Probing & tuning Rydberg excitons/polaritons in 2D semiconductors via a magnetic field

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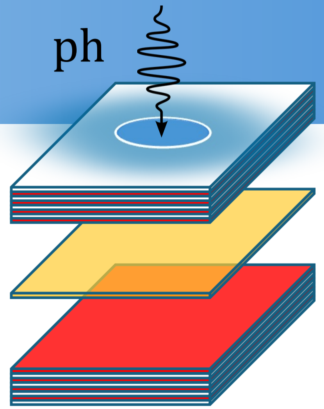
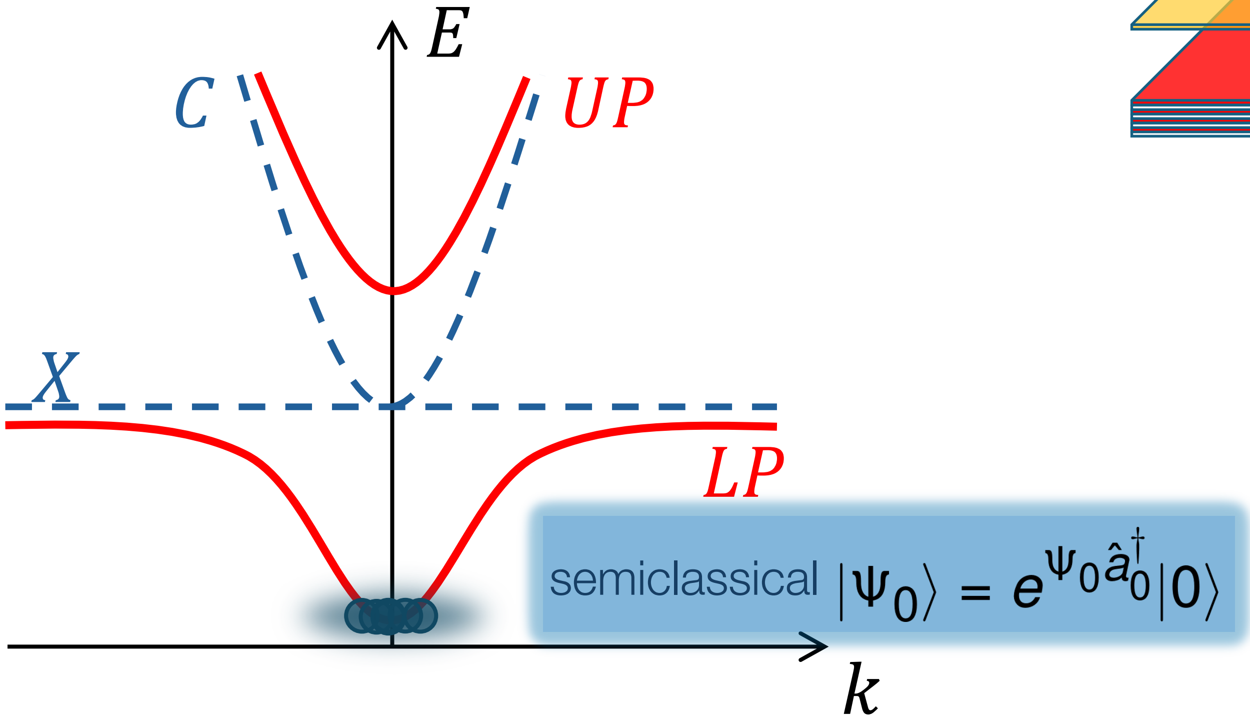
IFiMaC

Quantum polaritonics in 2D heterostructures?



$|polaritons\rangle = |matter\rangle + |light\rangle$



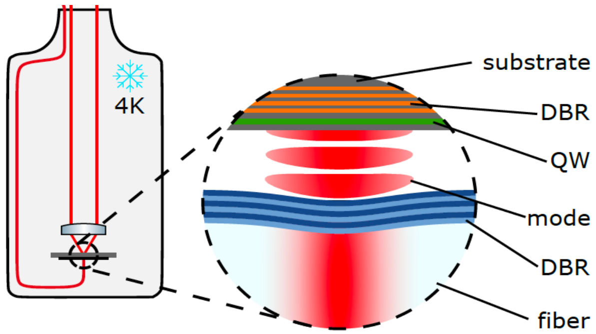
$$i\partial_t \Psi_0 = \left[-\frac{\nabla^2}{2m} + V(\mathbf{r}) - i\left(\gamma - \Gamma|\Psi_0|^2\right) + g|\Psi_0|^2 \right] \Psi_0$$

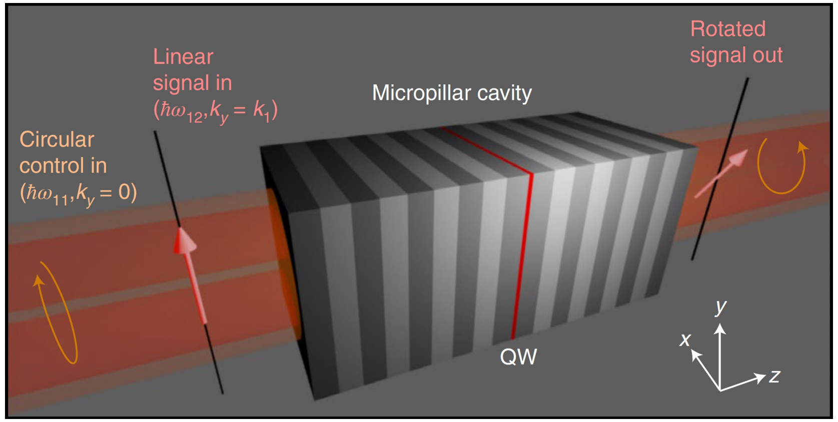
PHENOMENOLOGY OF NON-LINEAR CLASSICAL WAVES

Towards quantum polaritonics $g_{PP} > \gamma$

▷ Enhancing non-linearities by full confinement

fiber 0D cavity [Delteil et al. Nature Mat (2019)]
[Munoz-Matutano et al. Nature Mat(2019)]

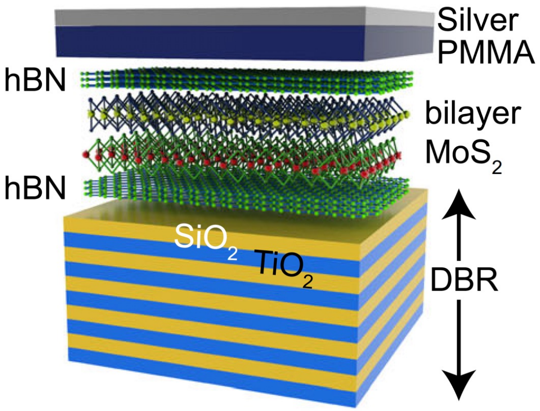




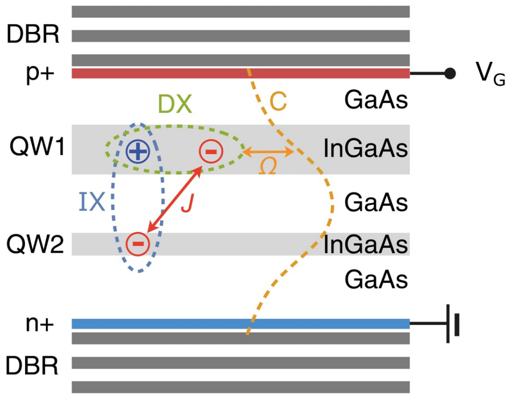
[Kuriakose et al. Nat Phot (2022)]

▷ Dipolar polaritons

[Cristofolini et al. Science (2012)]



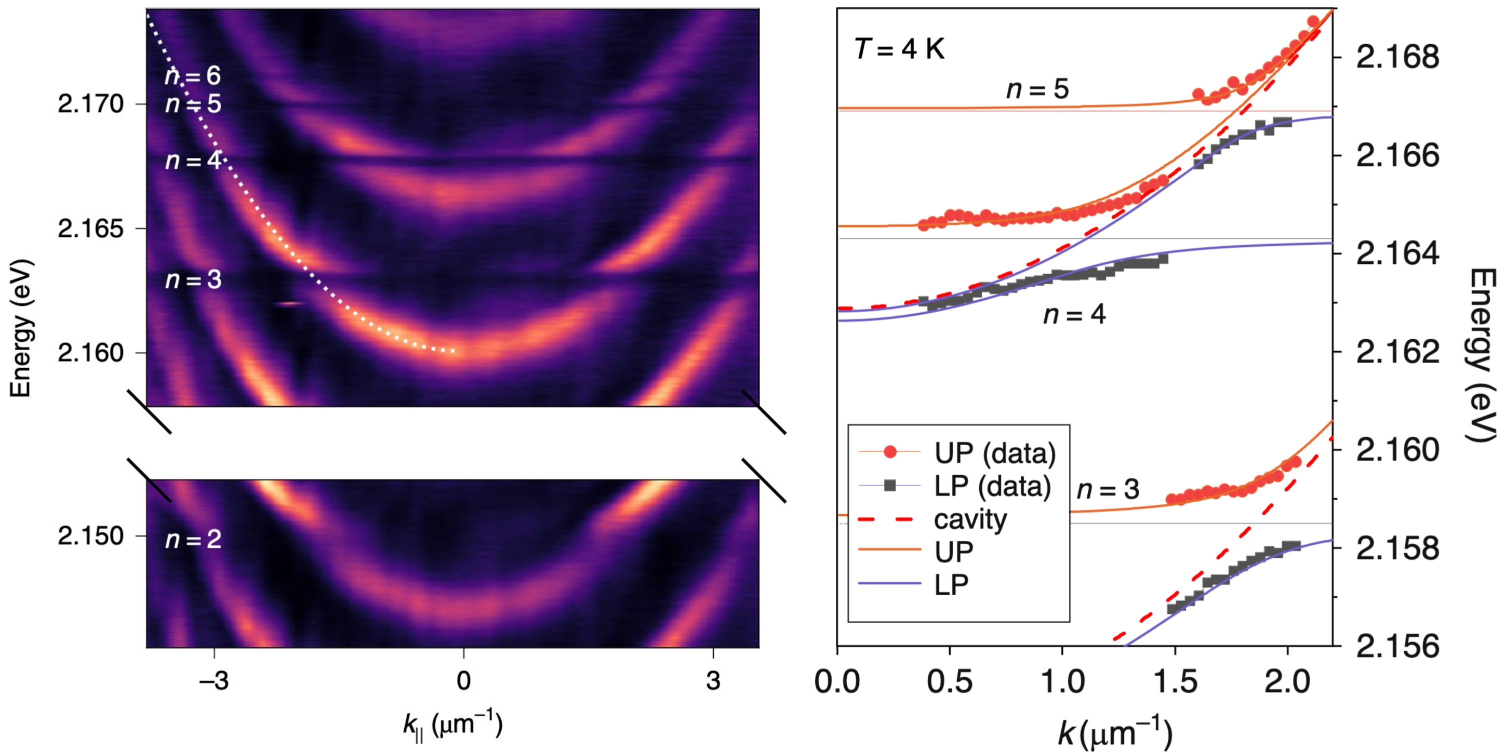
[Datta et al. Nature Comm (2022)]
[Louca et al. Nature Comm (2023)]

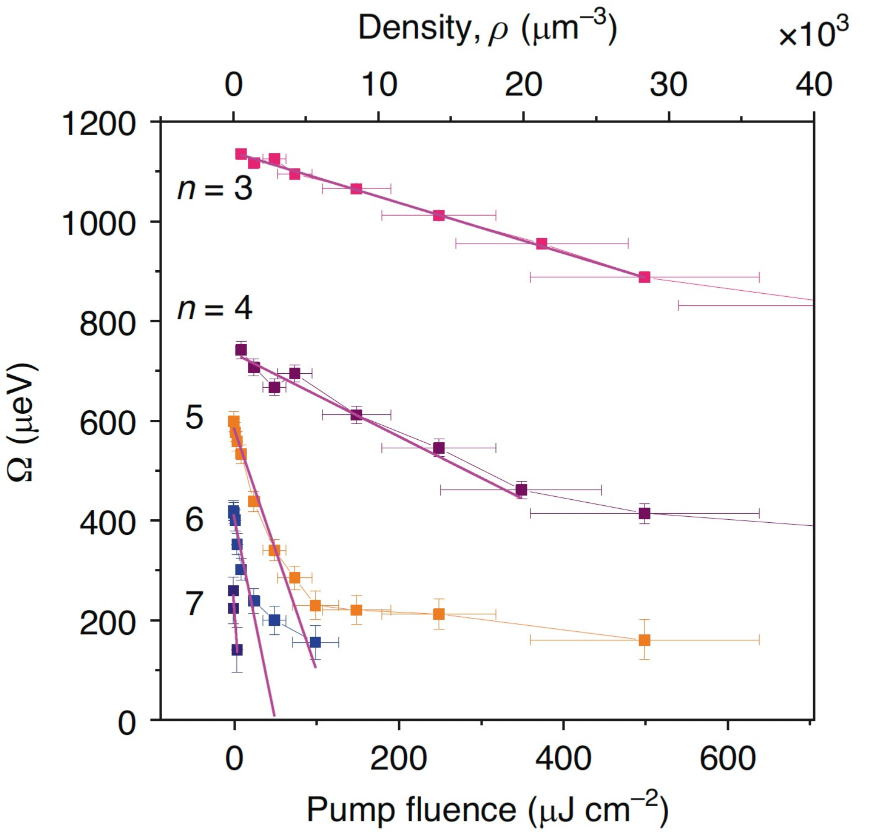


[Togan et al. PRL (2018)]

Rydberg polaritons with Cu_2O crystals

▷ Strong non-linearity (Rydberg blockade)



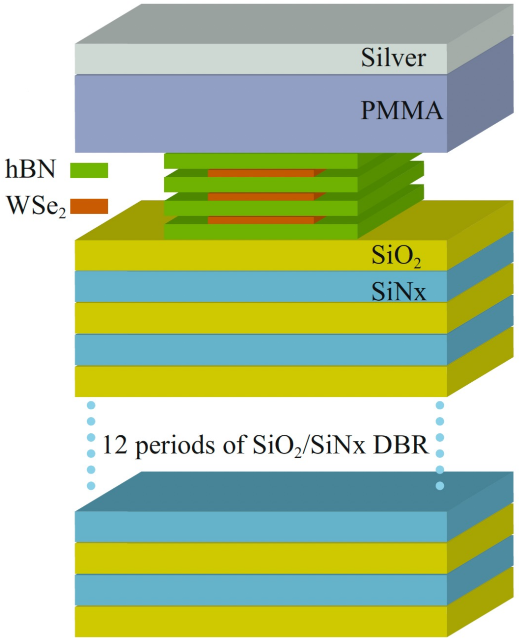


[Orfanakis et al. Nat Mat (2022)]

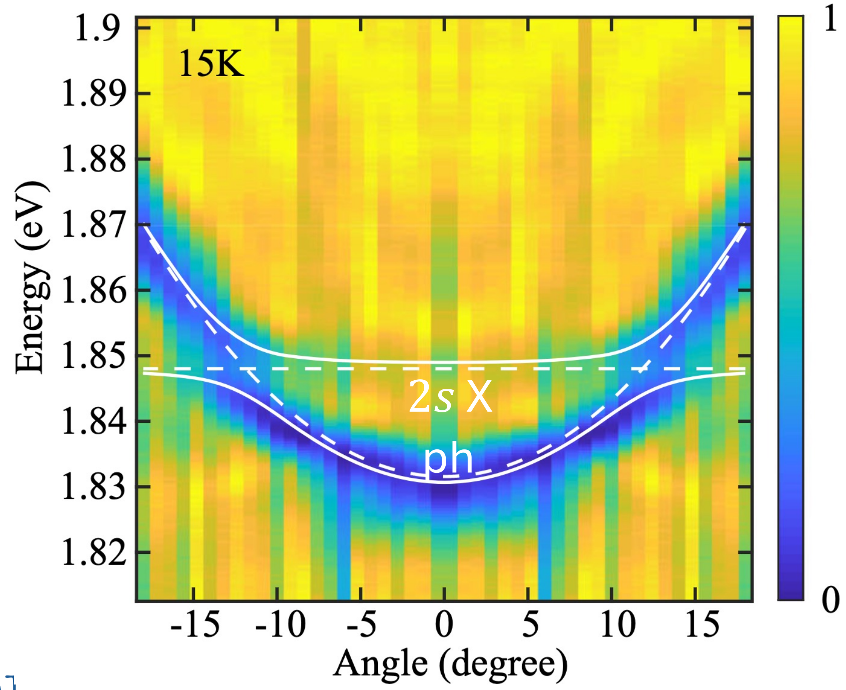
[Makhonin et al. *Light: Sci&App* (2024)]

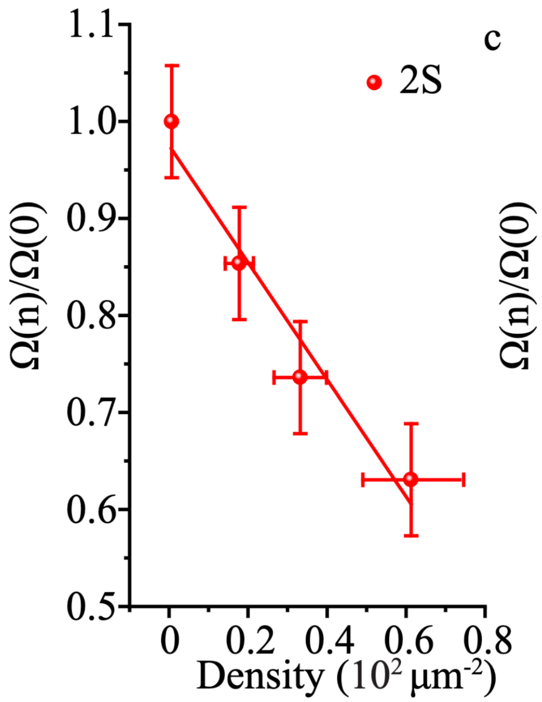
Rydberg polaritons with TMD monolayers

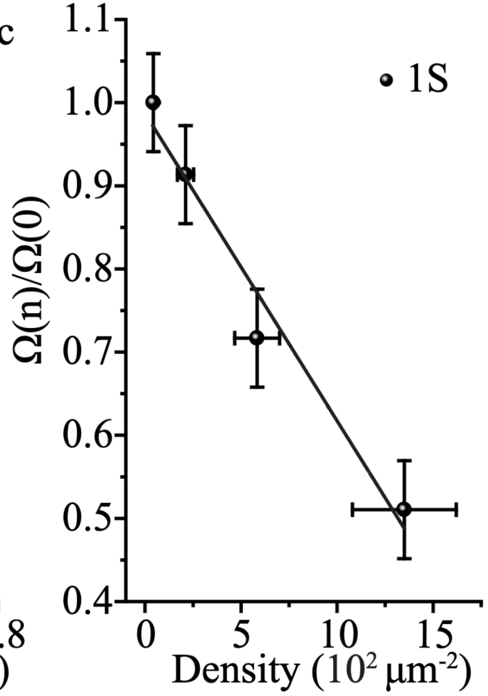
▷ Tuning resonance by temperature ▷ Strong non-linearity (polariton blockade)



[Gu et al. Nat Comm (2021)]

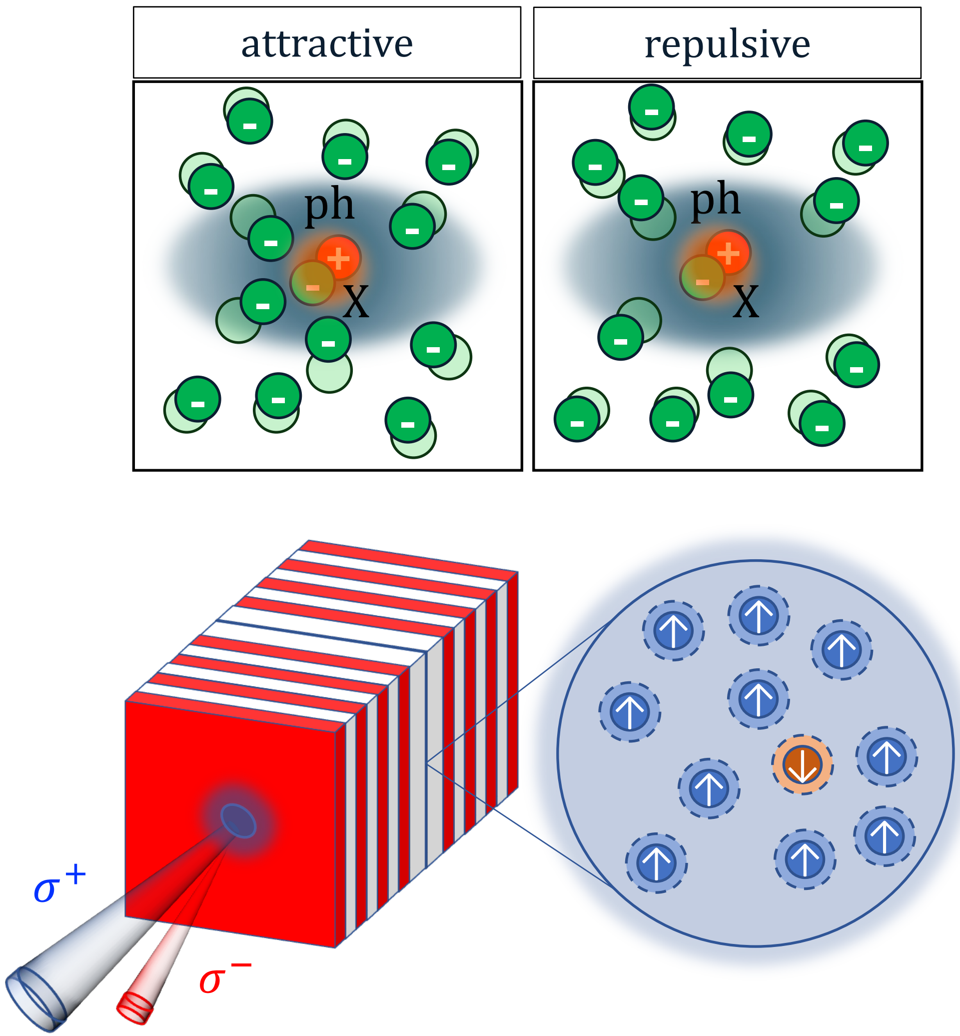




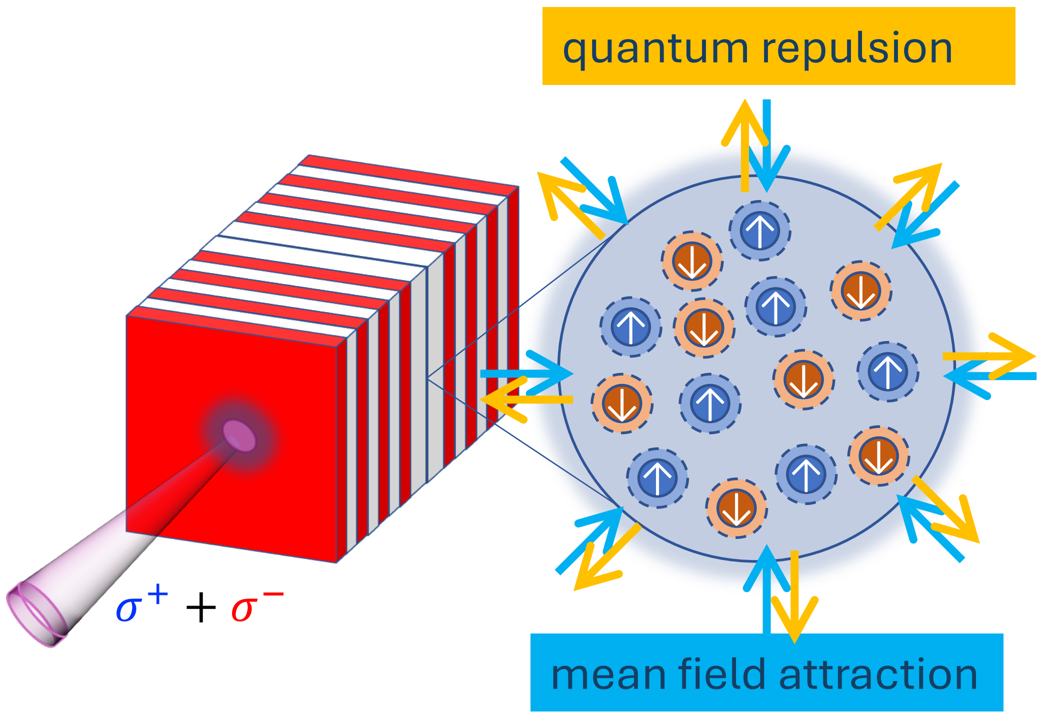


Towards quantum polaritonics: beyond mean-field effects

▷ Fermi & Bose polaron polaritons

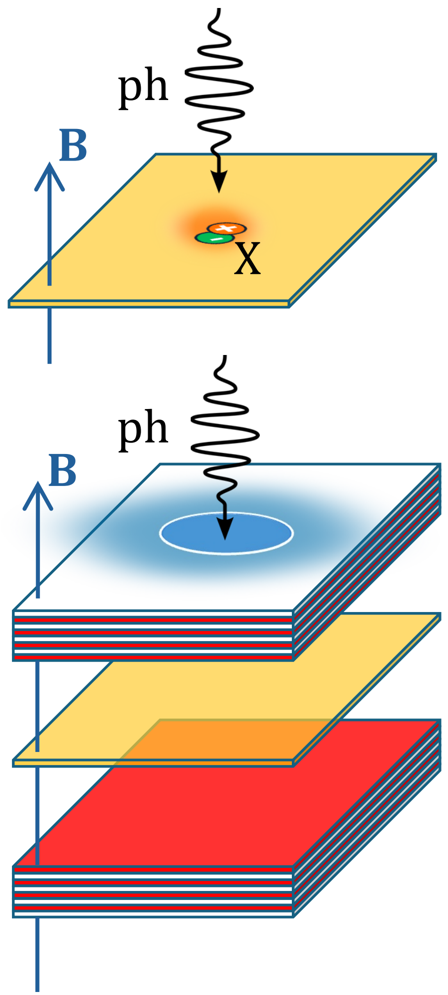


▷ Quantum droplets of light



Outline

1. Magneto-optical spectroscopy
   → probing & tuning exciton properties
   → enhanced exciton binding & coupling to light
2. Cavity polaritons

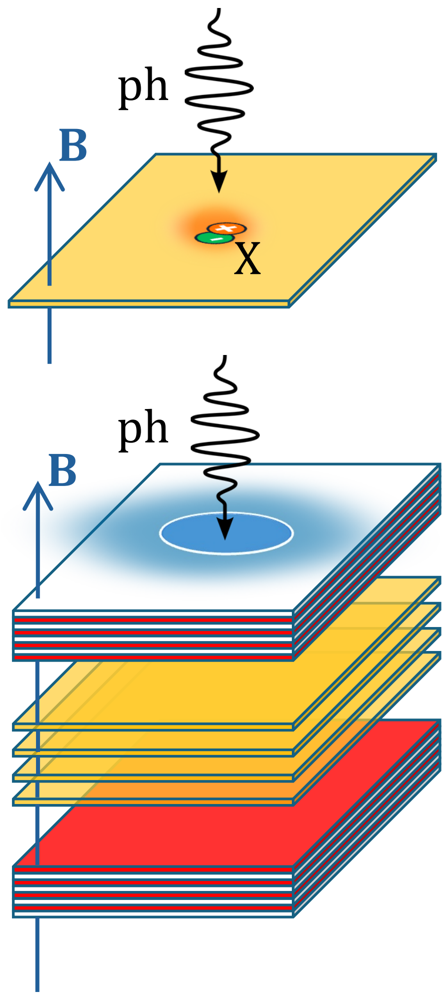


[Laird et al. PRB (2022)]
[de la Fuente Pico et al. PRB (2025)]

Outline

1. Magneto-optical spectroscopy
   → probing & tuning exciton properties
   → enhanced exciton binding & coupling to light
2. Cavity polaritons: Very-strong coupling $\Omega{\sim}E_B$
   → hybridization of multiple excitonic states
   → NON-PERTURBATIVE MICROSCOPIC THEORY
3. g_{XX} & g_{PP} interaction strengths

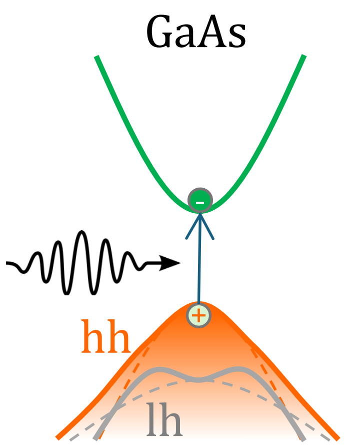
see David de la Fuente Pico's poster!

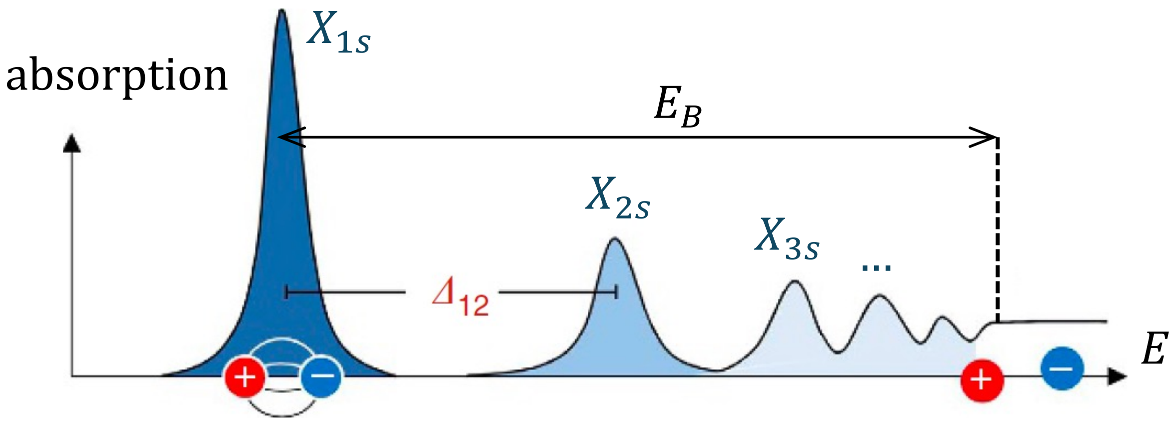


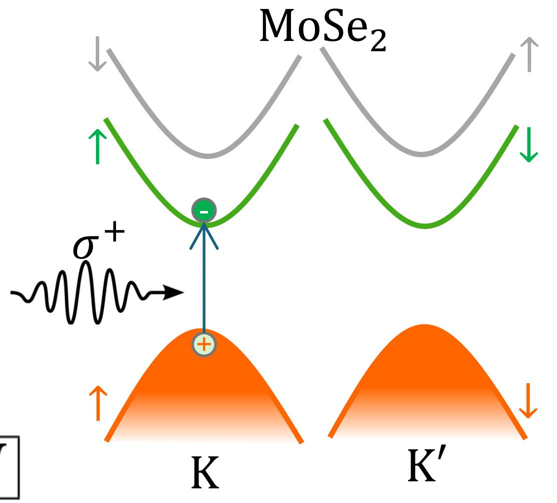
[Laird et al. PRB (2022)]
[de la Fuente Pico et al. PRB (2025)]

excitons in QWs

excitons in TMD monolayers







Coulomb

$$V_q^C = -\frac{2\pi e^2}{\varepsilon q}$$

2D hydrogen

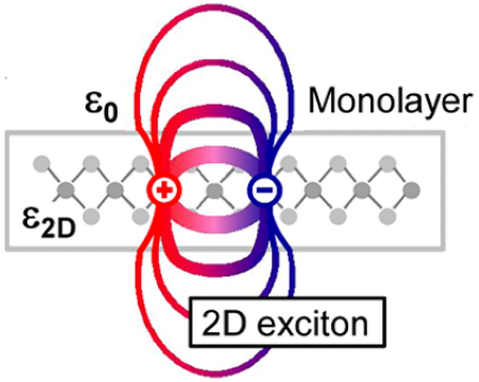
	hBN-WS_2	GaAs QW
Effective dielectric constant ε	4.35	12.9
Screening length r_0 (nm)	0.78	0
Reduced mass μ (m_0)	0.175	0.041
exciton binding energy E_B (meV)	178.8	13.5
exciton radius $\sqrt{\langle r^2 \rangle_{1s}}$ (nm)	1.7	10.2

Rytova-Keldysh

$$V_q^{RK} = -\frac{2\pi e^2}{\varepsilon q} \frac{1}{1 + r_0 q}$$

exciton Rydberg & Bohr radius

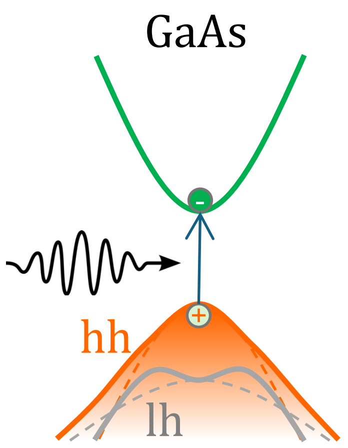
$$R_X = \frac{2\mu e^4}{\varepsilon^2} = \frac{1}{2\mu a_X^2}$$

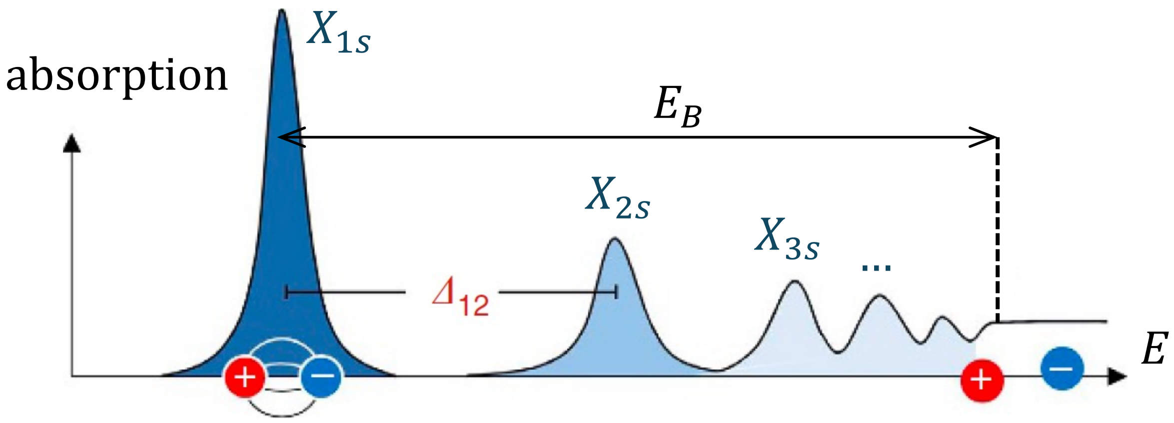


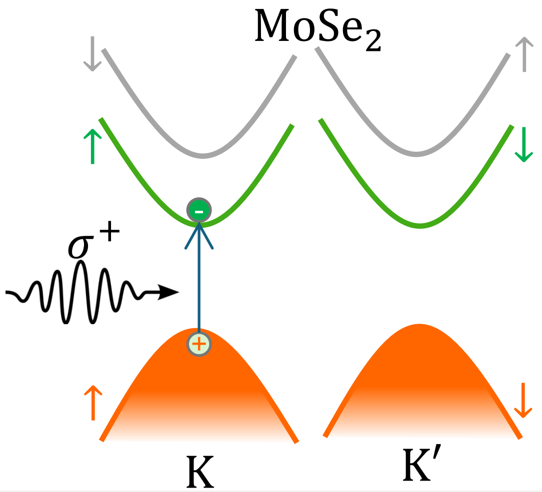
[Chernikov et al., PRL (2014)]

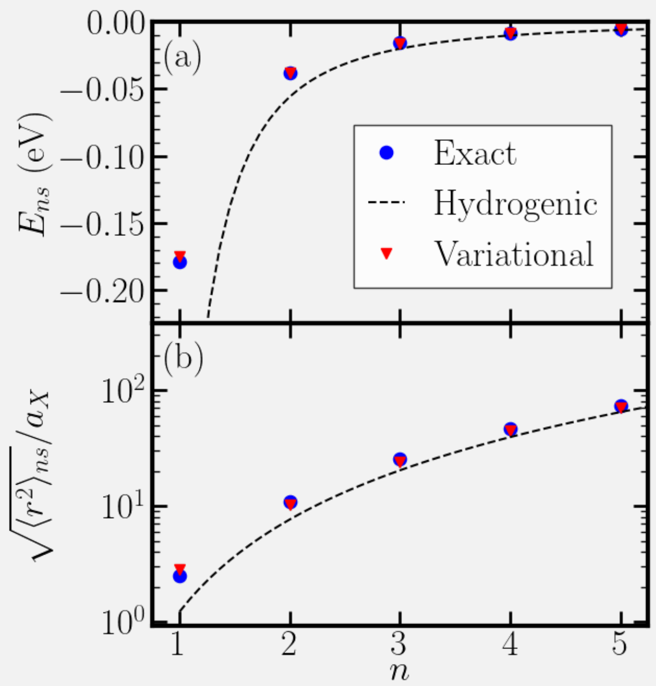
excitons in QWs

excitons in TMD monolayers



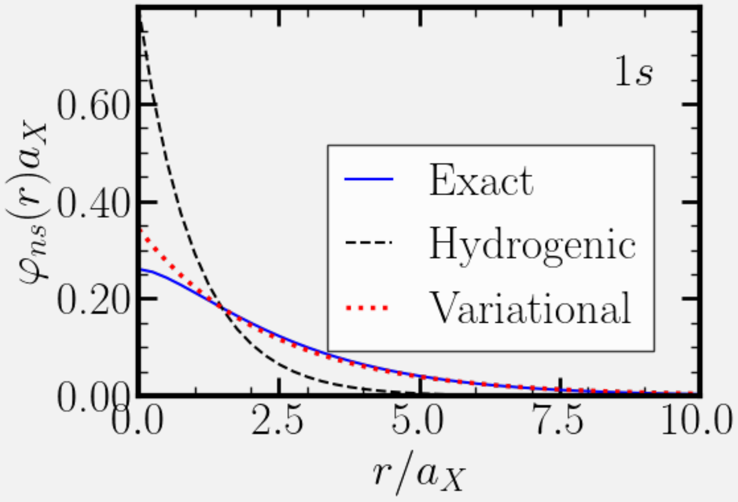


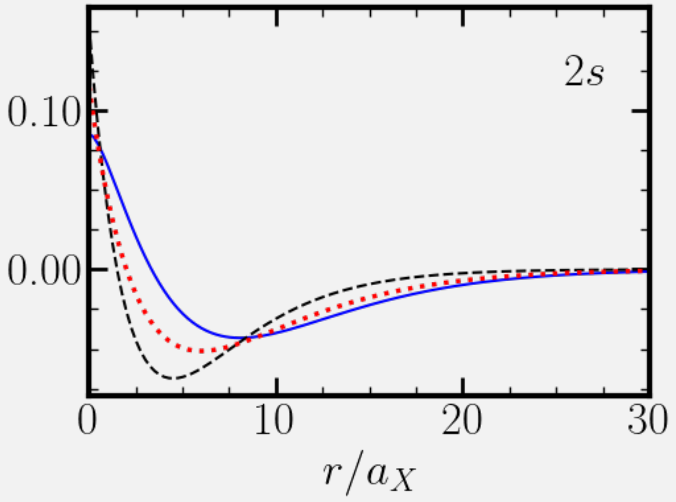




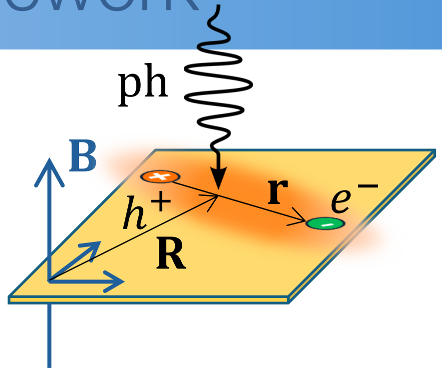
hBN encapsulated WS_2
$E_B = 179$ meV

[Goryca et al., Nat Comm (2019)]
[de la Fuente Pico et al., PRB (2025)]





Exciton magneto-optical spectroscopy: theoretical framework



- Total magnetic momentum $\hat{\mathbf{K}} = -i\nabla_{\mathbf{R}} - \frac{e}{2c}\mathbf{B}\times\mathbf{r}$
- Center of mass frame: Lamb transformation

$$\hat{X}^\dagger_{\mathbf{K}} = \frac{1}{\sqrt{A}}\int d\mathbf{r}_e d\mathbf{r}_h e^{i\left(\mathbf{K}+\frac{e}{2c}\mathbf{B}\times\mathbf{r}\right)\cdot\mathbf{R}}\varphi_{\mathbf{K}}(\mathbf{r})\hat{\Psi}^\dagger_e(\mathbf{r}_e)\hat{\Psi}^\dagger_h(\mathbf{r}_h)$$

[Gor'kov & Dzyaloshinskii, Sov J ETP (1968)]

$$\langle 0|\hat{X}_{\mathbf{K}}|\hat{H}_m|\hat{X}^\dagger_{\mathbf{K}}|0\rangle = \int d\mathbf{r}\varphi^*_{\mathbf{K}}(\mathbf{r})\hat{H}'_m\varphi_{\mathbf{K}}(\mathbf{r})$$

$$\hat{H}'_m = \left[-\frac{\nabla^2_{\mathbf{r}}}{2\mu} \underbrace{- i\frac{e\eta}{2\mu c}\mathbf{B}\cdot(\mathbf{r}\times\nabla_{\mathbf{r}})}_{\propto (m_e-m_h)\hat{L}_z} + \underbrace{\frac{e^2}{8\mu c^2}(\mathbf{B}\times\mathbf{r})^2}_{\frac{\mu\omega_c^2}{2}r^2} + \frac{e}{Mc}(\mathbf{K}\times\mathbf{B})\cdot\mathbf{r} + \frac{\mathbf{K}^2}{2M} + V(r)\right]$$

- Numerically efficient & exact solution at arbitrary field strength

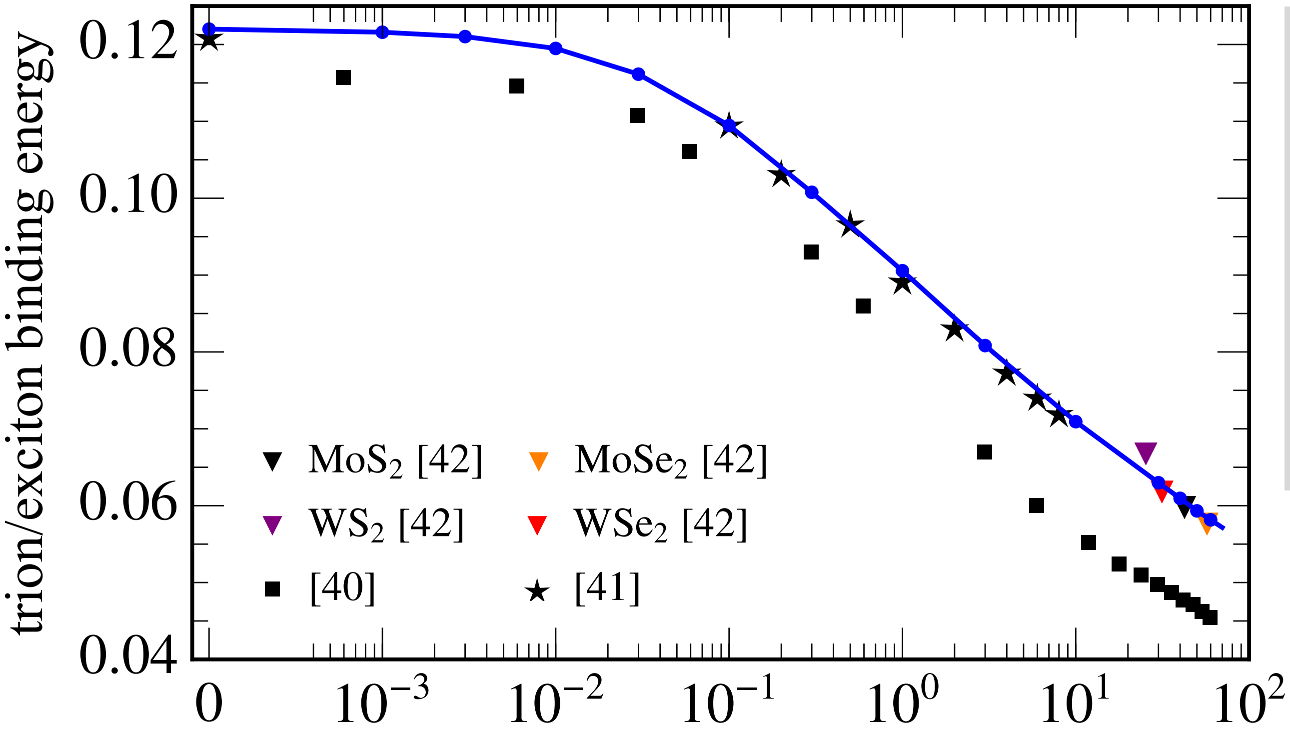
[Laird et al. PRB (2022)]
[de la Fuente Pico et al. PRB (2025)]

  → avoids manipulation of derivatives

  → mapping 2D harmonic oscillator – 2D hydrogen problem [Duru&Kleinert FdP (1982)]

  → (Landè) subtraction scheme to cancel the pole of the potential $\begin{cases}\text{Coulomb}\\ \text{Rytova-Keldysh}\end{cases}$

(Distinguishable) trions in TMD monolayers



r_0/a_X

screening length

[Kumar et al. PRB (2025)]

Variational
[Courtade et al, PRB (2017)]

quantum Monte Carlo
[Szyniszewski et al, PRB (2017)]

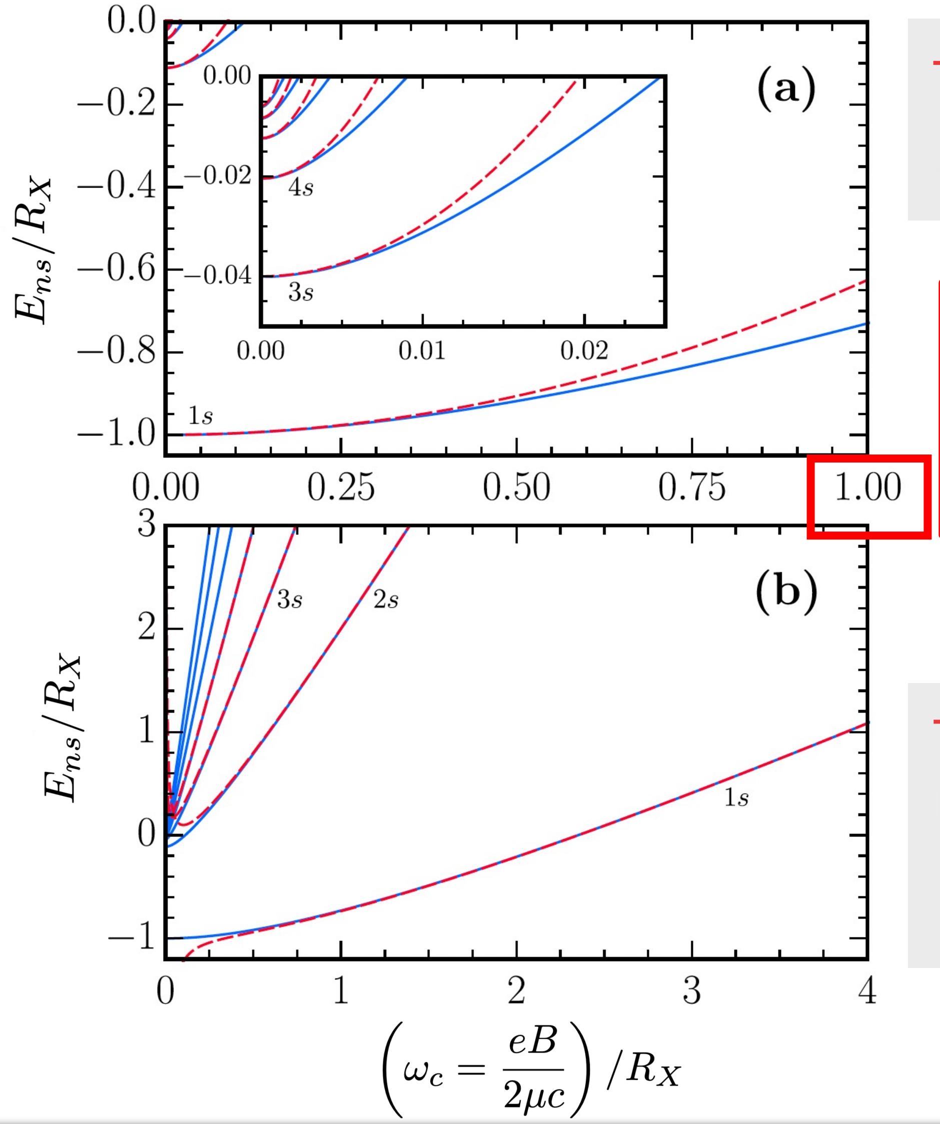
exact diagonalisation
[Fey et al. PRB (2020)]

Rytova-Keldysh potential

$$V_q^{RK} = -\frac{2\pi e^2}{\varepsilon q}\frac{1}{1 + r_0 q}$$

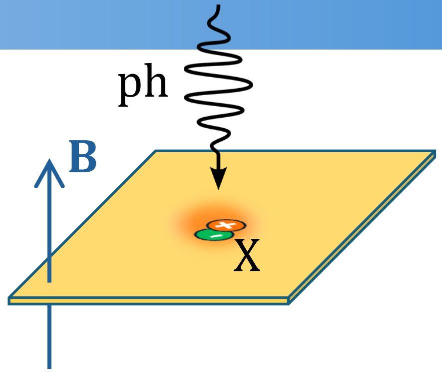
GaAs QWs: exciton diamagnetic shift

[Laird et al. PPB (2022)]



low-field

$$E_{ns} \simeq E_{ns}^{hyd} + \frac{1}{2}\mu\omega_c^2\langle r^2\rangle_{ns}^{hyd}$$



GaAs QW
$R = 13.5$ meV
$\mu = 0.041 m_0$
$B = 10$ T

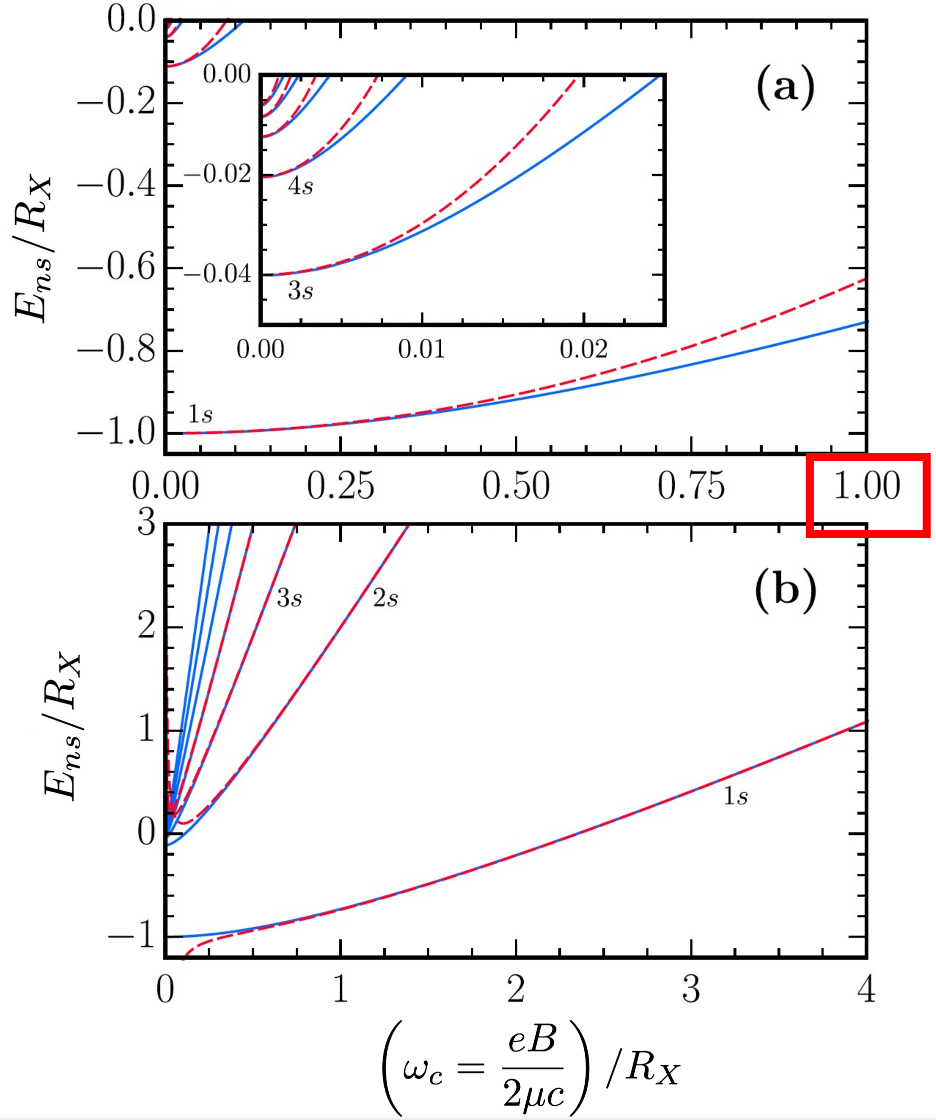
high-field

$$E_{ns} \simeq \underbrace{\omega_c(2n-1)}_{E_{ns}^{Landau}} + O(\sqrt{\omega_c})$$

[MacDonald & Ritchie, PRB (1986)]

GaAs QWs: exciton diamagnetic shift & binding energies

[Laird et al. PPB (2022)]



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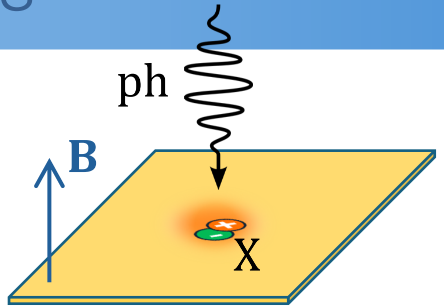
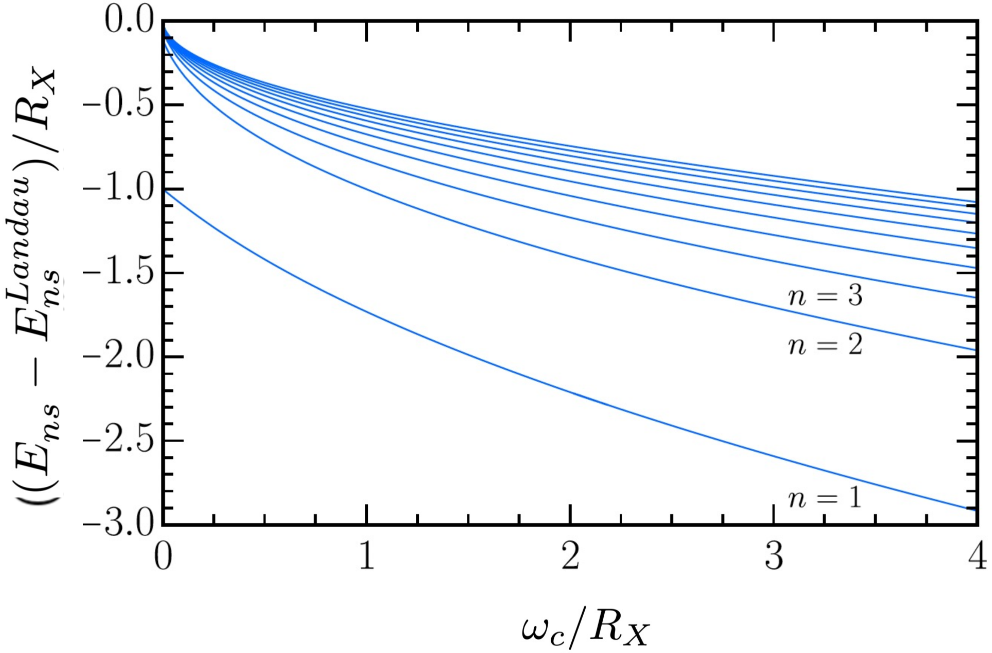
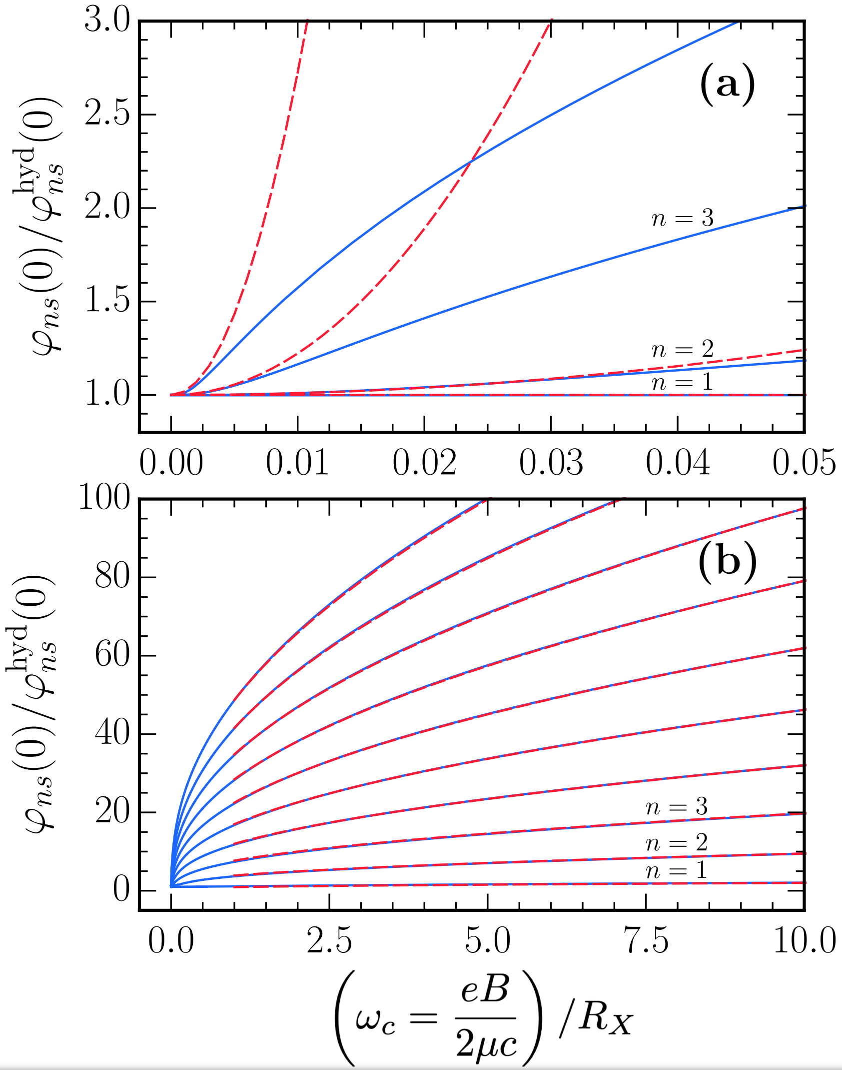
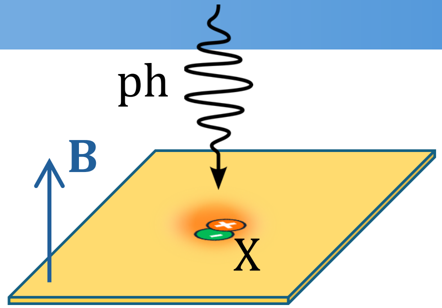


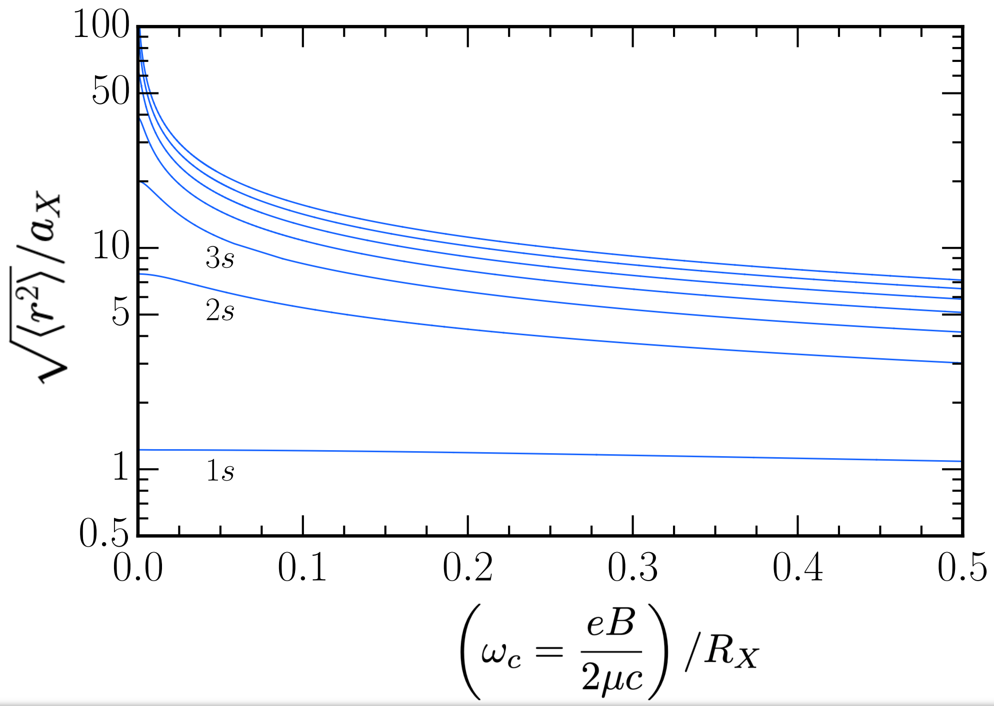


GaAs QWs: exciton oscillator strength & size

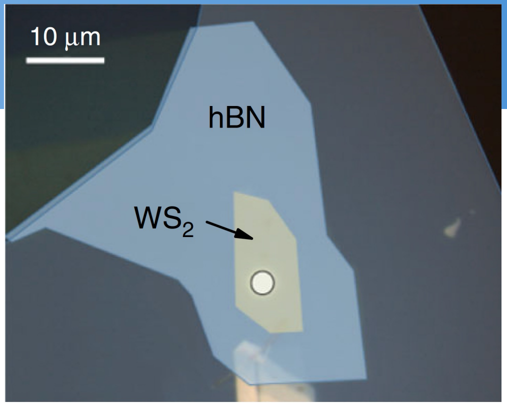
[Laird et al. PPB (2022)]



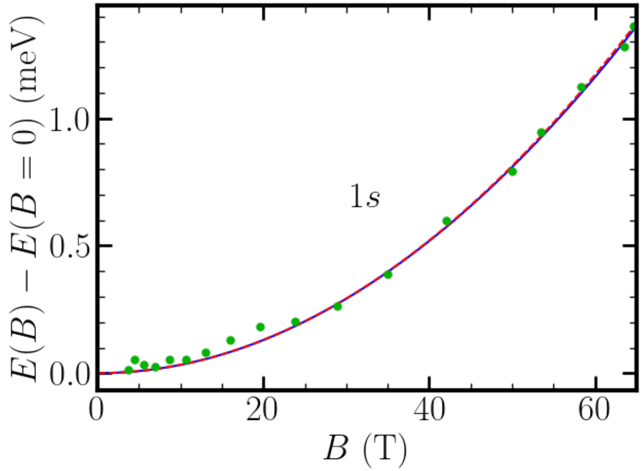




TMD monolayers (experiments Scott Crooker's group)

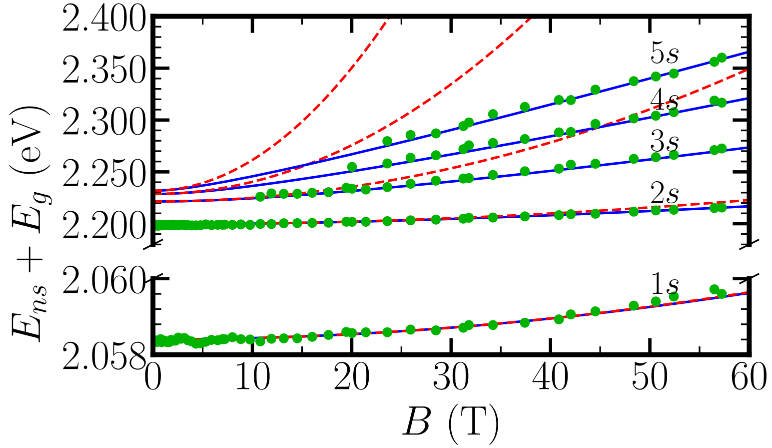


no encapsulated WS_2 $E_B = 410$ meV



[Stier et al. Nat Comm (2016)]

hBN encapsulated WS_2 $E_B = 178.8$ meV



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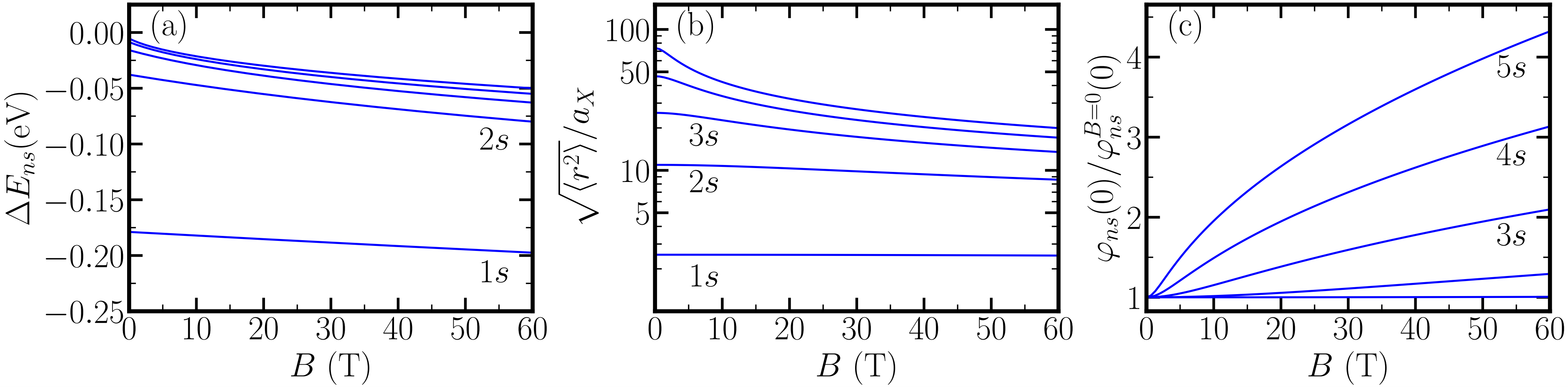
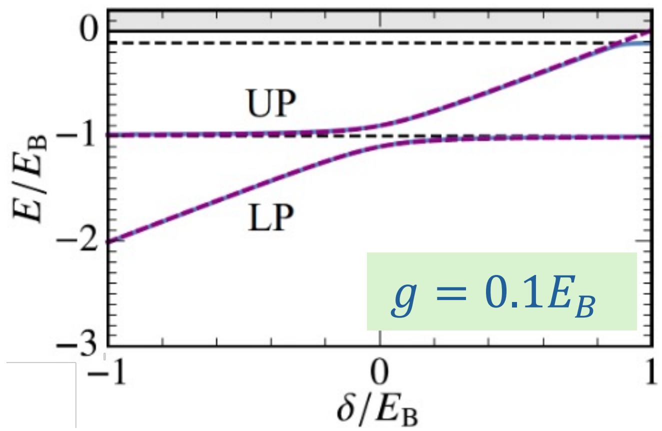


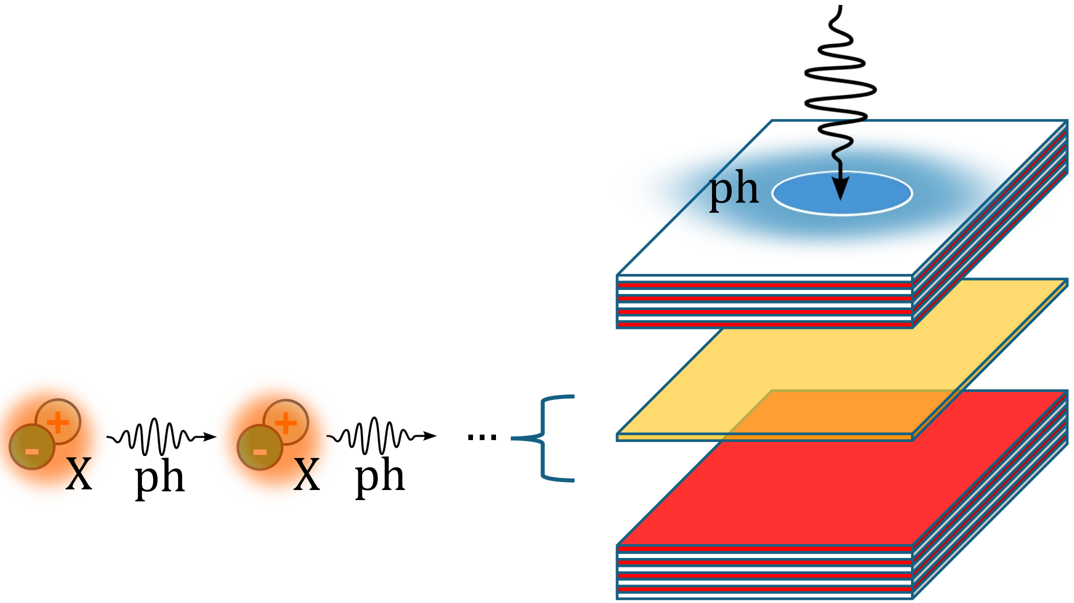
Polaritons: strong & very strong light-matter coupling

▷ Light-matter coupling $g = \frac{\Omega}{2}$

$g > \gamma$	strong	energy transfer between excitons and photons

[Levinsen et al. PPR (2019)]





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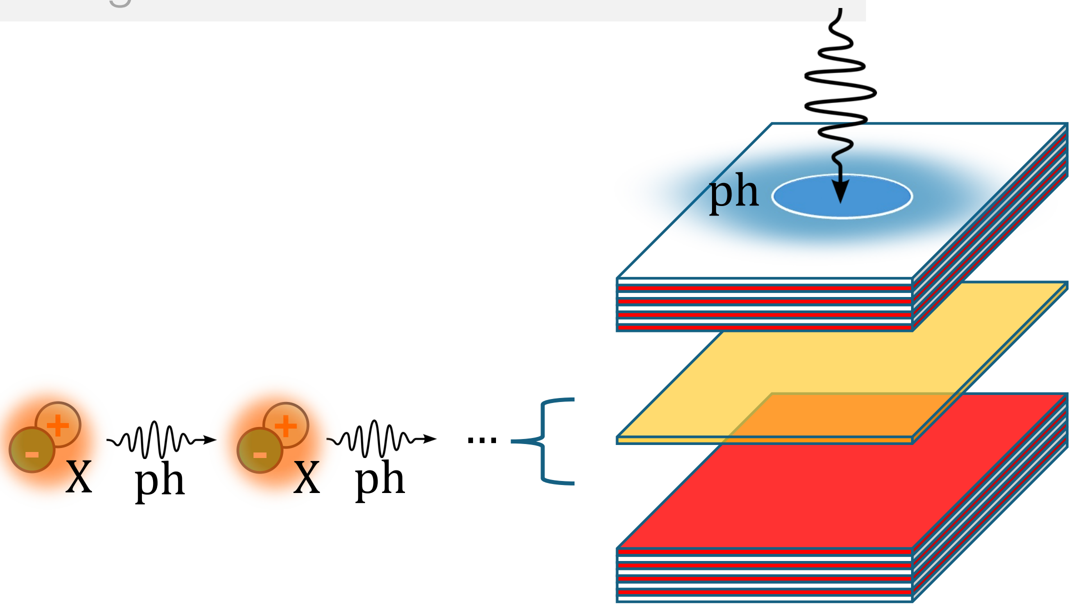
$g > \gamma$	strong	energy transfer between excitons and photons
$g \sim E_B$	very strong	hybridization of different excitonic levels
$g \sim E_g$	ultra strong	hybridization with different number of excitations

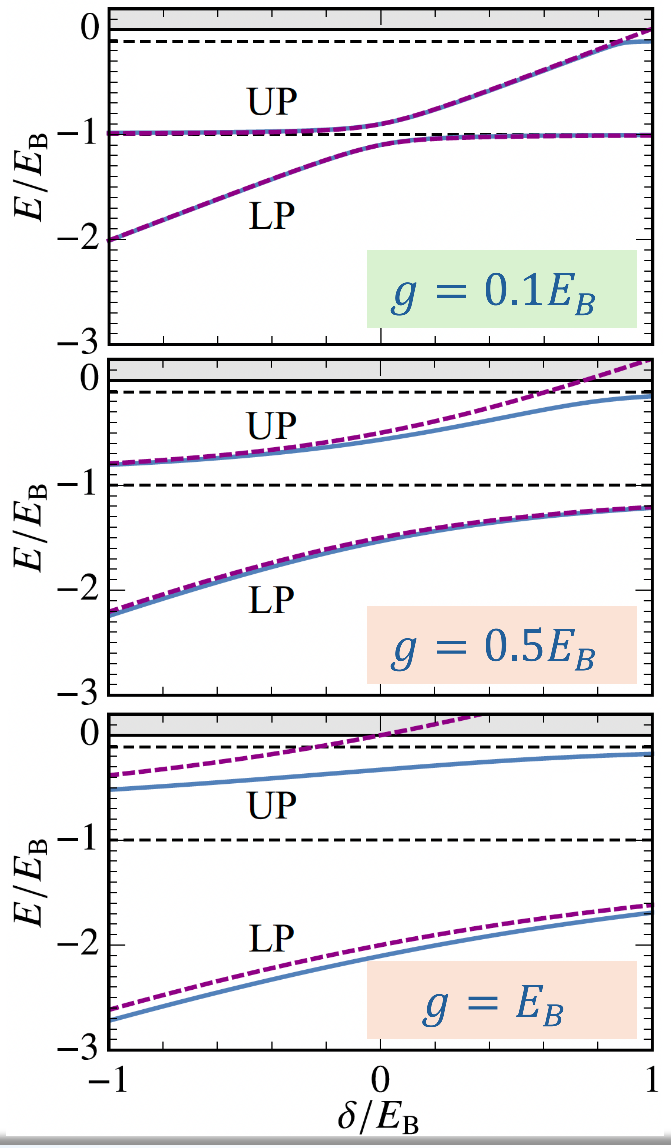
g

[Khurgin SSC (2001)]

beyond coupled-oscillator description



[Levinsen et al. PPR (2019)]



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g ↓

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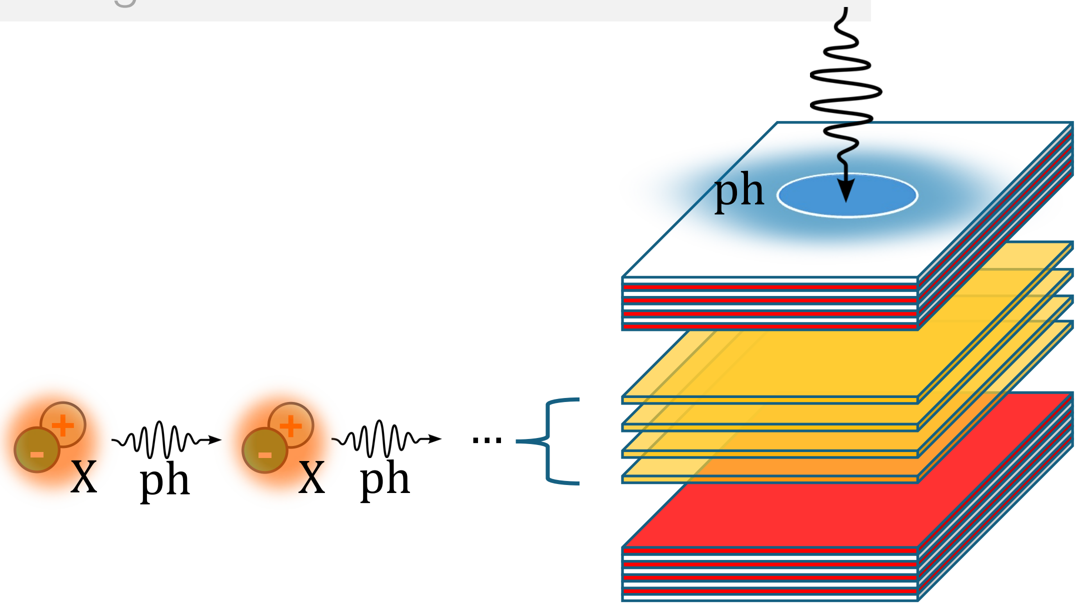
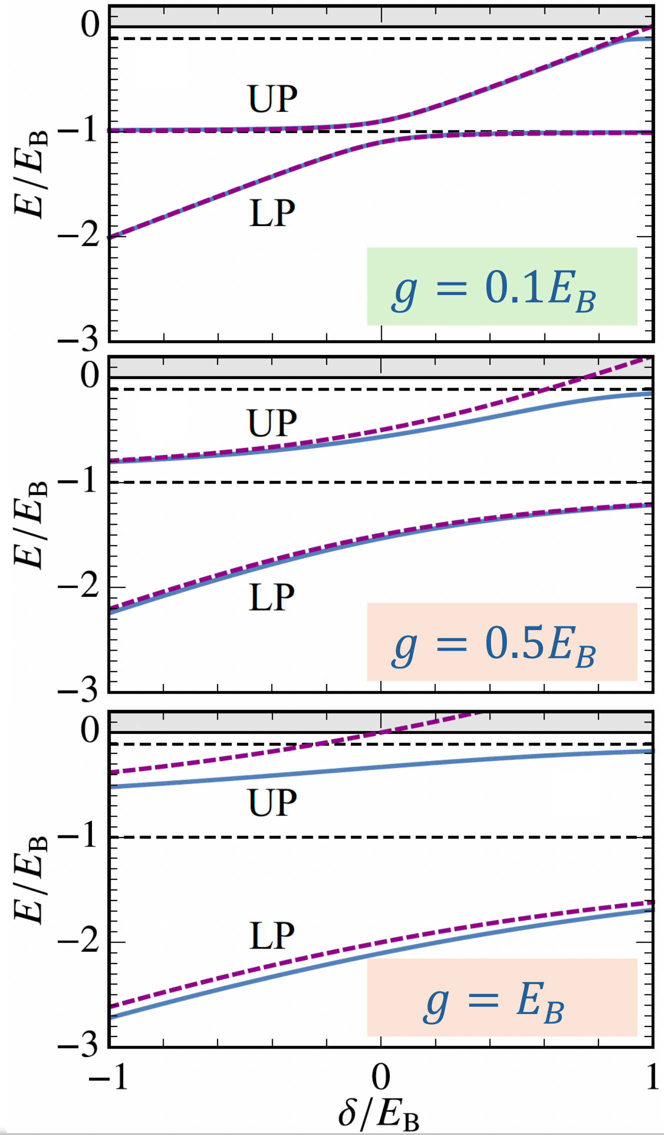


[Levinsen et al. PPR (2019)]



Polaritons: strong & very strong light-matter coupling

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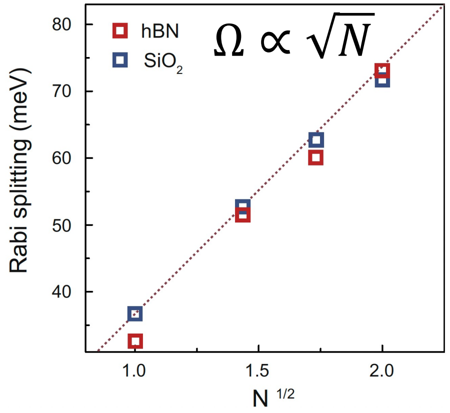
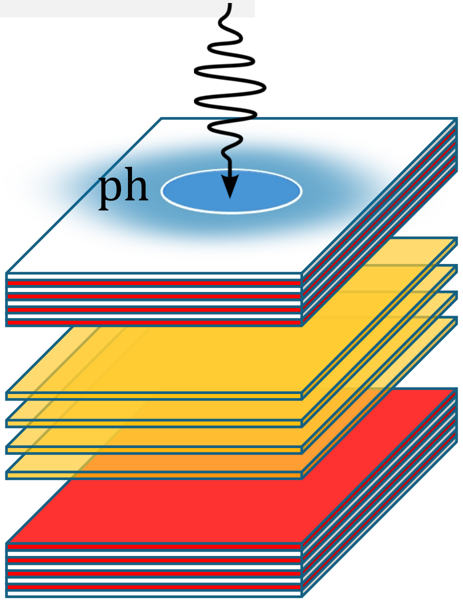
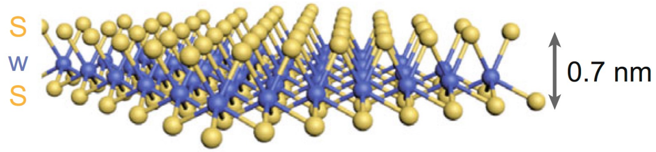
g ↓



[Zhao et al. Nat Comm (2023)]



$4 \times$



$\Omega = 71$ meV

$E_B \sim 180$ meV

$\frac{g}{E_B} \sim 0.2$

[Brodback et al. PRL (2017)]

7nm $28 \times$ GaAs QWs

$\Omega = 17.4$meV

$E_B = 13.5$ meV

$\frac{g}{E_B} \sim 0.64$

Magnetopolaritons: very strong light-matter coupling

▷ Light-matter coupling $g = \frac{\Omega}{2}$

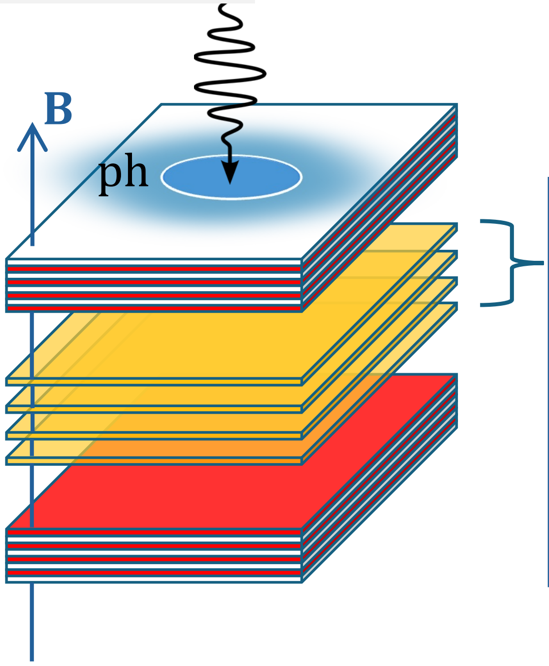
g

$g > \gamma$	strong	energy transfer between excitons and photons
$g \sim E_B$	very strong	hybridization of different excitonic levels
$g \sim E_g$	ultra strong	hybridization with different number of excitations

▷ + magnetic field $\mathbf{B}$

Use diamagnetic shift to verify very strong coupling effects: [Yang et al. NJP (2015)]'s proposal

Probe the modifications of the e-h wavefunction due to very-strong coupling to light

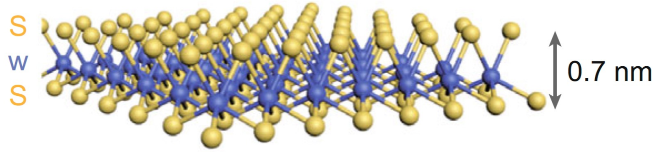


[Brodback et al. PRL (2017)]

7nm 28 × GaAs QWs
$\Omega = 17.4\text{meV}$
$E_B = 13.5\text{ meV}$
$\frac{g}{E_B} \sim 0.64$

[Zhao et al. Nat Comm (2023)]

4 ×



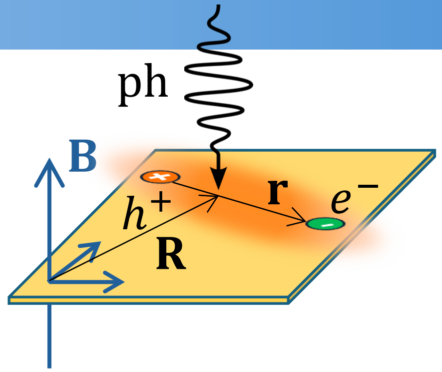
$\Omega = 71\text{ meV}$
$E_B \sim 180\text{ meV}$
$\frac{g}{E_B} \sim 0.2$

Polaritons in a magnetic field

[Laird et al. PPB (2022)]
[de la Fuente et al., PRB (2025)]

▷ Light-matter coupling $\hat{H}_{Cm} = \frac{g}{\sqrt{\mathcal{A}}}\left[\hat{a}\int d\mathbf{r}\hat{\Psi}_e^\dagger(\mathbf{r})\hat{\Psi}_h^\dagger(\mathbf{r}) + \text{h.c.}\right]$

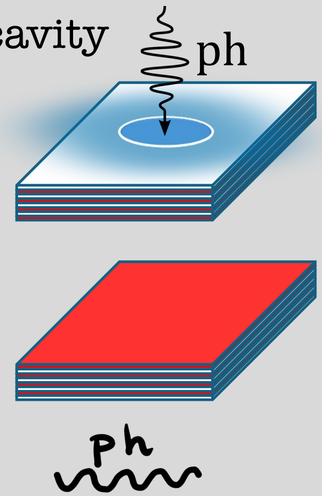
▷ Polariton state $\hat{P}^\dagger = \frac{1}{\sqrt{\mathcal{A}}}\int d\mathbf{r}_e d\mathbf{r}_h e^{i\frac{e}{2c}(\mathbf{B}\times\mathbf{r})\cdot\mathbf{R}}\varphi_0(\mathbf{r})\hat{\Psi}_e^\dagger(\mathbf{r}_e)\hat{\Psi}_h^\dagger(\mathbf{r}_h) + \gamma\hat{a}^\dagger$



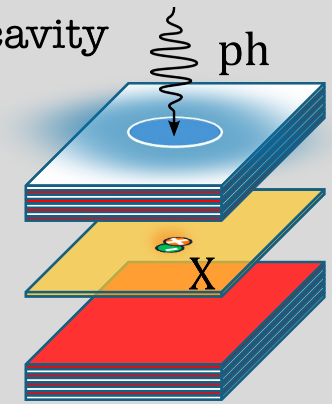
- Allows matter properties to be modified by light & vice versa
  → access the very-strong coupling regime

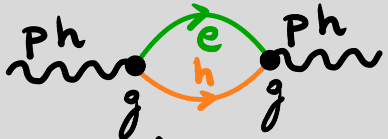
▷ Renormalization scheme [Levinsen et al. PPR (2019)]

passive cavity



active cavity





$E = \omega$

$E = \omega - g^2 \sum_{\mathbf{k}} \frac{1}{\varepsilon_B + \varepsilon_{\mathbf{k}e} + \varepsilon_{\mathbf{k}h}}$

→ energy shift depends on microscopic details (UV cut-off)

Magnetopolaritons: strong coupling $g = \frac{\Omega}{2} \ll E_B$

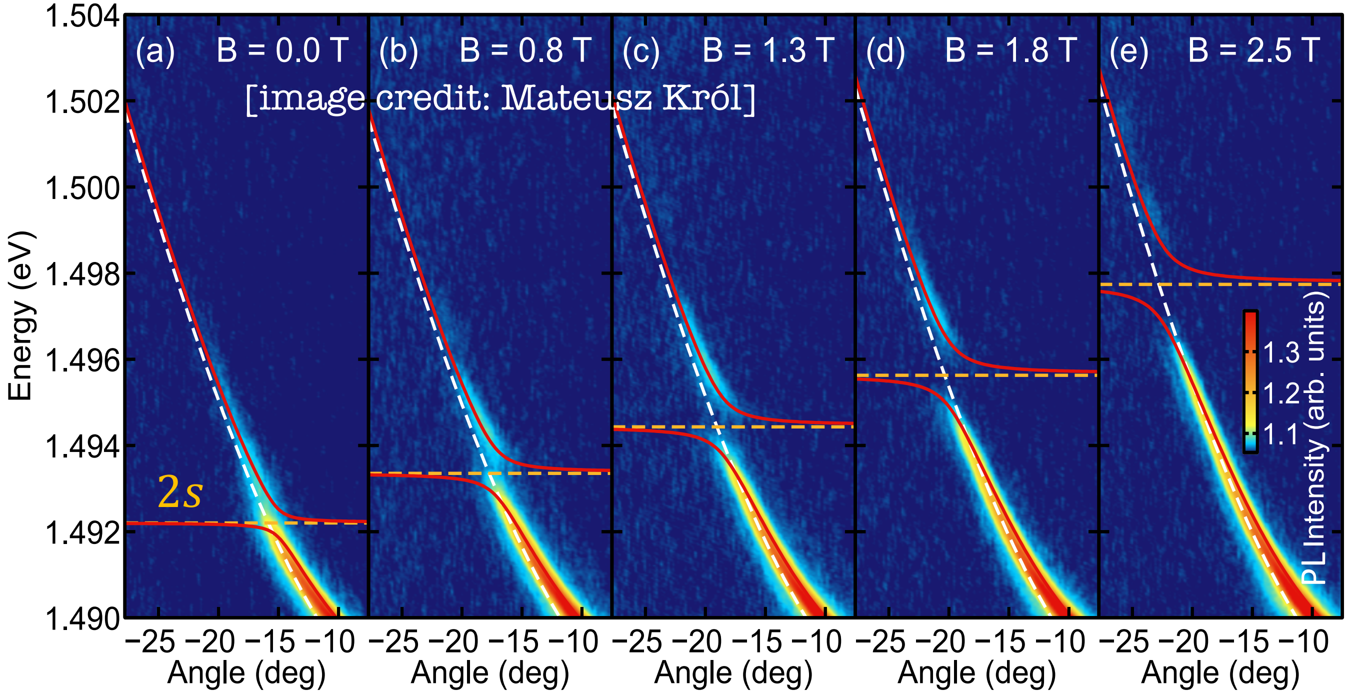
8 nm InGaAs 1 QW
$E_B = 7$ meV
$\Omega = 3.5$ meV
$\frac{g}{E_B} \sim 0.25$
$\mu = 0.046 m_0$

[Piętka et al. PRB (2015)]
[Piętka et al. PRB (2017)]

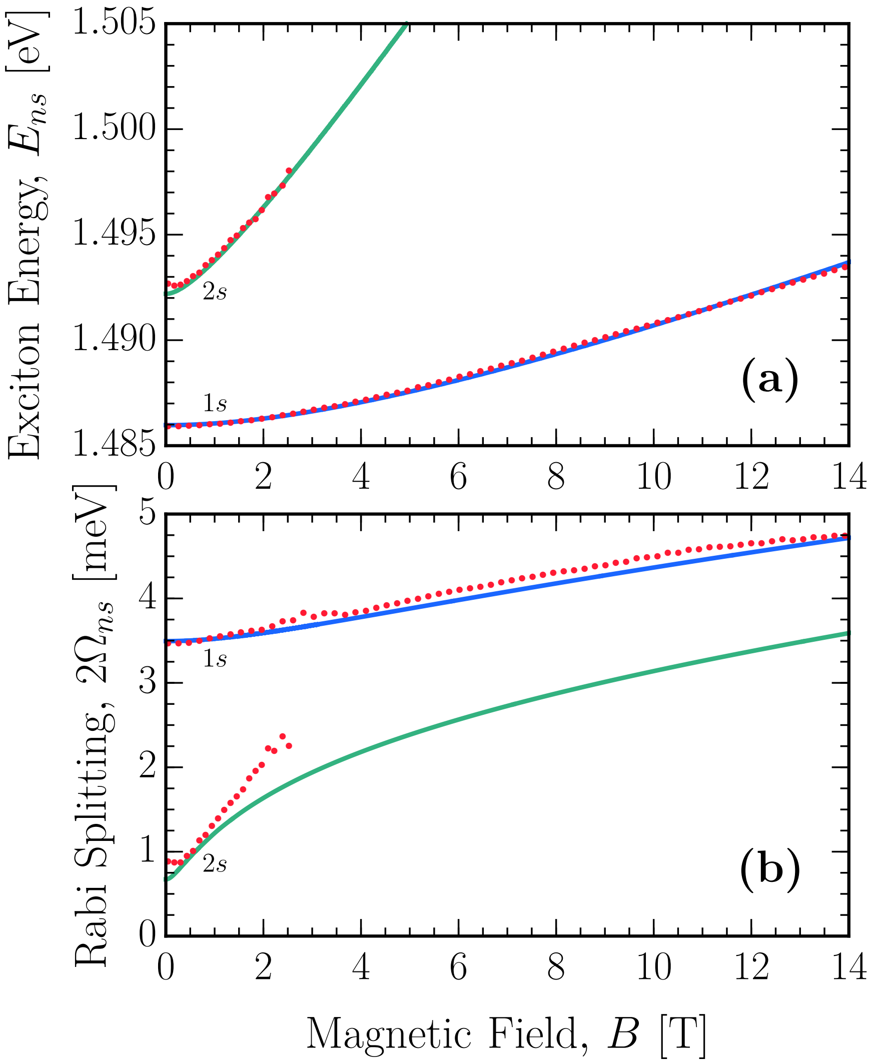
▷ perturbative light-matter

$$\Omega_{ns} = \varphi_{ns}(0)/\varphi_{ns}^{hyd}(0)$$

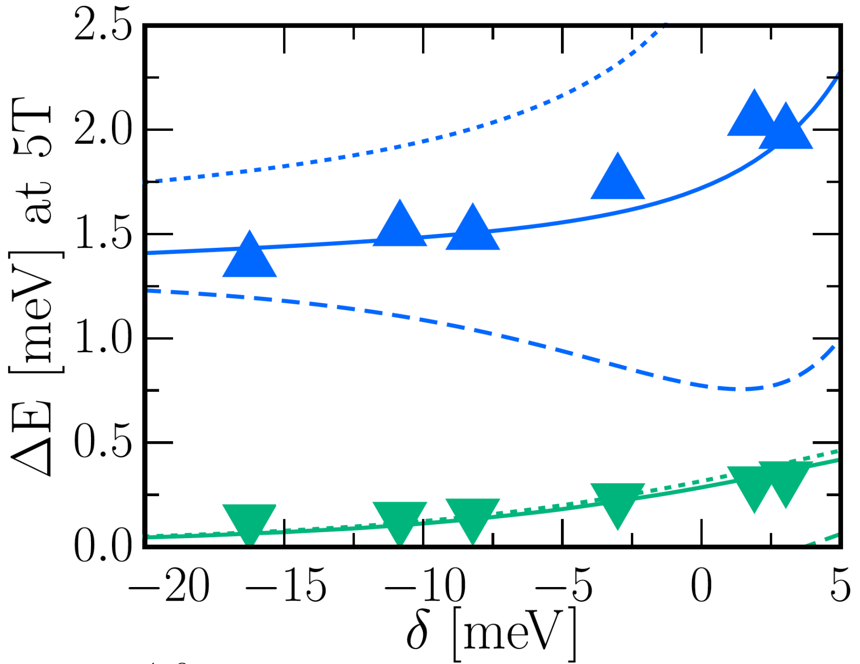
$$\mathbb{H}_{3\text{-COM}} = \begin{pmatrix} \delta + E_{1s} & \Omega_{1s}/2 & \Omega_{2s}/2 \\ \Omega_{1s}/2 & E_{1s} & 0 \\ \Omega_{2s}/2 & 0 & E_{2s} \end{pmatrix}$$

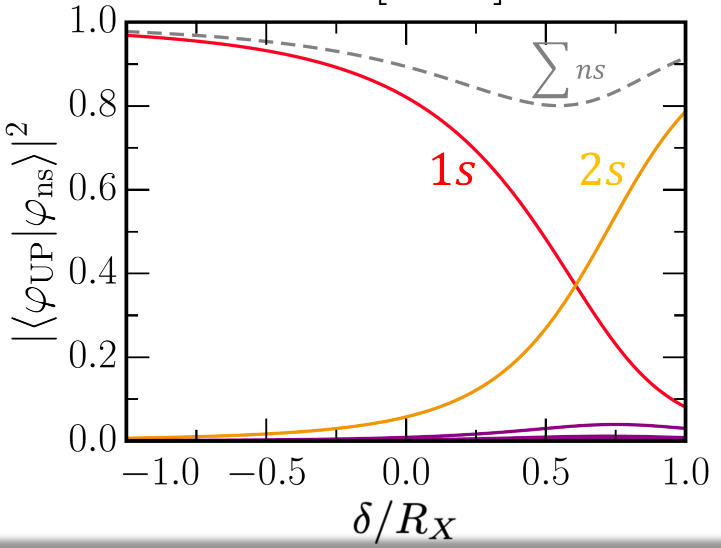


[Laird et al. PPB (2022)]



Magnetopolaritons: very-strong coupling $g = \frac{\Omega}{2} \sim E_B$





7nm 28 × GaAs QWs [Brodback et al. PRL (2017)]

$\Omega = 17.4$meV

$E_B = 13.5$ meV

$\frac{g}{E_B} \sim 0.64$

▼LP▲UP data

exact theory [Laird et al. PPB (2022)]

3 COM (perturbative light-matter)

$\frac{1}{2}\mu\omega_c^2\langle r^2\rangle_{\substack{LP\\UP}} = \frac{1}{2}\mu\omega_c^2(1-\gamma^2)\langle r^2\rangle_{\varphi,\substack{LP\\UP}}$ (perturbative magnetic field)

Probe the modifications of the e-h wavefunction due to very-strong coupling to light

- LP: light-induced shrinkage of matter component → smaller eh separation in LP than 1s exciton
- UP: importance of the admixture of exciton Rydberg states

XX & PP interaction strength [de la Fuente et al., PRB (2025)]

▷ Born Approximation

$$\left|P^2\right\rangle = \hat{P}^\dagger \hat{P}^\dagger \left|0\right\rangle$$

$$\frac{g_{PP}}{\mathcal{A}} = \frac{\left\langle P^2\right| \hat{H} \left|P^2\right\rangle}{\left\langle P^2|P^2\right\rangle} - 2E$$

1. analytical
2. $g_{XX} = g_{PP}|_{g=0=\Omega}$
3. $g_{XX}^{B=0}$ recovers same result from [Ciuti et al. PRB (1998)] [Tassone & Yamamoto PRB (1999)]
4. $g_{PP}^{B=0}$ recovers same results from [Levinsen et al. PRR (2019)]

→ strict upper bound on the interaction strength [Li et al., PRB (2021)]

→ no momentum transfer: exchange interactions only

XX & PP interaction strength [de la Fuente et al., PRB (2025)]

▷ Born Approximation

$$|P^2\rangle = \hat{P}^\dagger \hat{P}^\dagger |0\rangle$$

$$\frac{g_{PP}}{\mathcal{A}} = \frac{\langle P^2 | \hat{H} | P^2 \rangle}{\langle P^2 | P^2 \rangle} - 2E$$

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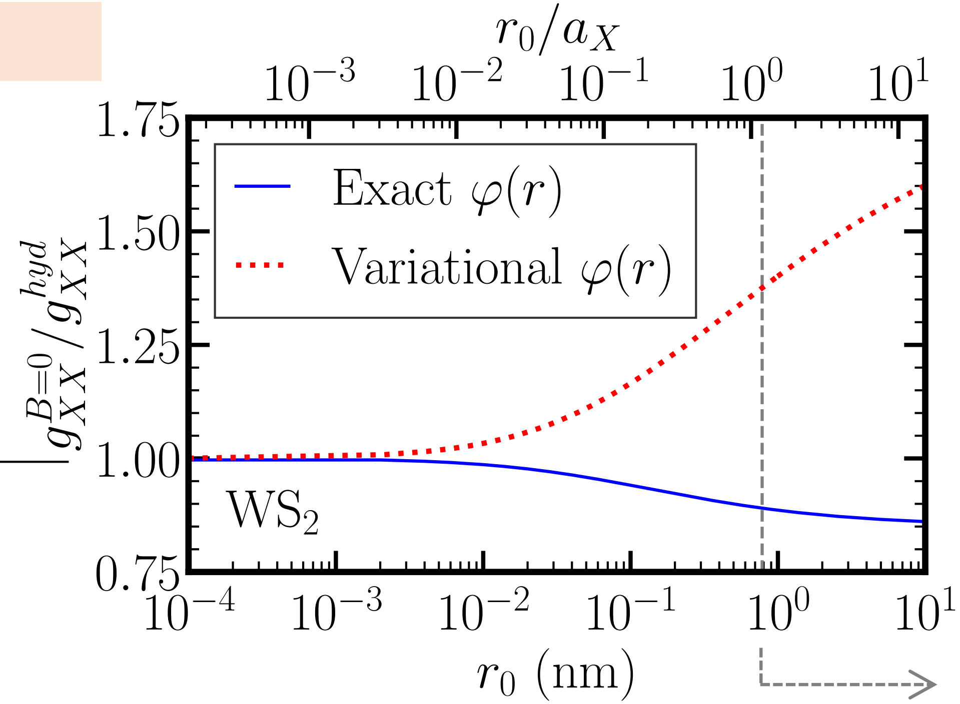
▷ TMD vs QW $g_{XX}^{B=0}$

Coulomb (universal)

$$\frac{g_{XX}^{hyd}}{\mathcal{A}} = \frac{6.0566}{2\mu}$$



▷ Rytova-Keldysh:
→ screening reduces $g_{XX}^{B=0}$
→ qualitatively incorrect results from variational approach [Shahnazaryan et al. PRB (2017)]
→ sensitivity on non-hydrogenic exciton wavefunction shape

$$\frac{g_{XX}^{B=0}|_{WS_2}}{\mathcal{A}} = \frac{5.39}{2\mu}$$

XX & PP interaction strength [de la Fuente et al., PRB (2025)]

▷ Born Approximation

$$\left|P^2\right\rangle = \hat{P}^\dagger \hat{P}^\dagger \left|0\right\rangle$$

$$\frac{g_{PP}}{\mathcal{A}} = \frac{\left\langle P^2\right| \hat{H} \left|P^2\right\rangle}{\left\langle P^2 | P^2\right\rangle} - 2E$$

1. analytical
2. $g_{XX} = g_{PP}|_{g=0=\Omega}$
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4. $g_{PP}^{B=0}$ recovers same results from [Levinsen et al. PRR (2019)]

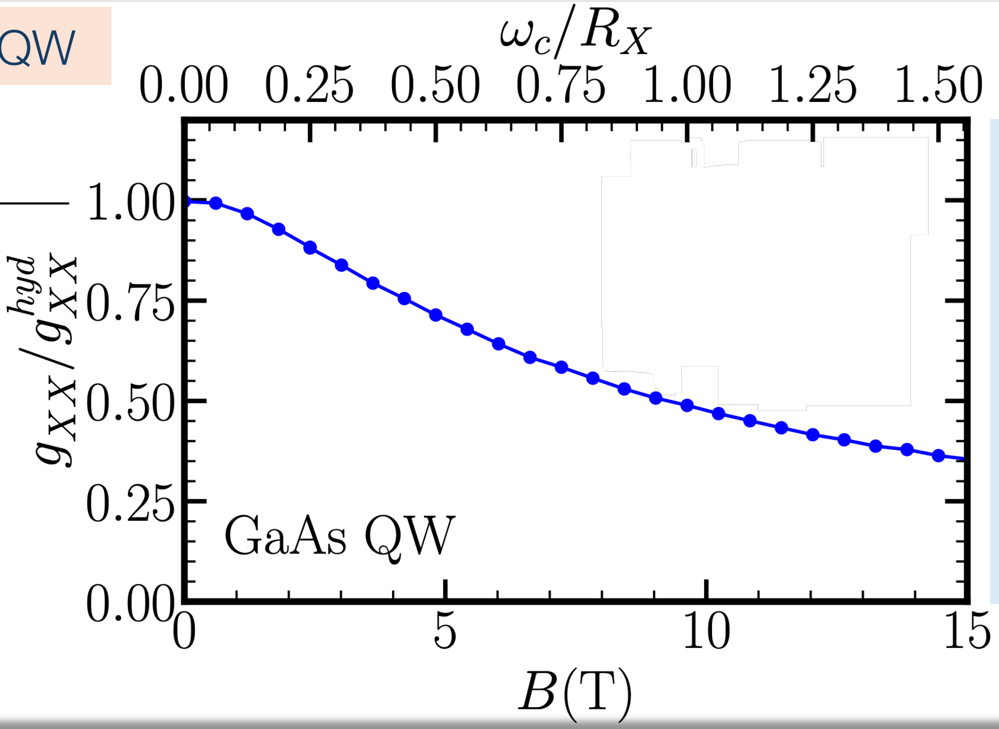
→ strict upper bound on the interaction strength [Li et al., PRB (2021)]

→ no momentum transfer: exchange interactions only

▷ B-dependence in QW

Coulomb (universal)

$$\frac{g_{XX}^{hyd}}{\mathcal{A}} = \frac{6.0566}{2\mu}$$



▷ Coulomb

→ decrease of g_{XX} with B due to the reduction of the exciton size

▷ Rytova-Keldysh

→ weak dependence on B

$$\frac{g_{XX}^{B=60\ \mathrm{T}}\big|_{\mathrm{WS_2}}}{\mathcal{A}} = \frac{5.04 \pm 0.02}{2\mu}$$

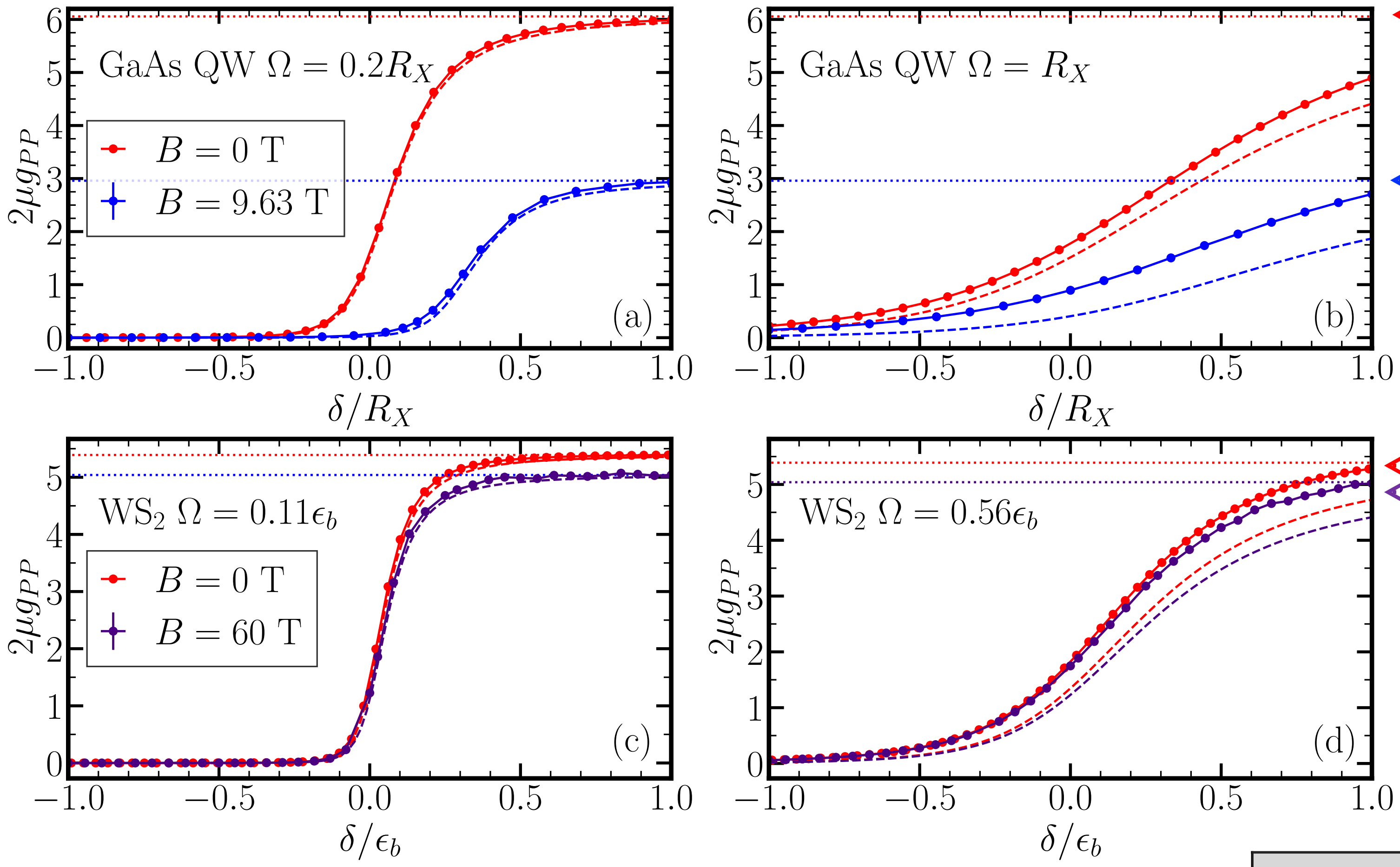
PP interaction strength [de la Fuente et al., PRB (2025)]

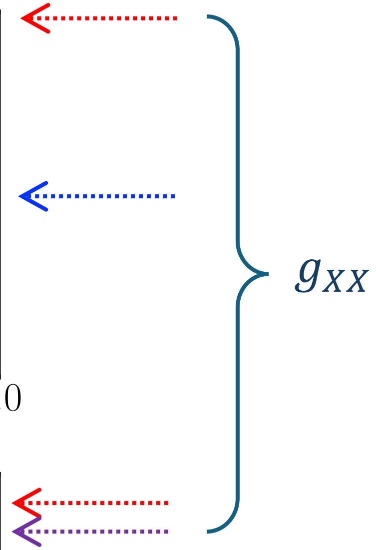
perturbative light-matter regime

very strong light-matter regime

Coulomb

Rytova-Keldysh



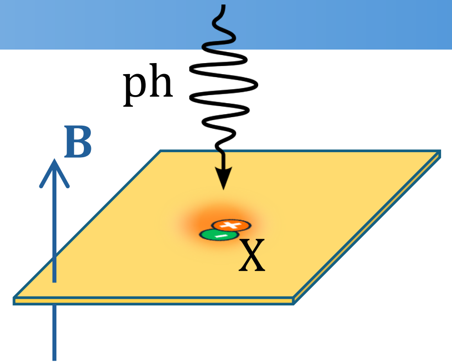


stronger non-perturbative light-matter effects with increasing magnetic field

- - - -	$g_{PP}^{(0)} = \beta^4 g_{XX}^{(0)}$
——	full numerics

Take 🏡 messages

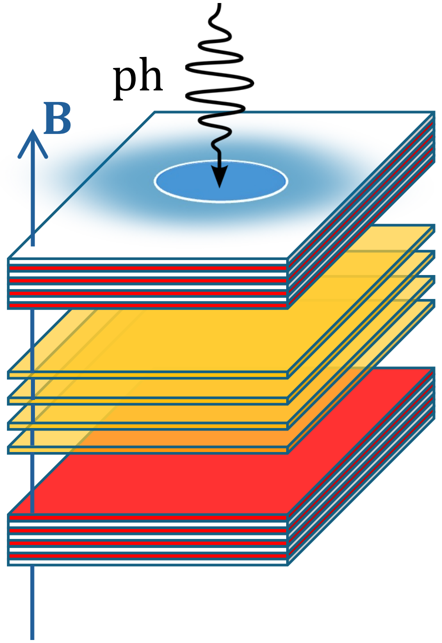
1. Magneto-optical spectroscopy
   → probing & tuning exciton properties
   → enhanced exciton binding & coupling to light



NON-PERTURBATIVE MICROSCOPIC THEORY

2. Cavity polaritons: Very-strong coupling $\Omega \sim E_B$
   → hybridization of multiple excitonic states
   → unambiguous signatures in the diamagnetic shift
   → probe the modifications of the e-h wavefunction due to very-strong coupling to light
   → agreement with experiments

3. g_{XX} & g_{PP} interaction strengths
   → $g_{XX}^{B=0}$ TMD monolayer: failure of variational approaches
   → magnetic field weakens interactions
   → light-induced modifications can enhance g_{PP} beyond g_{XX}



[Laird et al. PRB (2022)]
[de la Fuente Pico et al. PRB (2025)]

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