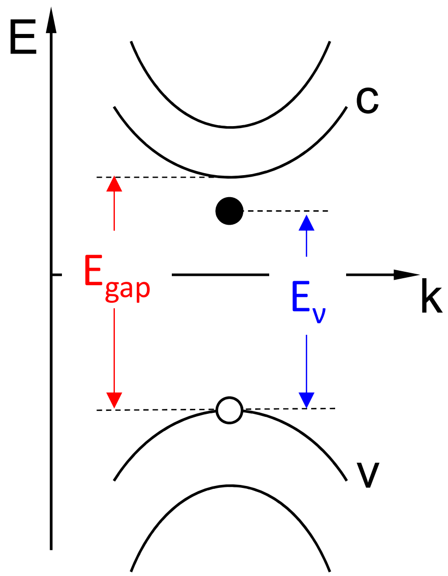


Electron and hole form bound state: Exciton

$$H_{\mathrm{int}} \sim \int d^3r \int d^3r \hat{\Psi}^\dagger_s(r)\hat{\Psi}^\dagger_{s'}(r')\frac{e^2}{|r-r'|}\hat{\Psi}_s(r)\hat{\Psi}_{s'}(r')$$

$$\hat{\Psi}_s(r) = \sum_k \left(\psi_{c,k,s}(r)\hat{c}_{c,k,s} + \psi_{v,k,s}(r)\hat{c}_{v,k,s}\right)$$

Excitons in 2D



i.e. in GaAs

$E_{1s} - E_{gap} \approx 20meV$

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...

$\Longrightarrow$ Hydrogen like equation:

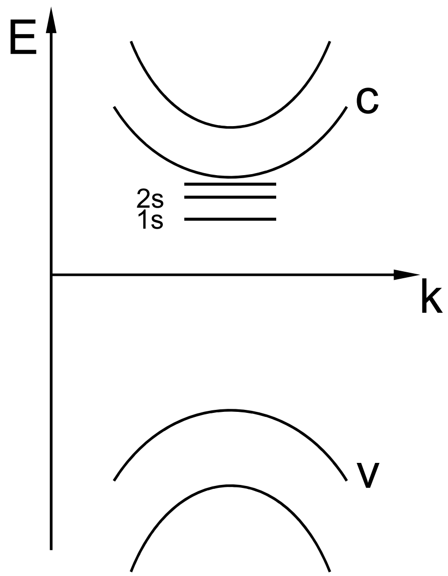
$$(E_\nu - E_{gap})\tilde{\psi}_\nu(r) = \left(\frac{-\hbar^2\nabla}{2m_r} - \frac{e^2}{\epsilon r}\right)\tilde{\psi}_\nu(r)$$

For different states: $\nu = 1s, 2s, 2p...$

i.e.: $\tilde{\psi}_{1s}(r) = \sqrt{\frac{8}{\pi}}\frac{1}{a_B}e^{\frac{-2r}{a_B}}$

With the Bohr-radius $a_B = \frac{4\pi\epsilon_0\epsilon\hbar^2}{e^2 m_r}$

Excitons in 2D



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i.e. in GaAs

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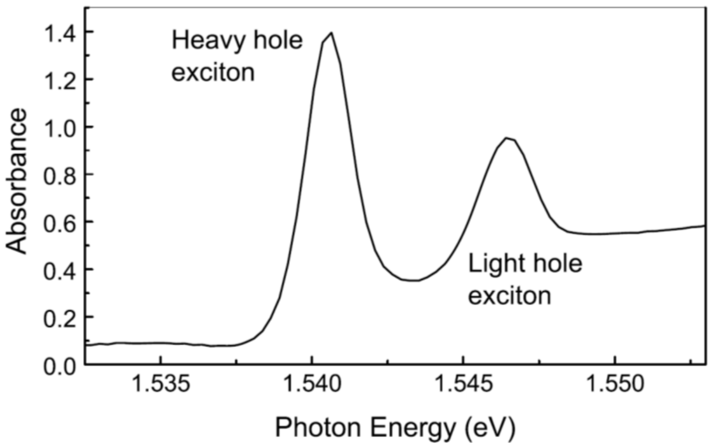


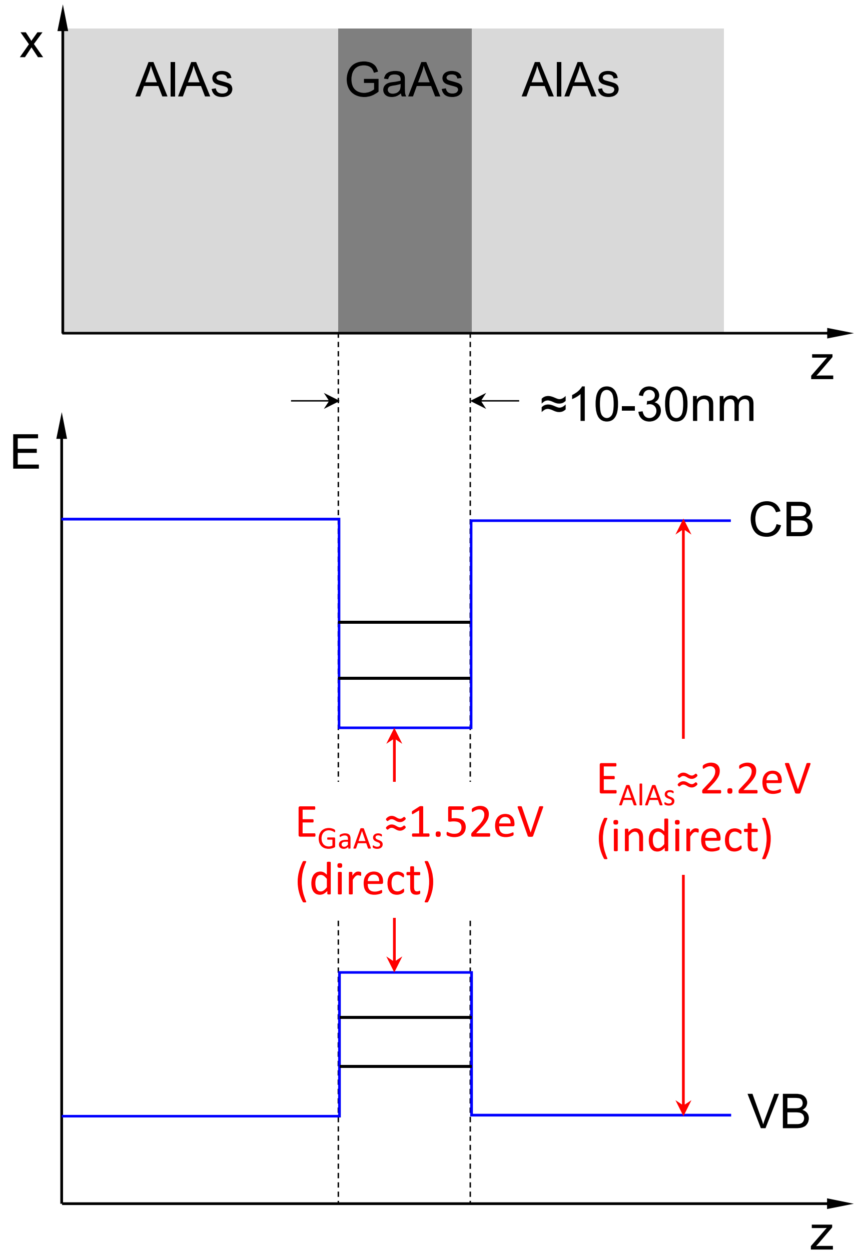
S.T. Cundiff, Optics Express 16, 4639 (2008)

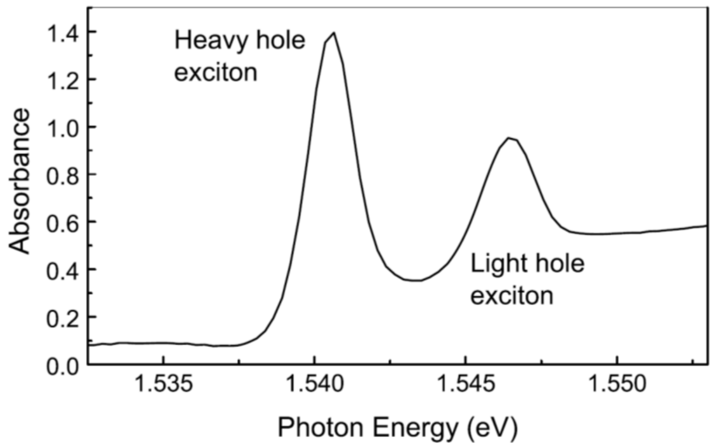
2D Semiconductors

Epitaxially grown heterostructures.

Material: III-V or II-VI

i.e. AlAs/GaAs quantum well





S.T. Cundiff, Optics Express 16, 4639 (2008)

2D Semiconductors - Disorder

Epitaxially grown heterostructures.

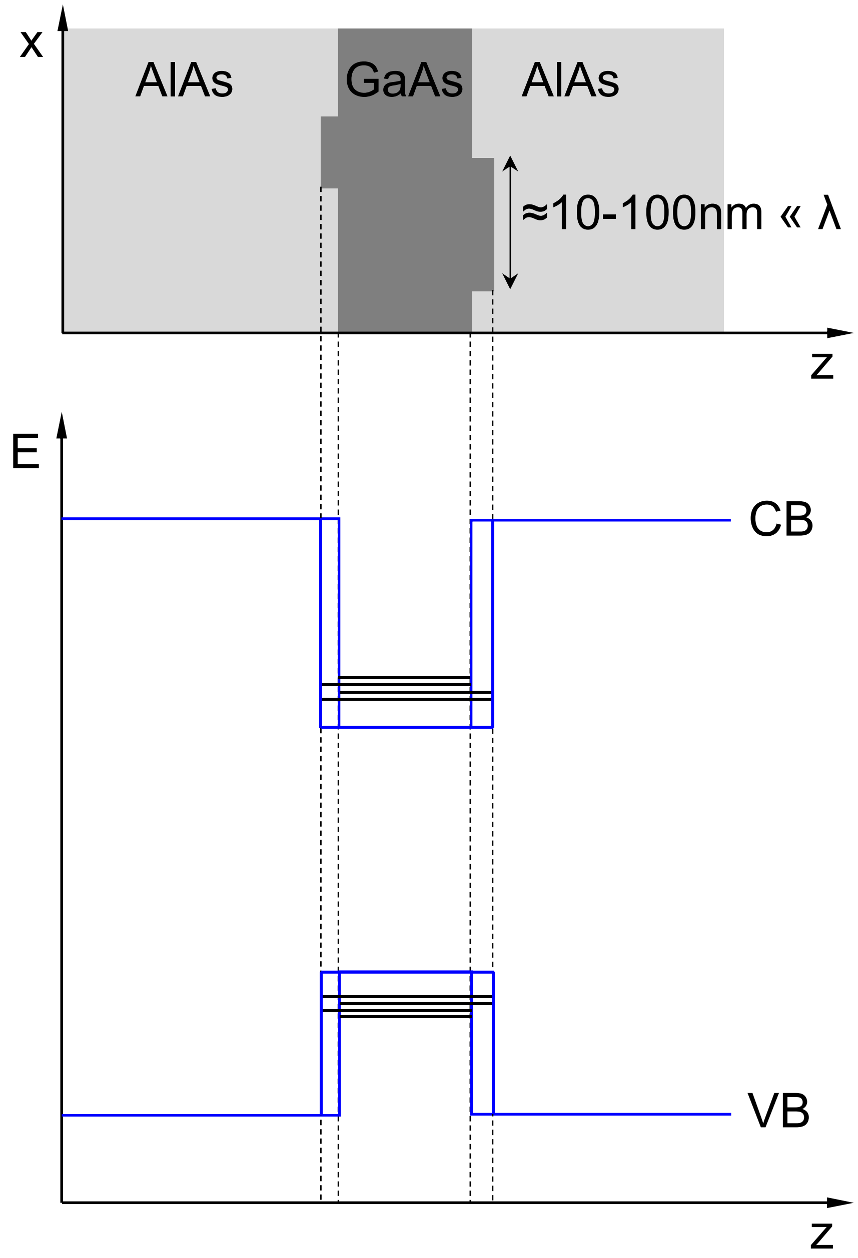
Few atom layer thickness variation

⟹ Several different exciton transition energies in a diffraction limited spot.

⟹ Inhomogeneous broadening of optical resonance.

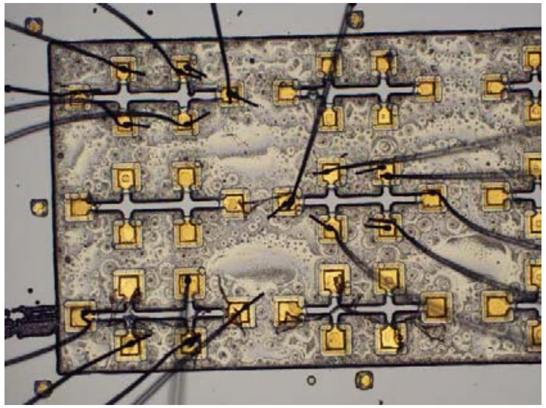
Homogeneous (lifetime limited) line width: 30μeV

Typical measured line width: ~300 μeV

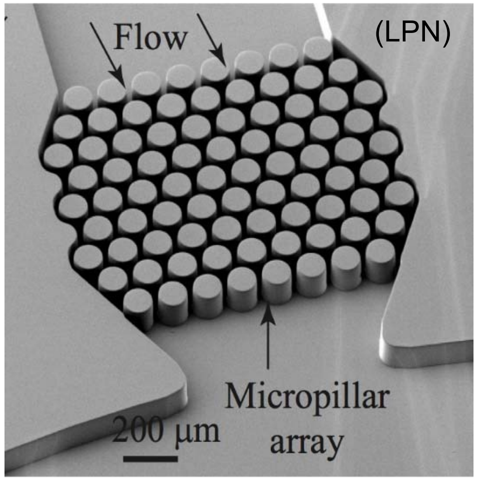


2D Semiconductor Technology

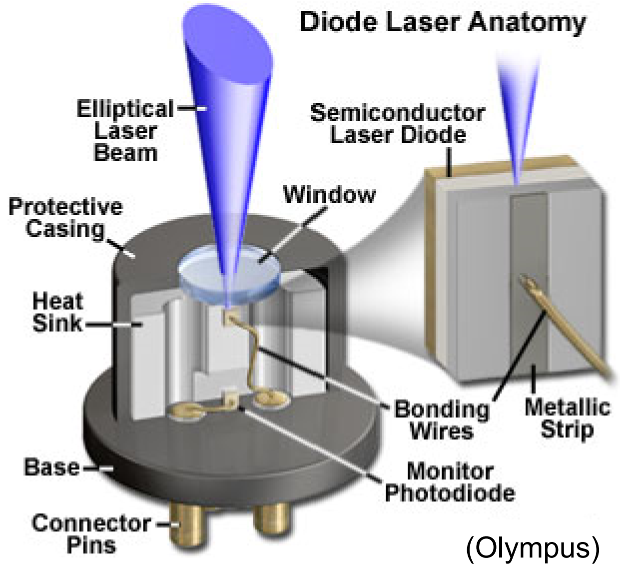
Electro optical devices



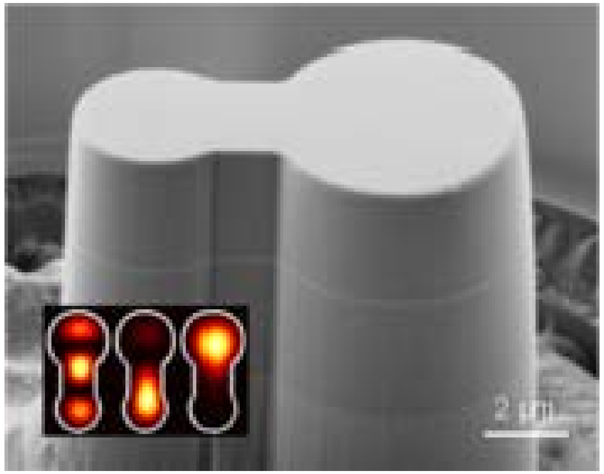
Hybrid devices



Laser diodes



Photonic devices



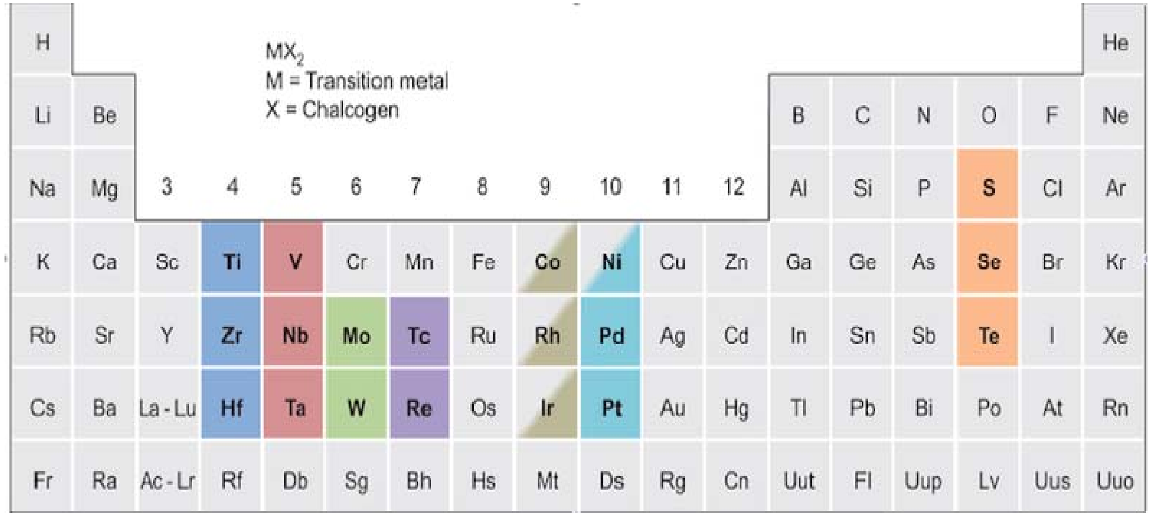
High quality material (optical and electrical properties)

Sophisticated growth and processing techniques

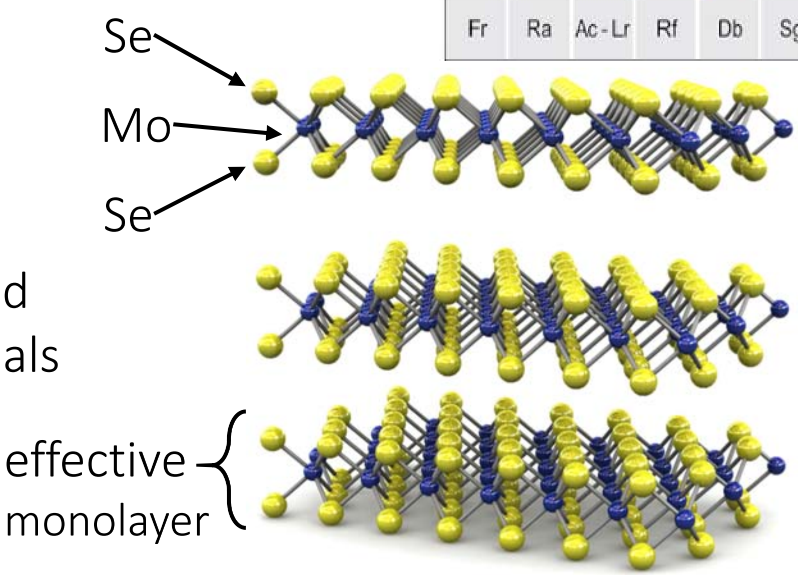
A New Type of 2D Semiconductors

Transition metal dichalcogenides (TMD):
A new class of truly two dimensional semiconductors

Formula: MX_2
M = Transition metal
X = Chalcogen



Layered materials



Electrical property	**Material**
Semiconducting	MoS_2 $MoSe_2$ WS_2 WSe_2 $MoTe_2$ WTe_2
Semimetallic	TiS_2 $TiSe_2$
Metallic, Superconducting	$NbSe_2$ NbS_2 $NbTe_2$ TaS_2 $TaSe_2$ $TaTe_2$

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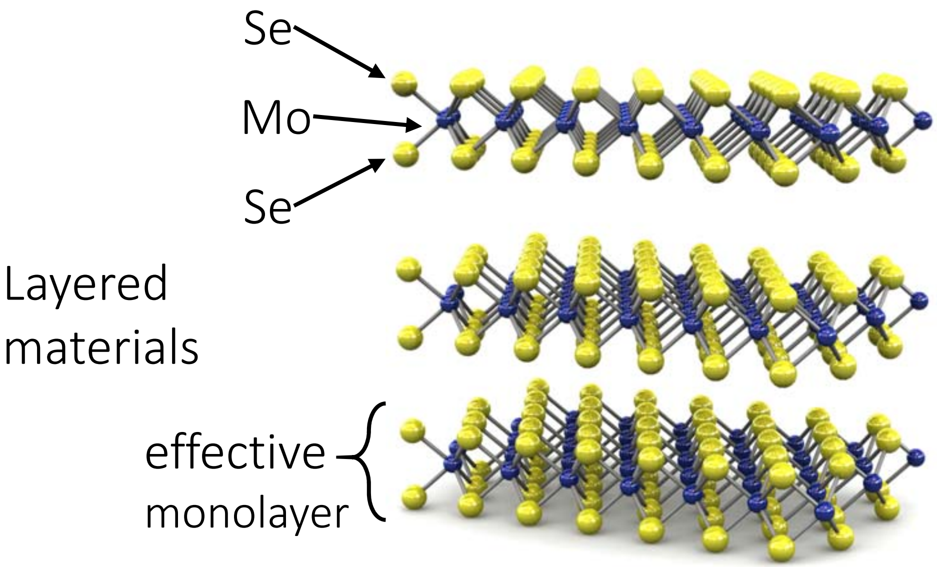

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$MoSe_2$

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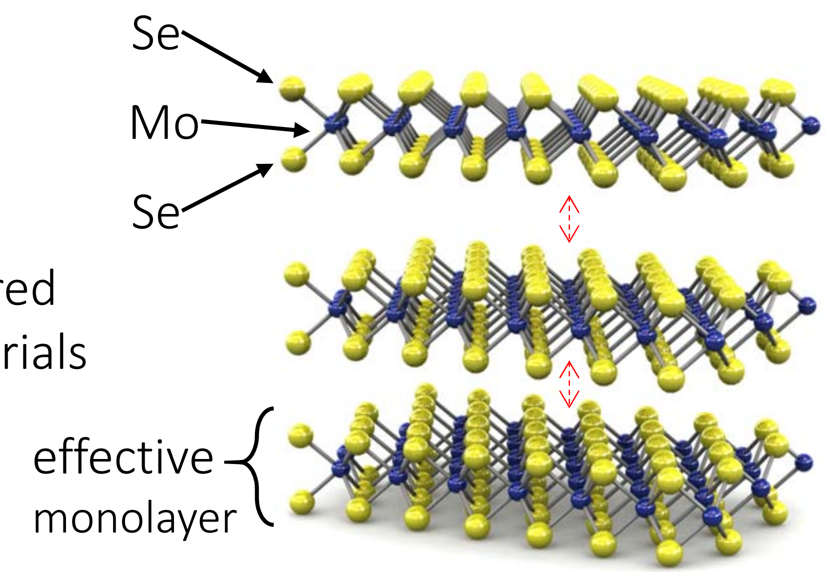
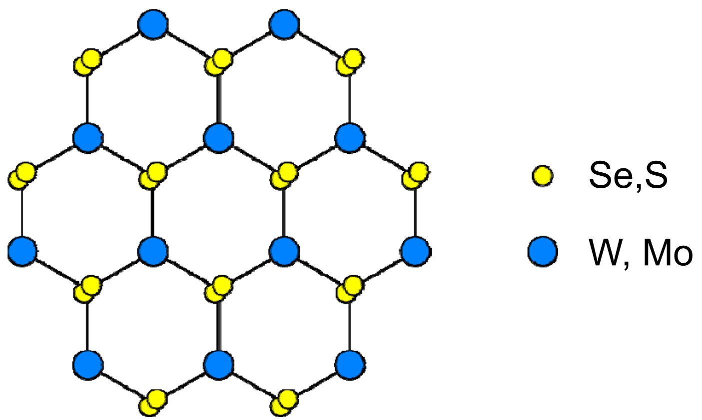


Van der Waals bonds between layers
⟹ Single layers obtained by exfoliation

TMD Crystal Structure

- Monolayer has a honeycomb lattice



TMD Crystal Structure

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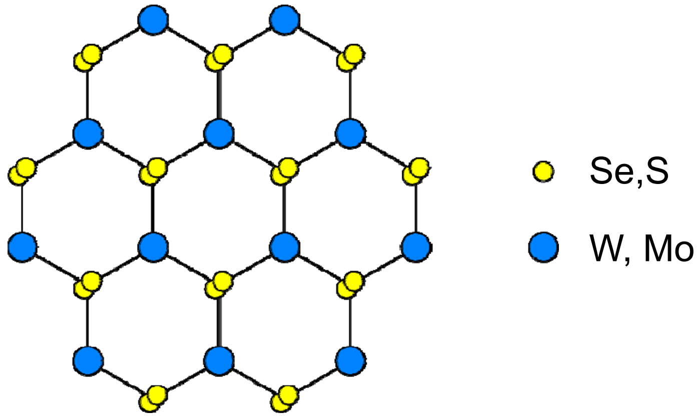
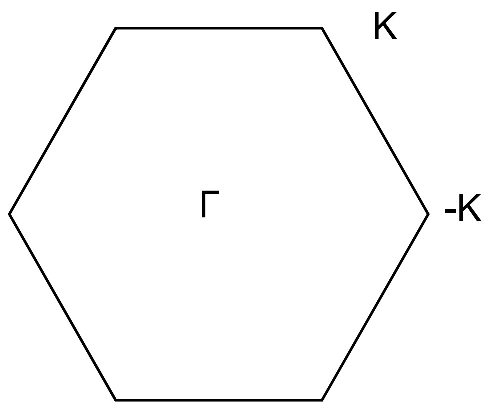
- Valley semiconductor: physics at ±K





TMD Crystal Structure

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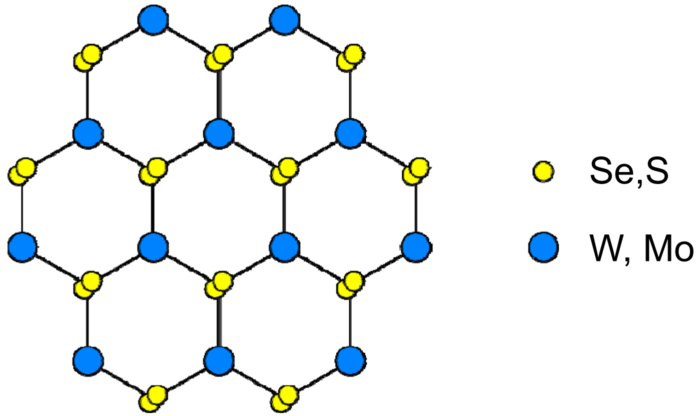
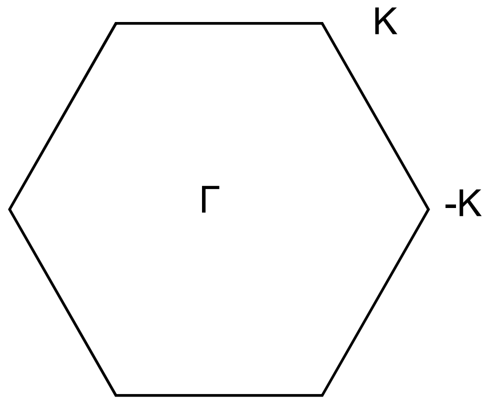
- Valley semiconductor: physics at ±K

- Broken inversion symmetry → band gap

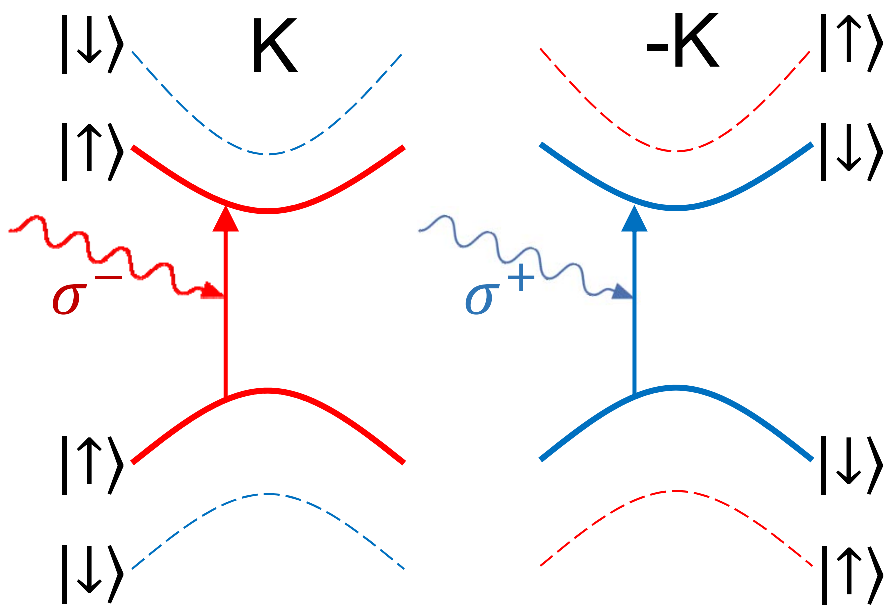




Optical Addressing of Valleys in $MoSe_2$

±K valleys respond to ±σ polarized light
⟹ valley addressability like spin

protection of spin coherence due to spin-valley locking?



Mak et. al., Nat. Nanotech. **7**, 494 (2012).

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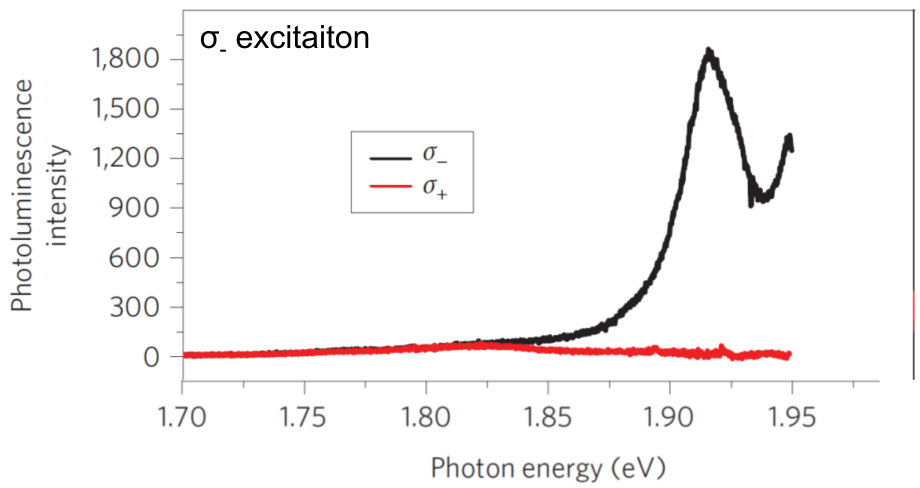

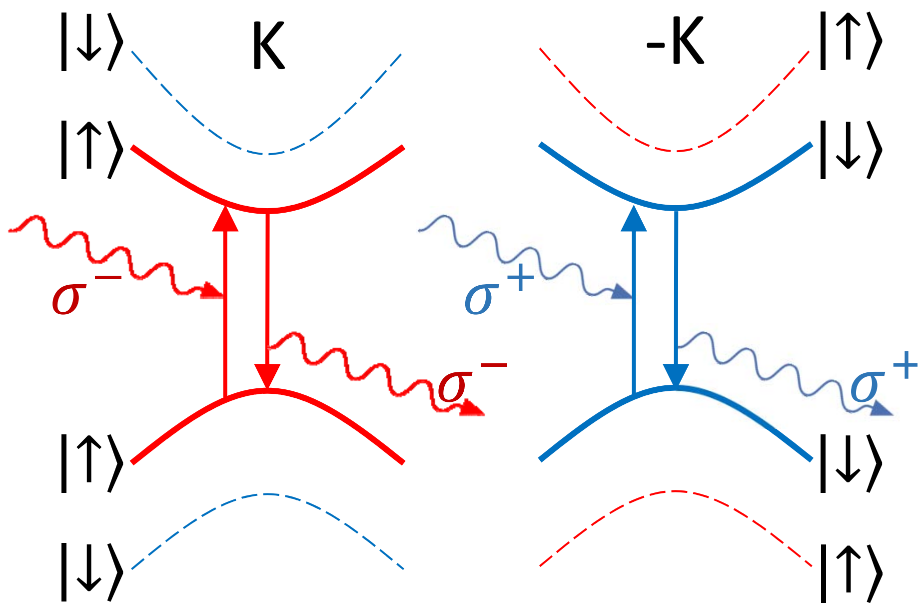

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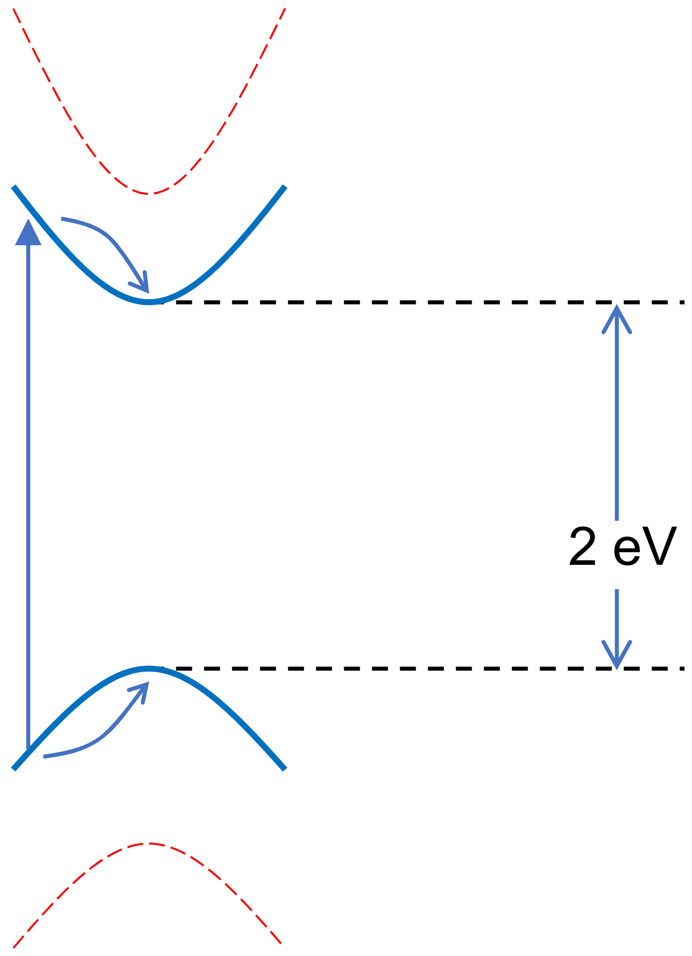
Photoluminescence of TMD Monolayers

- Excitation of high-energy free electron-hole pairs

- Strong Coulomb interaction due to lack of screening (truly 2D)



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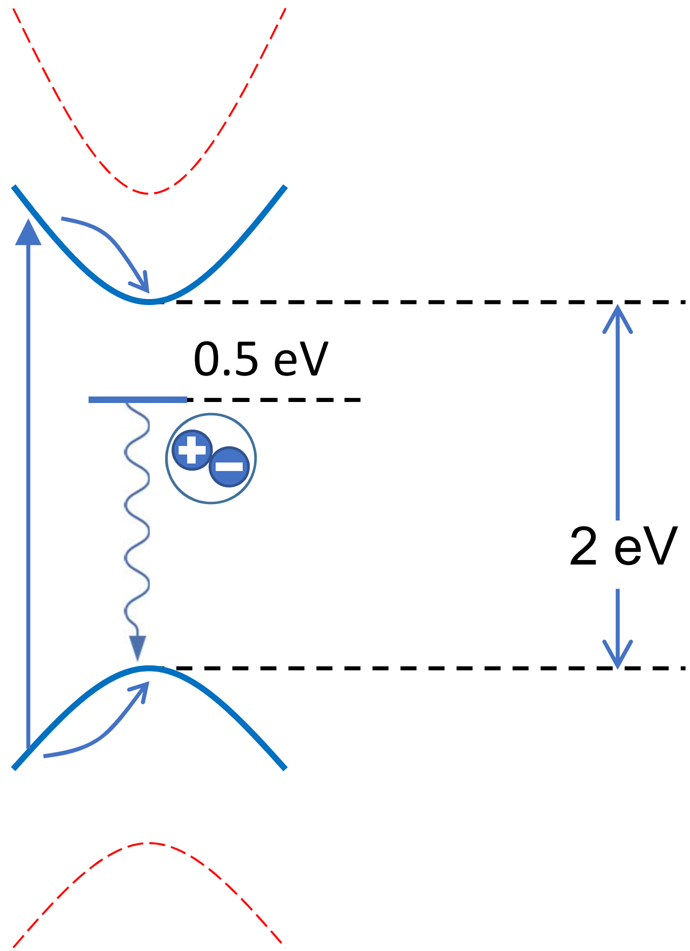

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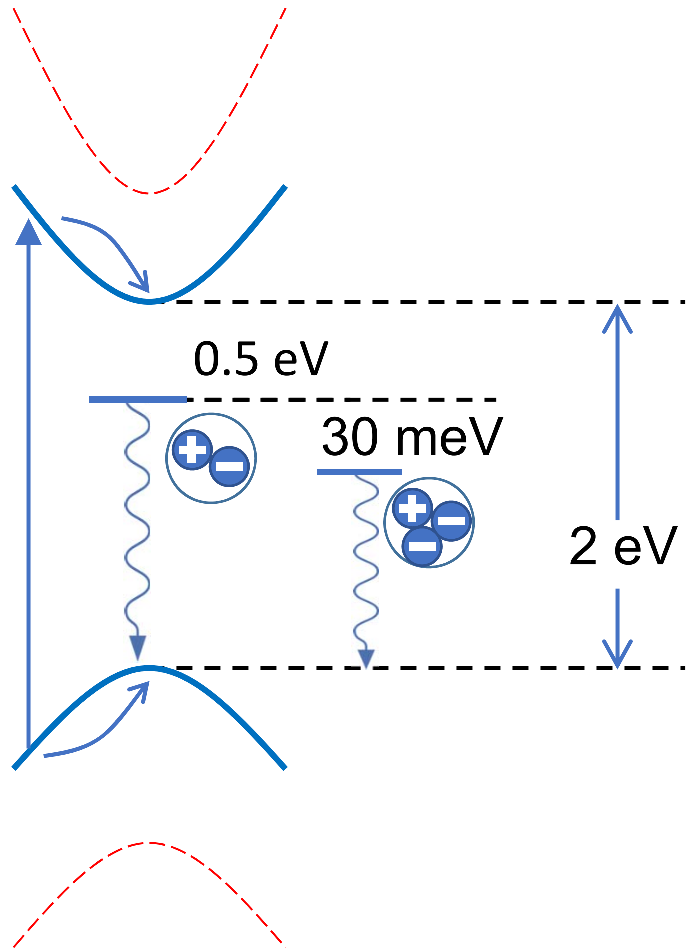
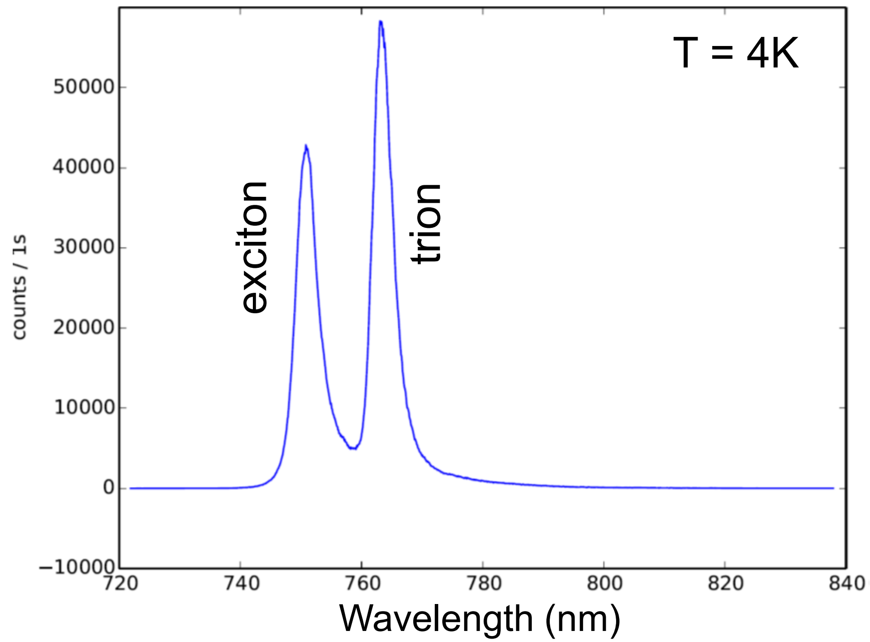
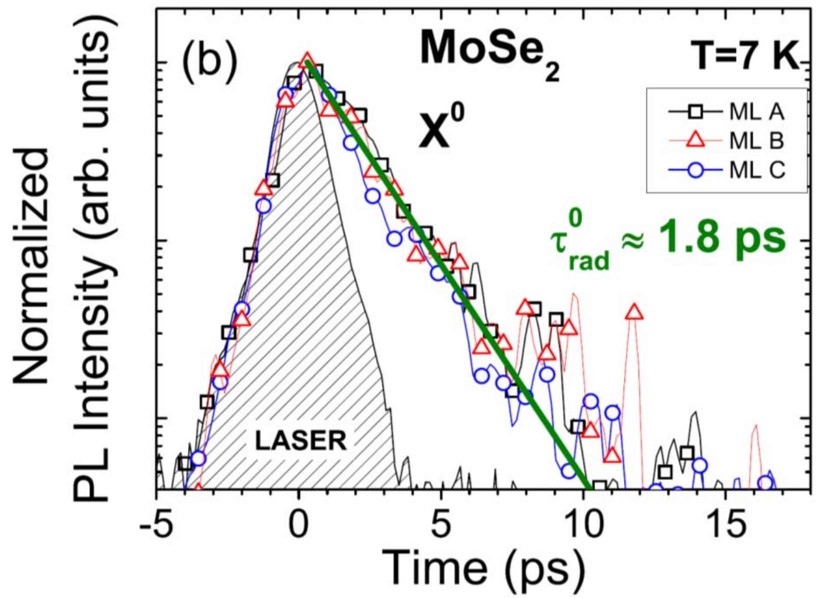
- Excitons form with huge binding energy of 500 meV
- PL is dominated by decay from exciton and trion



Photoluminescence of TMD Monolayers

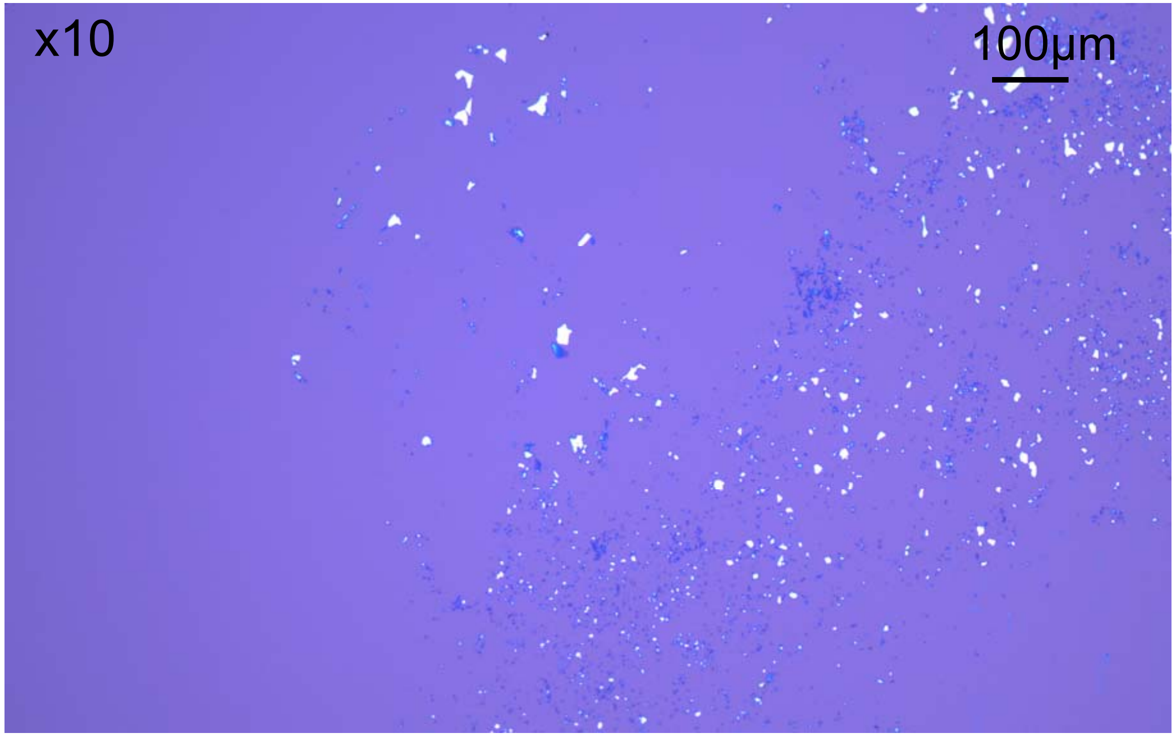
- PL dominated by exciton and trion peaks
- Most flakes are electron doped hence the trion peak
- Smallest linewidth: ΔE= 3 meV
- Radiative lifetime: $\Gamma_{rad} \geq$ 1 meV
- Small exciton Bohr radius → strong light-matter coupling



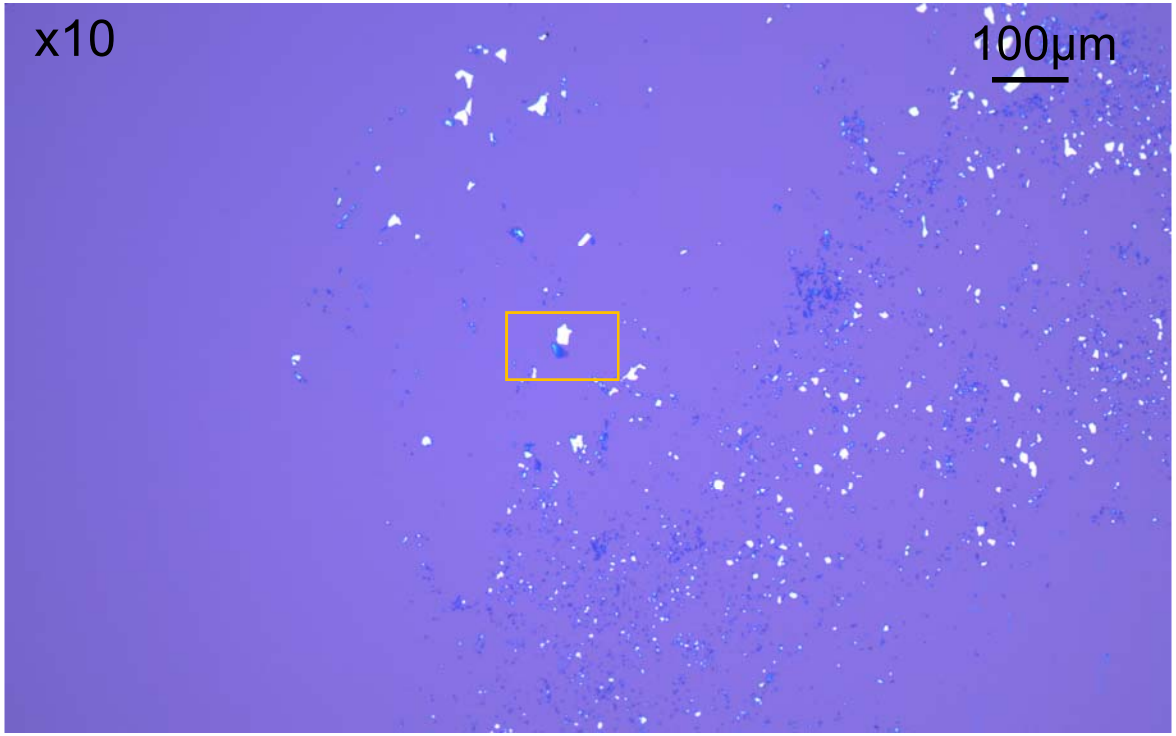


C. Robert et. al., PRB **93**, 205423 (2016).

Exfoliation (and finding) of a monolayer: Optical microscopy image



Exfoliation (and finding) of a monolayer: Optical microscopy image



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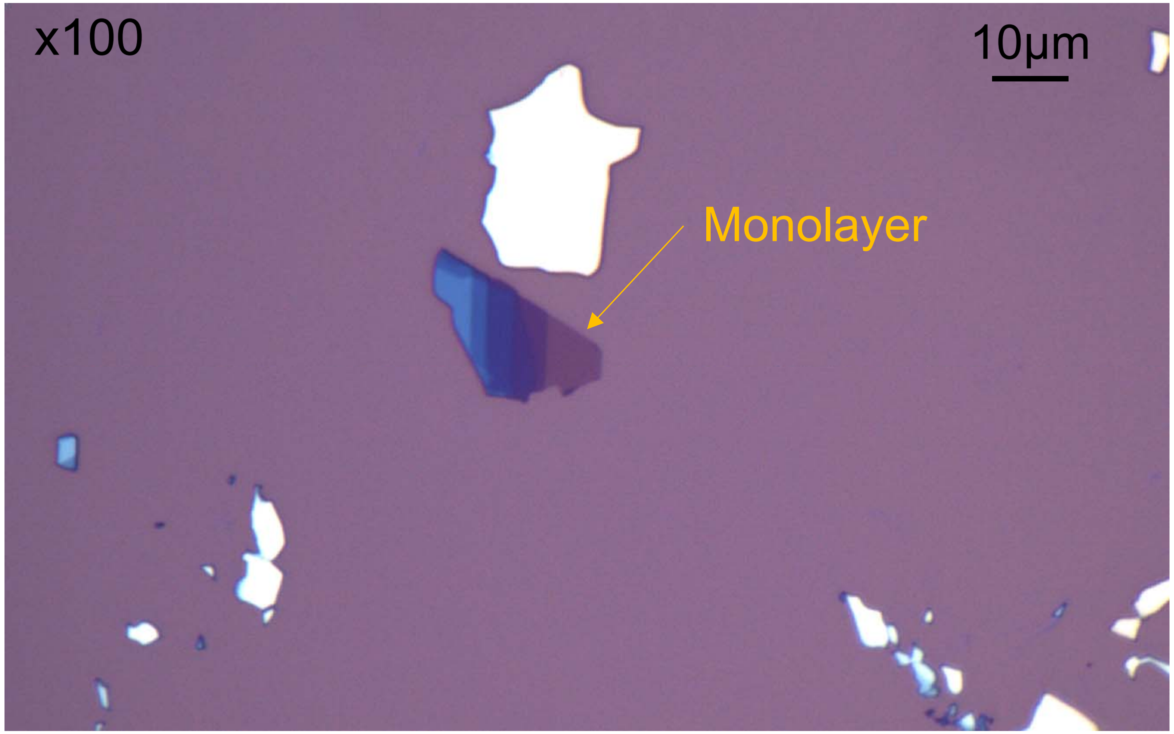
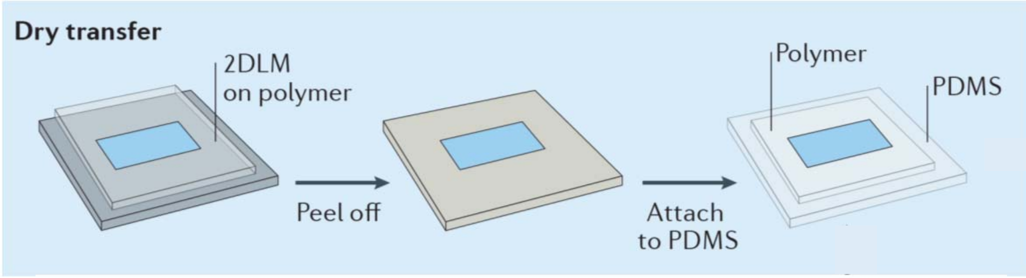
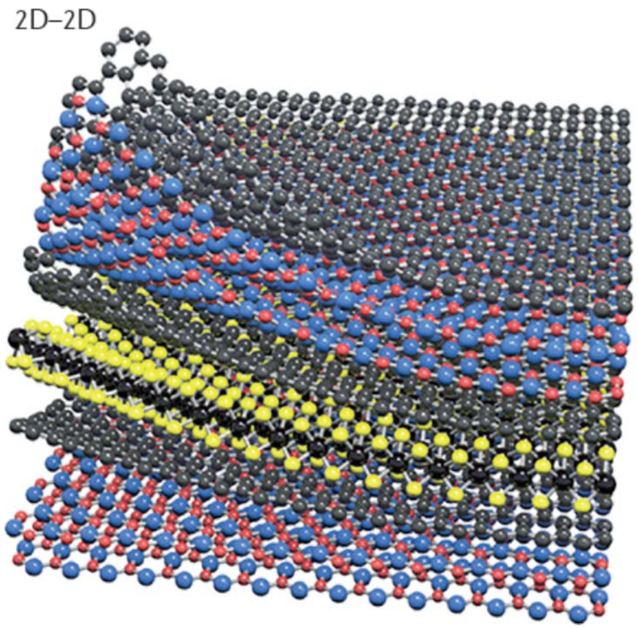


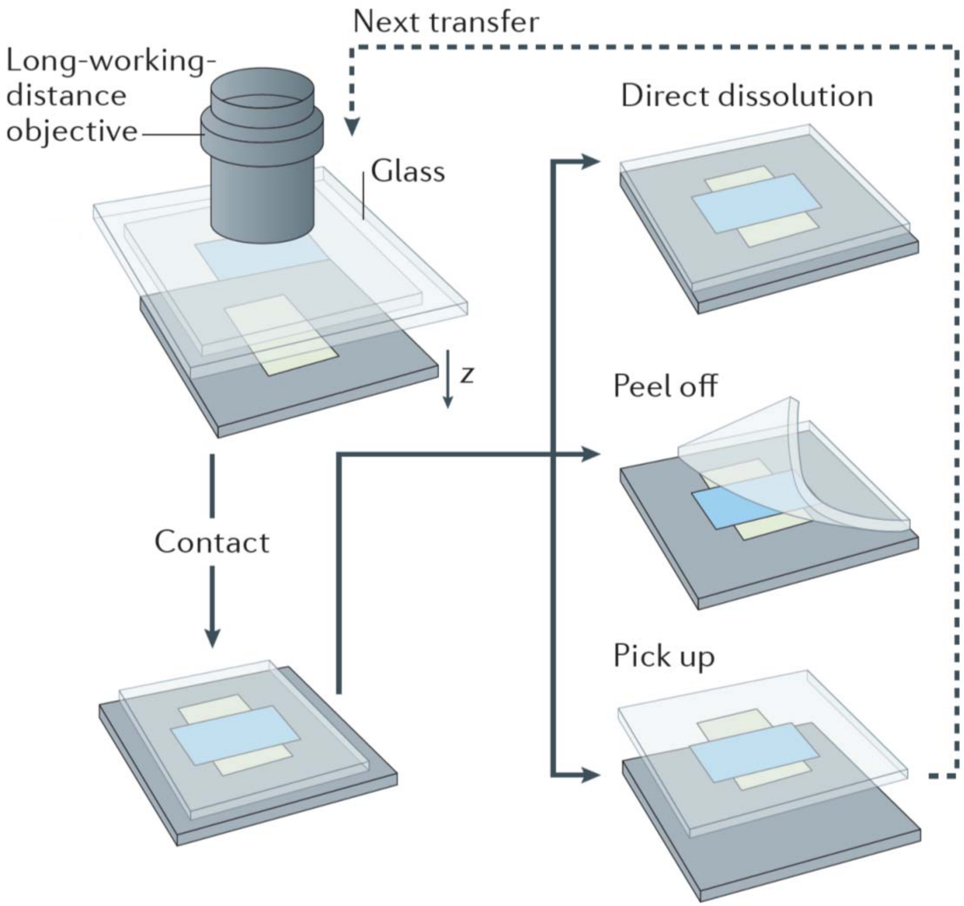
Van-der Waals Heterostructure

Exfoliation and dry pick-up and transfer

⟹ Build heterostructures







Y. Liu et. al., Nat. Rev. Mat **1**, 1 (2016).

Van der Waals Heterostructure

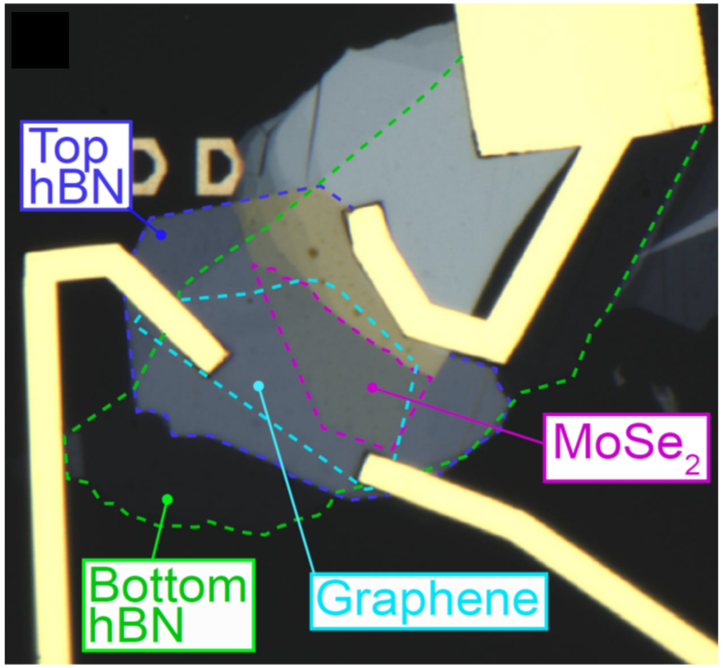
- $MoSe_2$ monolayer

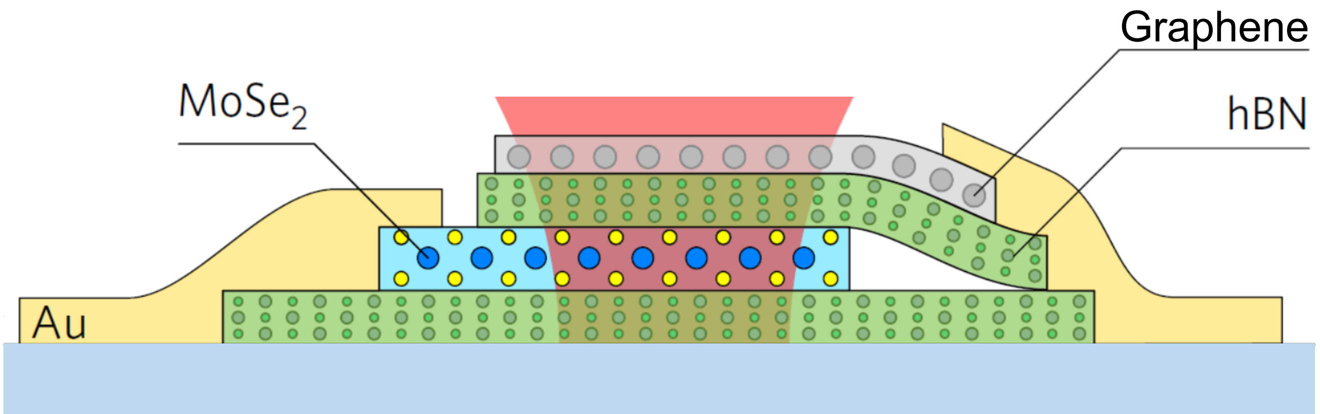
- Encapsulated in hBN

⟹ Match crystal structure
Protect against contamination

- Graphene gate electrode

⟹ Control electron density

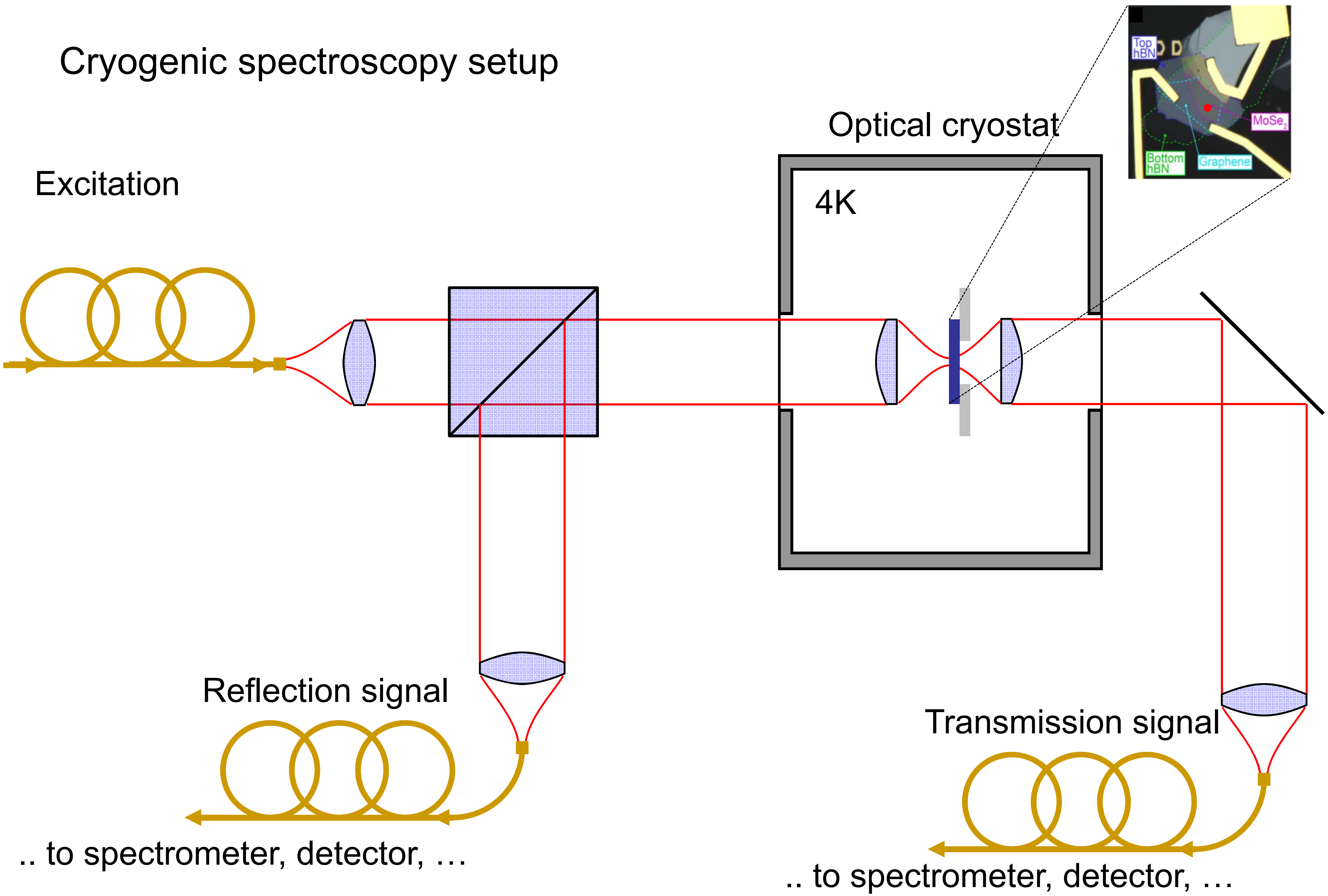




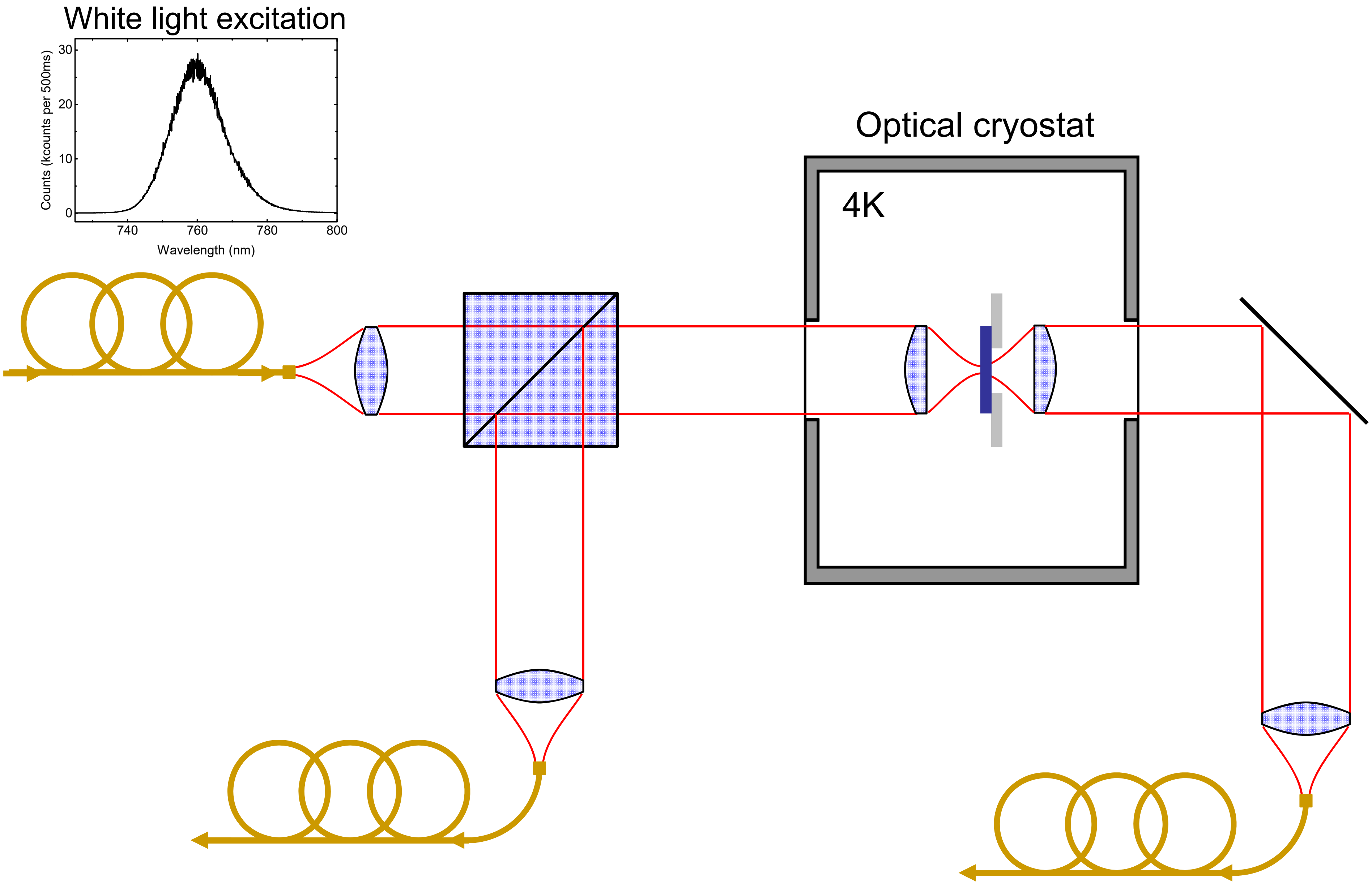
M. Sidler, et al., Nat. Phys. **13**, 255 (2017), P. Back, et al., arXiv 1705.07317

Reflection and Transmission Spectroscopy

Cryogenic spectroscopy setup



Optical Response of a $MoSe_2$ Monolayer



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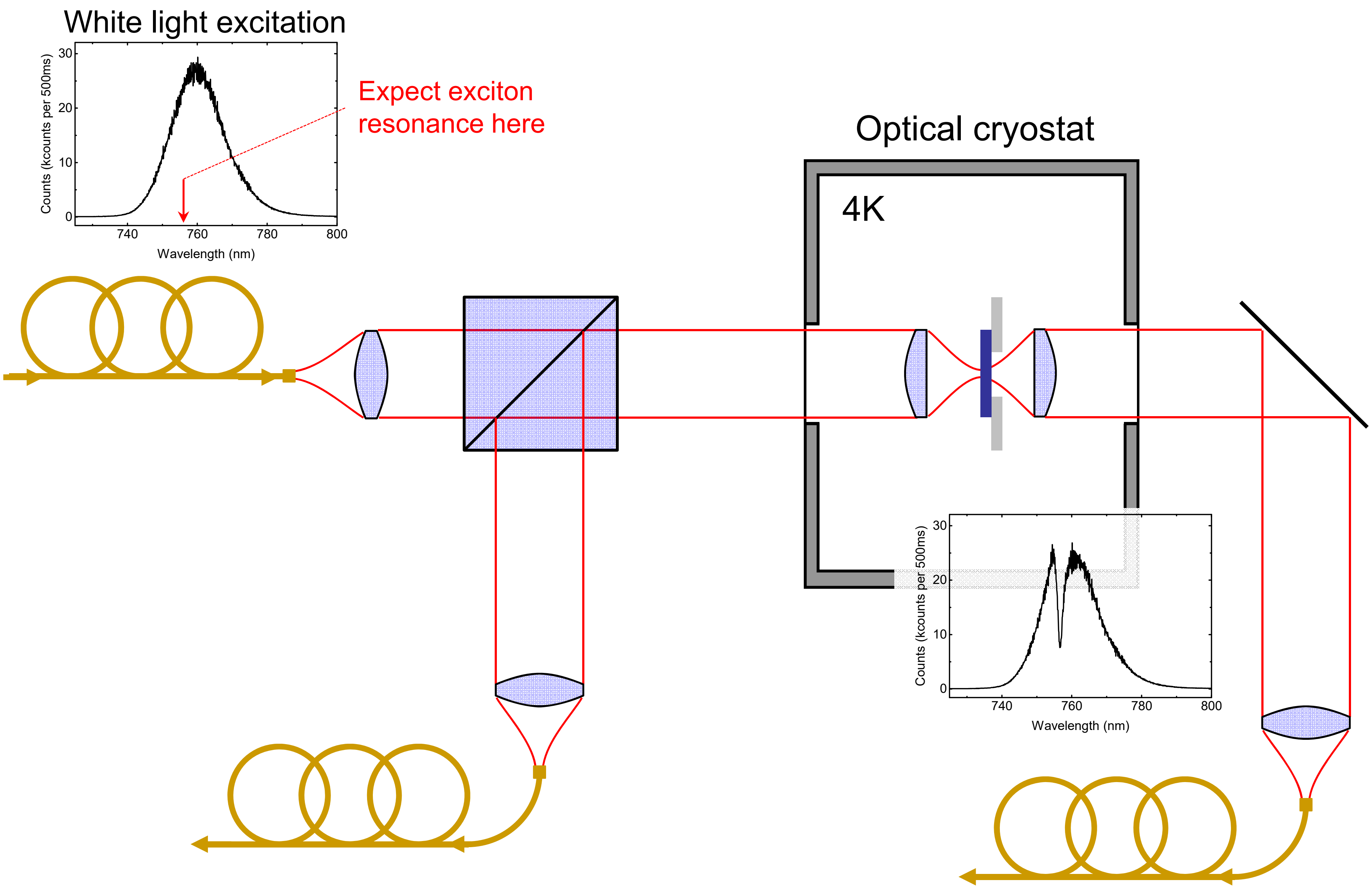

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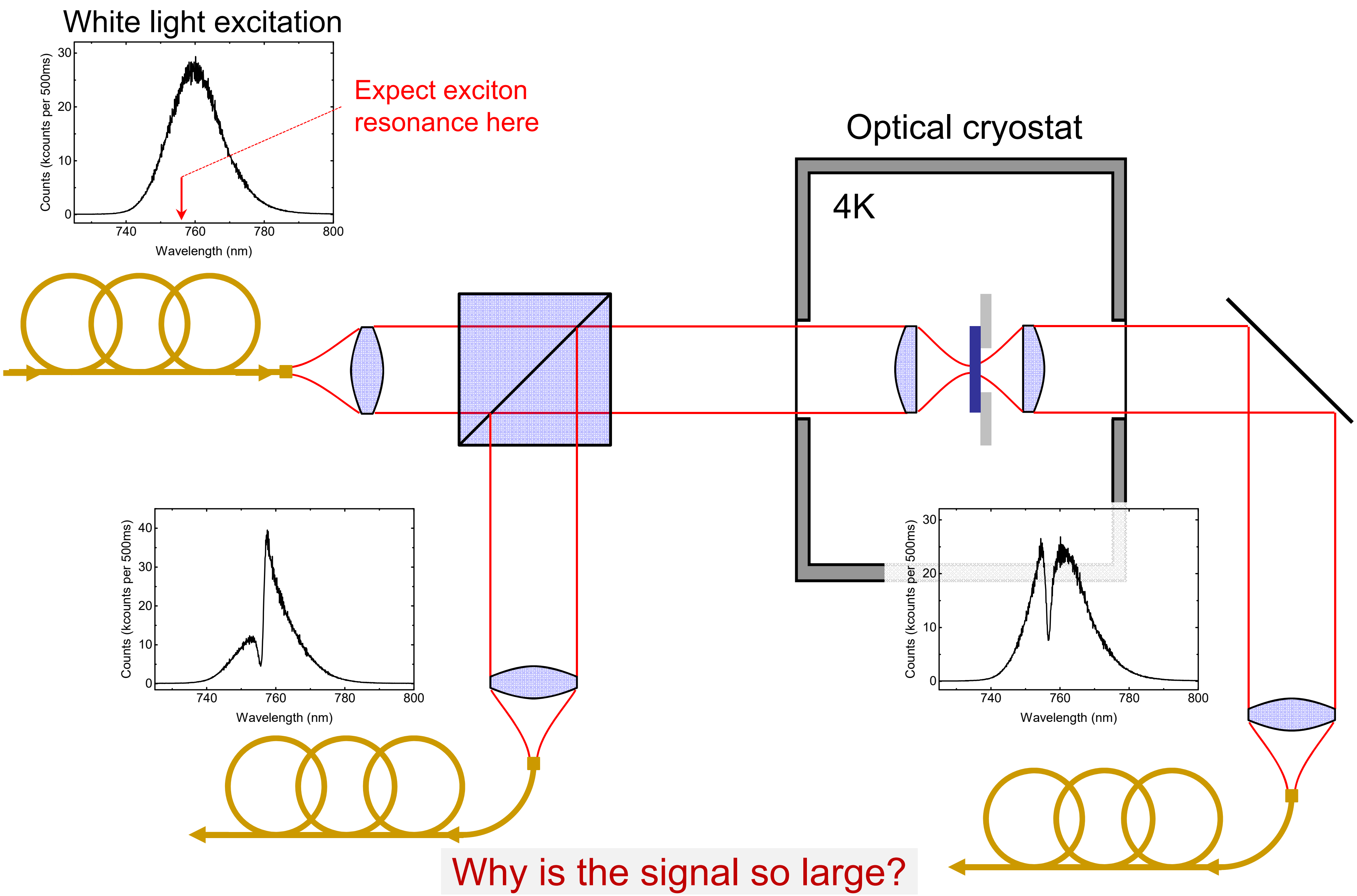
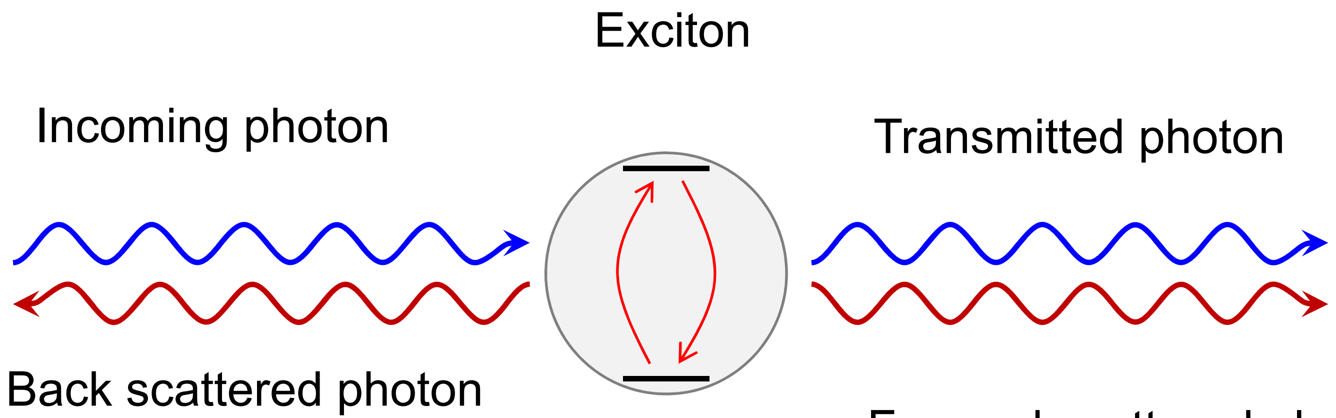
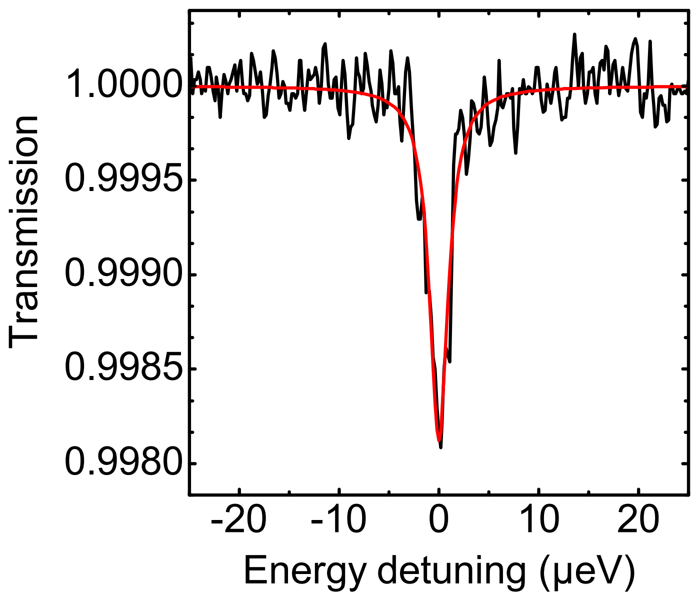


Optical Response of an Exciton



Forward scattered photon
(π phase shift)

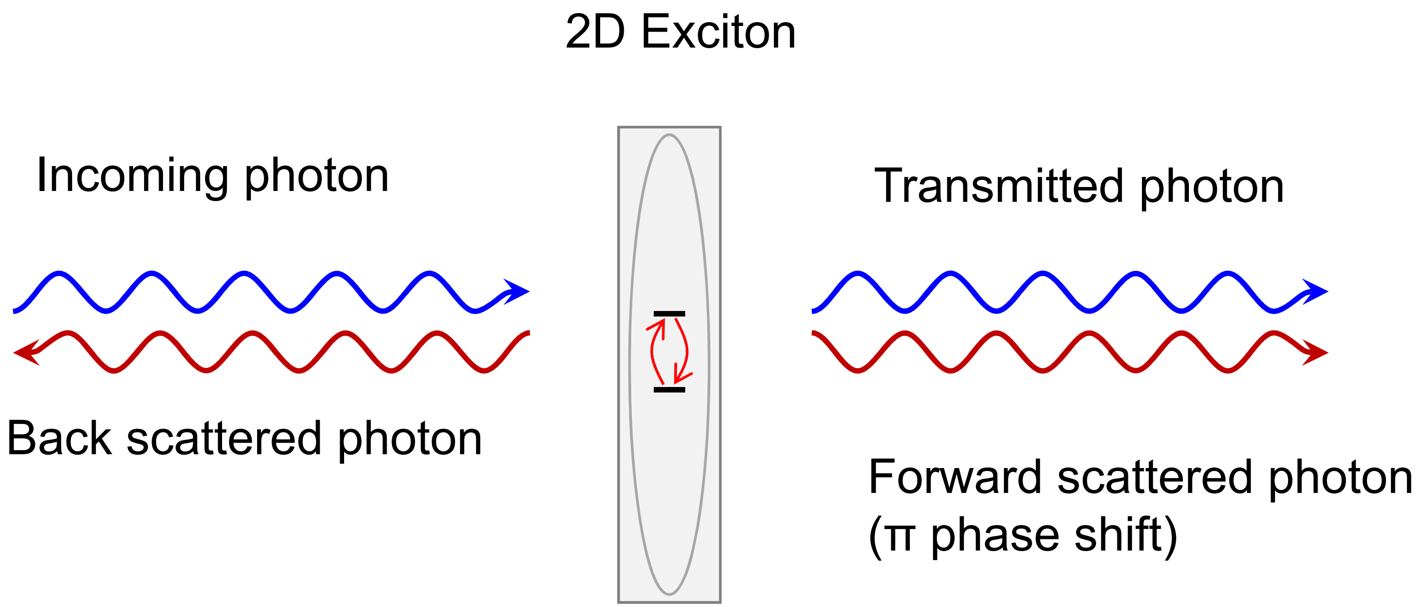
➡ Destructive interference

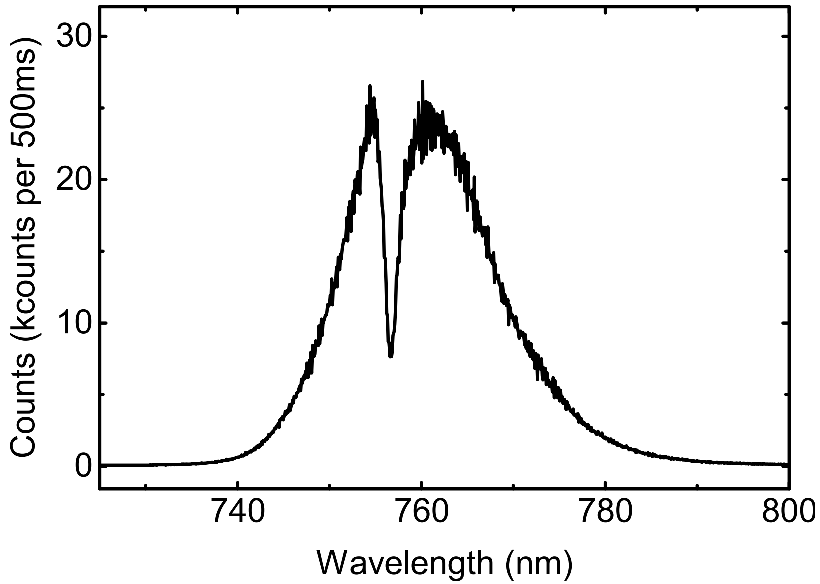


Remember: Quantum dot contrast 0.1 to 10%

- Signal scales with ratio of scattering cross section and focus spot size.
- Dipole emission.

Optical Response of a 2D Exciton



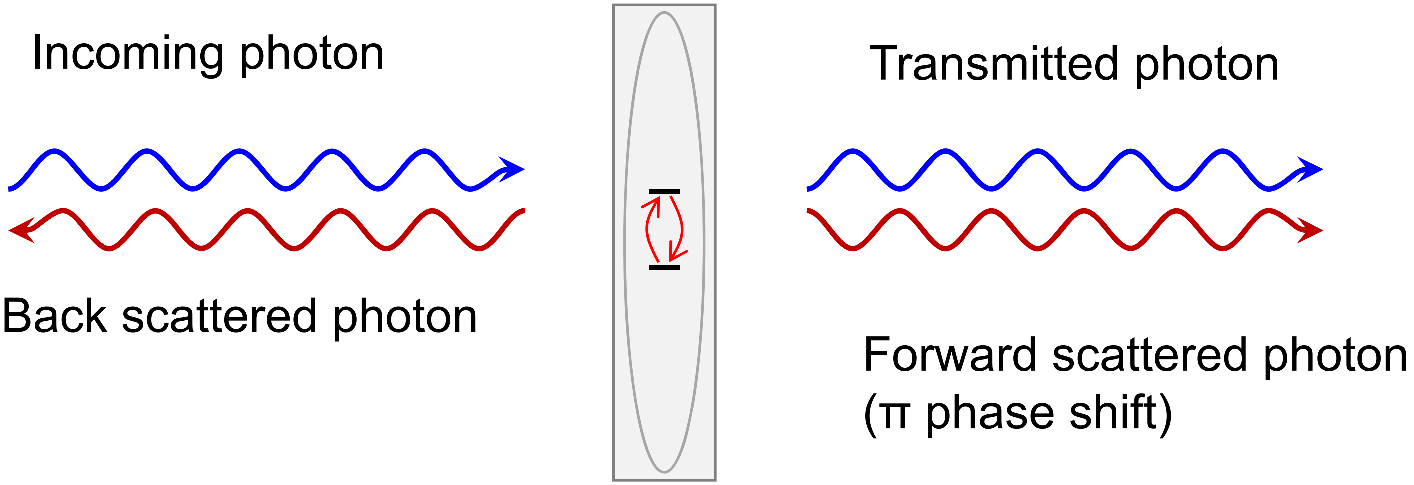


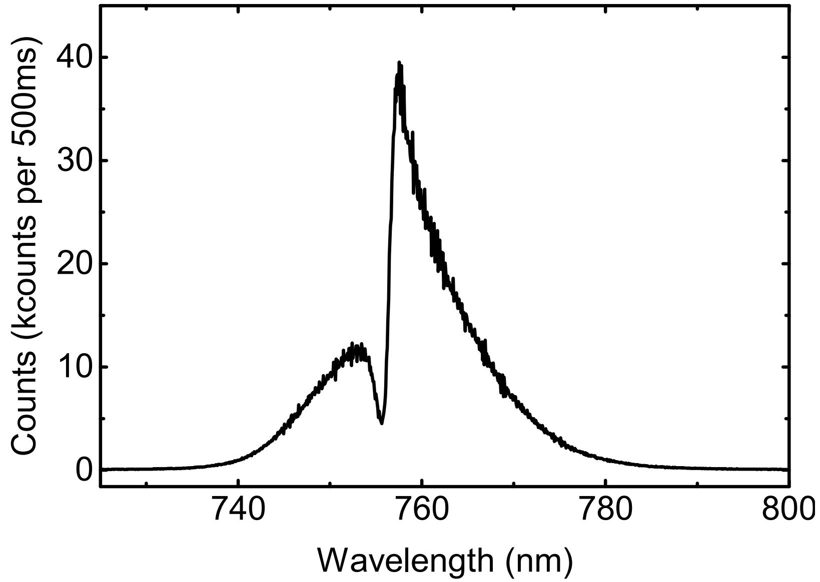
In-plane momentum is conserved
⇨ Scattered mode is identical to excitation mode

➡ Perfect mode overlap
➡ Perfect destructive interference

The “Perfect” Mirror

2D Exciton



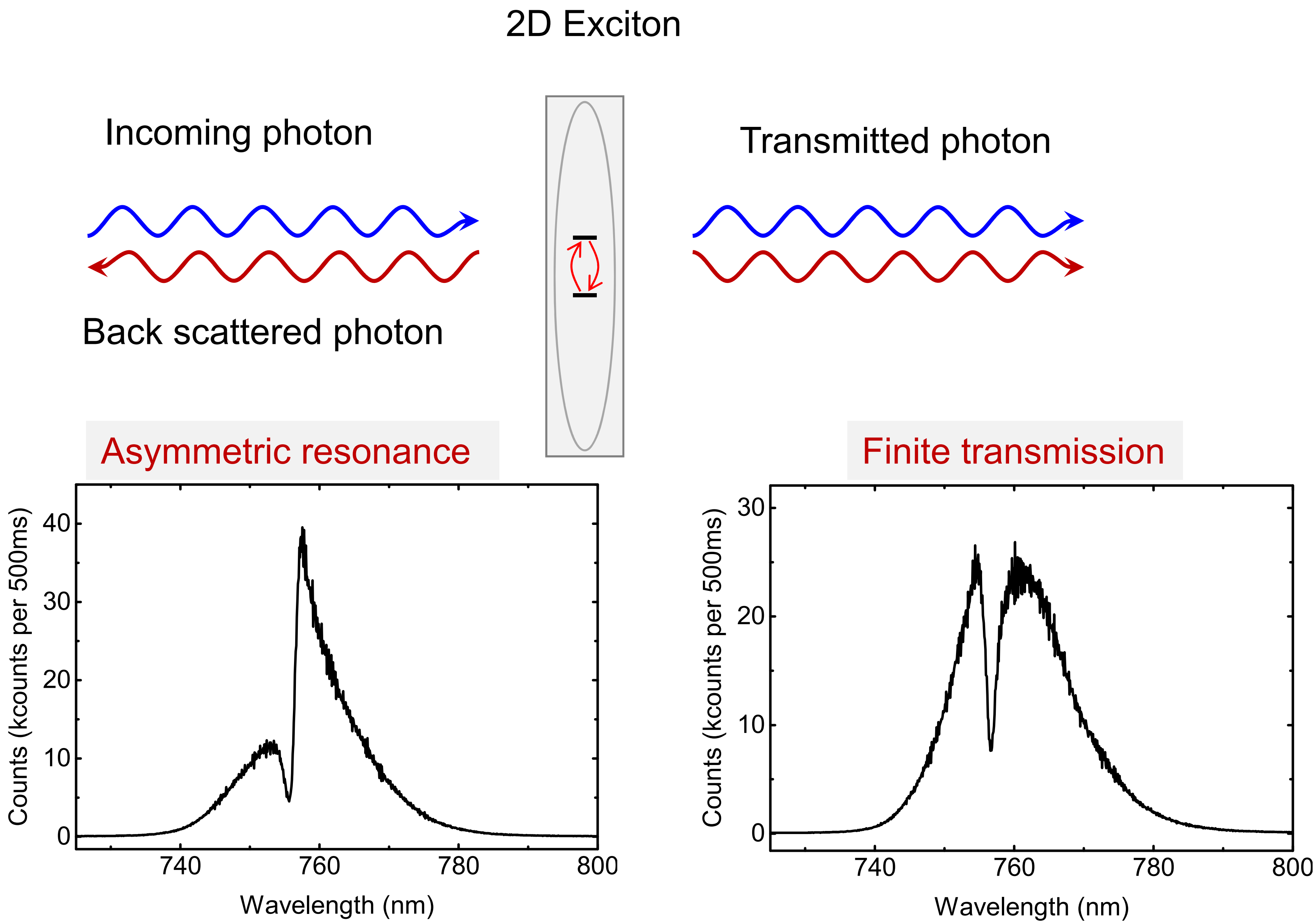


In-plane momentum is conserved
⇨ Scattered mode is identical to excitation mode

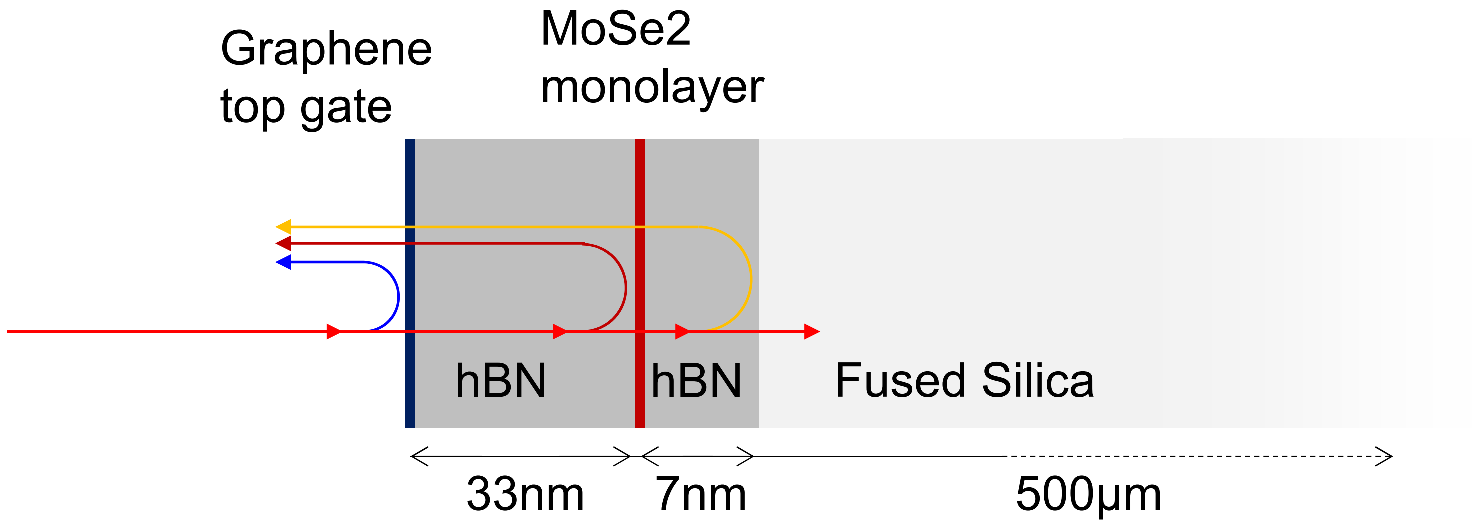
➡ Perfect mode overlap
➡ Perfect destructive interference

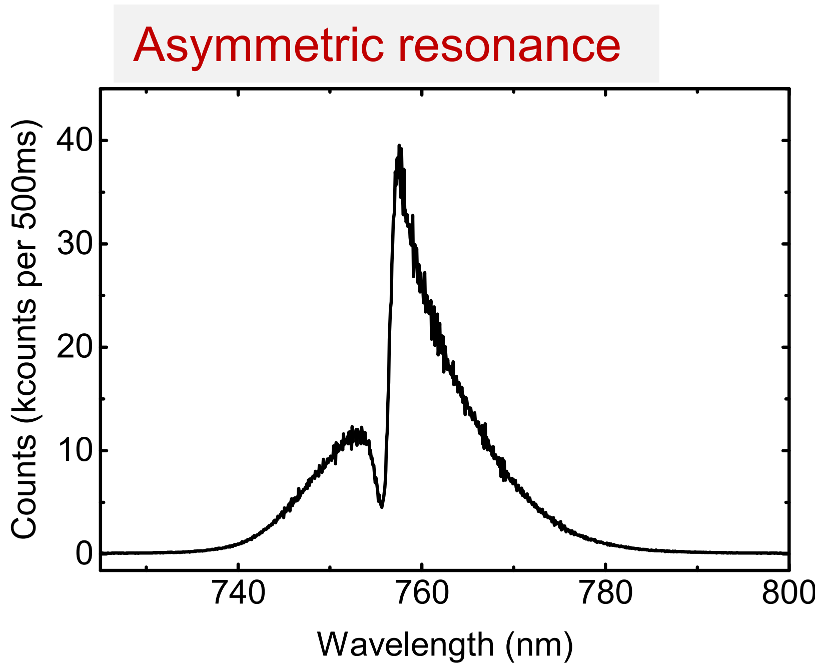
Energy is conserved
➡ All the light is reflected
➡ "Perfect" mirror

The “Perfect” Mirror



The Perfect Mirror

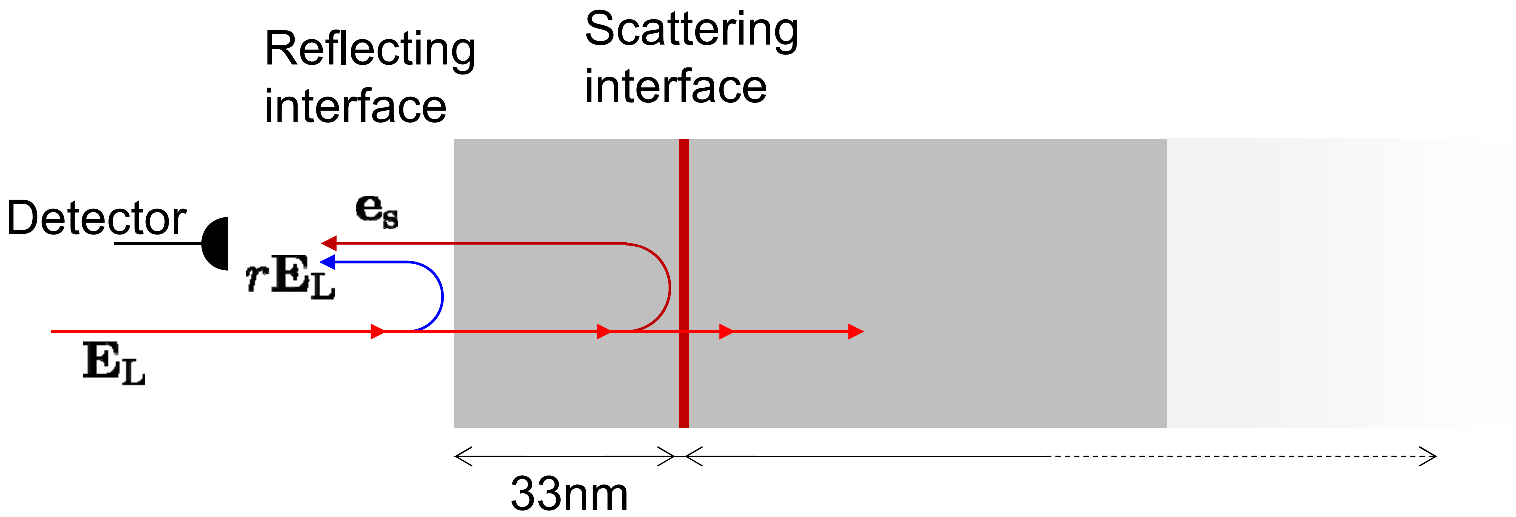




Interference between light reflected from

- Vacuum – hBN interface
- $MoSe_2$ monolayer
- hBN – Fused Silica substrate

Reflection Spectrum



Coherently scattered field: $\mathbf{e}_\mathrm{s} = \mathbf{E}_\mathrm{L} \frac{\delta\gamma + i\gamma^2}{\delta^2 + \gamma^2}$

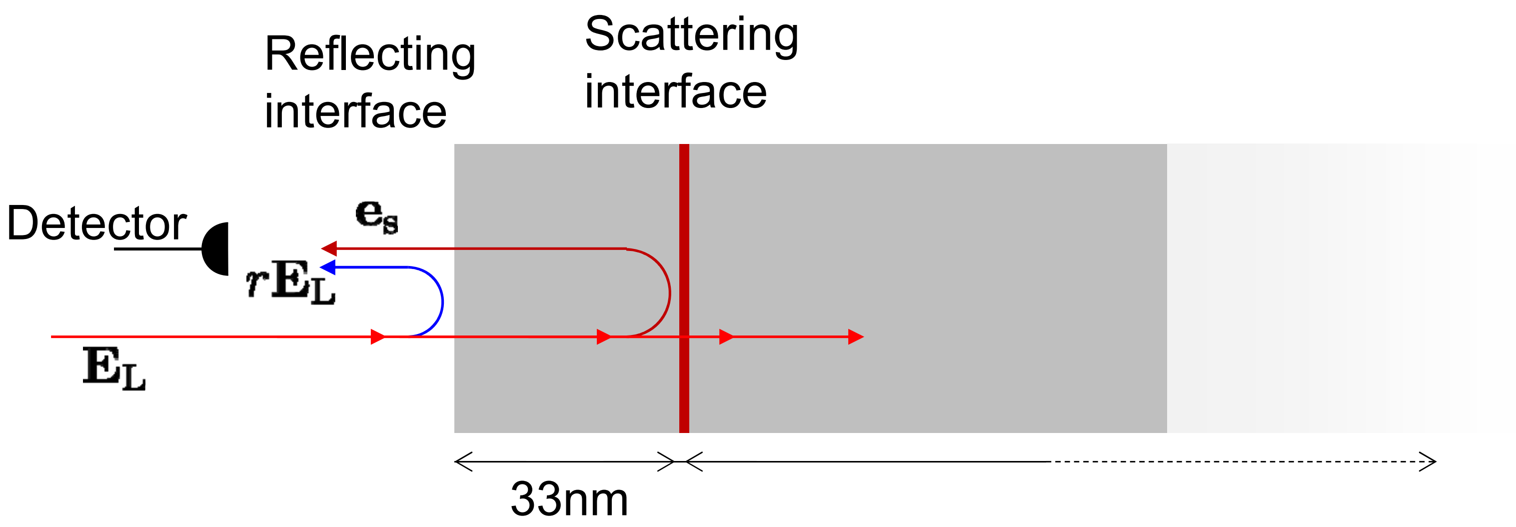
Reflected field: $\Delta R = |r\mathbf{E}_\mathrm{L} + \mathbf{e}_\mathrm{s}|^2 = |r\mathbf{E}_\mathrm{L}|^2 + 2r\mathbf{e}_\mathrm{s}\mathbf{E}_\mathrm{L} + |\mathbf{e}_\mathrm{s}|^2$

Interference term: $\Delta R_\mathrm{I} = 2r\mathbf{e}_\mathrm{s}\mathbf{E}_\mathrm{L}$

Path length difference between $r\mathbf{E}_\mathrm{L}$ and $\mathbf{e}_\mathrm{s}$ on the detector: $L = 2 \times 33\mathrm{nm}$

$\Longrightarrow$ Phase shift: $\phi = \frac{2L}{\lambda} 2\pi$

Reflection Spectrum

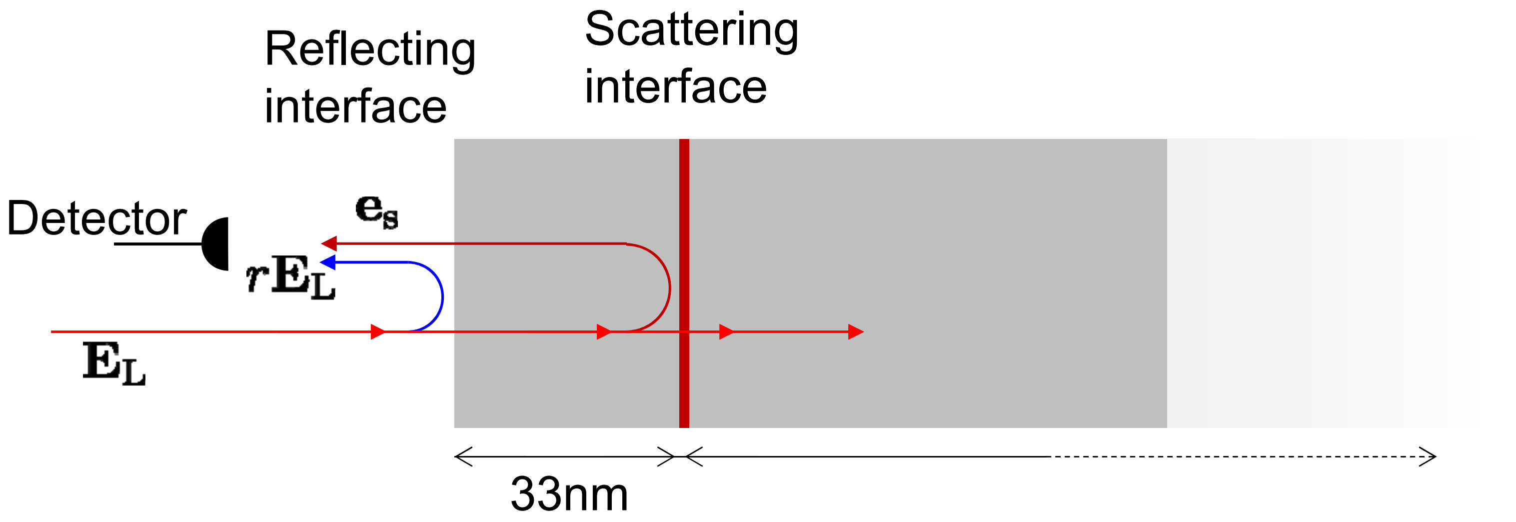


Coherently scattered field: $\mathbf{e}_s = \mathbf{E}_L \frac{\delta\gamma + i\gamma^2}{\delta^2 + \gamma^2}$

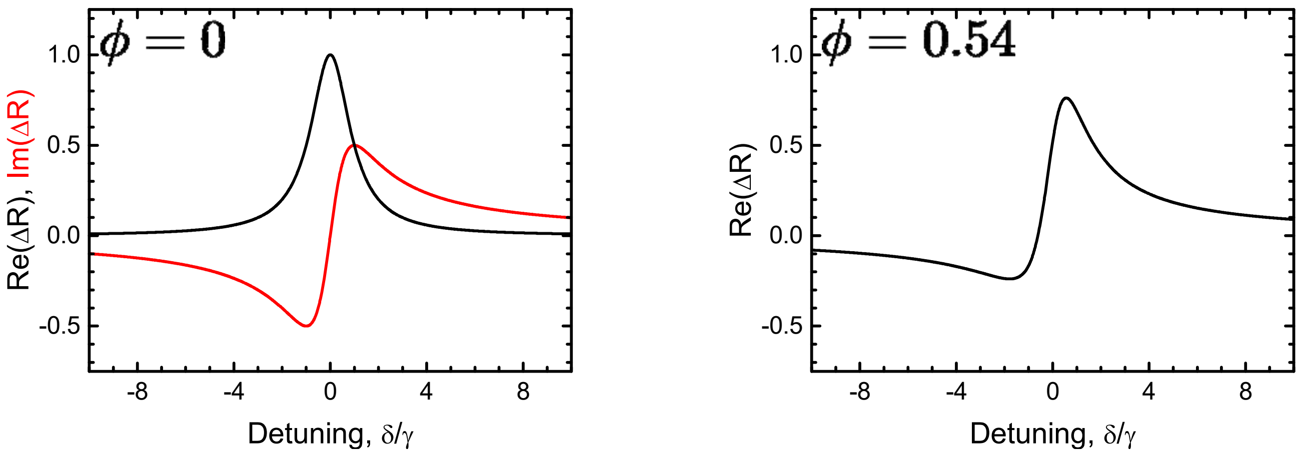
Contribution of the interference term on the detector (only the real part):

$$\Delta R_I = 2r\mathbf{e}_s\mathbf{E}_L = 2re^{i\phi}\frac{\delta\gamma + i\gamma^2}{\delta^2 + \gamma^2} = (\delta\gamma\cos(\phi) - \gamma^2\sin(\phi))\frac{1}{\delta^2 + \gamma^2}$$

Reflection Spectrum



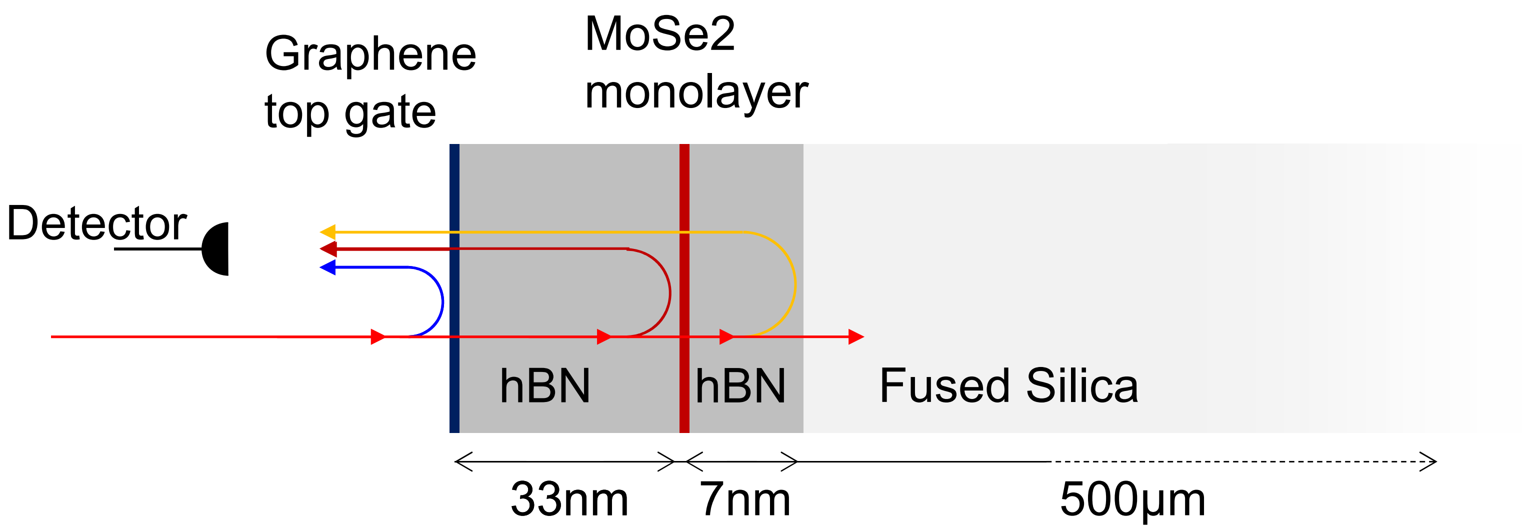
$$\Delta R_I = 2r\mathbf{e}_s\mathbf{E}_L = 2re^{i\phi}\frac{\delta\gamma + i\gamma^2}{\delta^2+\gamma^2} = (\delta\gamma\cos(\phi) - \gamma^2\sin(\phi))\frac{1}{\delta^2+\gamma^2}$$



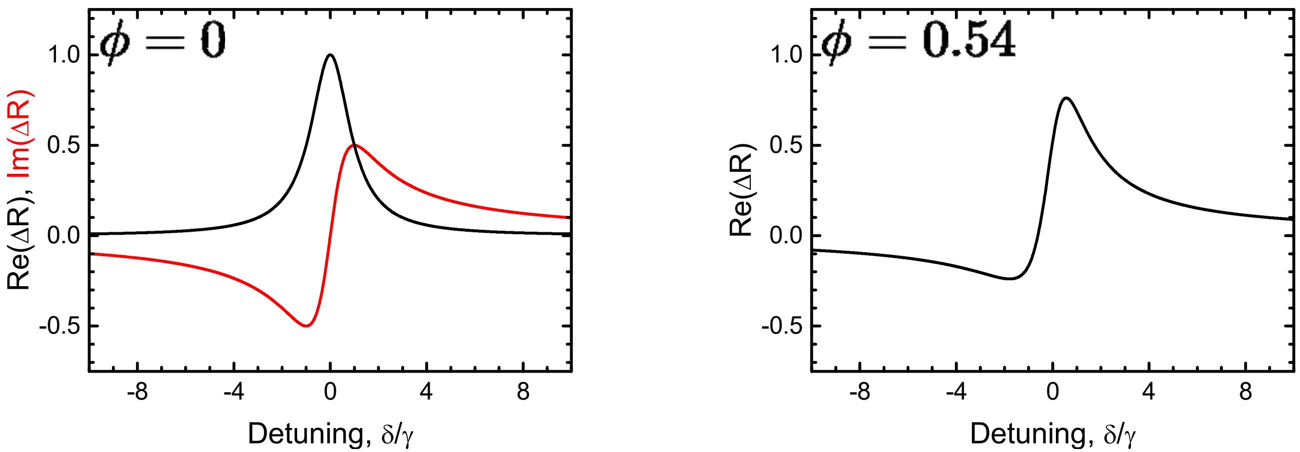
Knowing all thicknesses and refractive indices:

⟹ Calculate normalized transmission and reflection spectra

Reflection Spectrum

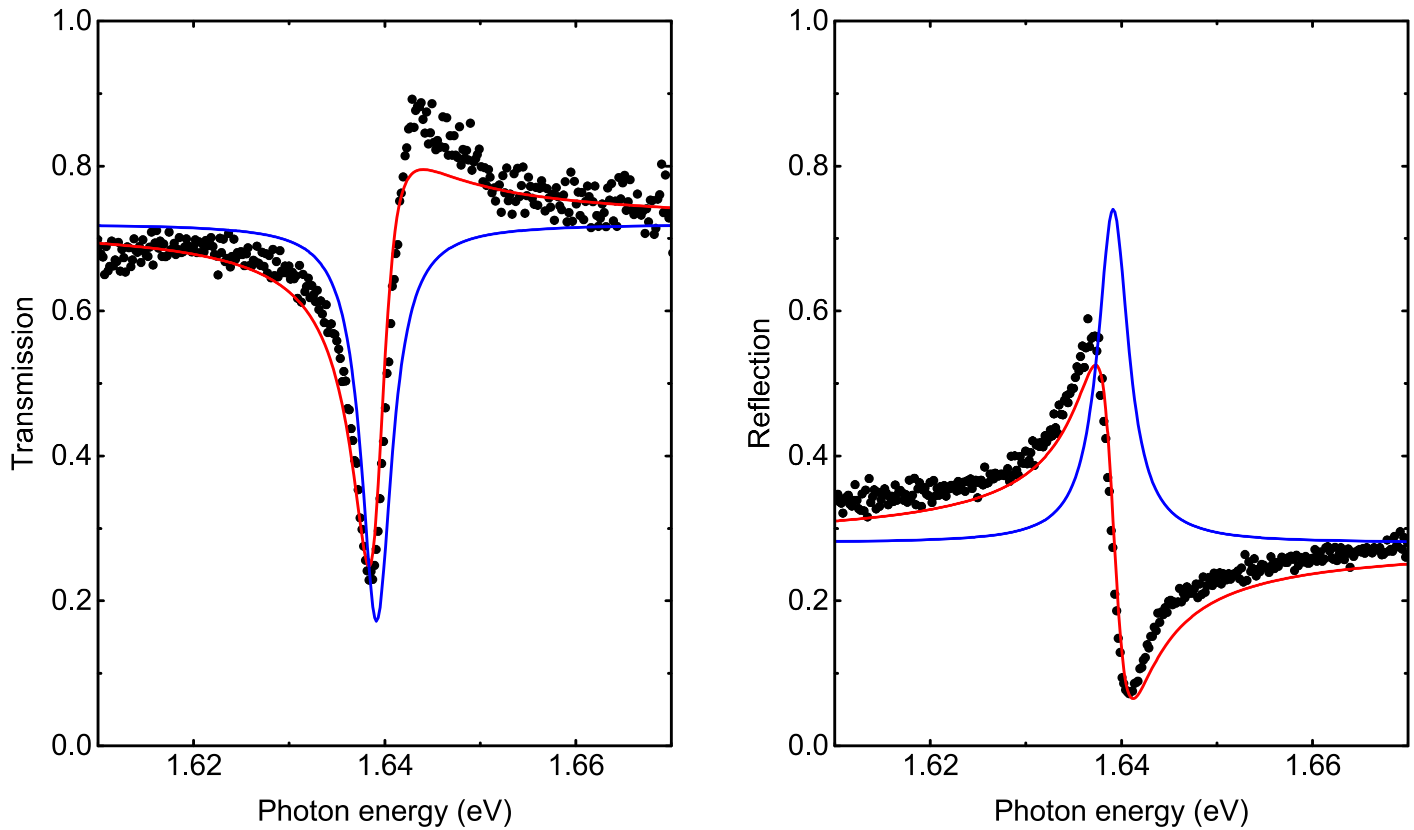


$$\Delta R_{\mathrm{I}} = 2r\mathbf{e}_{\mathrm{s}}\mathbf{E}_{\mathrm{L}} = 2re^{i\phi}\frac{\delta\gamma + i\gamma^2}{\delta^2+\gamma^2} = (\delta\gamma\cos(\phi) - \gamma^2\sin(\phi))\frac{1}{\delta^2+\gamma^2}$$



Knowing all thicknesses and refractive indices:
⟹ Calculate normalized transmission and reflection spectra

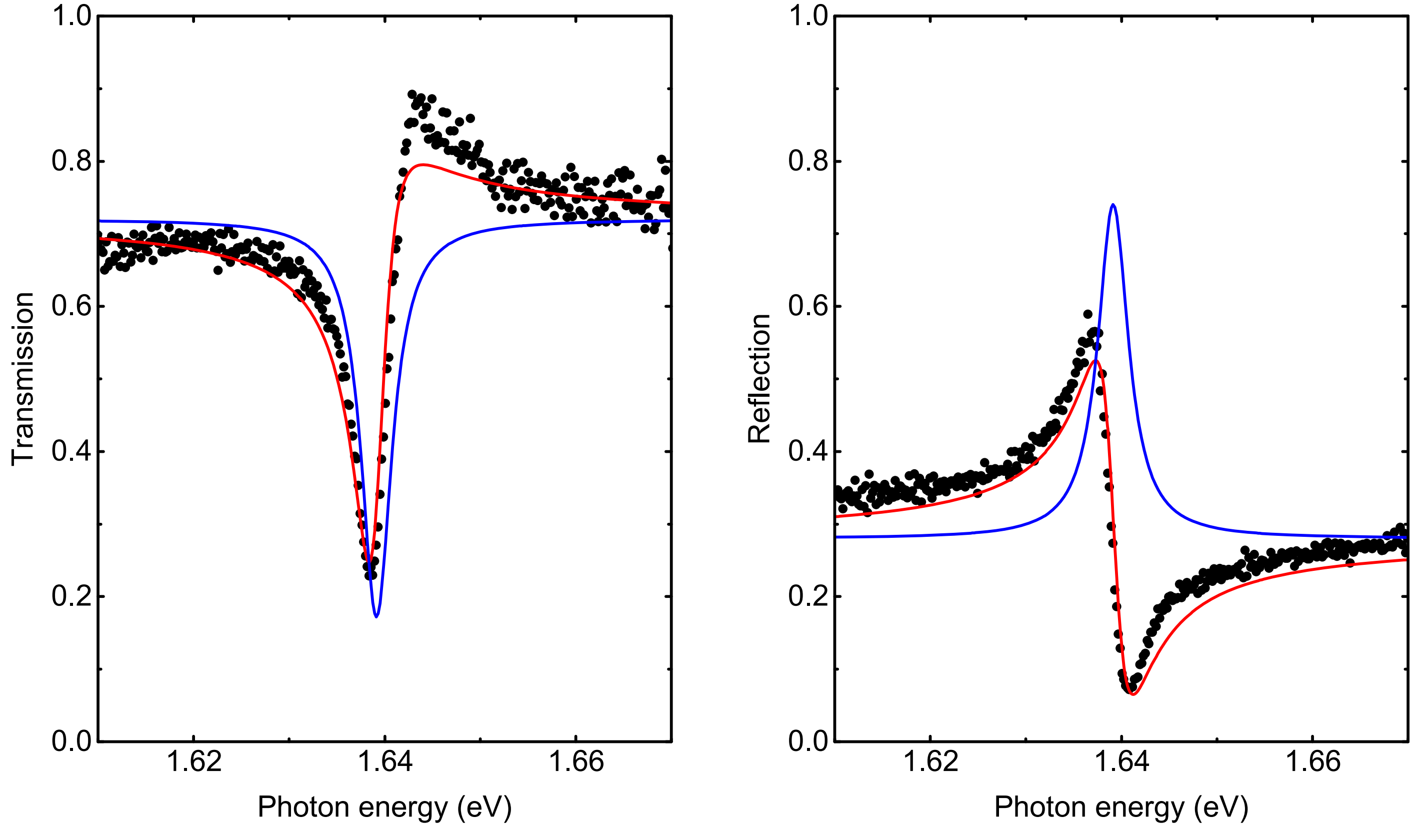
An Atomically Thin Mirror



Knowing all thicknesses and refractive indices:

⟹ Calculate normalized transmission and reflection spectra

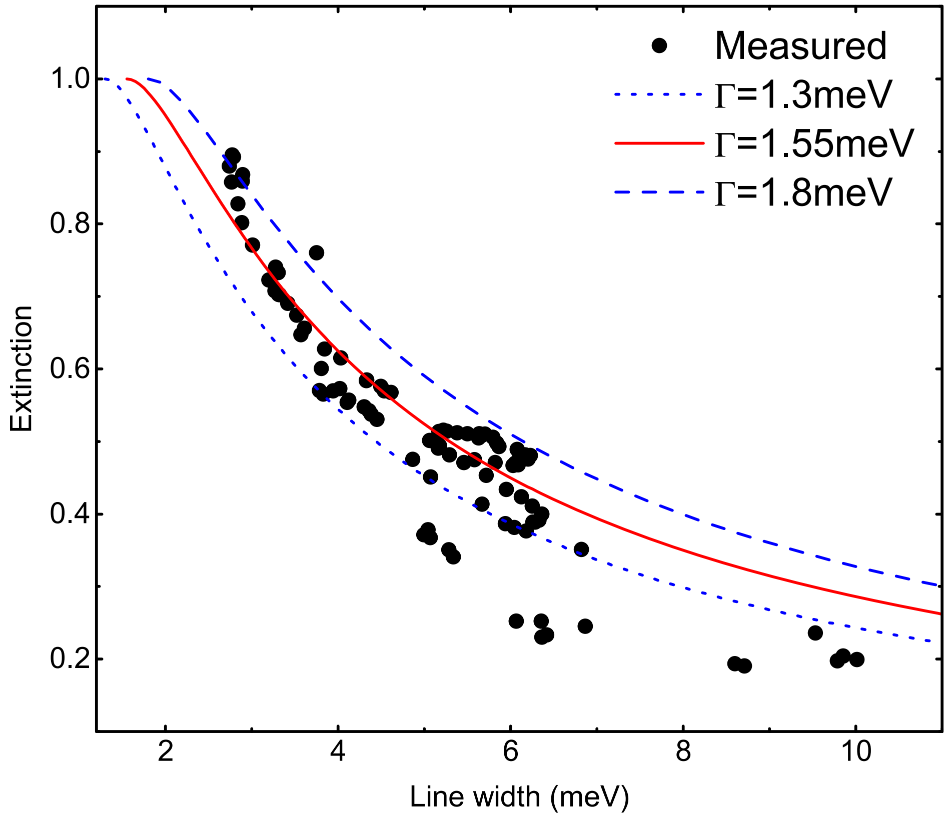
An Atomically Thin Mirror

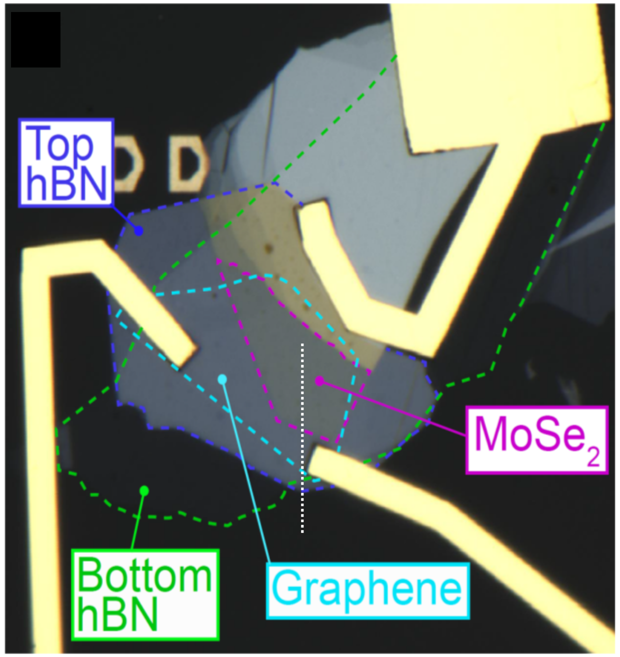


Maximum extinction: 76%
Maximum reflection: 46%

Limited by inhomogeneous broadening (disorder).
Disorder on sample leads to shifts in resonance and fluctuations in line width.

Influence of Disorder





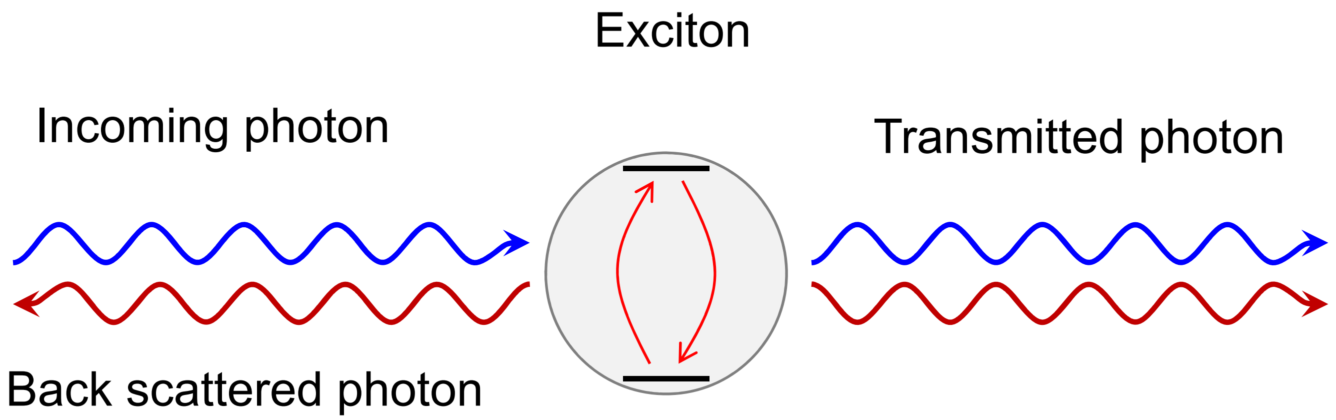
Extinction:

$$1 - T_{\mathrm{min}} = 1 - \frac{(\gamma_{\mathrm{tot}} - \Gamma)^2}{\gamma_{\mathrm{tot}}^2}$$

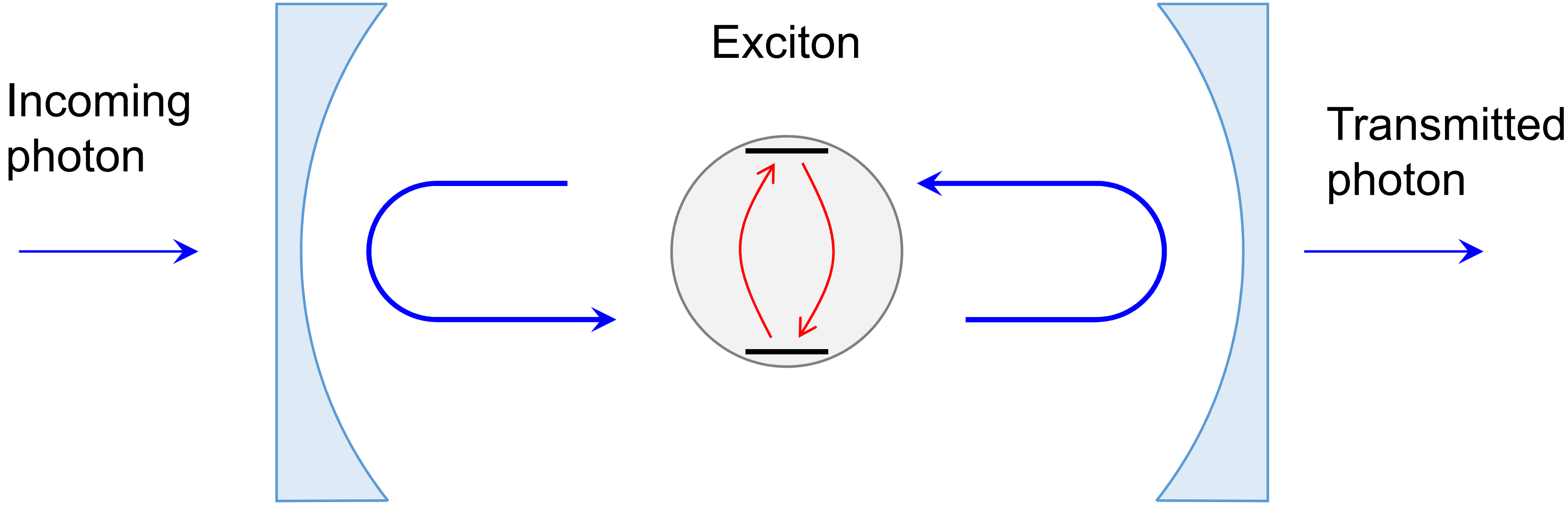
Extrapolation of line with to Γ=1.3-1.8meV

Limited by inhomogeneous broadening (disorder).
Disorder on sample leads to shifts in resonance and fluctuations in line width.

P. Back, arXiv 1705.07317


Exciton
Incoming photon
Transmitted photon
Back scattered photon

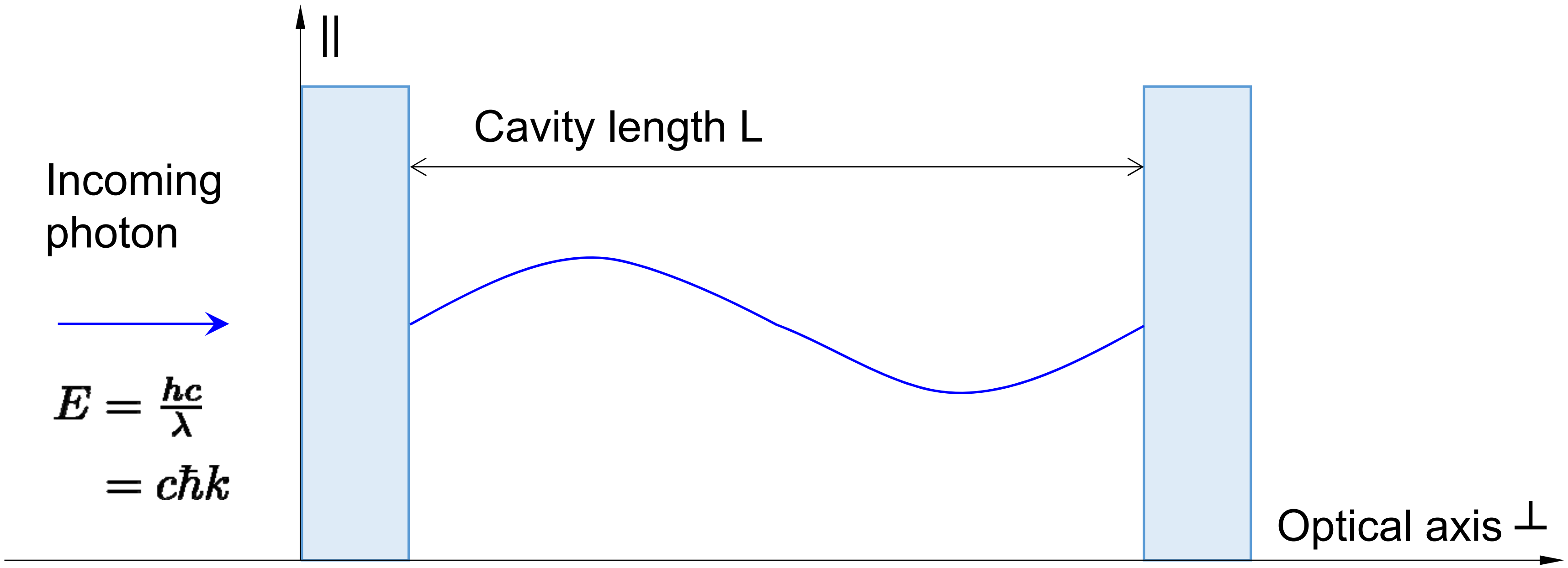
Cavity Quantum Electrodynamics



Confine photon to cavity mode

Photon can be scattered by the exciton multiple times

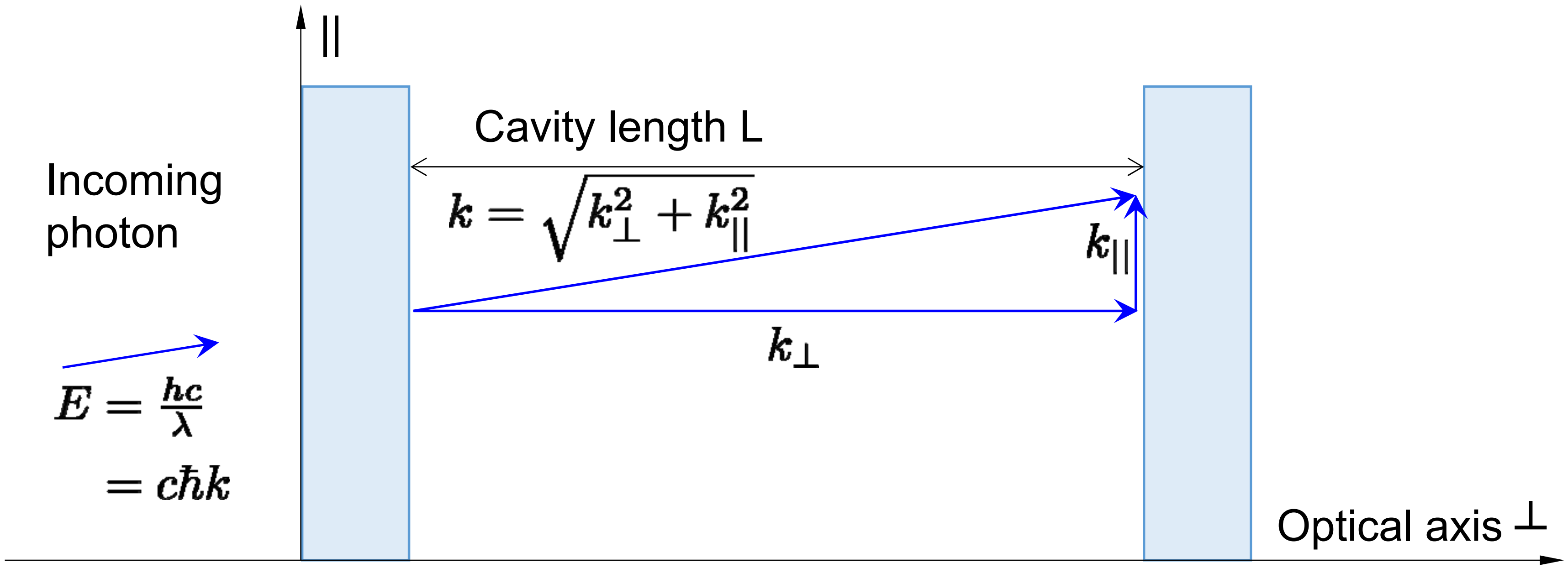
Planar 2D Cavity



Cavity length defines resonance wavelength: $2L \equiv n\lambda \qquad n = 1, 2, \ldots$

$\Longrightarrow k_\perp = \frac{n\pi}{L}$ Cavity mode has well defined $k_\perp$

Planar 2D Cavity



Cavity length defines resonance wavelength: $2L \equiv n\lambda \qquad n = 1, 2, \ldots$

$\Longrightarrow k_\perp = \frac{n\pi}{L}$ Cavity mode has well defined $k_\perp$

In general:

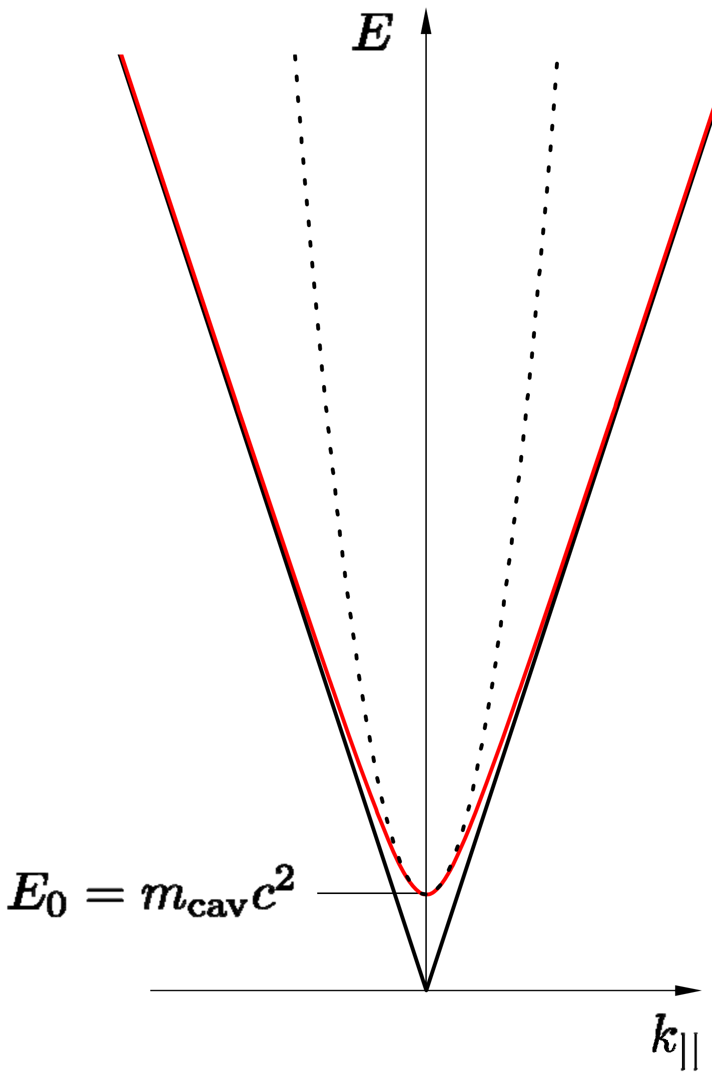
Energy of a cavity photon: $E = \hbar\omega = c\hbar k = c\hbar\sqrt{k_\perp^2 + k_{||}^2}$

$$(\hbar\omega)^2 = \left(\frac{n\pi\hbar}{Lc}c^2\right)^2 + \left(\hbar c k_{||}\right)^2$$

With: $m_{\mathrm{cav}} = \frac{n\pi\hbar}{Lc}$

$$\boxed{\hbar\omega \approx m_{\mathrm{cav}}c^2 + \frac{\hbar^2 k_{||}^2}{2m_{\mathrm{cav}}}}$$

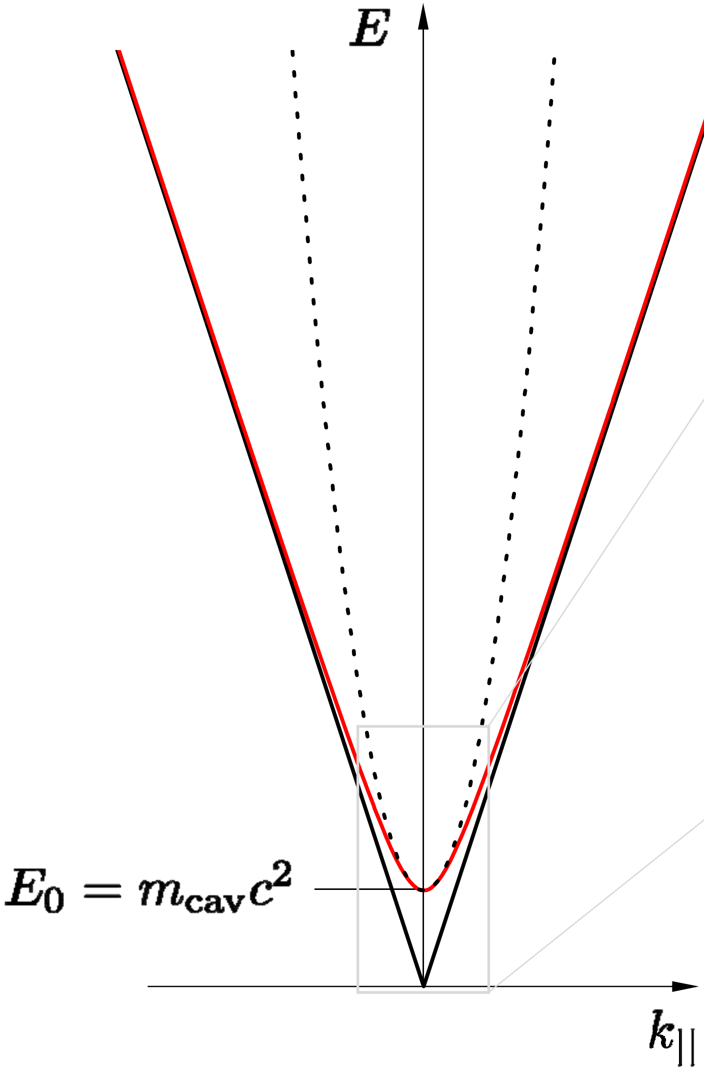
Cavity Dispersion

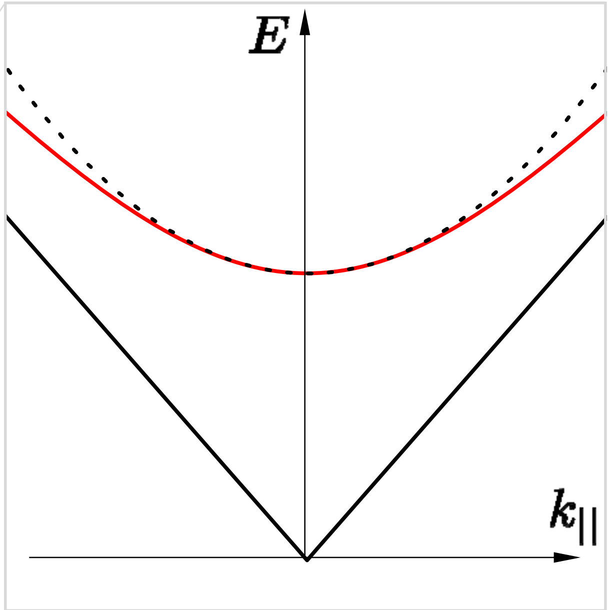


$$(\hbar\omega)^2 = \left(\frac{n\pi\hbar}{Lc}c^2\right)^2 + \left(\hbar c k_{||}\right)^2$$

$$\boxed{\hbar\omega \approx m_{\rm cav}c^2 + \frac{\hbar^2 k_{||}^2}{2m_{\rm cav}}}$$

Cavity Dispersion

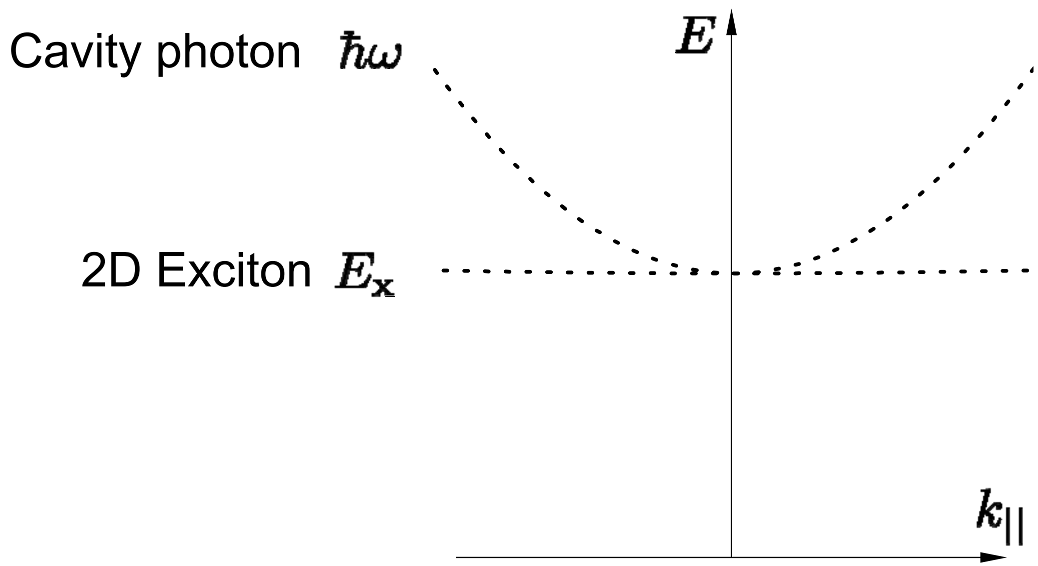




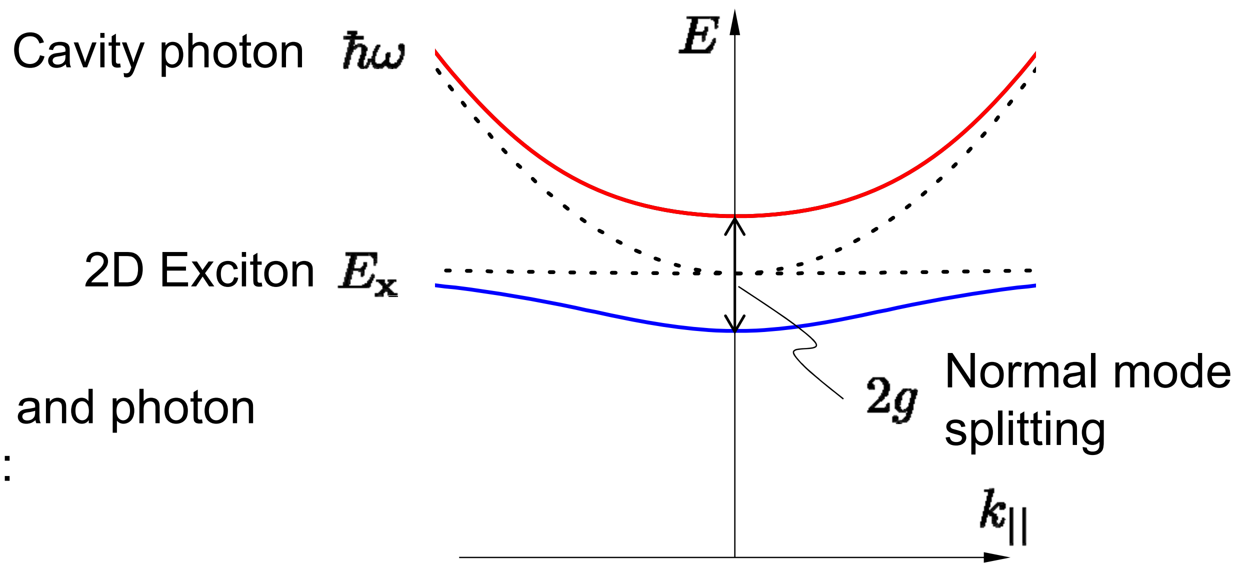
$$(\hbar\omega)^2 = \left(\frac{n\pi\hbar}{Lc} c^2\right)^2 + \left(\hbar c k_{||}\right)^2$$

$$\hbar\omega \approx m_{\mathrm{cav}} c^2 + \frac{\hbar^2 k_{||}^2}{2 m_{\mathrm{cav}}}$$

Cavity Mode Coupled to 2D Exciton



Cavity Mode Coupled to 2D Exciton



Coupling between exciton and photon
(coupled oscillator model):

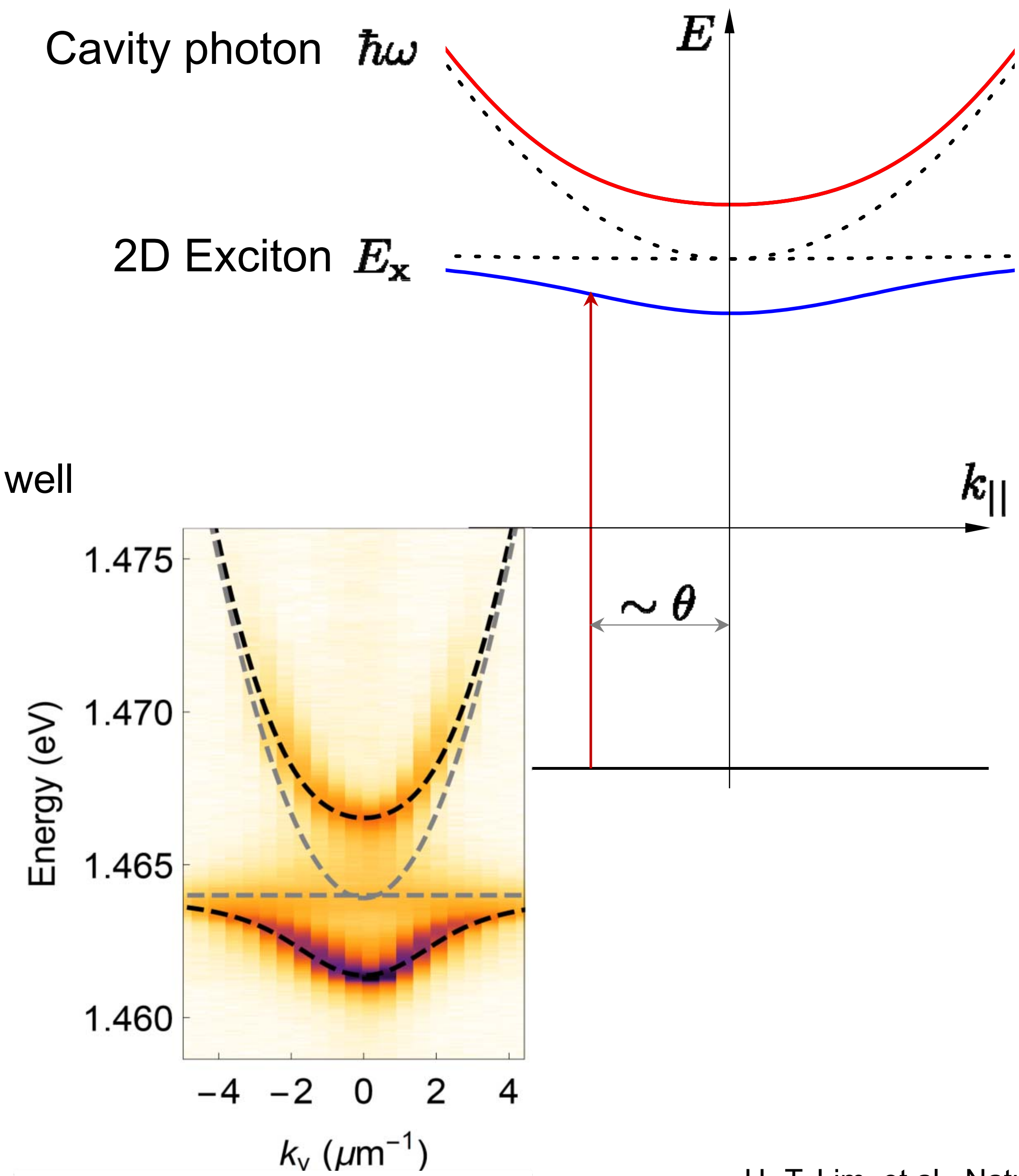
$$\hat{H} = \begin{pmatrix} \hbar\omega & g \\ g & E_{\mathbf{x}} \end{pmatrix}$$

Coupling constant g

New eigenmodes: $$E_{\pm} = \frac{1}{2}\left(\hbar\omega + E_{\mathbf{x}} \pm \sqrt{4g^2 + (\hbar\omega - E_{\mathbf{x}})}\right)$$

E_+ ≡ Upper polariton
E_- ≡ Lower polariton

Cavity Mode Coupled to 2D Exciton



Resonant laser absorption:

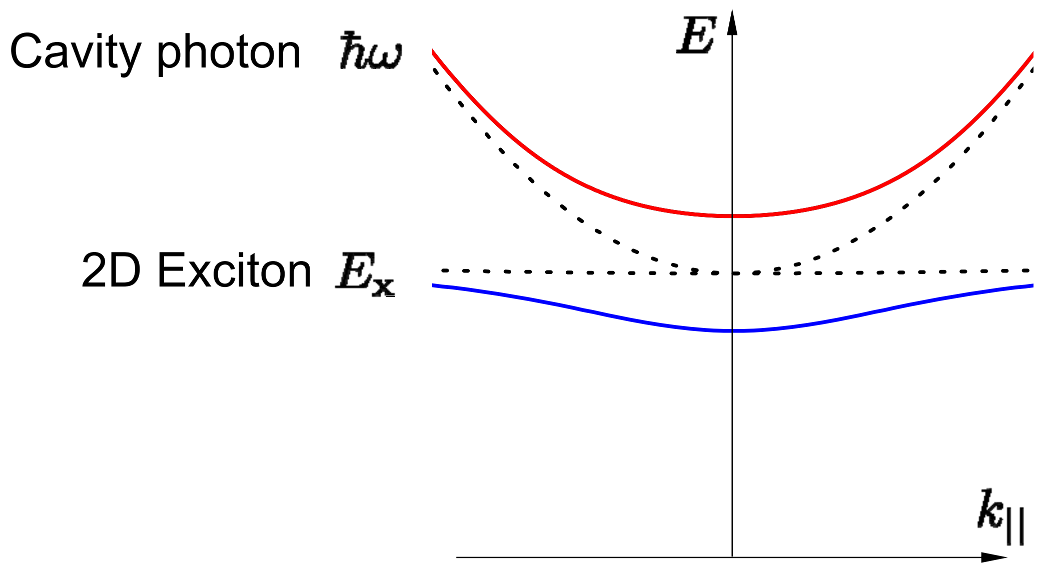
⇨ Probe dispersion with well defined:

- Photon energy $E_{\mathbf{Laser}}$

- In-plane momentum
  $k_{||} = k \sin\theta \approx \frac{2\pi\theta}{\lambda}$

H.-T. Lim, et al., Nature Comm. (2016)

Cavity Mode Coupled to 2D Exciton



Lower polariton:

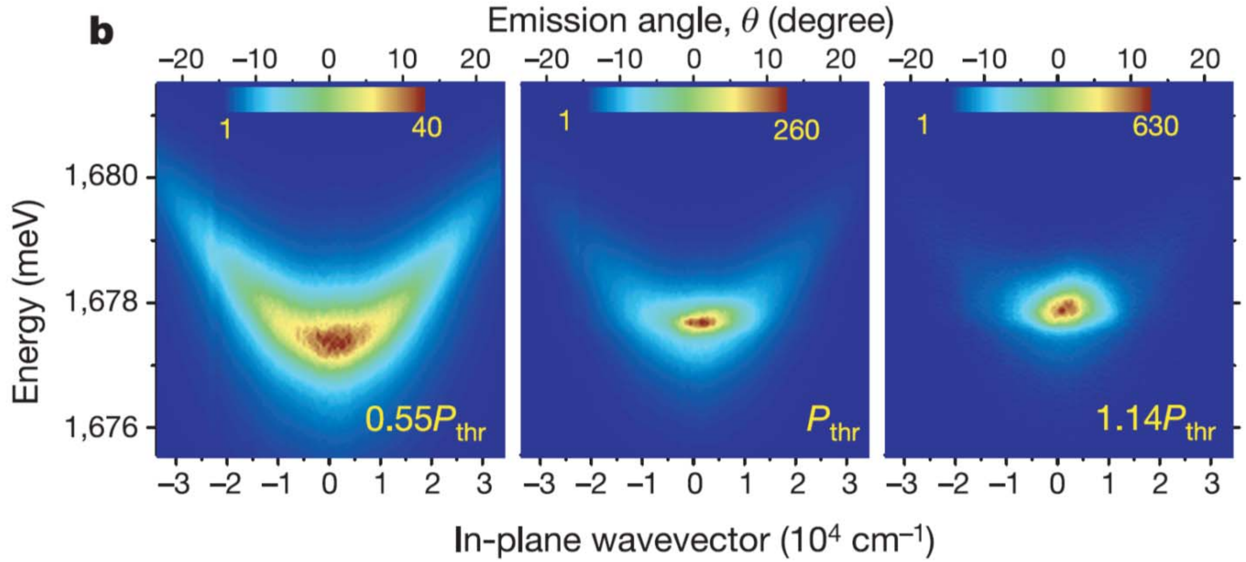
Lower mass than exciton

$m_{\mathbf{cav}} \approx 1 \times 10^{-}5 m_e$

At resonance: $\frac{1}{m_{\mathbf{pol}}} = \frac{1}{2m_{\mathbf{cav}}}$

⇨ Bose-Einstein condensation of polaritons. (up to room temperature)

$T_c \sim \frac{1}{m}$

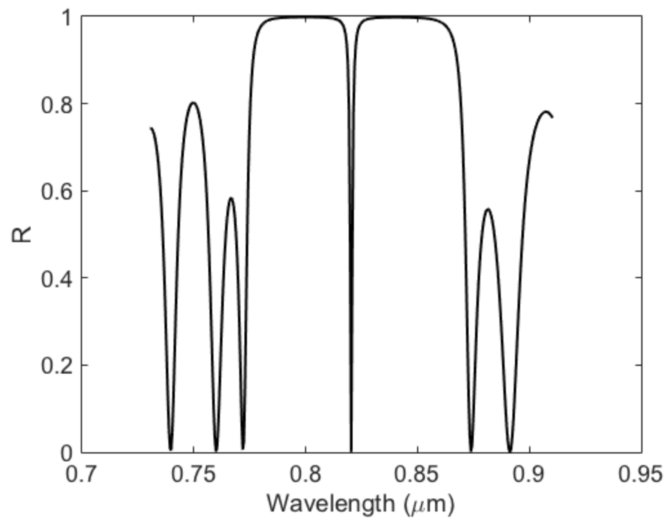


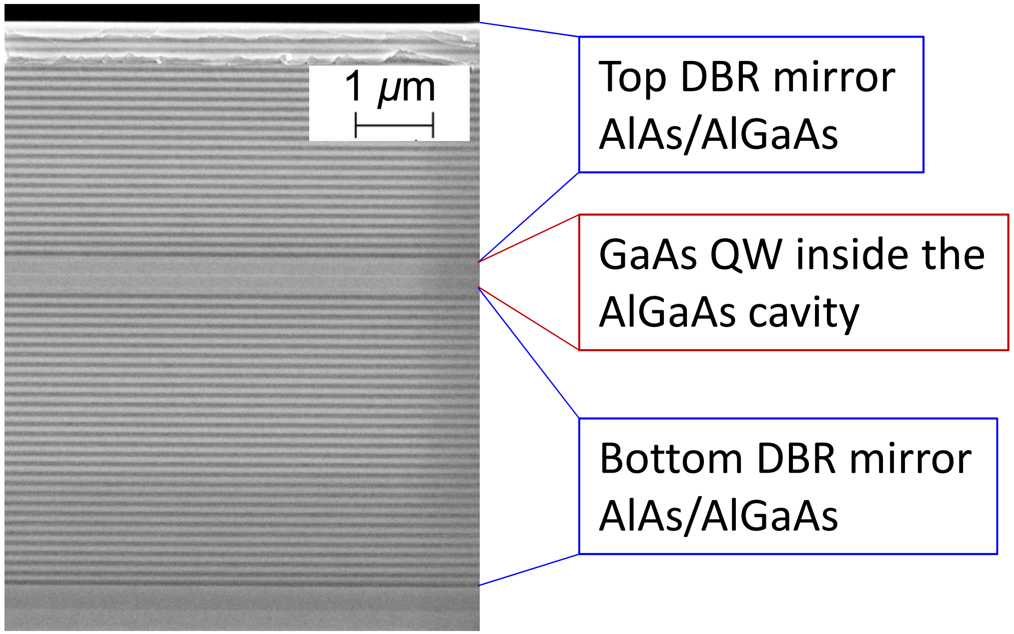
J. Kasprzak, et al., Nature (2006)

2D Polaritons

Example: Epitaxially grown planar microcavity

Reflection spectrum of a cavity with Bragg mirrors:



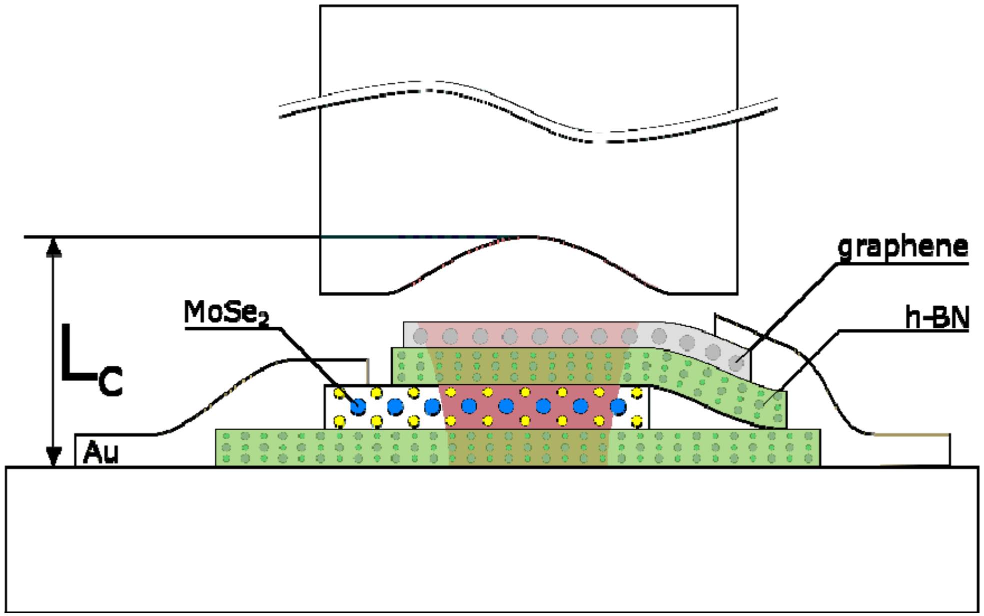


Cross section SEM image

High quality, versatile, robust micro cavity
Basis for most quantum photonic devices.

TMD Coupled to a 0D Cavity Mode

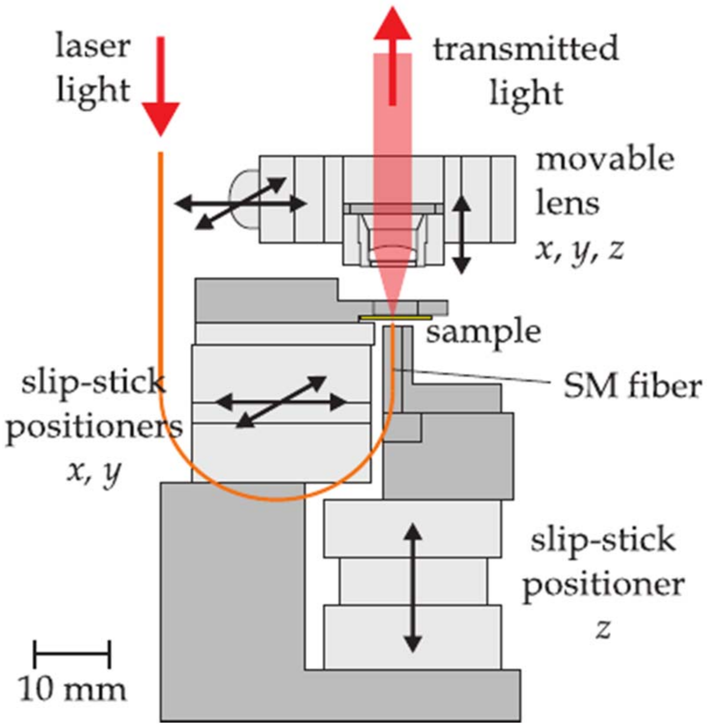
Open optical-fiber based cavity setup:

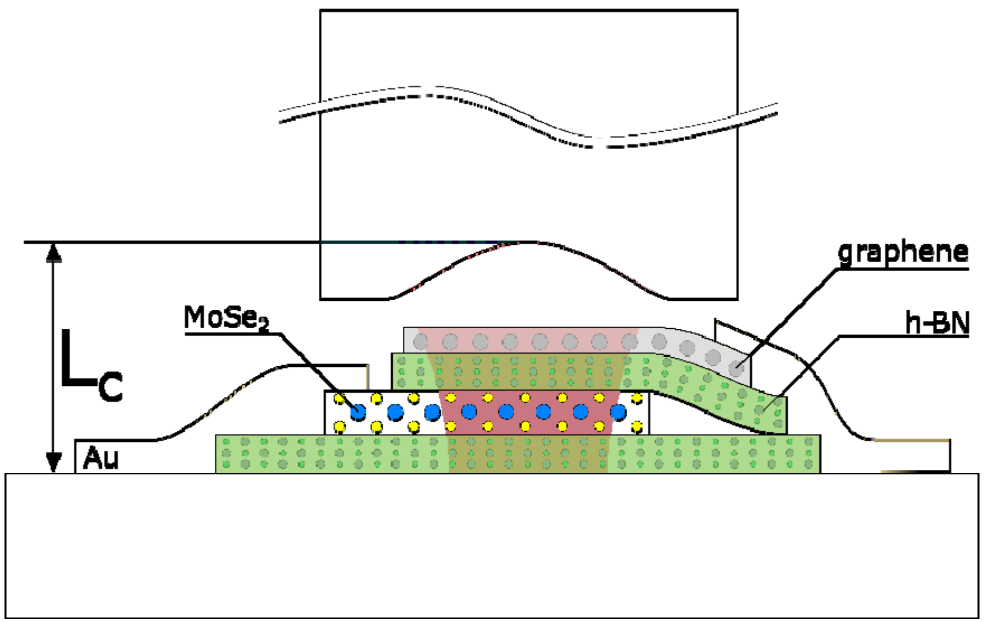


D. Hunger, NJoP, (2010), S. Dufferwiel, Nat.Phot. (2017), M. Sidler, Nat.Phys. (2017)

TMD Coupled to a 0D Cavity Mode

Open optical-fiber based cavity setup:

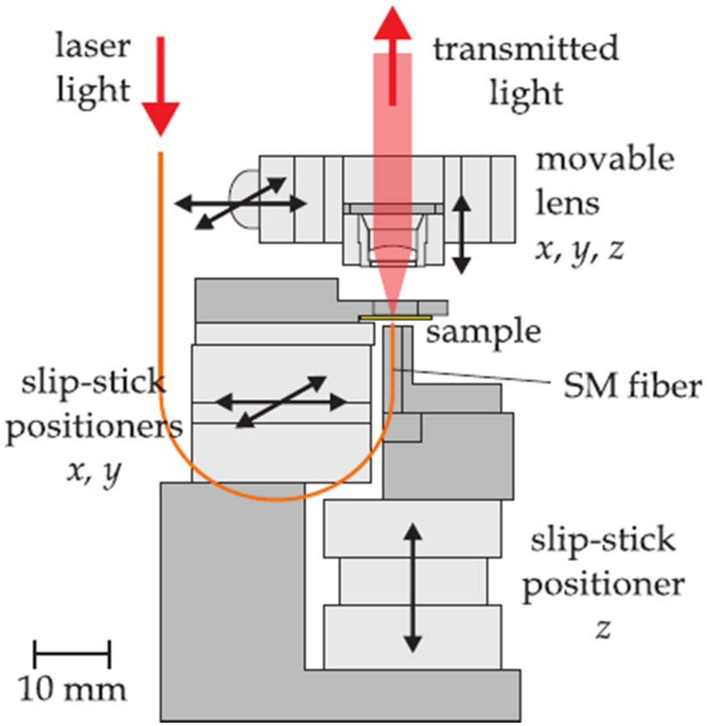


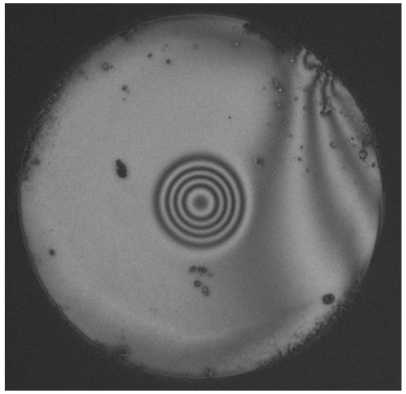


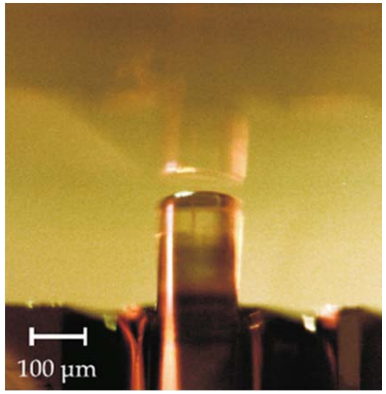
M. Sidler, Nat.Phys. (2017)

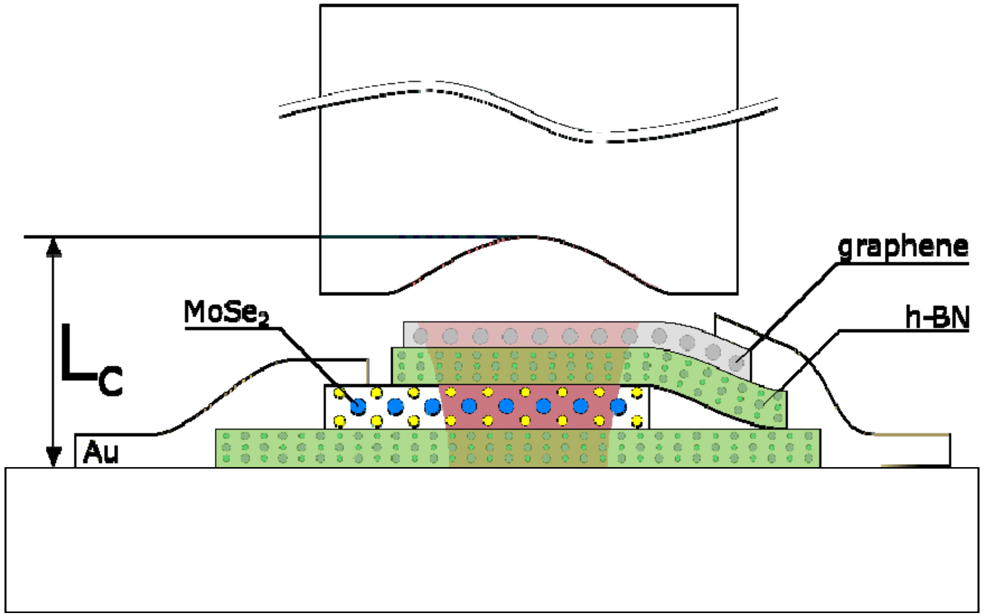
TMD Coupled to a 0D Cavity Mode

Open optical-fiber based cavity setup:







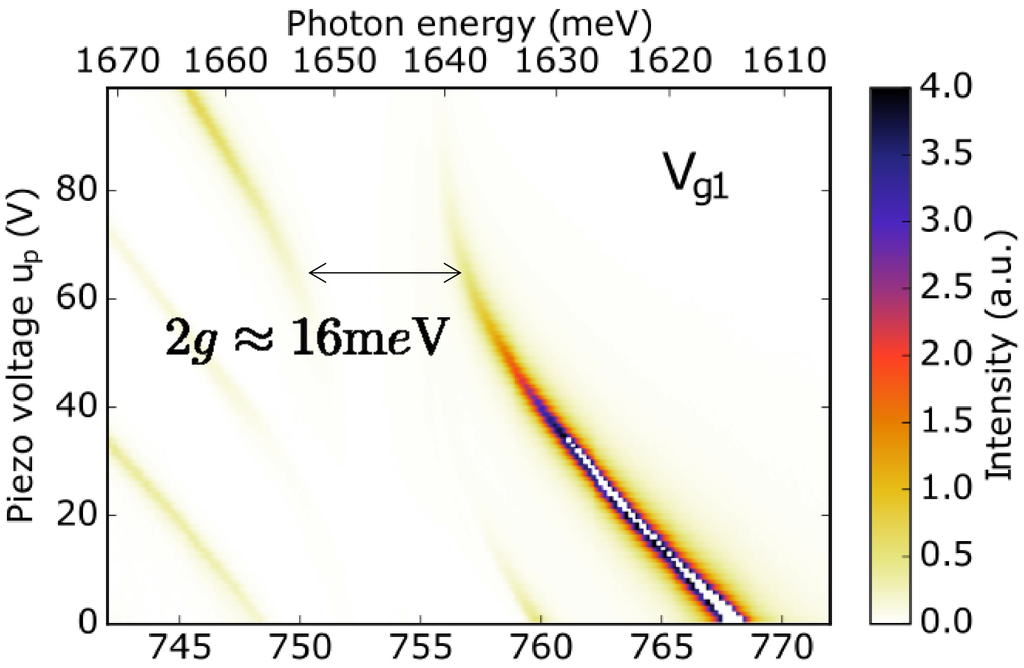


M. Sidler, Nat.Phys. (2017)

Strong Coupling of a TMD Exciton to a 0D Cavity Mode

Open optical-fiber based cavity setup:
- Scan cavity mode to sample position
- Tune cavity length

Transmission spectrum as function of cavity length:

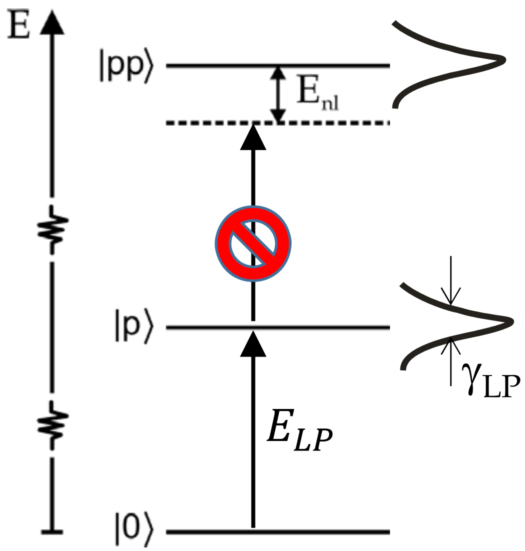


Normal mode splitting: Direct measurement of light matter coupling

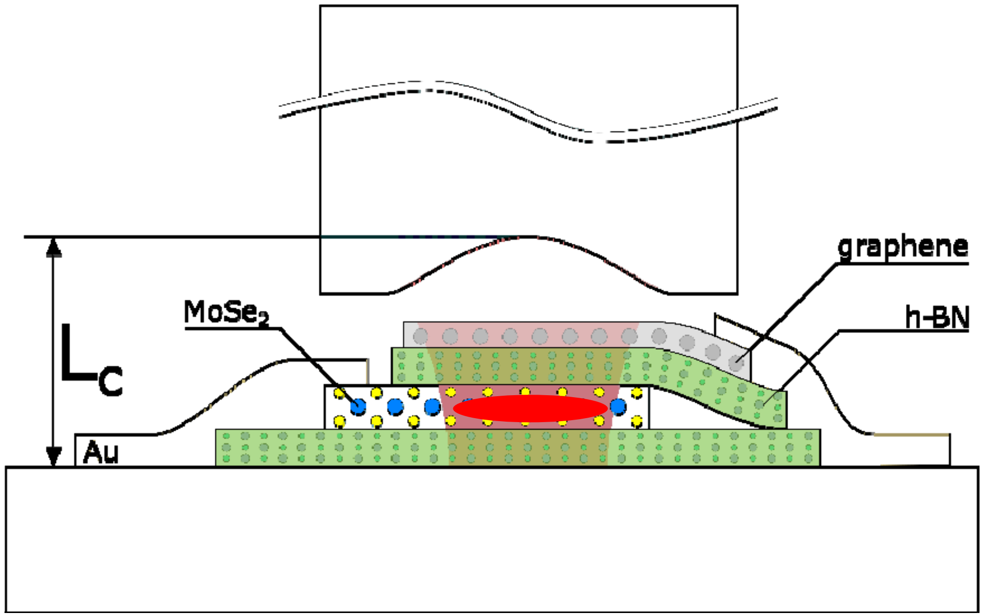
M. Sidler, Nat.Phys. (2017)

Single Photon Interactions

0D Cavity mode
⟹ confined polariton state



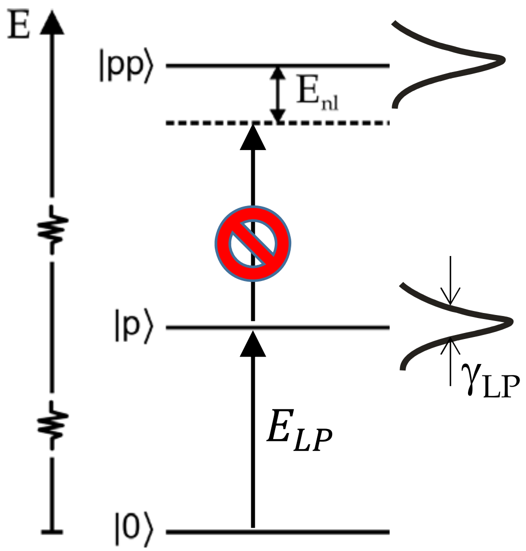
Interactions > Linewidth
⟹ Second polariton can not be created resonantly

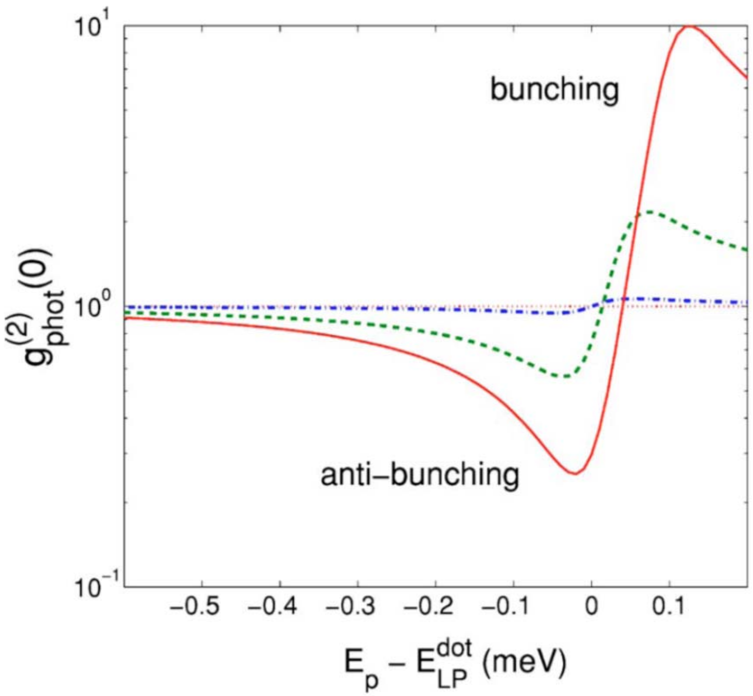


A. Verger, Phys. Rev. B (2006)

Single Photon Interactions

0D Cavity mode
⟹ confined polariton state





Interactions > Linewidth
⟹ Second polariton can not be created resonantly

Experimental signature:
Non classical light emission – anti bunching

A. Verger, Phys. Rev. B (2006)