EXCITON-POLARITONS IN 2D TRANSITION METAL DICHALCOGENIDE LAYERS PLACED IN A PLANAR MICROCAVITY

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Outline

1. Introduction

- Transition metal dichalcogenides (TMDs);
- Excitons in TMDs and the optical conductivity

2. TMD layer inserted in a planar microcavity

- Dispersion relations for TE and TM waves;
- Polariton lifetime;
- Photoluminescence
  (calculated results vs experiment)

3. Two TMD layers in a microcavity

- Dispersion relations;
- Hopfield coefficients

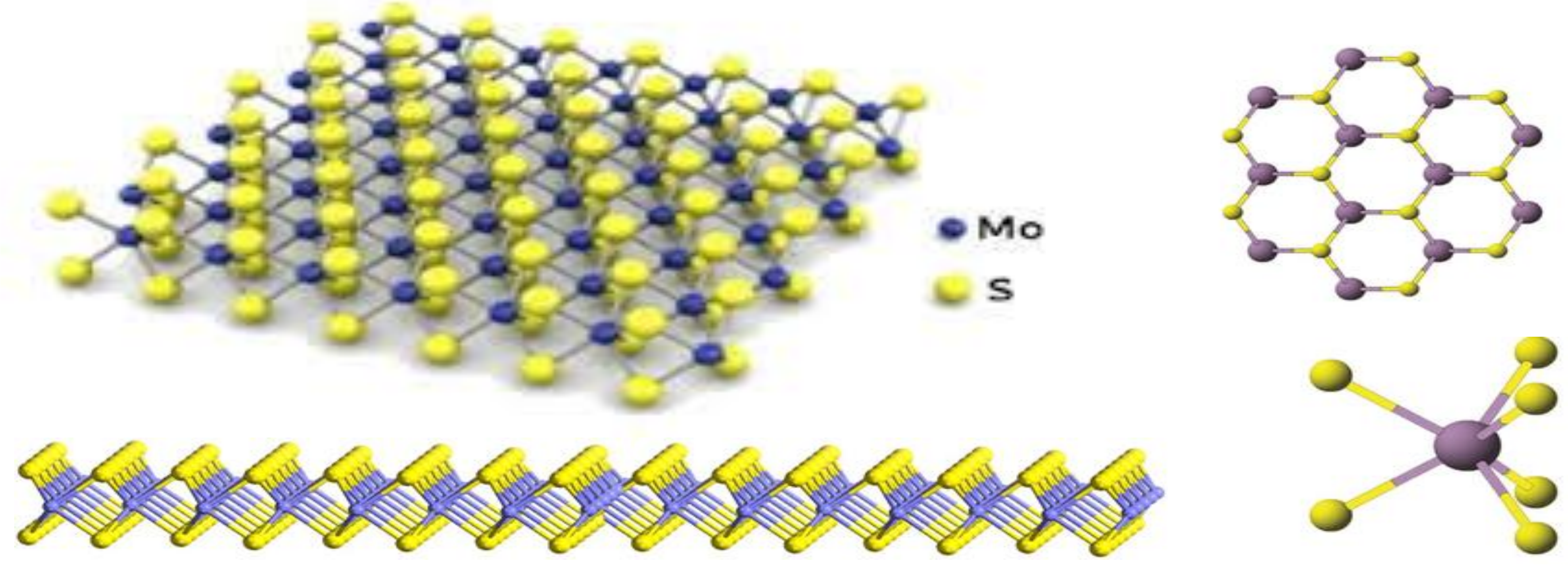
4. Polariton-polariton interaction and Gross-Pitaevskii equation

6. Summary

1. Introduction

-TMDs (transition metal dichalcogenides)

TMDs are layered materials with hexagonal crystal lattice that are now being produced in single-layer or few-layer form (MoS_2, $MoSe_2$, WS_2, WSe_2,…)

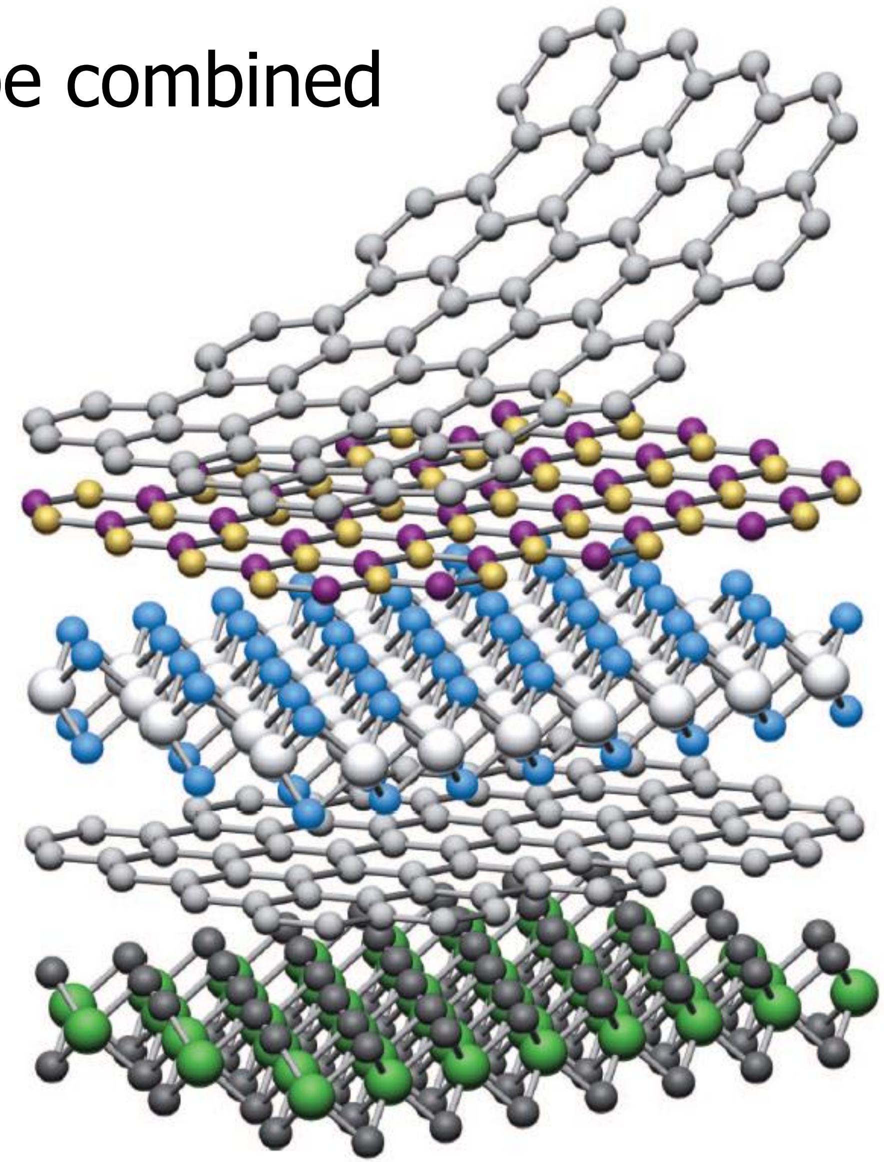


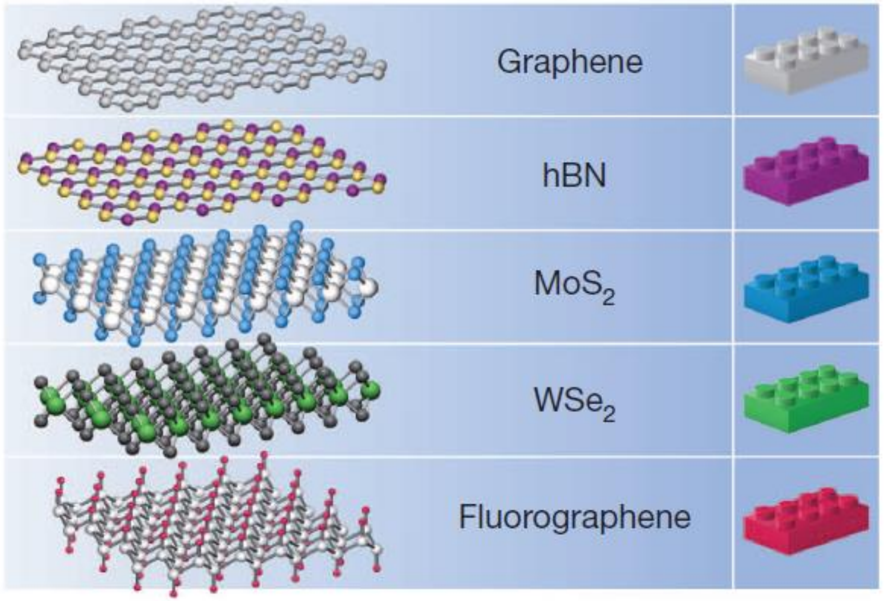
Different views of single-layer MoS_2 crystal lattice

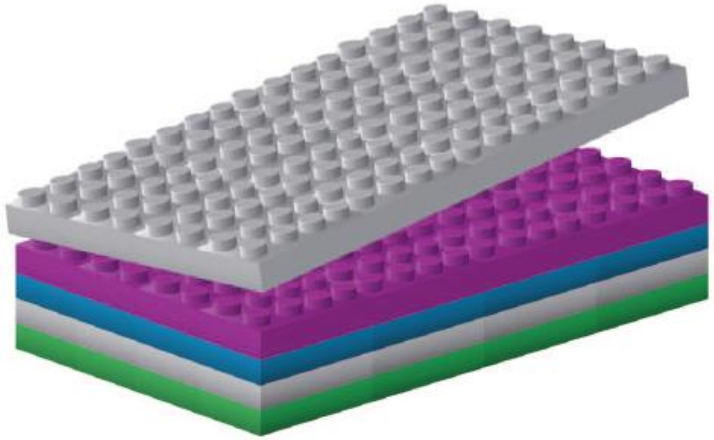
Combination of 2D materials

2D materials can be combined into van der Walls heterostructures.

No epitaxy required!



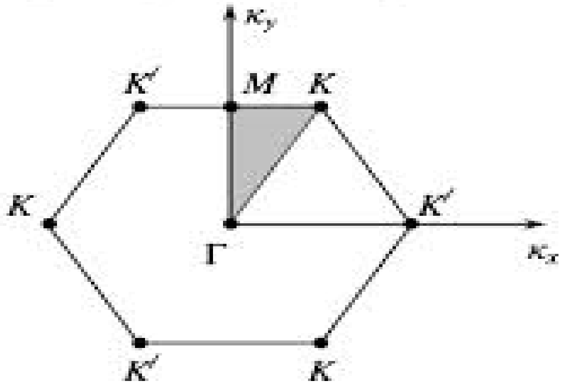




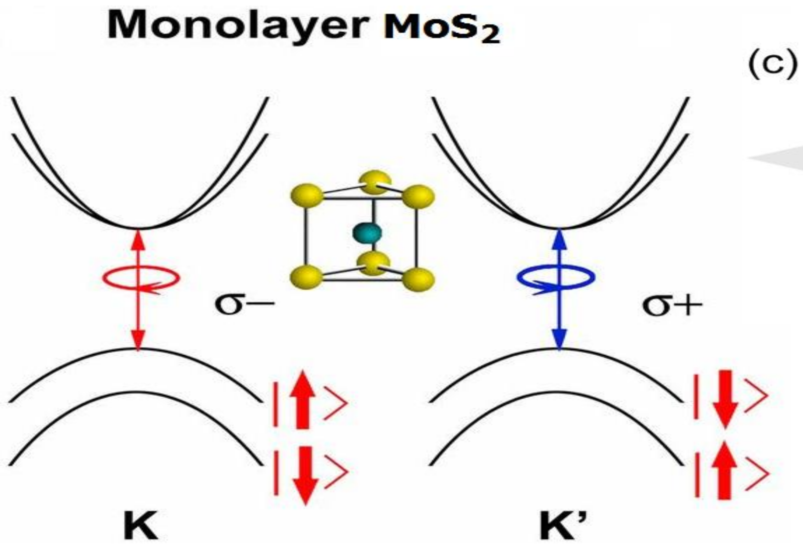
[A. Geim & I. Grigorieva, Nature, 499 (2013) 419 - 425]

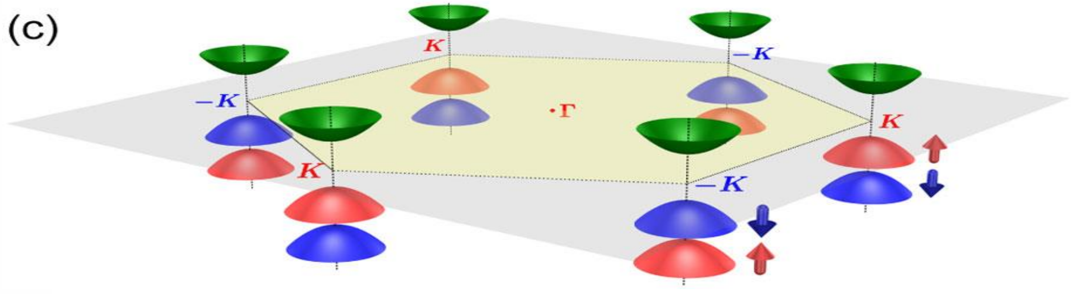
TMDs – 2D semiconductors (2DSCs)

In contrast with graphene (semimetal, E_g=0), TMDs are semiconductors (E_g>0).



1-st Brillouin zone





Schematic drawing of the band structure at the band edges

The Brillouin zone is the same as for graphene. Also, as in graphene, CB minima and VB maxima are located in K and K’ points at the border of the Brillouin zone.

<u>**Contrary to graphene, there is a band gap (of the order of 1.5 – 2.5 eV) between the CB and the VB. The spectrum near the minima (maxima) is parabolic, not linear. So, $m^* \neq 0$.**</u>

Because of the spin-orbit interaction, two valence bands corresponding to different spin orientations are split by some energy of the order of 0.5 eV.

[Di Xiao et al., Phys. Rev. Lett. 108 (2012) 196802]

-Excitons in TMD monolayers

J. Phys.: Condens. Matter **27** (2015) 315301

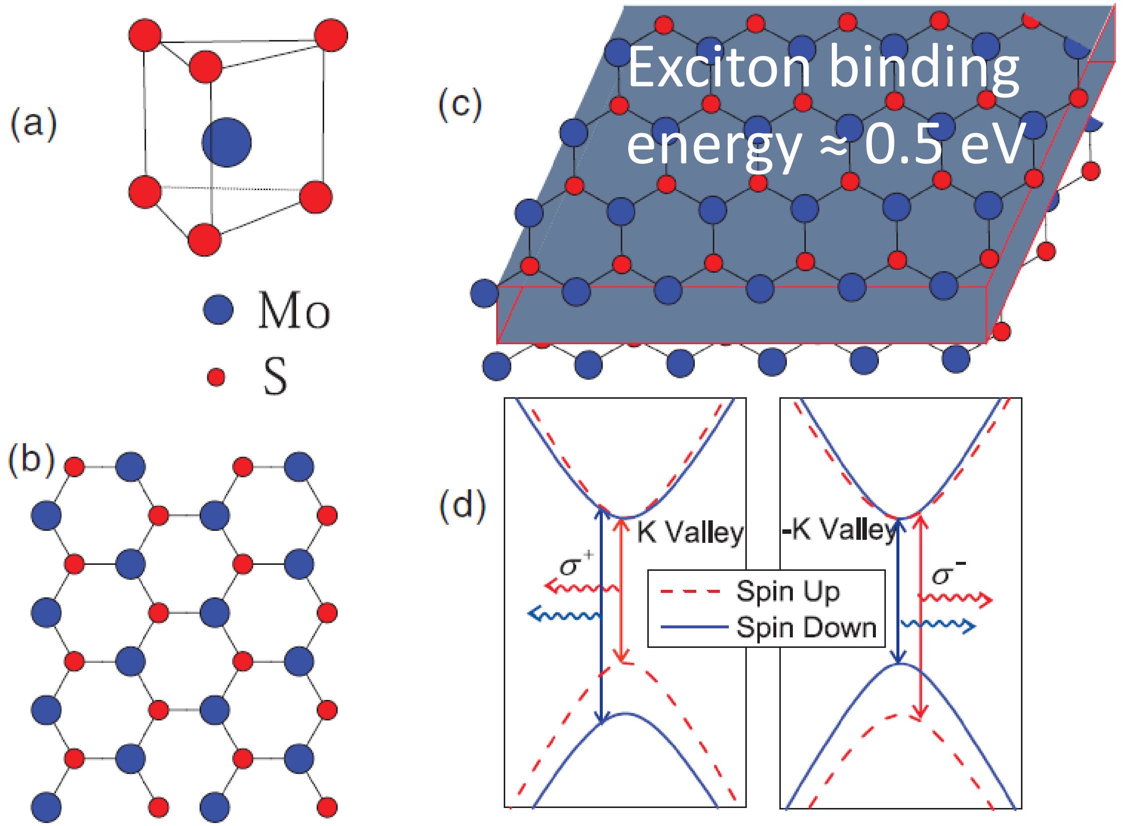


Figure 1. (a) The unit cell of monolayer MoS_2, (b) top view of the MoS_2 monolayer. (c) Schematic of two layers MoS_2 separated by a dielectric layer. (d) The band structure of monolayer MoS_2 at the band edges located at the K, $-K$ points.

PHYSICAL REVIEW B **89**, 205436 (2014)

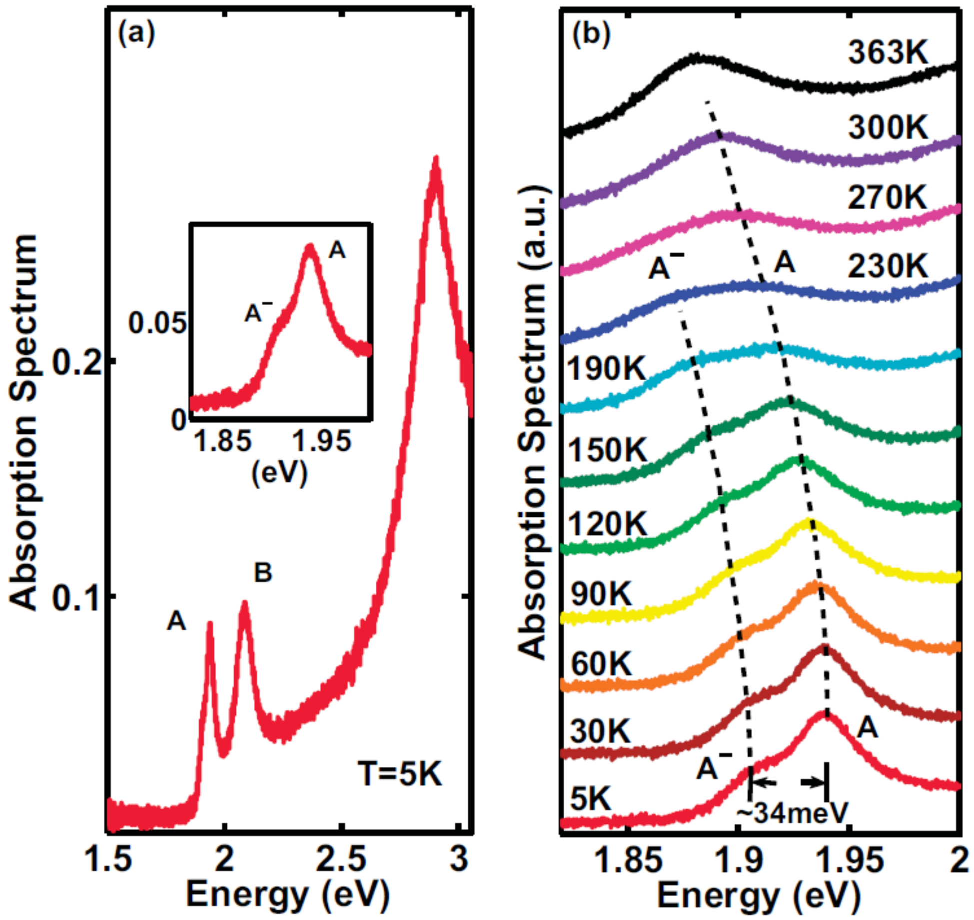


FIG. 2. (Color online) (a) Measured broadband absorption spectrum $A(\omega)$ of a MoS_2 monolayer at $T = 5$ K is plotted. Two main absorption resonances, A and B, attributed to excitons are visible together with a smaller A^- attributed to trions. The inset shows the A and A^- peaks in greater detail. (b) Measured absorption spectrum of a MoS_2 monolayer near the A and A^- peaks is plotted for different temperatures. The curves for different temperatures are given offsets

Optical conductivity due to excitons

$$\chi_{2D} = 2|\Psi_{ex}^{2D}(0)|^2 \langle |\mathbf{e}\cdot\mathbf{d}_{ex}|^2\rangle (E_{ex} - \hbar\omega - i\hbar\gamma)^{-1}$$

Envelope wavefunction (H-like model): $|\Psi_{ex}^{2D}(0)|^2 = 2/(\pi a_{ex}^2)$

Interband matrix element: $\mathbf{P}_{cv}(\mathbf{q}\approx 0) = m_0 v(\tau\mathbf{e}_x + i\,\mathbf{e}_y)$ $(\tau = \pm 1)$

$$\boxed{\sigma_{2D} = -i\omega\chi_{2D}}$$

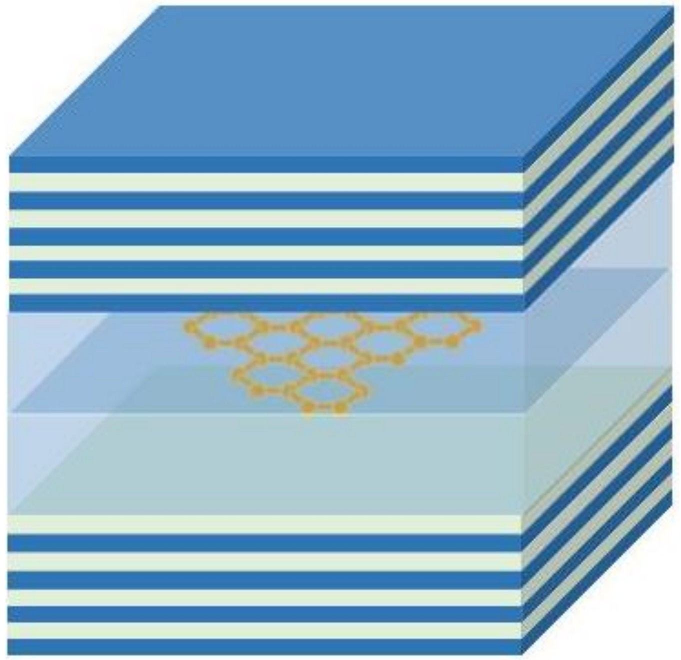
$$\boxed{= \frac{4e^2v^2}{\pi a_{ex}^2\omega}\sum_{A,B}\frac{-i}{E_{A,B} + \hbar^2q^2/(2m_{ex}) - \hbar\omega - i\hbar\gamma_{A,B}}}$$

MoS_2 : $E_A \approx 1.9\,\text{eV}$, $E_B \approx 2.1\,\text{eV}$, $a_{ex} \approx 0.8\,\text{nm}$, $v \approx 0.55\cdot 10^8\ \text{cm/s}$, $m_{ex} \approx m_0$

2. TMD layer inserted in a microcavity

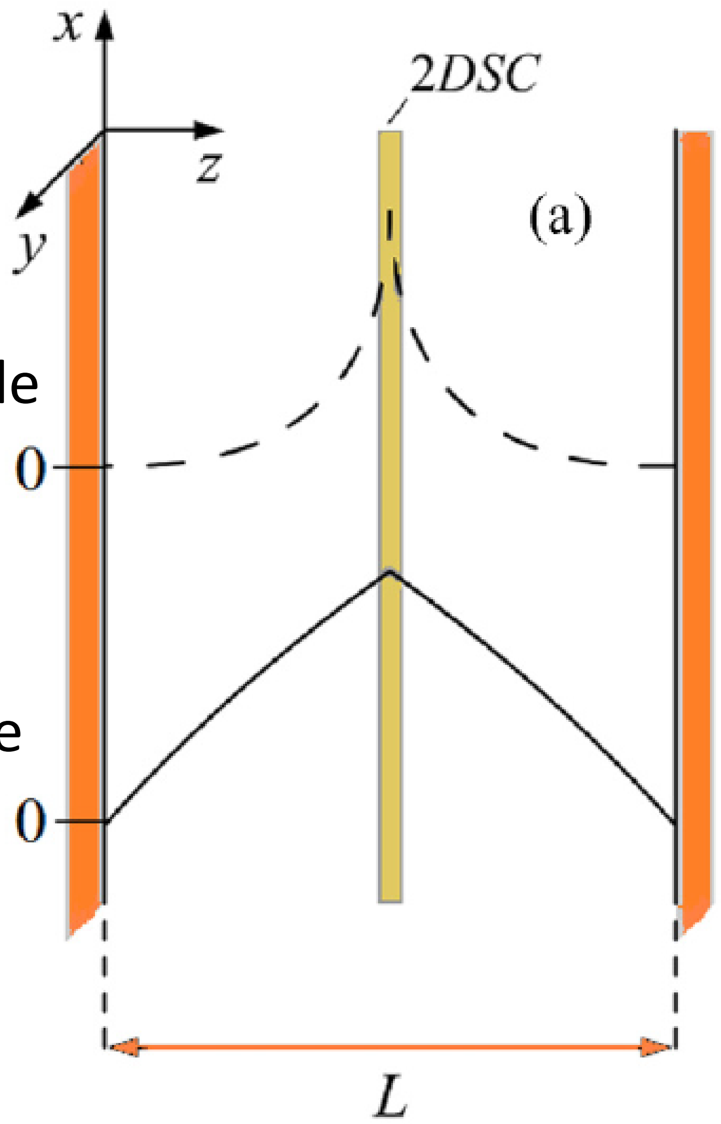
Monolithic cavity

with a TMD layer placed in the middle



Bragg mirrors
made of e.g.
SiO_2/TiO_2 SL

Model cavity



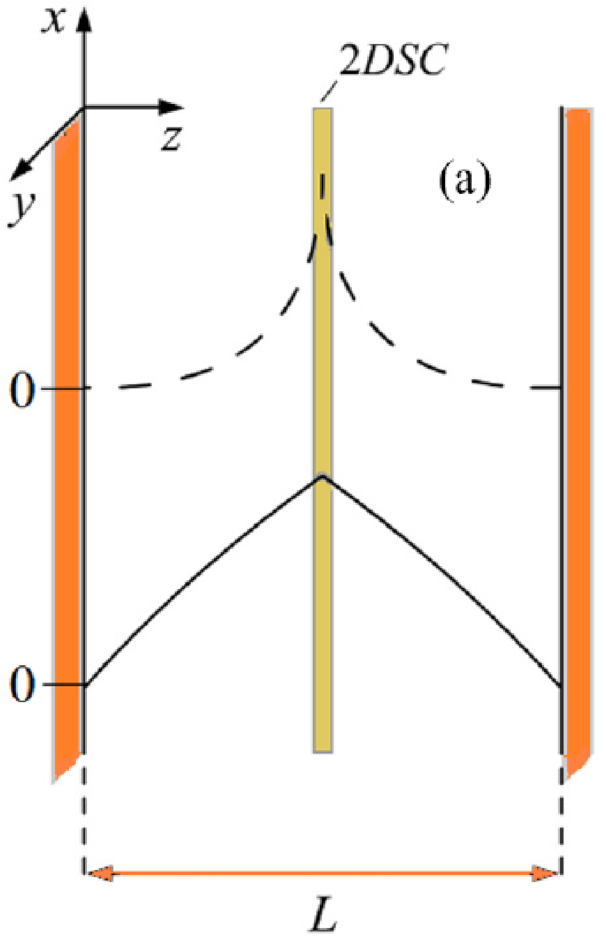
Surface mode

MC mode

[N. Lundt et al.,arXiv:1604.03916; L. Flatten et al.,arXiv:1605.04743]

TMD layer inserted in a microcavity

$$k_z = \sqrt{\varepsilon \frac{\omega^2}{c^2} - q^2}$$



TE waves

$$E_y = \sin(k_z z) e^{iqx}, \quad z \leqslant L/2,$$

$$E_y = \sin[k_z(L - z)] e^{iqx}, \quad z \geqslant L/2,$$

B. c. on TMD: $$H_x|_{z=\frac{L}{2}+0} - H_x|_{z=\frac{L}{2}-0} = \frac{4\pi\sigma_{2D}}{c} E_y.$$

TM waves

$$H_y = -\cos(k_z z) e^{iqx}, \quad z \leqslant L/2,$$

$$H_y = \cos[k_z(L - z)] e^{iqx}, \quad z \geqslant L/2.$$

TMD layer inserted in a microcavity

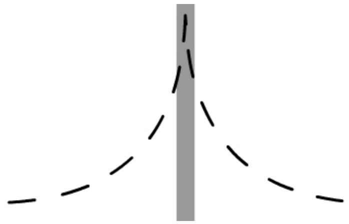
-Dispersion relations and curves

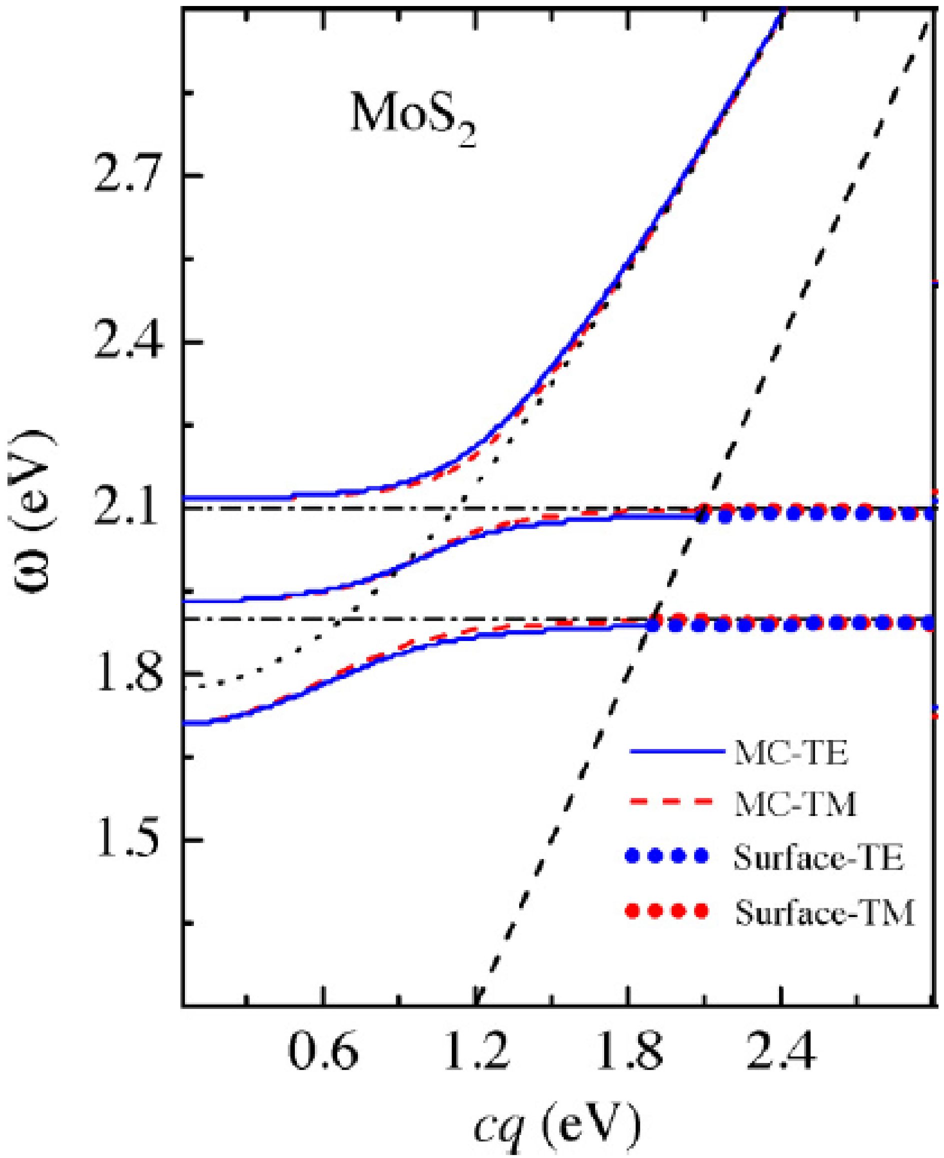
TE waves $$\tilde{k}_z \mathrm{Cot}\tilde{k}_z = \frac{\pi\omega L}{c}\tilde{\chi}_{2\mathrm{D}}(\omega,q),$$

TM waves $$\frac{\mathrm{Cot}\tilde{k}_z}{\tilde{k}_z} = \frac{4\pi c}{\varepsilon\omega L}\tilde{\chi}_{2\mathrm{D}}(\omega,q),$$

$$\tilde{\chi}_{2\mathrm{D}} = i\sigma_{2\mathrm{D}}/c,\ \tilde{k}_z = \tfrac{1}{2}k_z L.$$

Modes on the right of the light line are surface polaritons





PHYSICAL REVIEW B **92**, 245435 (2015)

TMD layer inserted in a microcavity

-Exciton-polariton lifetime

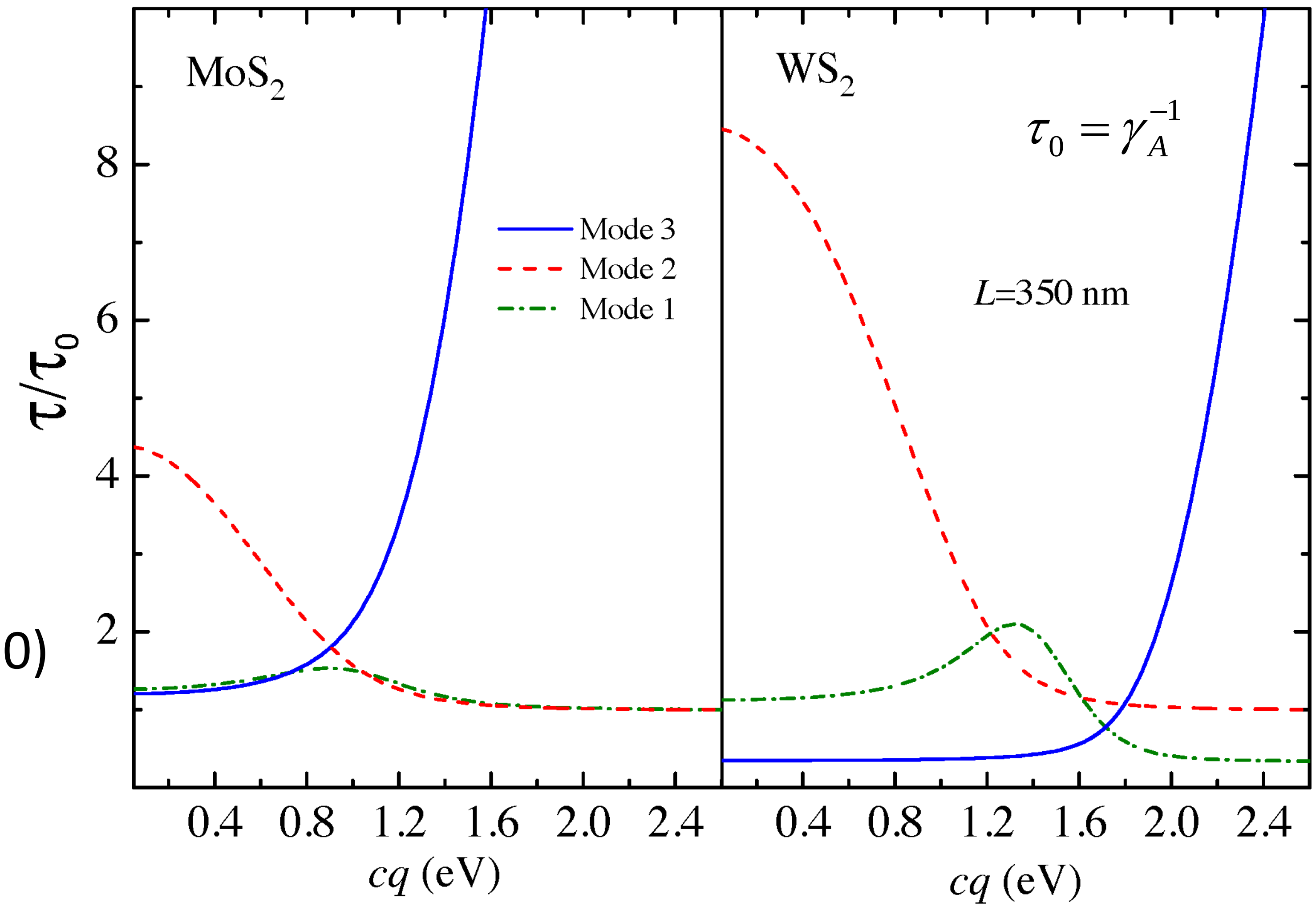
If the damping is included, exciton-polariton modes have complex frequencies (for real *q*),

$$\tau = -\left[\operatorname{Im}\omega_i(q)\right]^{-1}$$

For the lowest order mode (*q* -> 0)

$$\tau = \gamma_A^{-1}\left(1 + \frac{\pi L \omega_{LT}}{c}\right) ,$$

$$\omega_{LT} = \frac{4e^2v^2}{\pi a_{ex}^2 c E_A}$$



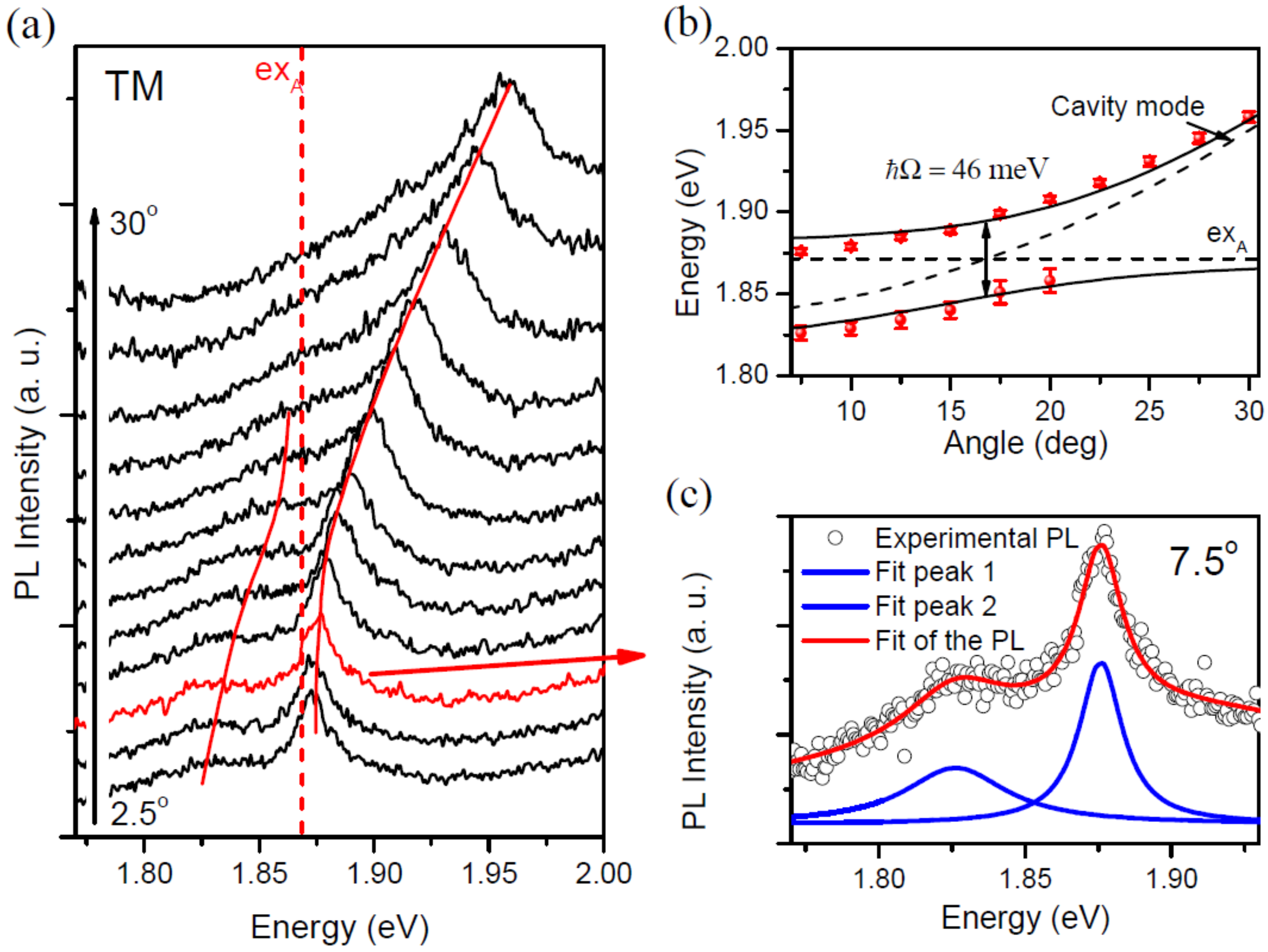
PHYSICAL REVIEW B **92**, 245435 (2015)

TMD layer inserted in a microcavity

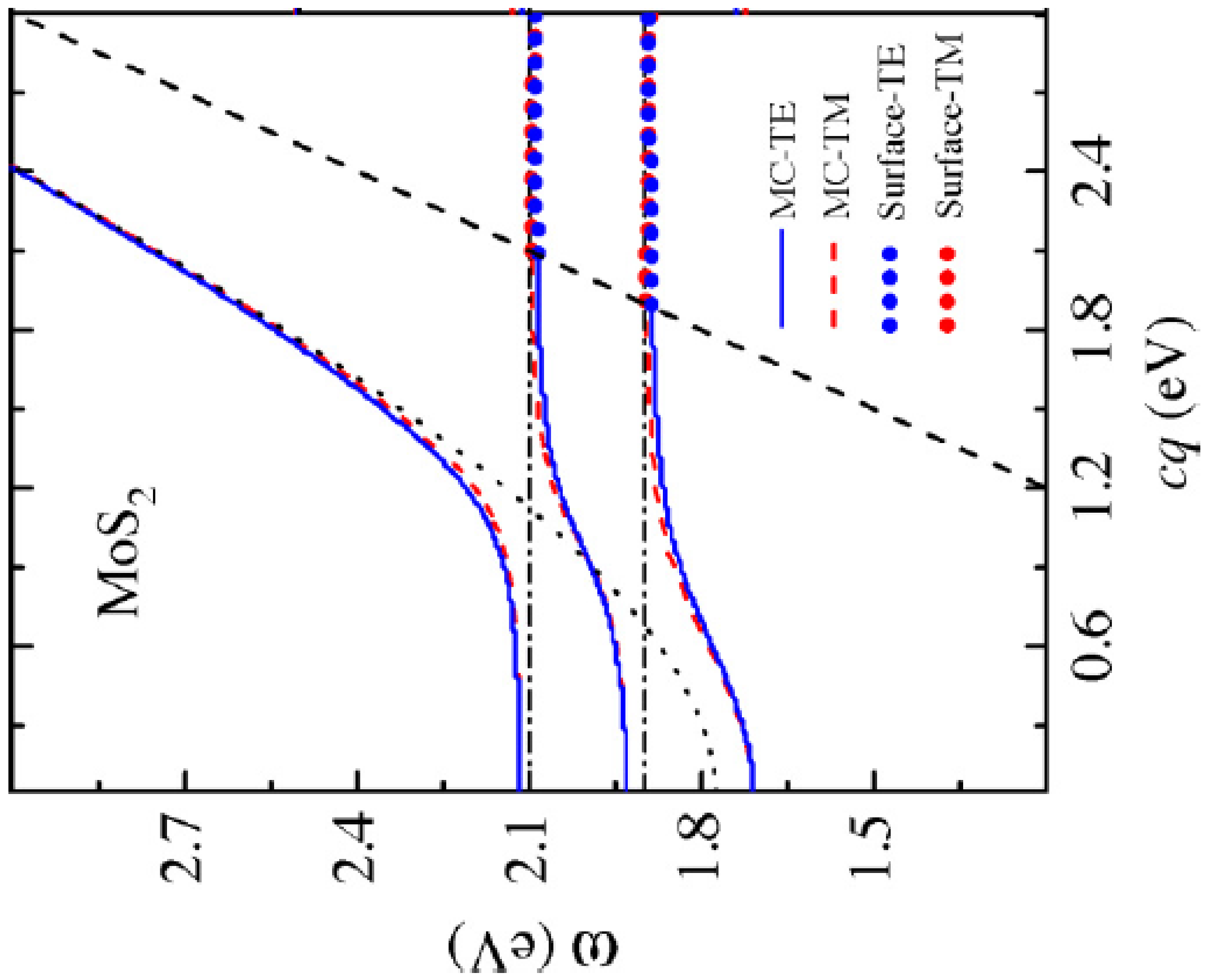
Experiment: angle-resolved PL spectra from a SiO_2/Si_3N_4 MC with embedded MoS_2 layer

Two modes are observed, which show the typical avoided crossing behavior.

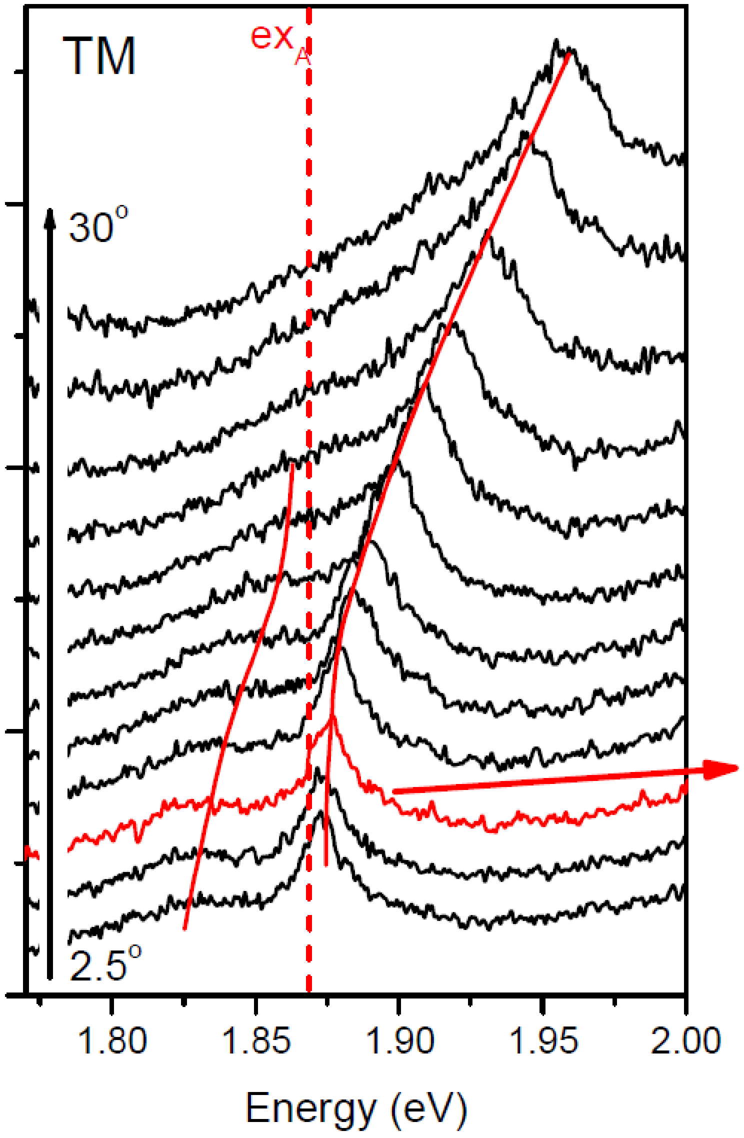
Rabi splitting ≈ 50 meV.



[X. Liu et al,, Nature Photonics 9, 30 (2015)]


MoS$_2$
MC-TE
MC-TM
Surface-TE
Surface-TM
2.7
2.4
2.1
1.8
1.5
ω (eV)
0.6
1.2
1.8
2.4
cq (eV)

(a)


TM
ex$_A$
30°
2.5°
PL Intensity (a. u.)
1.80
1.85
1.90
1.95
2.00
Energy (eV)

3- Two TMD layers inserted in a microcavity

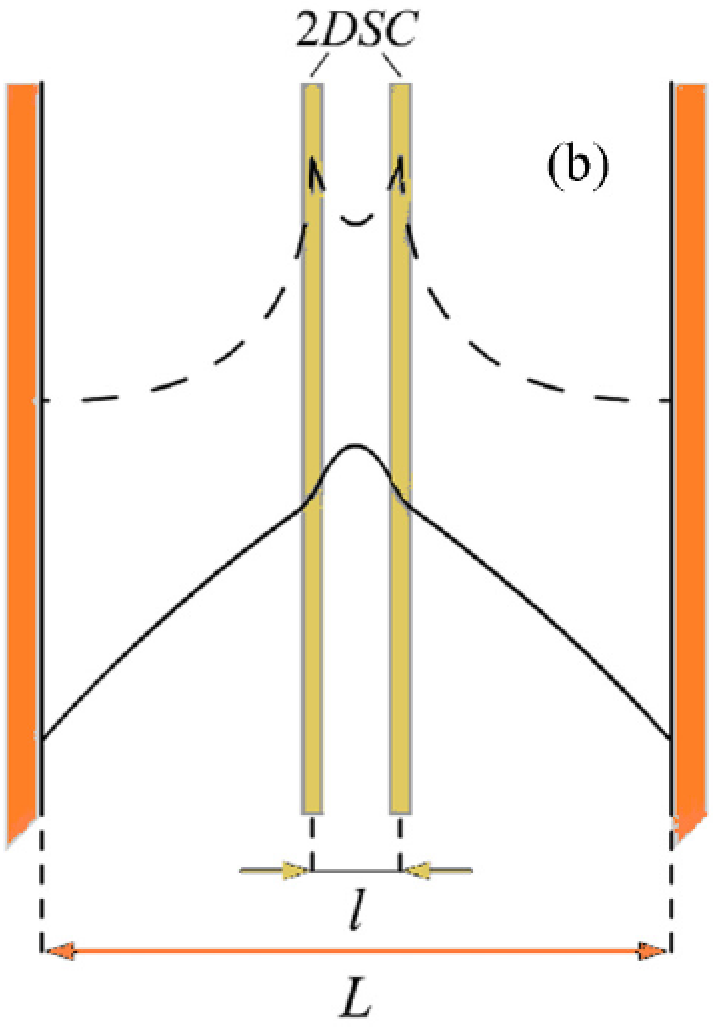
$$k_z^{(i)} = \sqrt{\varepsilon_i \frac{\omega^2}{c^2} - q^2}, \quad i = 1,\, 2$$

TE waves

$$E_y = \sin\left(k_z^{(1)} z\right) e^{iqx}, \quad 0 \leqslant z \leqslant z_1,$$

$$E_y = \begin{bmatrix} a\cos\left[k_z^{(2)}(z - L/2)\right] \\ b\sin\left[k_z^{(2)}(z - L/2)\right] \end{bmatrix} \times e^{iqx}, z_1 \leqslant z \leqslant z_2,$$

$$E_y = \pm\sin\left[k_z^{(1)}(L - z)\right] e^{iqx}, \quad z_2 \leqslant z \leqslant L,$$



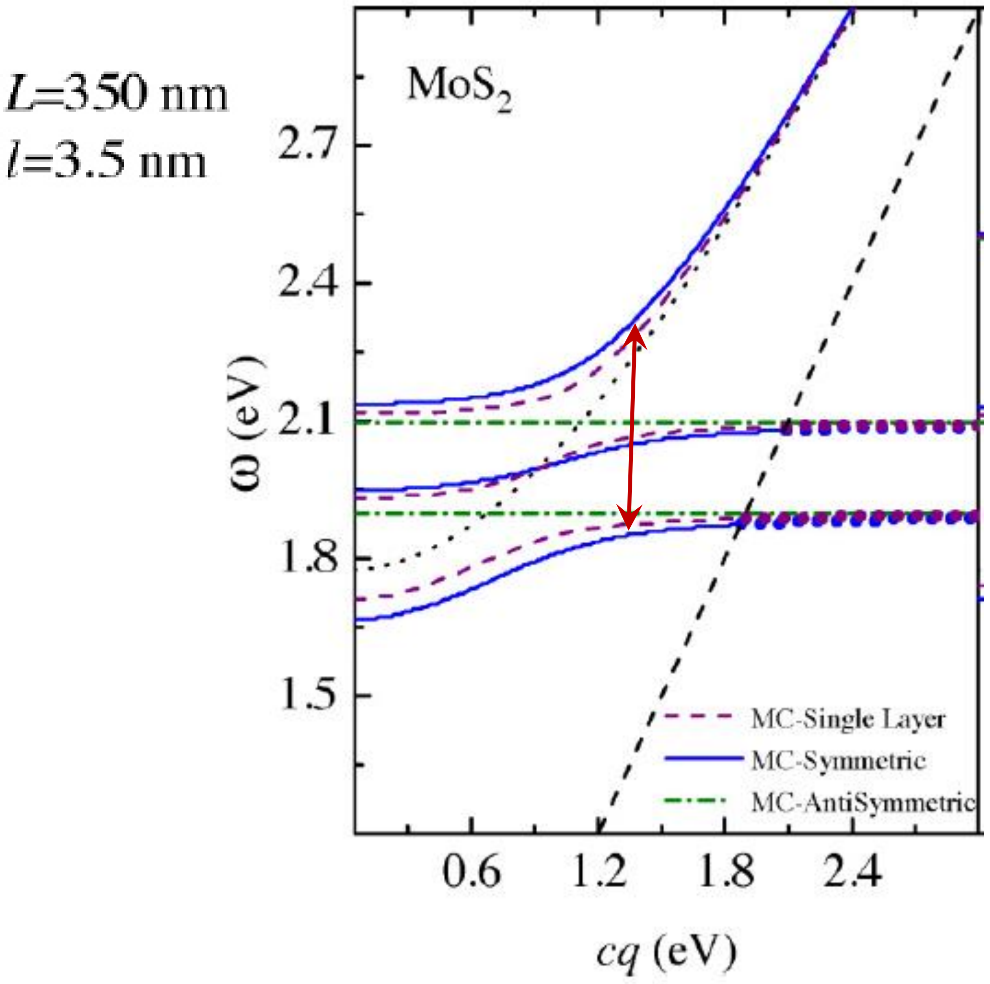
Dispersion relation (symmetric modes)

$$\frac{ck_z^{(1)}}{\omega} - \frac{ck_z^{(2)}}{\omega} \tan\left(k_z^{(1)} z_1\right) \tan\left(k_z^{(2)} l/2\right)$$

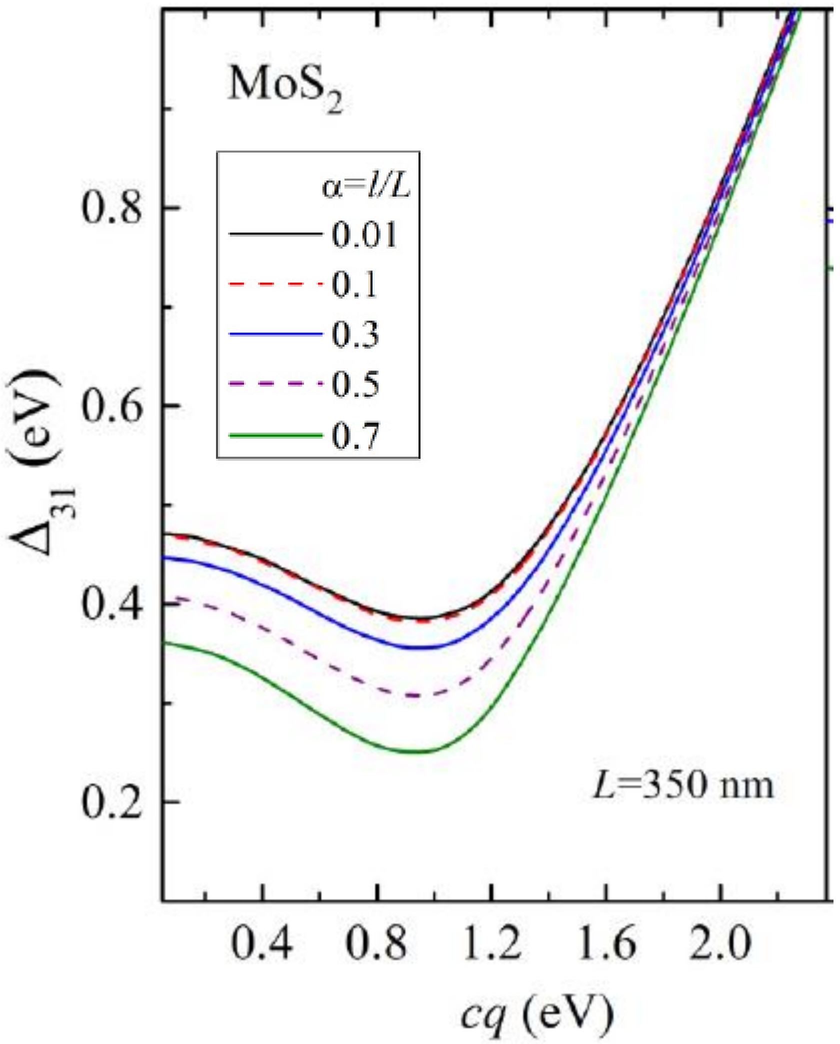
$$= \frac{4\pi i \sigma_{2\mathrm{D}}}{c} \tan\left(k_z^{(1)} z_1\right),$$

-Two TMD layers inserted in a microcavity

Dispersion curves

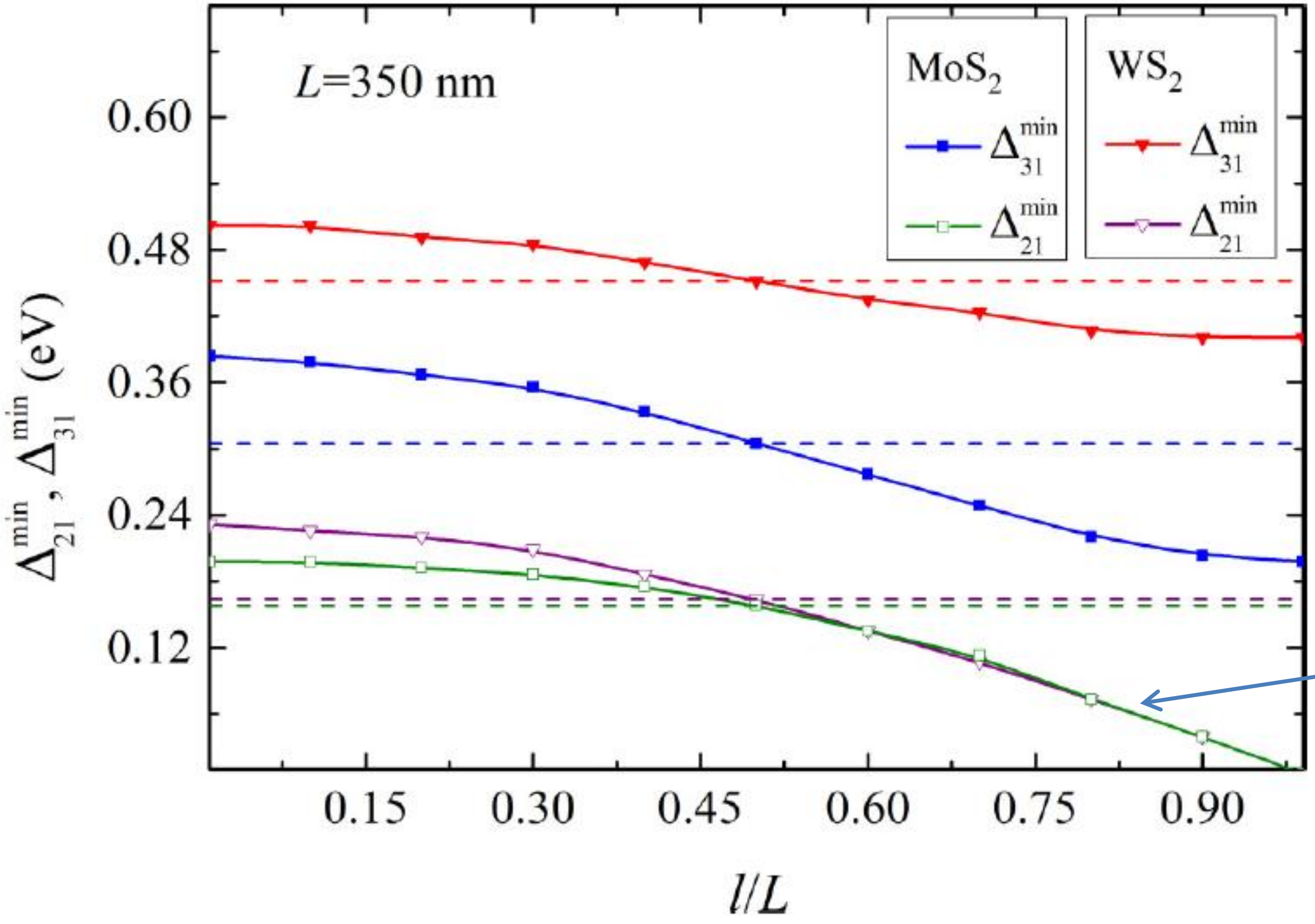


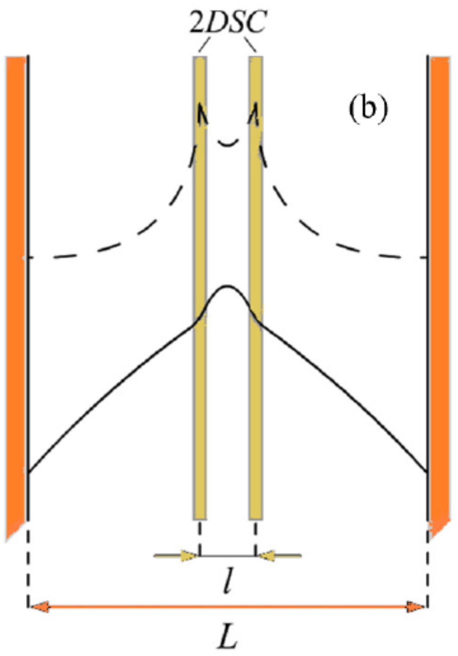
Overall splitting for different interlayer spacing values



Two TMD layers inserted in a microcavity

Calculted Rabi splitting dependence on inter-layer spacing



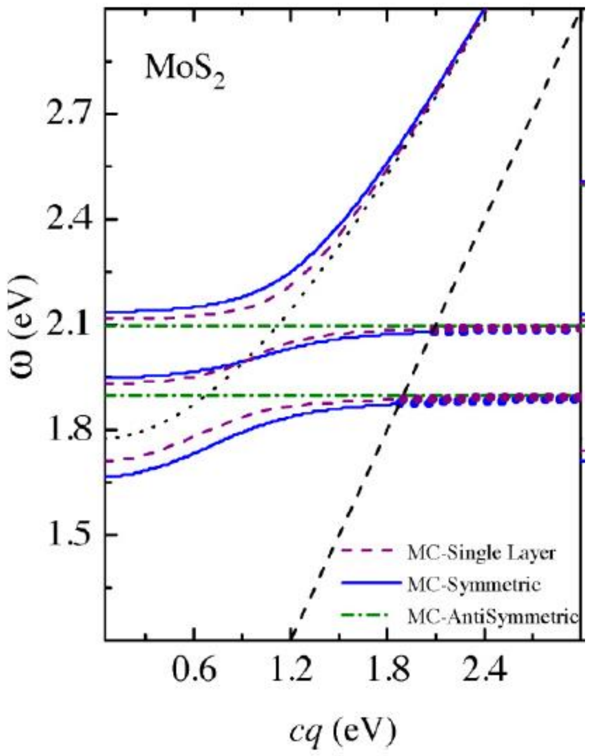


Rabi splitting for *A* exciton-polaritons neglecting *B* excitons

PHYSICAL REVIEW B **92**, 245435 (2015)

-Hopfield coefficients

$$H = \sum_{\mathbf{q}} \big[E_c(\mathbf{q}) P_{\mathbf{q}}^\dagger P_{\mathbf{q}} + E_A(\mathbf{q}) A_{\mathbf{q}}^\dagger A_{\mathbf{q}} + E_B(\mathbf{q}) B_{\mathbf{q}}^\dagger B_{\mathbf{q}} + g_{A\text{-}ph}(\mathbf{q}) P_{\mathbf{q}}^\dagger A_{\mathbf{q}} + g_{B\text{-}ph}(\mathbf{q}) P_{\mathbf{q}}^\dagger B_{\mathbf{q}} + \text{H.c.} \big],$$



Transformation to new bosonic operators

$$\alpha_{\mathbf{q}}^{(i)} = \kappa_{ph}^{(i)}(\mathbf{q}) P_{\mathbf{q}} + \kappa_B^{(i)}(\mathbf{q}) B_{\mathbf{q}} + \kappa_A^{(i)}(\mathbf{q}) A_{\mathbf{q}}, \quad i = 1,\, 2,\, 3,$$

The exciton-photon interaction parameters in the Hamiltonian can be expressed in terms of the (classically calculated) polariton dispersion curves:

$$g_{A\text{-}ph}^2(q) = \Delta_A^{(1)}(q) \Delta_A^{(2)}(q) \frac{\Delta_B^{(1)}(q)[E_c(q) - E_1(q)] - \Delta_B^{(2)}(q)[E_c(q) - E_2(q)]}{\Delta_B^{(1)}(q)\Delta_A^{(2)}(q) - \Delta_B^{(2)}(q)\Delta_A^{(1)}(q)}$$

$$\Delta_{A,B}^{(i)}(q) = E_{A,B} - E_i$$

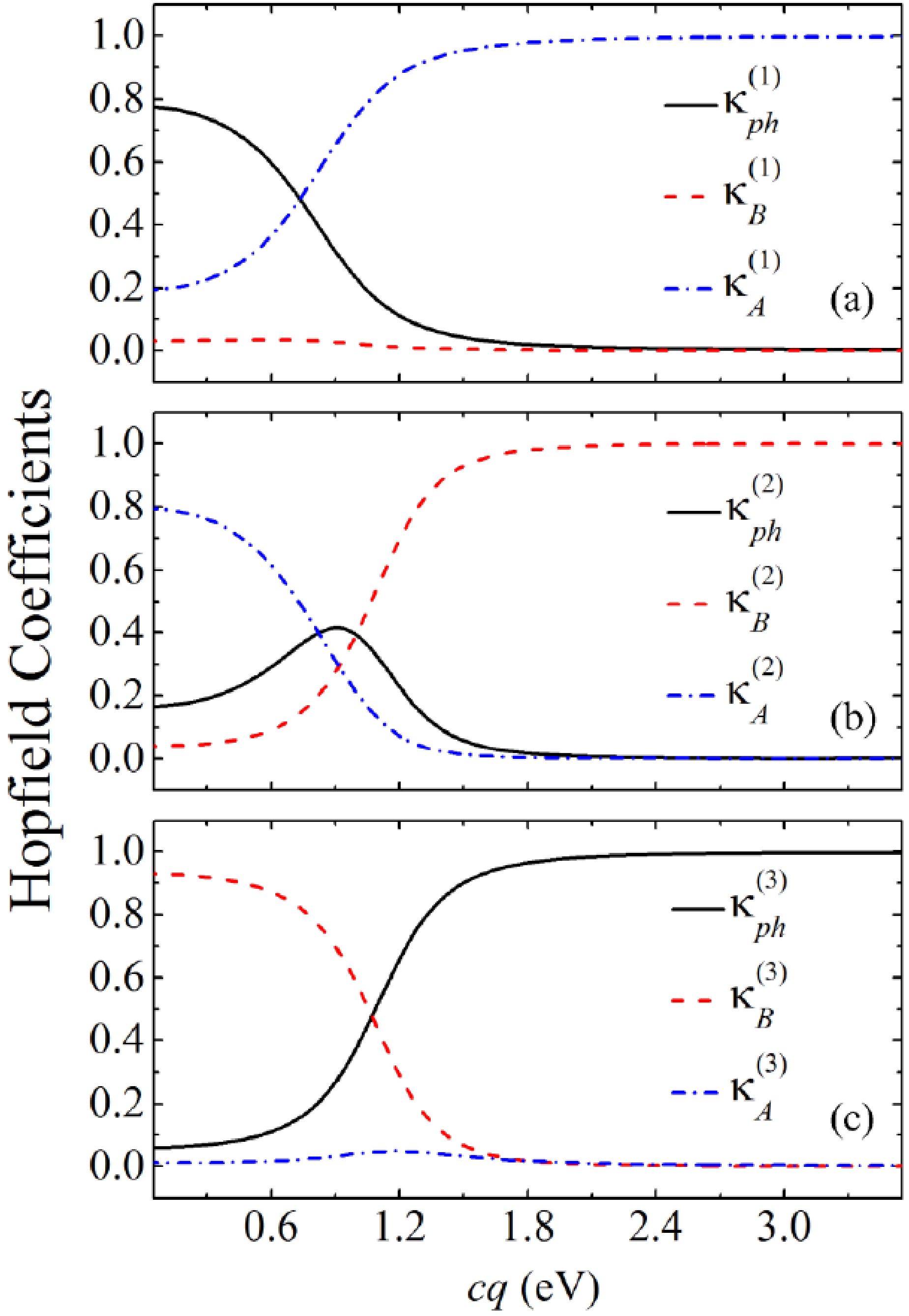
-Hopfield coefficients

$$(\kappa_{ph}^{(i)}(\mathbf{q}))^2 + (\kappa_B^{(i)}(\mathbf{q}))^2 + (\kappa_A^{(i)}(\mathbf{q}))^2 = 1$$

$$\kappa_{ph}^{(i)}(q) = \frac{\Delta_A^{(i)}(q)\Delta_B^{(i)}(q)}{\sqrt{\Delta_A^{(i)2}(q)\Delta_B^{(i)2}(q) + \Delta_B^{(i)2}(q)g_{A-ph}^2(q) + \Delta_A^{(i)2}(q)g_{B-ph}^2(q)}};$$

$$\kappa_A^{(i)}(q) = -\frac{\Delta_B^{(i)}(q)g_{A-ph}(q)}{\sqrt{\Delta_A^{(i)2}(q)\Delta_B^{(i)2}(q) + \Delta_B^{(i)2}(q)g_{A-ph}^2(q) + \Delta_A^{(i)2}(q)g_{B-ph}^2(q)}};$$

$$\kappa_B^{(i)}(q) = -\frac{\Delta_A^{(i)}(q)g_{B-ph}(q)}{\sqrt{\Delta_A^{(i)2}(q)\Delta_B^{(i)2}(q) + \Delta_B^{(i)2}(q)g_{A-ph}^2(q) + \Delta_A^{2(i)}(q)g_{B-ph}^2(q)}}$$

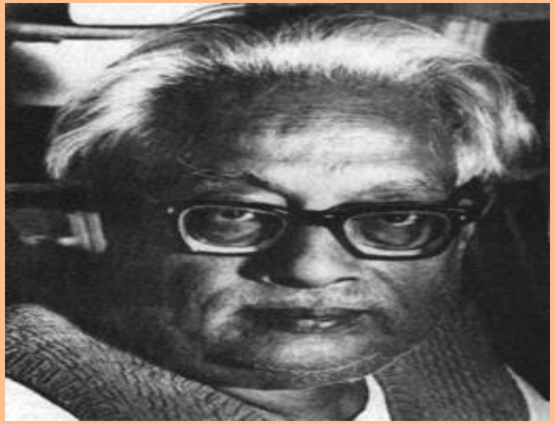


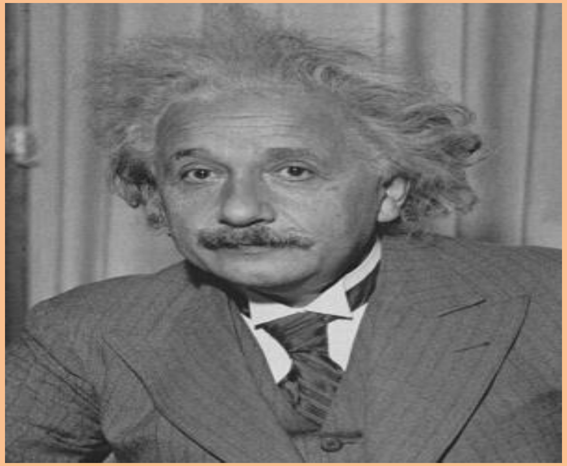
Bose-Einstein Condensation

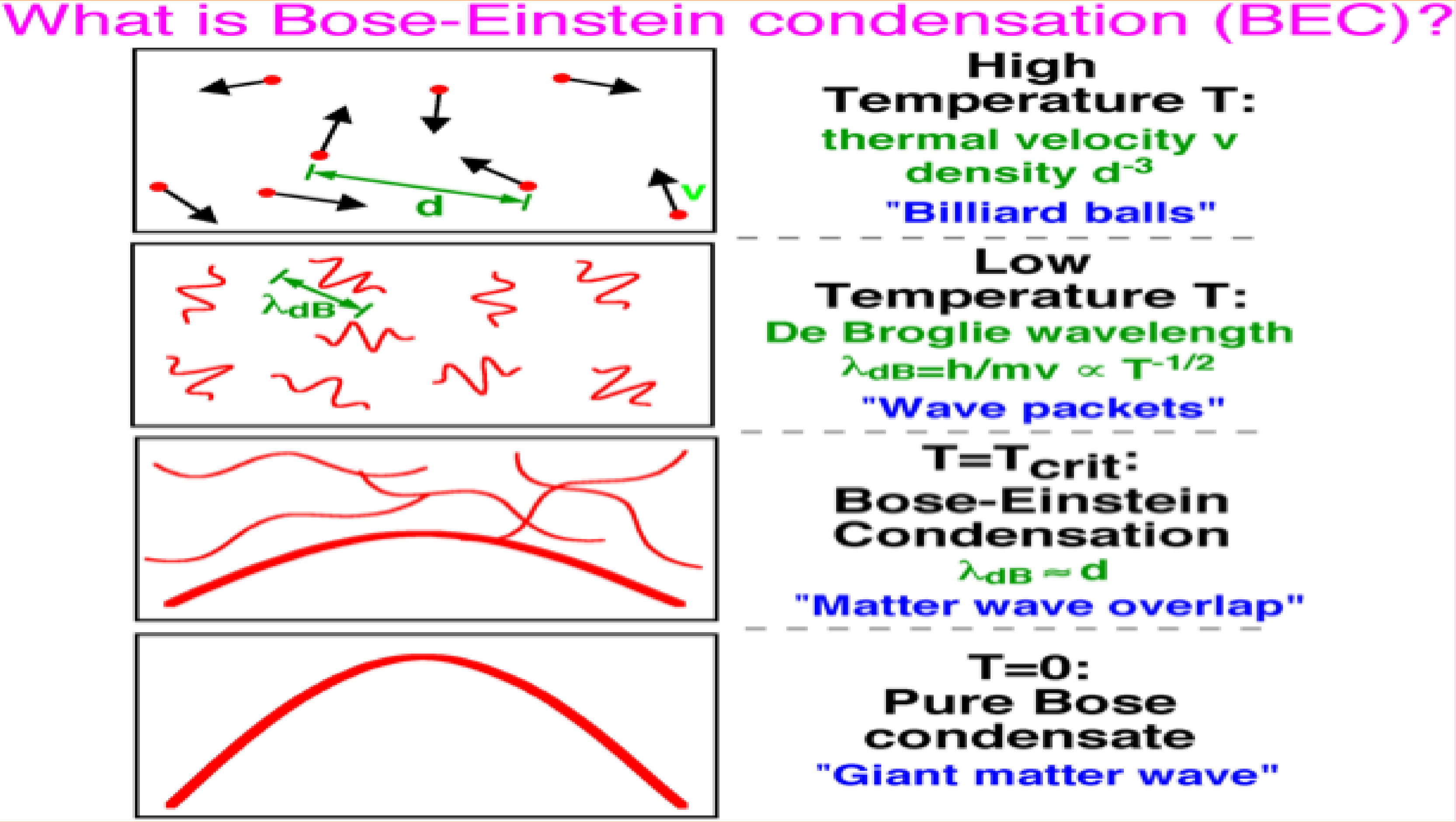
Satyendra Nath Bose y Albert Einstein

Boson: Statistics

A. Einstein, Sitzungsber. K. Preuss. Akad. Wiss. Phys. Math., 261, (1924).







Gross-Pitaievskii equation

$$-\frac{\hbar^2}{2m}\frac{d^2\Phi}{dx^2}+\frac{1}{2}m\omega^2x^2\Phi+\lambda\left|\Phi\right|^2\Phi=\mu\Phi$$

μ-the chemical potential
ω-trap frequency
m-the alkaline mass
λ-self-interaction parameter

L.K. Pitaevskii, Sov. Phys. JETP, 13, (1961), 451

- Bloch oscillations

Phys. Rev. Lett. 82, 2022 (1999)

- Superfluidity

- Dispersion and effective mass

Phys. Rev. Lett. 86, 4447 (2001)

- Josephson physics in optical lattices

- Mott-insulator transition

-Exciton-polariton condensates

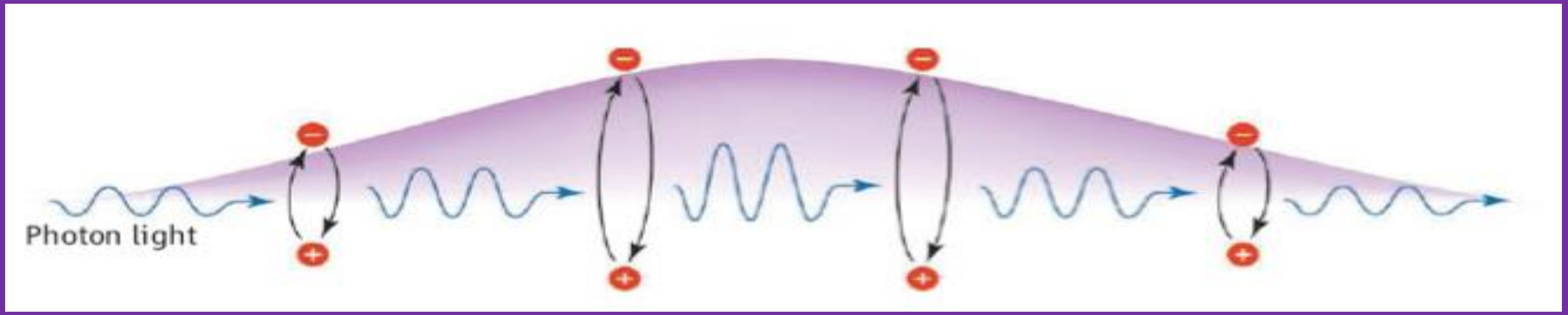
De Broglie wavelength

$\lambda_{dB} = h/mv \propto T^{-1/2}$

Atoms $T_c \sim 10^{-9} K$

polaritons-----m is 0.0001 electron mass

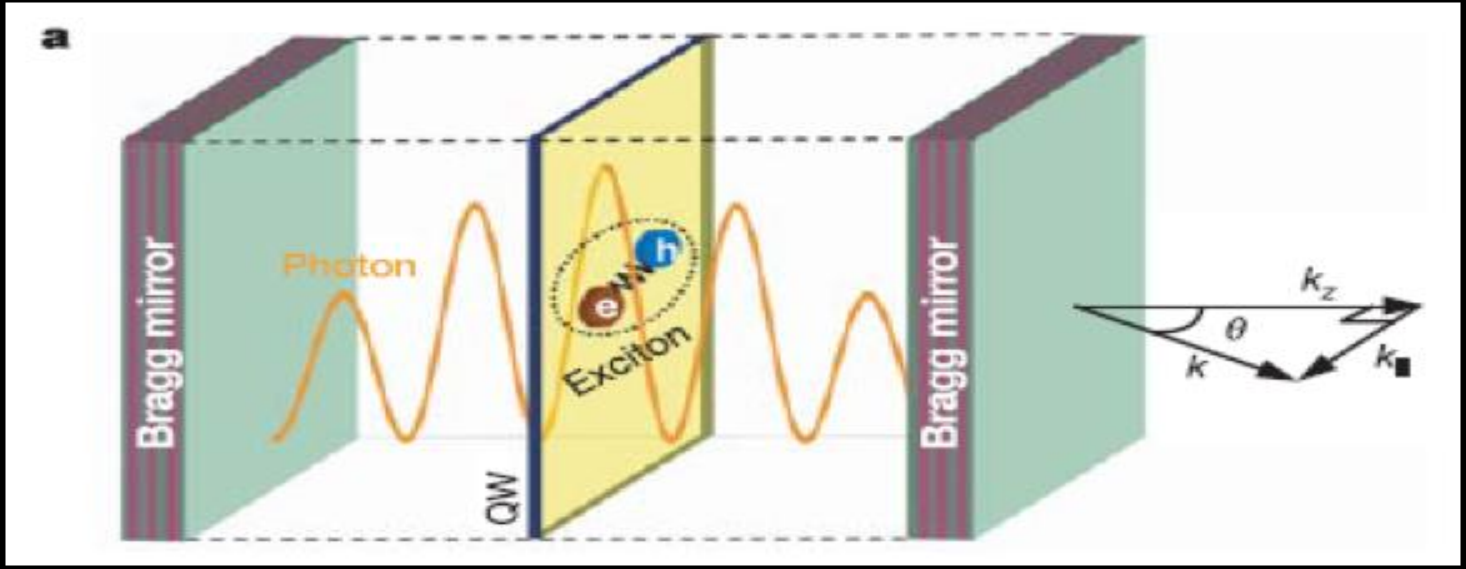
$T_c \sim 300 K$

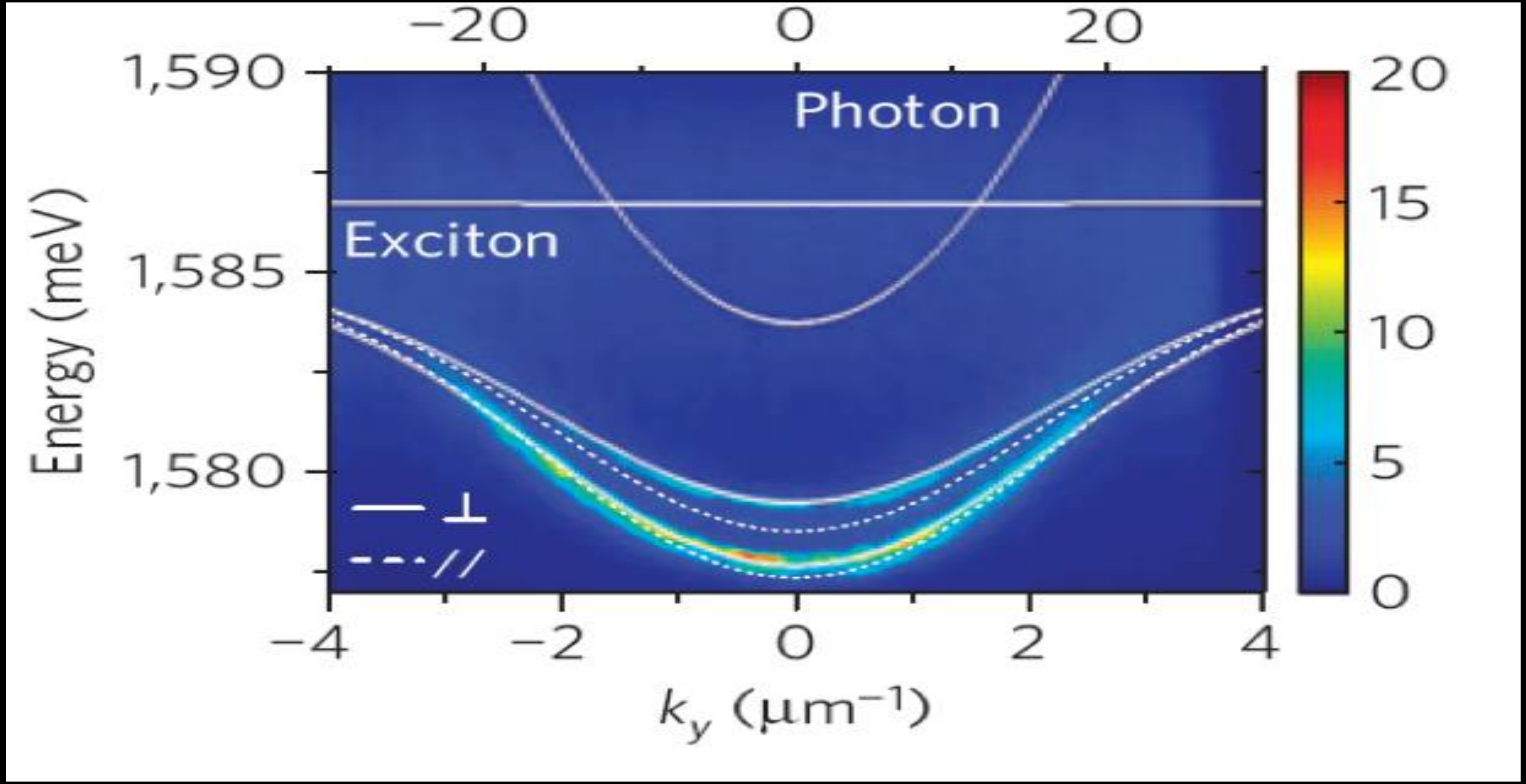


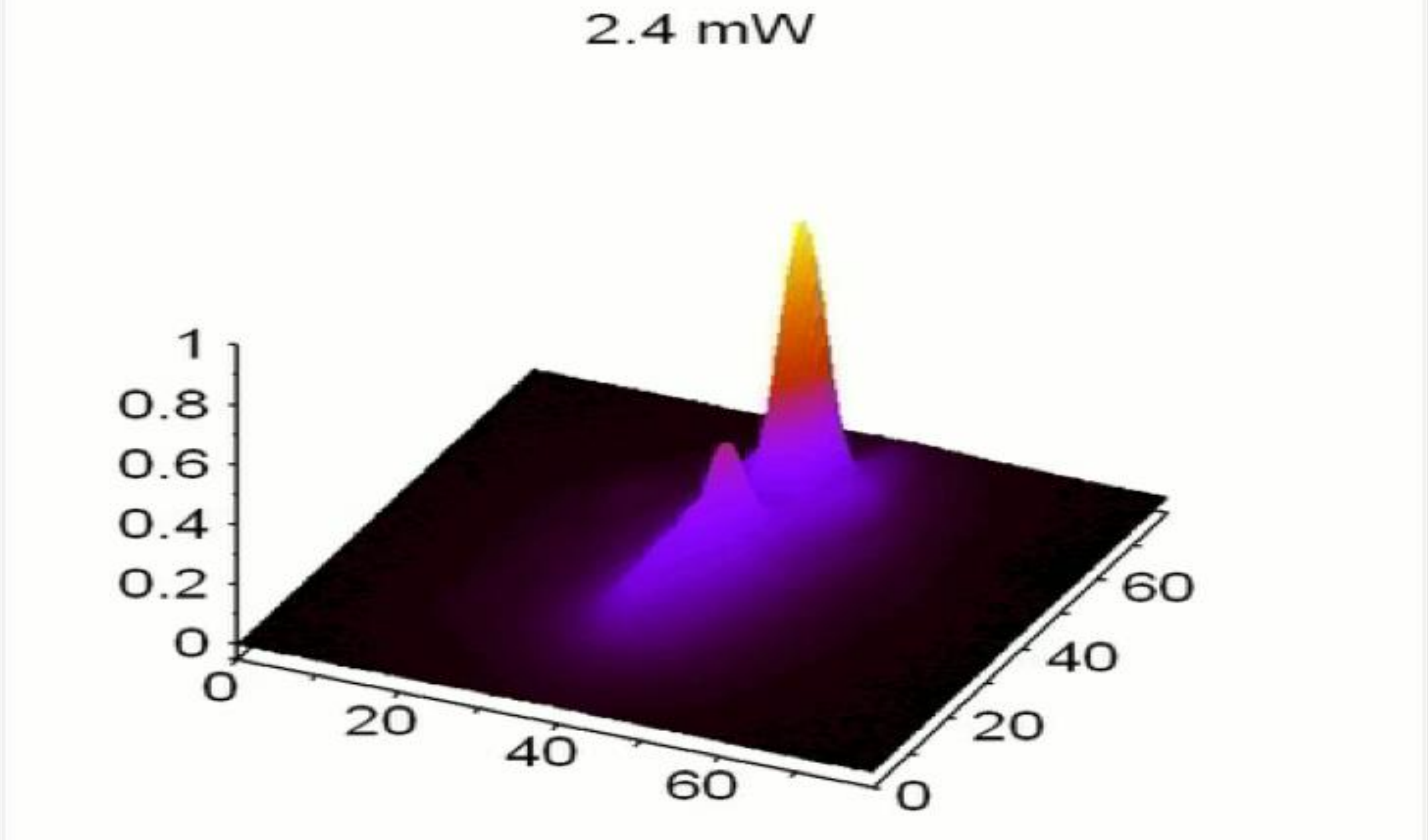
Science V. 316

2.- The excitons and photons interaction form a new quantum state= polariton.

1.- Photons from a laser create electron-hole pairs or excitons.


a
Bragg mirror
Photon
Exciton
QW
Bragg mirror
k_z
θ
k
$k_\parallel$

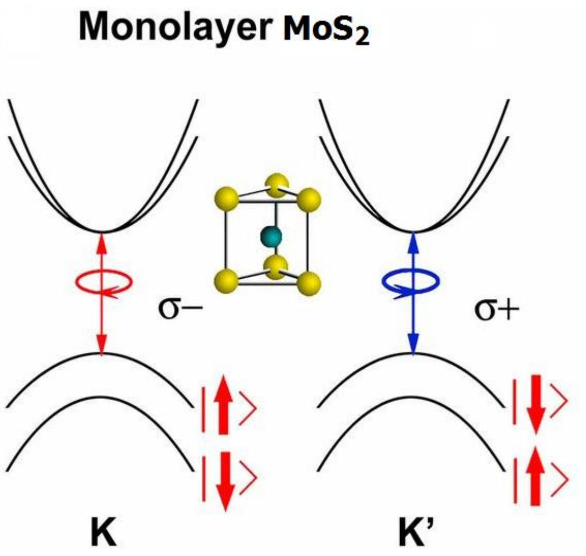

−20
0
20
1,590
1,585
1,580
Energy (meV)
Photon
Exciton
⊥
//
20
15
10
5
0
−4
−2
0
2
4
k_y (μm^{-1})


2.4 mW
1
0.8
0.6
0.4
0.2
0
0
20
40
60
0
20
40
60

-Non-linear regime

Hopfield coefficients determine the **polariton-polariton interaction**:

$\Lambda = \Lambda_{11} + \Lambda_{12}$ Λ_{11} and Λ_{12} are the interaction parameters, describing the interaction between polaritons having parallel and antiparallel spins.



I. Rought approximatio $V(\mathbf{r}-\mathbf{r}') = |X|^4 \int d\mathbf{q} \tilde{V}^{ex-ex}(\mathbf{q}) e^{i\mathbf{q}(\mathbf{r}-\mathbf{r}')}$

where $X \equiv \kappa_{ex}^{(i)}(0)$ If $\tilde{V}^{ex-ex}(\mathbf{q}) \approx const$ for $qa_{ex} \ll 1$,

$V(\mathbf{r}-\mathbf{r}') = \Lambda\delta(\mathbf{r}-\mathbf{r}')$, $\Lambda_{11} = 6R_a a_{ex}^2 |X|^4$

For linear polarization we have the contribut

II. Scattering approximation: $\Lambda_{11} = 6R_a a_{ex}^2 |X|^2$ Phys. Rev. B, **58**, 7926 (1998).

-Non-linear regime

$$\mathcal{H} = \int d\mathbf{r}\hat{\Psi}^\dagger(\mathbf{r})\left[-\frac{\hbar^2}{2m_p}\nabla^2_{\mathbf{r}} + V_c\right]\Psi(\mathbf{r}) + \frac{1}{2}\int d\mathbf{r}d\mathbf{r}'\hat{\Psi}^\dagger(\mathbf{r})\hat{\Psi}^\dagger(\mathbf{r}')V(\mathbf{r}-\mathbf{r}')\hat{\Psi}(\mathbf{r})\hat{\Psi}(\mathbf{r}')\ ,$$

-Hopfield coefficients

Describes two-particle interactions

$$\frac{1}{(2\pi)^4}\int d\mathbf{r}''d\mathbf{r}'''\int d\mathbf{k}_1 d\mathbf{k}_2 d\mathbf{q}\tilde{V}^{ex-ex}(\mathbf{q})\kappa^{(i)}_{ex}(\mathbf{k}_1)^*\kappa^{(i)}_{ex}(\mathbf{k}_2)^*\kappa^{(i)}_{ex}(\mathbf{k}_1-\mathbf{q})\kappa^{(i)}_{ex}(\mathbf{k}_2+\mathbf{q}) \times \exp\{i\left[\mathbf{q}(\mathbf{r}''-\mathbf{r}''')+\mathbf{k}_1(\mathbf{r}-\mathbf{r}'')+\mathbf{k}_2(\mathbf{r}'-\mathbf{r}''')\right]\}\ ,$$

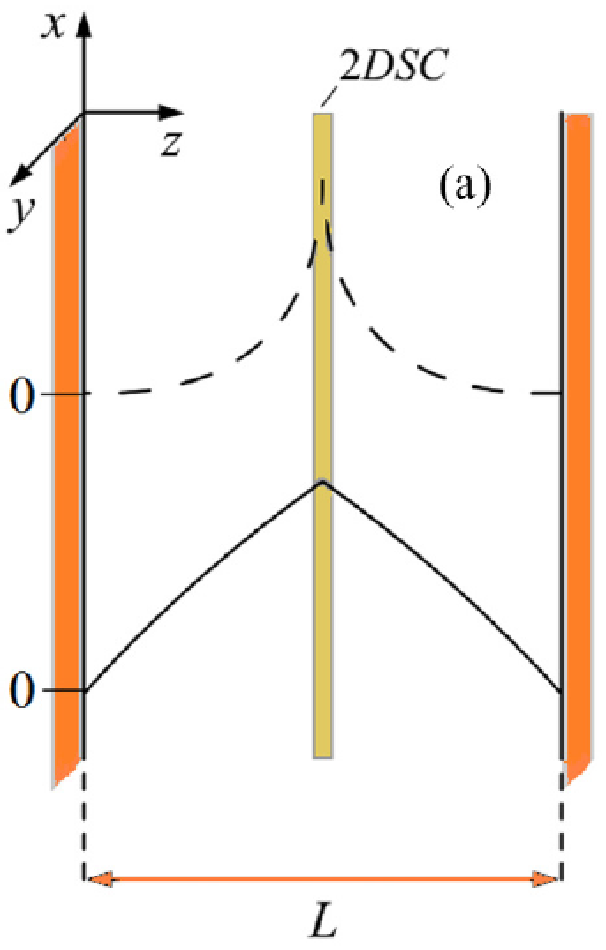
Fourier transform of the exciton-exciton interaction potential

Gross-Pitaevskii equation:

$$\boxed{-\frac{\hbar^2}{2m}\frac{d^2\Phi}{dx^2}+\frac{1}{2}m\omega^2x^2\Phi+\lambda\left|\Phi\right|^2\Phi=\mu\Phi}$$

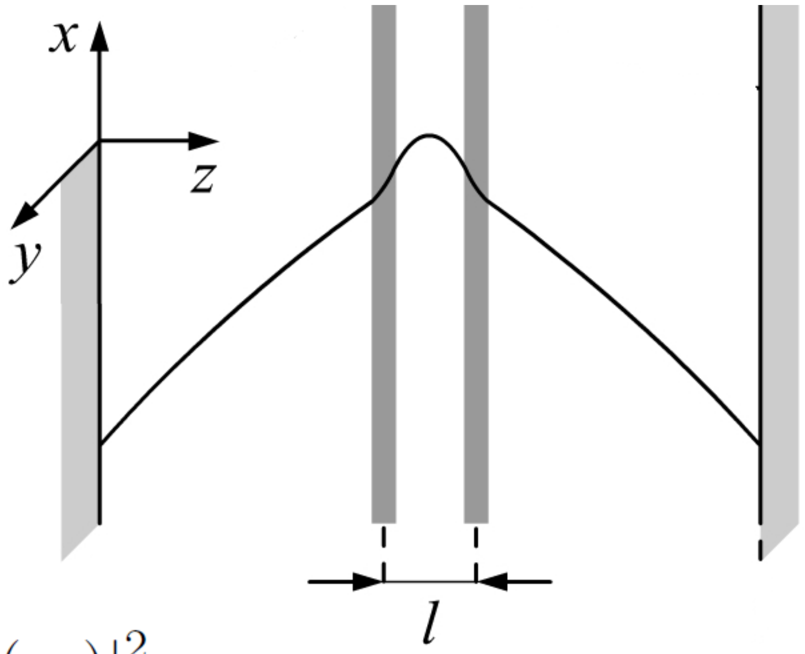
$\Lambda = \Lambda_{11} + \Lambda_{12}$

Λ proportional to the **Hopfield coefficients**



Non-linear regime

For the system with **two TMD layers**, two BECs can be created involving spatially separated excitons (1, 2= two layers). They are coupled via dipole-dipole interaction.



$$V_{12}^{ex-ex}(\mathbf{R}) = \frac{\mu_1 \cdot \mu_2 - 3(\mu_1 \cdot \mathbf{e_R})(\mu_2 \cdot \mathbf{e_R})}{\varepsilon R^3}$$

$$U_{12} = \frac{1}{2}|X|^4 \int d\mathbf{r_1} d\mathbf{r_2} |\Phi_1(\mathbf{r_1})|^2 V_{12}^{ex-ex}(\mathbf{r_1} - \mathbf{r_2} - l\mathbf{e}_z)|\Phi_2(\mathbf{r_2})|^2 ,$$

For a TE wave polarized along y

$$\mu_{1x} = \mu_{2x} = 0 ;$$

$$\mu_{1y} = \mu_{2y} = \alpha E_y^0|_{z=\frac{L}{2}\pm\frac{l}{2}}$$

The inter-condensate interaction term in the Gross-Pitaevskii equation:

$$\Lambda_{12} = \frac{4\pi}{15l}|X|^4 \left(\alpha E_y^0|_{z=L/2\pm l/2}\right)^2$$

It can be tuned by adjusting the inter-layer spacing l.

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Non-linear regime

Hopfield coefficients determine the **polariton-polariton interaction**:

Gross-Pitaevskii equation: $$i\hbar\frac{\partial}{\partial t}\mathbf{\Phi}(\mathbf{r},t) = \mathbf{L}\mathbf{\Phi}(\mathbf{r},t)\ ,$$ $1, 2 \equiv \uparrow, \downarrow$

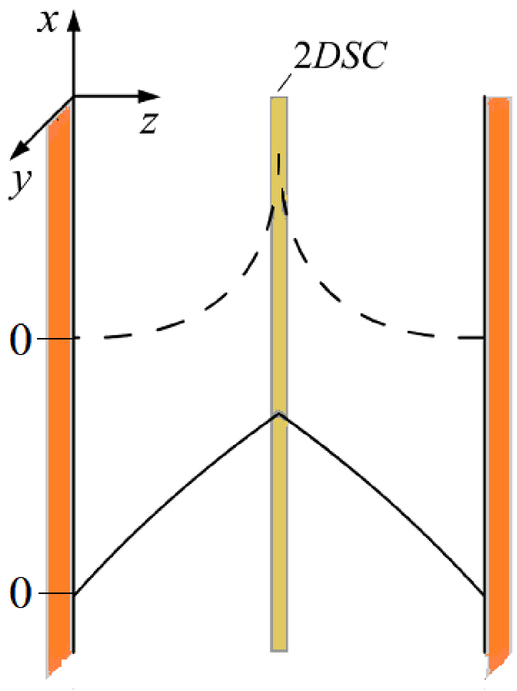
$$\mathbf{\Phi}(\mathbf{r},t) = \begin{bmatrix} \Phi_1(\mathbf{r},t) \\ \Phi_2(\mathbf{r},t) \end{bmatrix}$$

$$\mathbf{L} = \begin{bmatrix} -\frac{\hbar^2}{2m_p}\nabla^2_{\mathbf{r}} + V_c + \Lambda\left|\Phi_1\right|^2 & \Lambda_{12}\Phi_1\Phi_2^\star \\ \Lambda_{21}\Phi_1^\star\Phi_2 & -\frac{\hbar^2}{2m_p}\nabla^2_{\mathbf{r}} + V_c + \Lambda\left|\Phi_2\right|^2 \end{bmatrix}$$

It describes two interacting condensates involving excitons with opposite spins in different valleys (K/K’).

Summary

- We calculated the exciton-polariton dispersion curves, mode lifetimes, Rabi splittings, angle-resolved absorption and emission spectra, and Hopfield coefficients of structures consisting of a nearly 2D semiconductor sheet inserted in a planar microcavity for two TMD materials, MoS_2 and WS_2. Our results indicate that the strong coupling regime can be achieved in these materials, in accordance with recent experiments.
- We also considered a structure containing two TMD sheets separated by some atomic-scale distance (l) employing a nearly 2D dielectric (e.g., h-BN), which offers the possibility of tuning the interaction between two exciton-polariton Bose-Enstein condensates. We show that the dynamics of this structure are ruled by two coupled Gross-Pitaevskii equations with the adjustable dipole-dipole coupling parameter $\sim l^{-1}$.



Thank you!