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# Modification of Exciton-Polariton Transport Coupled to Phononic Bath

Saeed Rahmanian Koshkaki

References:

- 1. **SRK**, A. Manjalingal, L. Blackham, and A. Mandal. arXiv:2502.12933 (2025).
- 2. L. Blackham, A. Manjalingal, **SRK**, and A. Mandal. arXiv:2501.16622 (2025).

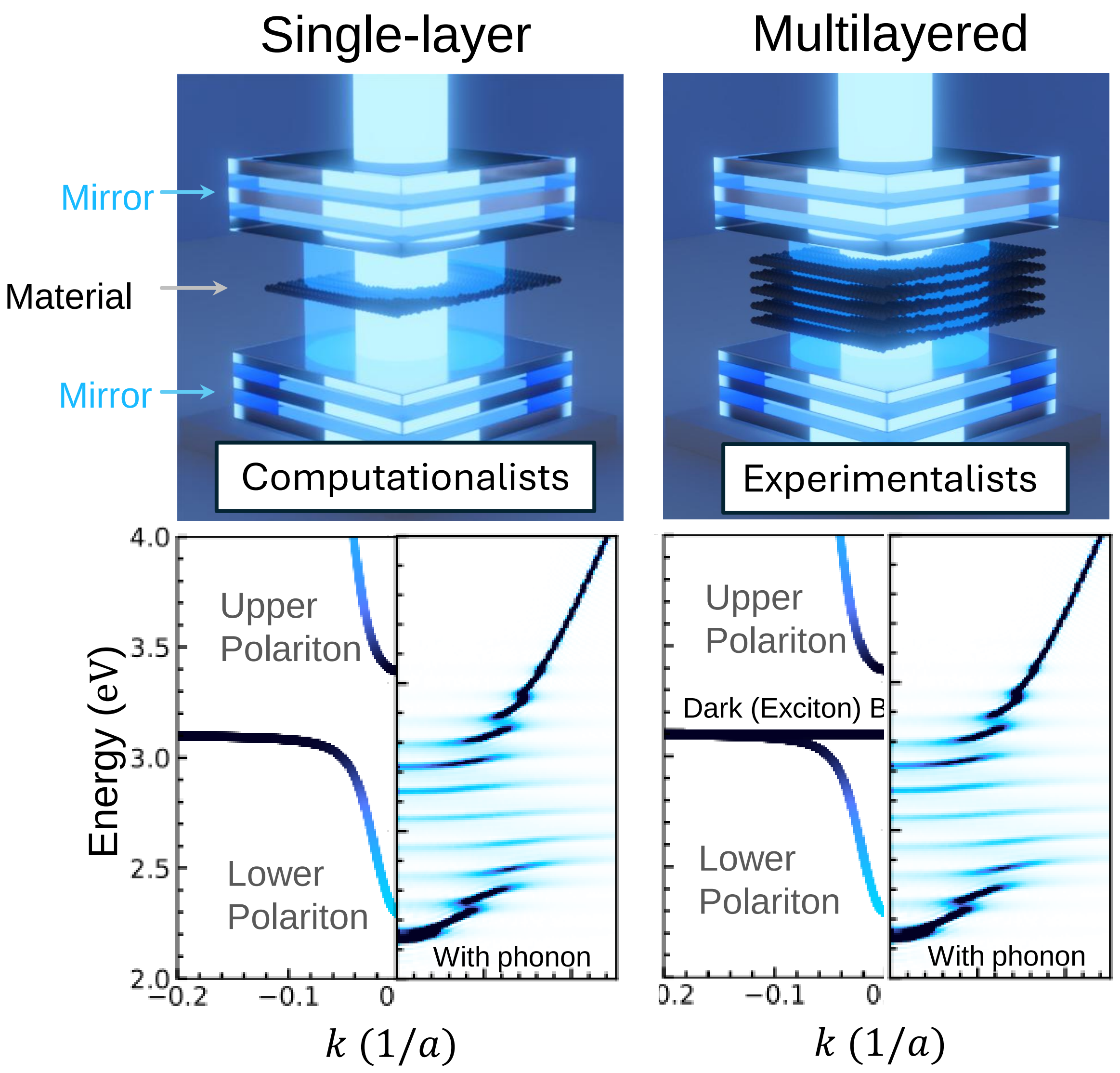
VISTA

April 2025

# Transport in Multilayered Materials

## Experiment vs Numerical simulations of exciton-polariton

- Experiments predominantly filled the entire optical cavity
- Theories mostly use a single layer to model the material inside a cavity
- Vibronic bands formation in the presence of exciton-phonon coupling



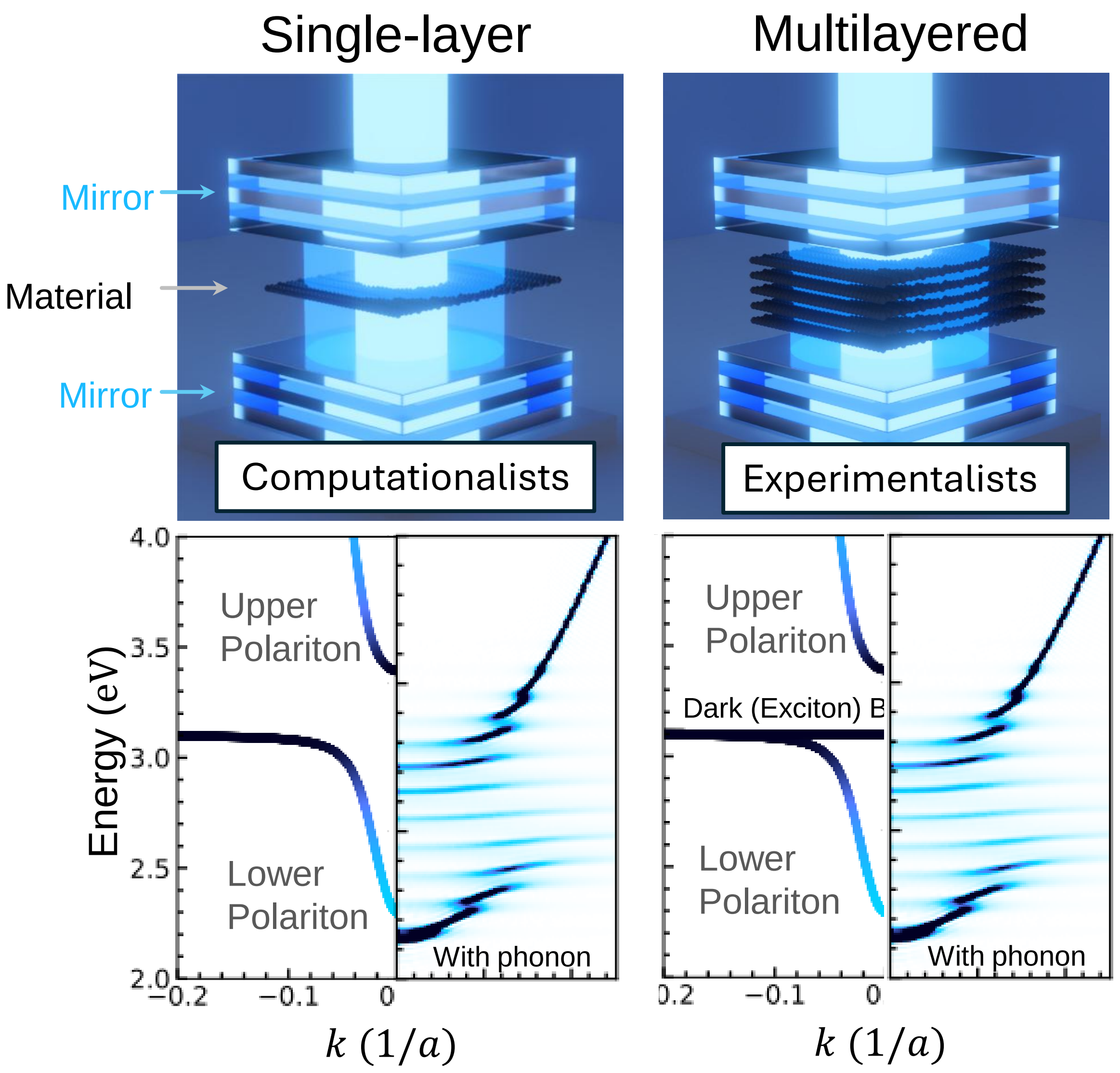

**SRK**, et al. arXiv:2502.12933 (2025).

# Main Results

1. We developed a mixed quantum classical method that allow us to propagate ~ 1,000,000 quantum states (Hilbert space size).

2. We developed a microscopic theory to describe the vibronic bands

3. We show that exciton-polariton dynamics is more coherent in multilayered materials.

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# Outline

## Model and Theory

$$\hat{H}_{\mathrm{LM}} = \hat{H}_{\mathrm{EP}} + \hat{H}_{\mathrm{env}}$$

$$|\Psi(t+\delta t)\rangle = e^{-i\hat{H}_{\mathrm{LM}}\delta t}|\Psi(t)\rangle$$

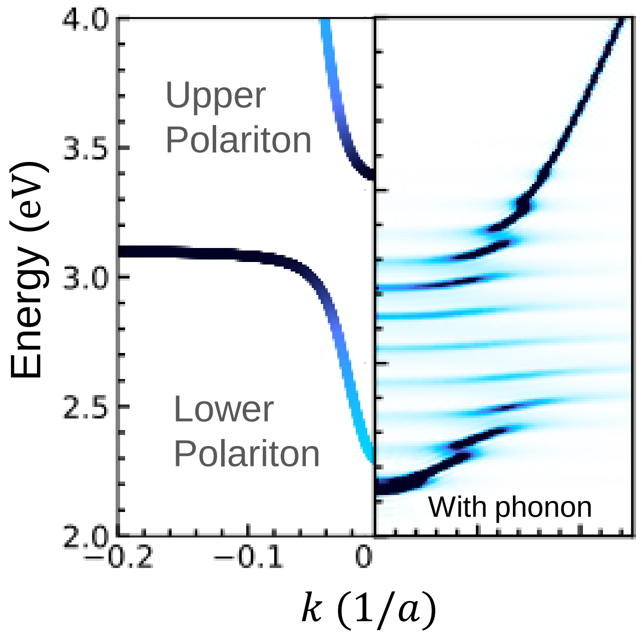
$$\dot{P}_n(t) = -\left\langle \Psi(t) \middle| \frac{\mathrm{d}\hat{H}_{\mathrm{LM}}}{\mathrm{d}R_n} \middle| \Psi(t) \right\rangle,$$

$$\dot{R}_n(t) = P_n(t)$$

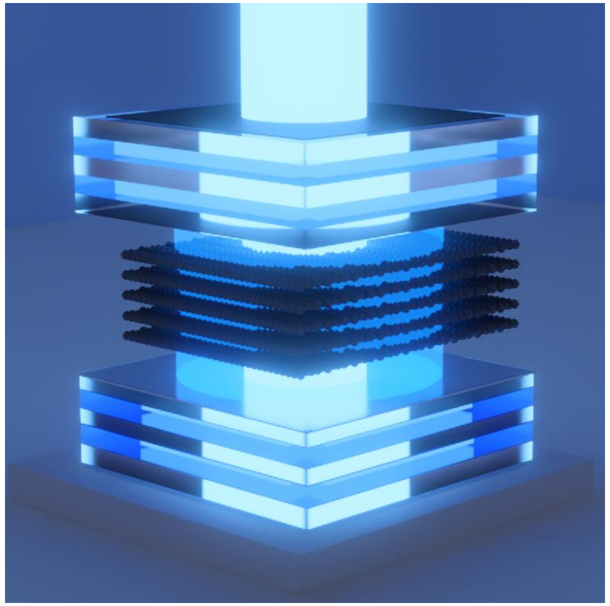
Bright Layer and Dark Layers

$$\hat{U}_{\mathrm{ft}}\hat{U}_{\mathrm{B}}$$

## Results 1: Vibronic Bands




## Results 1: Transport in Multilayered Materials



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## Results 1: Vibronic Bands




## Results 1: Transport in Multilayered Materials

# Model and Theory

Holstein-Tavis-Cummings Light-Matter (LM) Hamiltonian for an exciton-polariton system beyond the long-wavelength approximation

1. Exciton coupled to a single frequency phonon ensemble (same oscillation frequency)
2. There is no inter-layer hopping (for multilayered materials)

$$\hat{H}_{\mathrm{LM}} = \boxed{\hat{H}_{\mathrm{EP}}} + \boxed{\hat{H}_{\mathrm{env}}}$$

- Contains bare exciton, bare photon, and exciton-photon interaction terms (photon in ***k*-space**)

- Contains cavity photon loss and exciton-phonon interaction terms (photon in ***k*-space**)

# Model and Theory

We perform time evolution using mixed-quantum-classical mean field Ehrenfest (MFE) method.

1. Phonons are considered quasi-classically ($R_n(t)$ and $P_n(t)$)
2. Exciton and photon evolve quantum mechanically

↓

Mixed Quantum Classical Dynamics

$$\dot{P}_n(t) = -\left\langle \Psi(t) \middle| \frac{\mathrm{d}\hat{H}_{\mathrm{LM}}}{\mathrm{d}R_n} \middle| \Psi(t) \right\rangle, \quad \dot{R}_n(t) = P_n(t)$$

$$i|\dot{\Psi}(t)\rangle = \left[\hat{H}_{\mathrm{LM}} - \frac{1}{2}\sum_n \frac{P_n^2}{2}\right]|\Psi(t)\rangle$$

# Model and Theory

The exciton-photon state evolves using

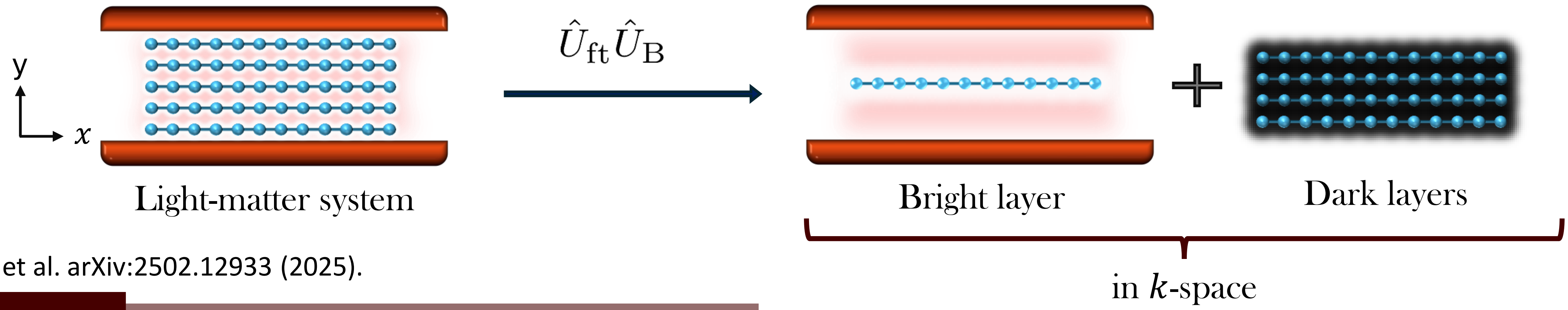
$$\hat{H}_{\mathrm{LM}} = \hat{H}_{\mathrm{EP}} + \hat{H}_{\mathrm{env}}$$

$$|\Psi(t+\delta t)\rangle = e^{-i\hat{H}_{\mathrm{LM}}\delta t}|\Psi(t)\rangle \approx e^{-i\hat{H}_{\mathrm{env}}\delta t/2} e^{-i\hat{H}_{\mathrm{EP}}\delta t} e^{-i\hat{H}_{\mathrm{env}}\delta t/2} |\Psi(t)\rangle$$

$$\approx e^{-i\hat{H}_{\mathrm{env}}\delta t/2} \hat{U}_{\mathrm{ft}}\hat{U}_{\mathrm{B}} \cdot e^{-i(\hat{U}_{\mathrm{B}}^{\dagger}\hat{U}_{\mathrm{ft}}^{\dagger}\hat{H}_{\mathrm{EP}}\hat{U}_{\mathrm{ft}}\hat{U}_{\mathrm{B}})\delta t} \hat{U}_{\mathrm{B}}^{\dagger}\hat{U}_{\mathrm{ft}}^{\dagger} e^{-i\hat{H}_{\mathrm{env}}\delta t/2} |\Psi(t)\rangle$$

Mapping to dark-bright layer

Fourier transform of exciton states

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Bright Layer and Dark Layers

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## Results 1: Vibronic Bands




## Results 1: Transport in Multilayered Materials

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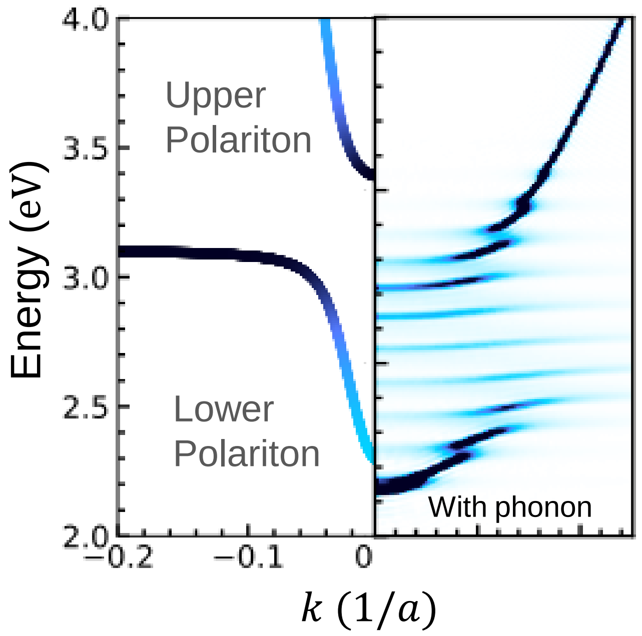



## Results 1: Transport in Multilayered Materials

# Vibronic Bands in Dispersion

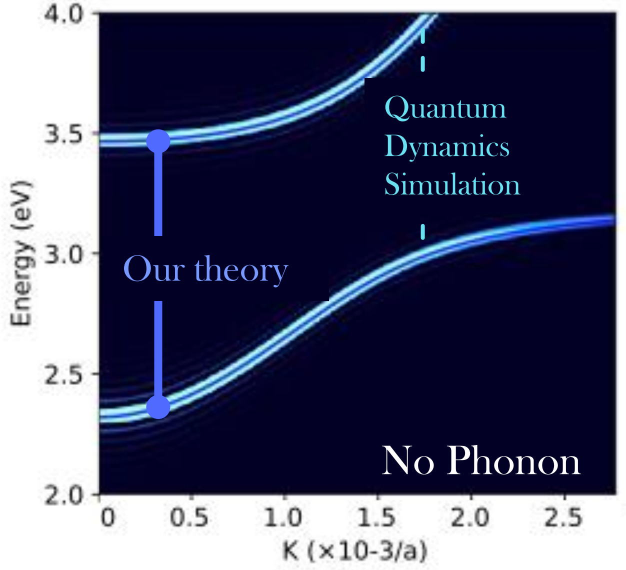
By treating phonon as Harmonic oscillators:

$$R_n(t) \approx R_n(0)\cos\omega t + \frac{1}{\omega}P_n(0)\sin\omega t$$

Floquet Model

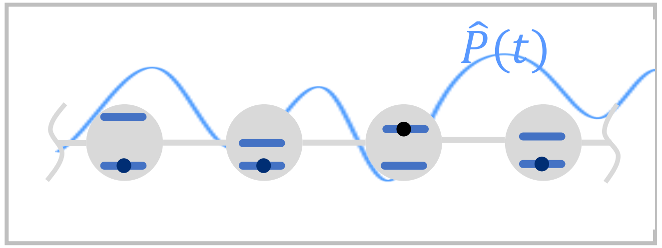
$$\hat{H}_{\mathrm{LM}} \rightarrow \hat{H}_{\mathrm{LM}}(t) = \hat{H}_{\mathrm{EP}} + \hat{P}e^{i\omega t} + \hat{P}^{\dagger}e^{-i\omega t}$$

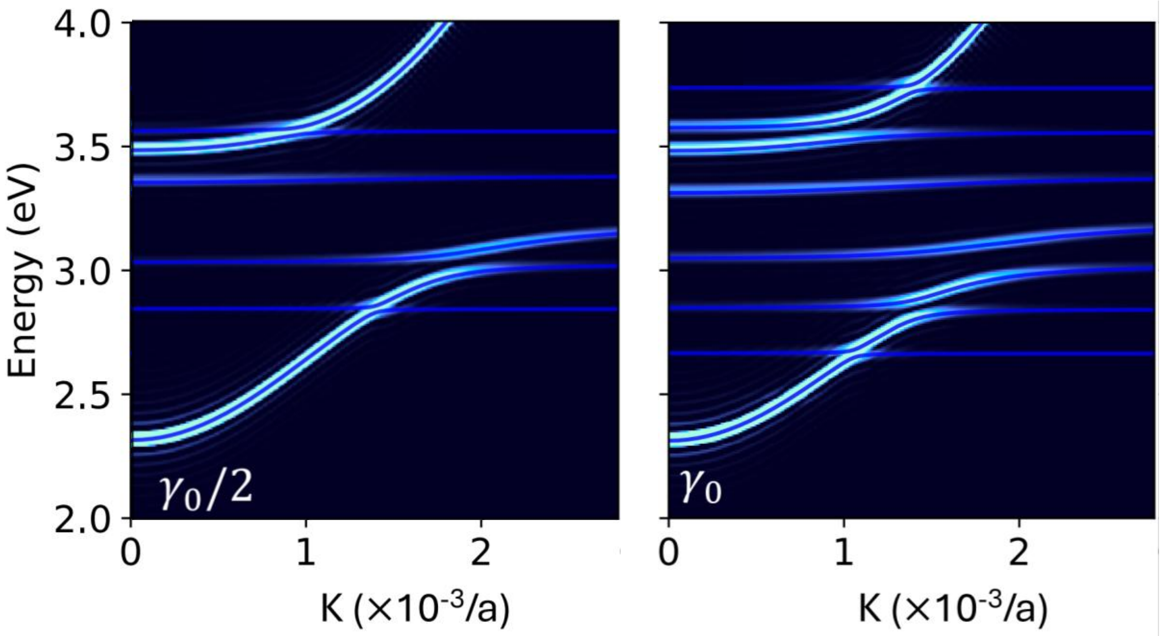
angle-resolved optical spectra




With Phonons

Exciton-Polariton is interacting *with a* time-dependent phonon field

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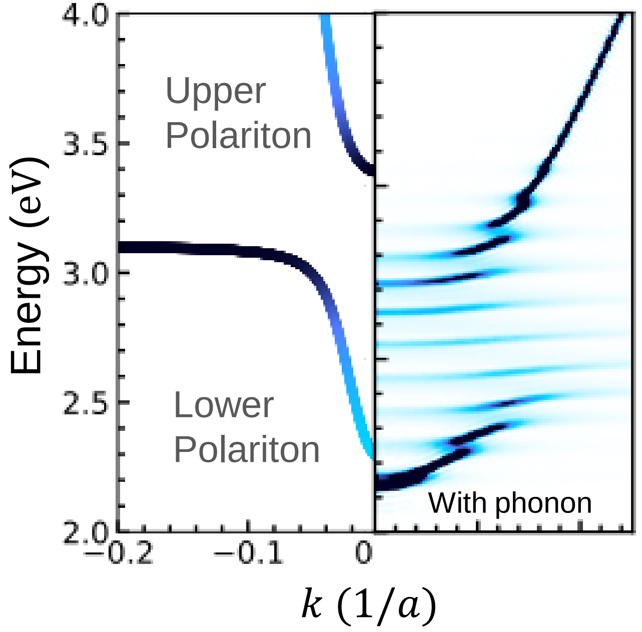


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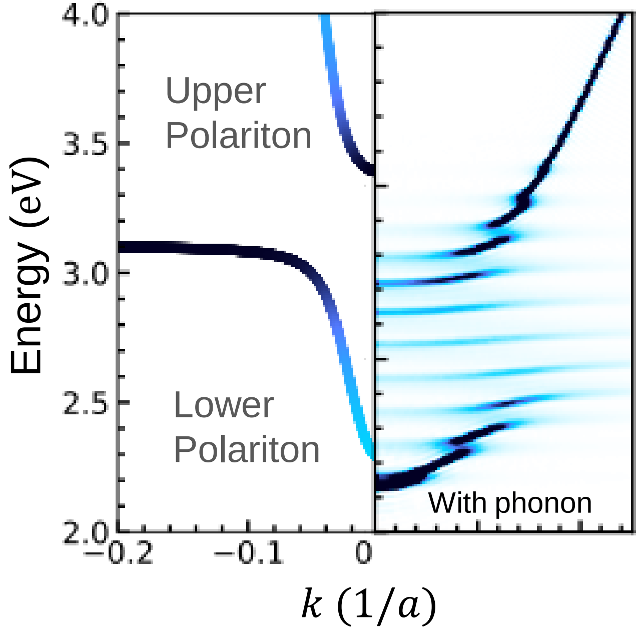


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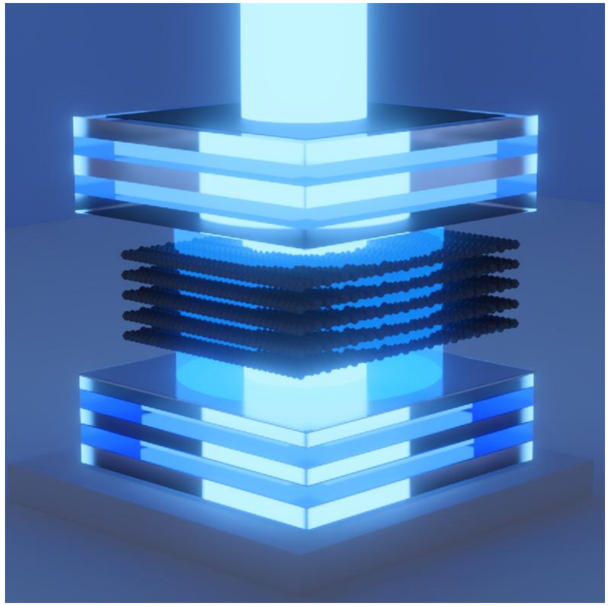
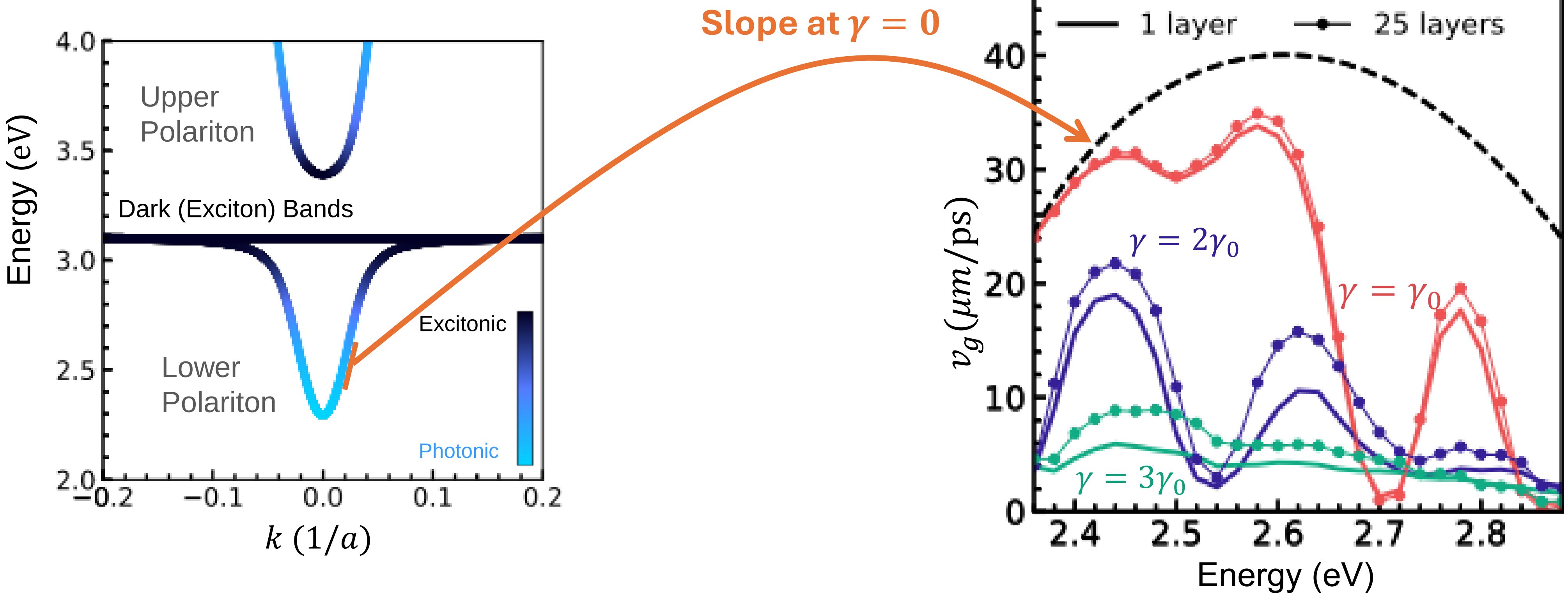


# Exciton group velocity

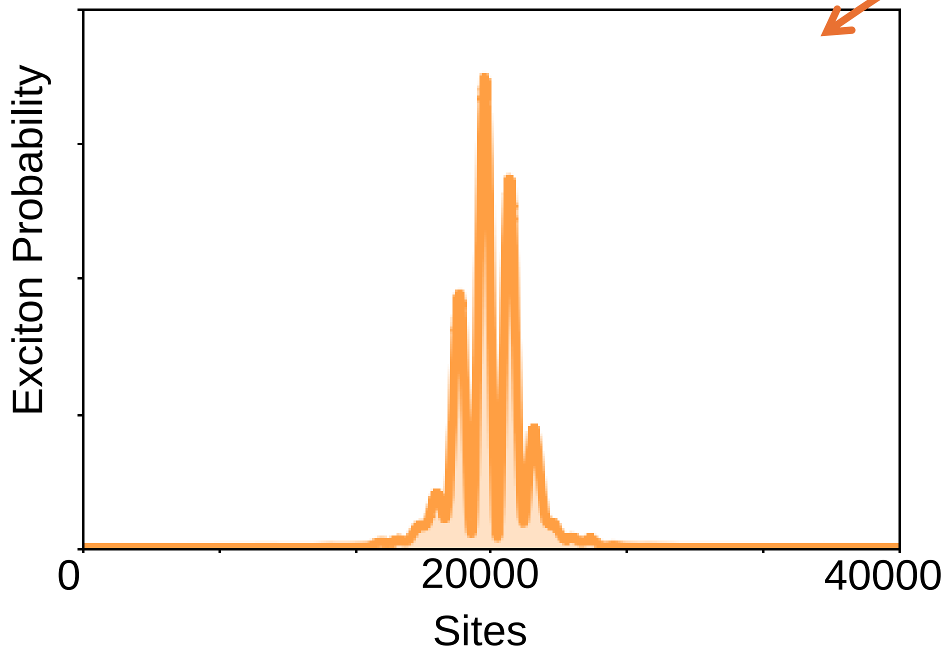
| | |
|---|---|
| $\gamma$: | Exciton-phonon coupling constant |
| $v_g$: | Group velocity |
| $E_0$: | Initial State energy |

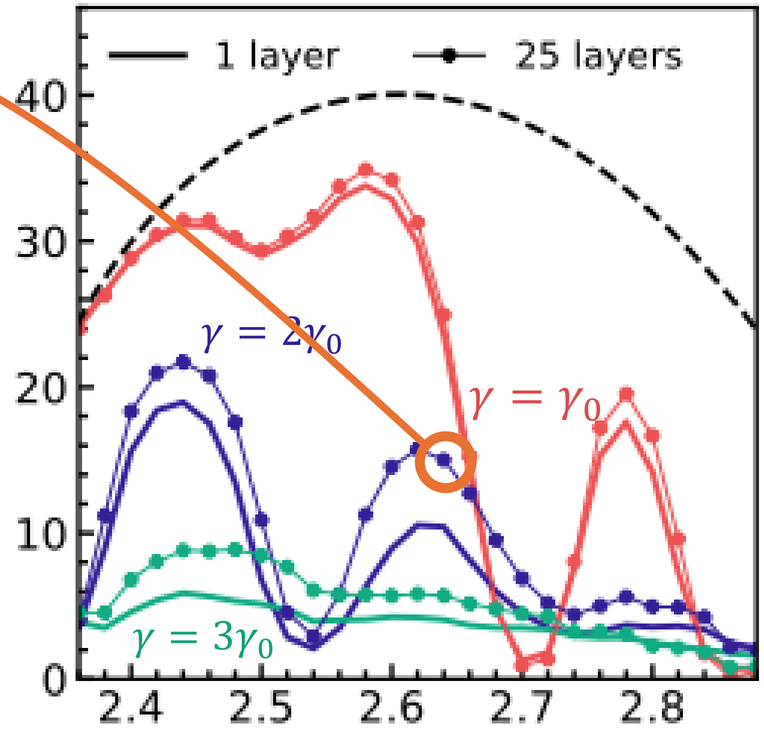



**SRK**, et al. arXiv:2502.12933 (2025).

# Exciton population transport over time

$\gamma$: Exciton-phonon coupling constant
$v_g$: Group velocity
$E_0$: Initial State energy

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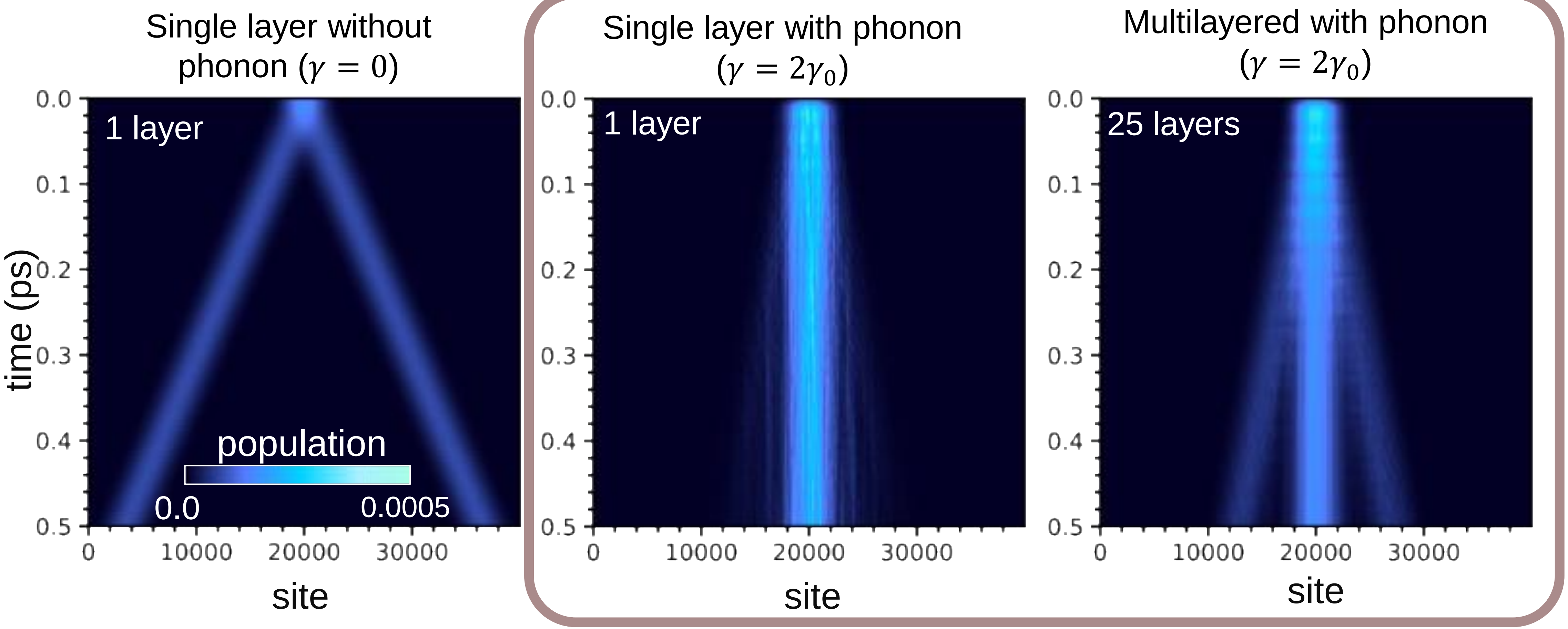


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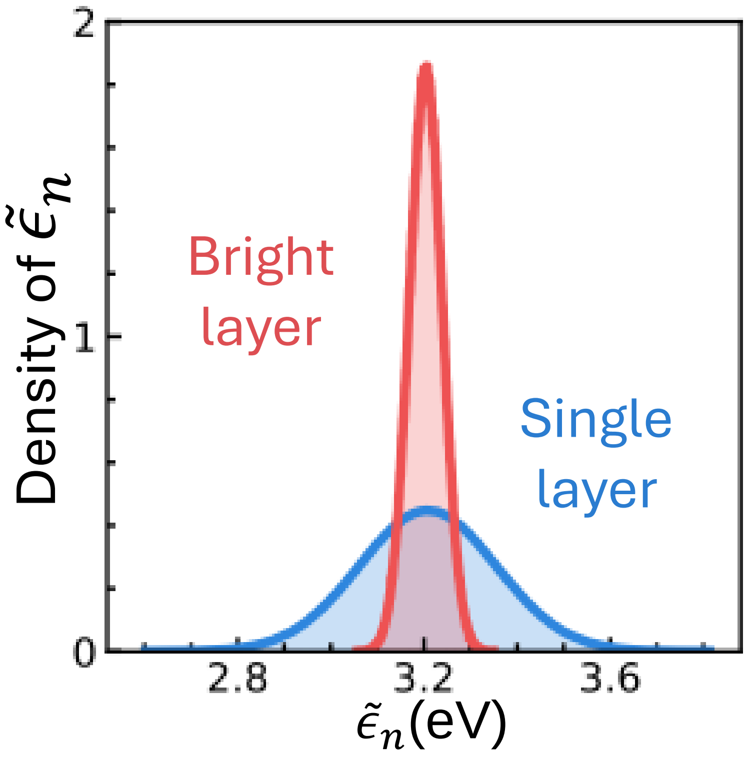
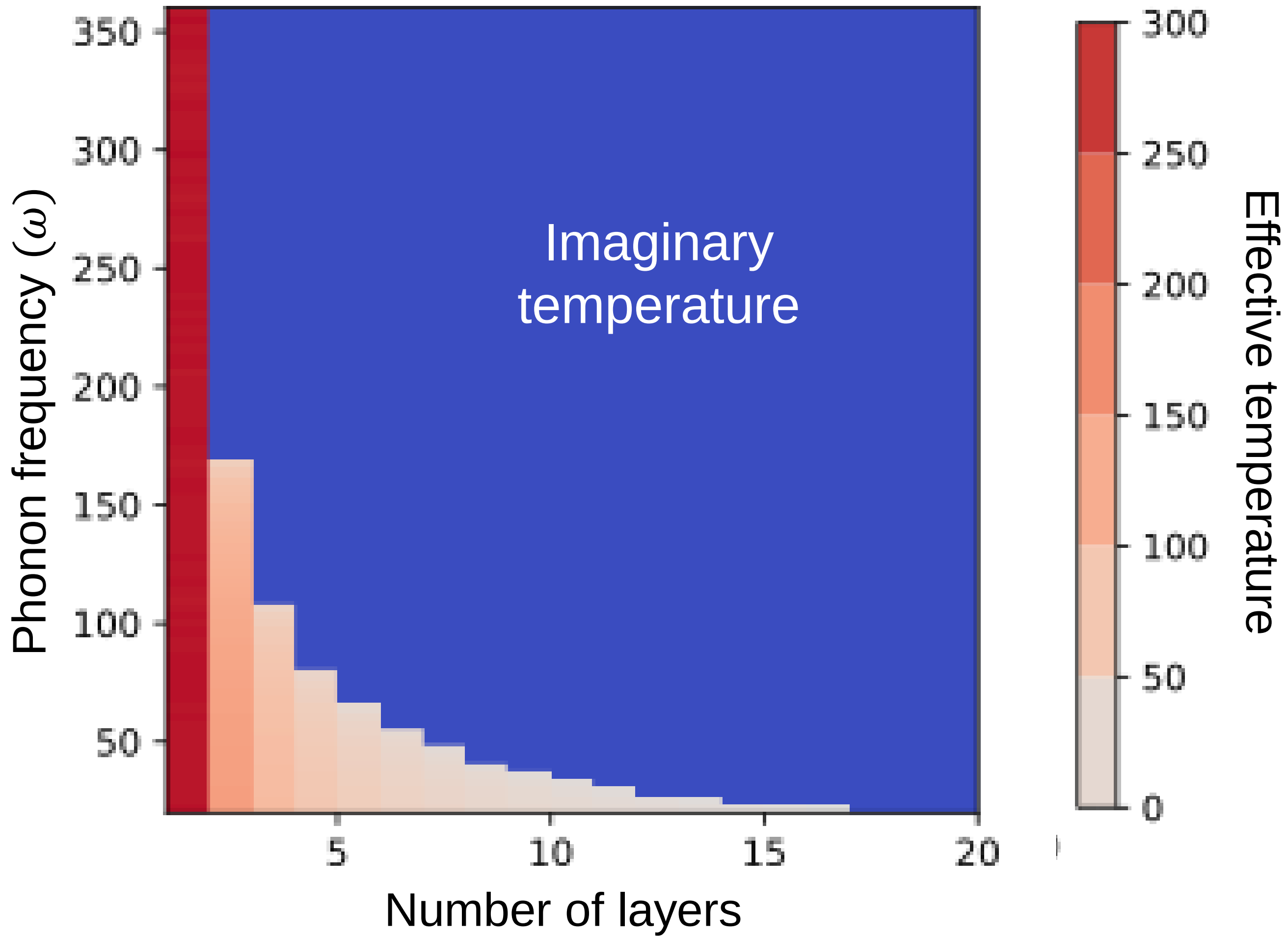
# Origin of enhanced transport

Phonon fluctuation synchronization in the Bright Layer

$\tilde{\epsilon}_n$ is the onsite energy modified by phonons

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# Conclusion

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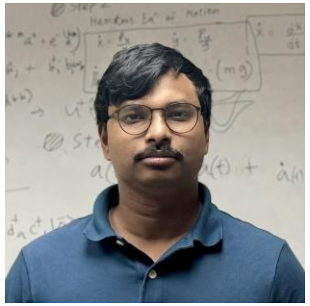
1. We developed a method enabling us to simulate exciton-polariton for an order of $10^7$ Hilbert space size.
2. Multilayered materials can be significantly different from single layer material, we studied the enhanced transport.
3. Multilayer materials show enhancement transport is even in the presence of cavity photon loss.
4. The enhancement of transport is multilayered material is due to a synchronization of phonon fluctuations
5. The effective temperature in multilayer materials is lower than single layer materials. There is possibly a phase transition happening when we increase the number of layers.



Arkajit Mandal, PI

Arshath Manjalingal, Graduate student

Logan Blackham, Graduate student

ACCESS

HPRC

# Thank you!



# Supporting Slides

# Main Results

1. <u>We developed a mixed quantum classical method</u> that allow us to propagate ~ 1,000,000 quantum states (Hilbert space size).

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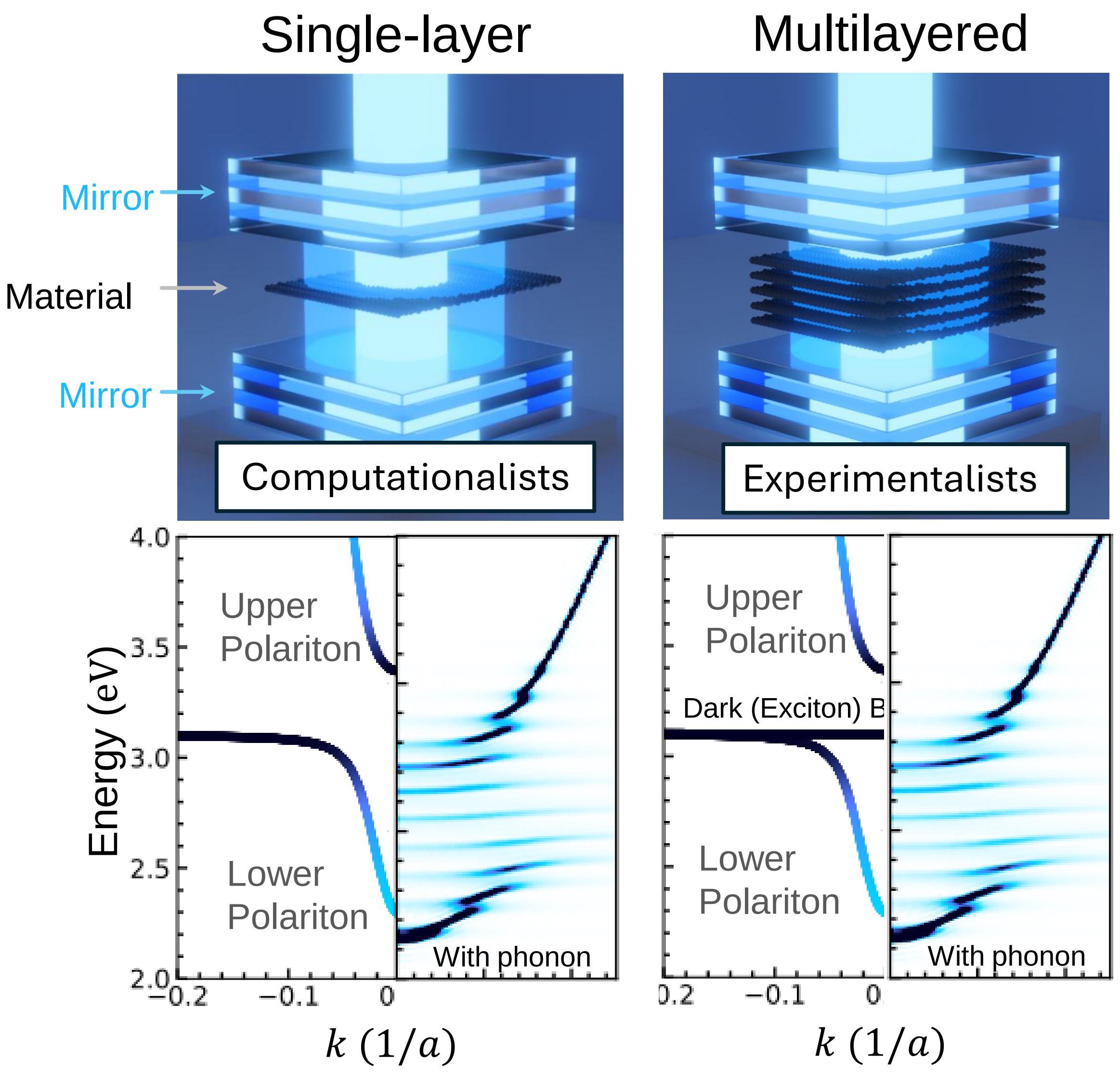


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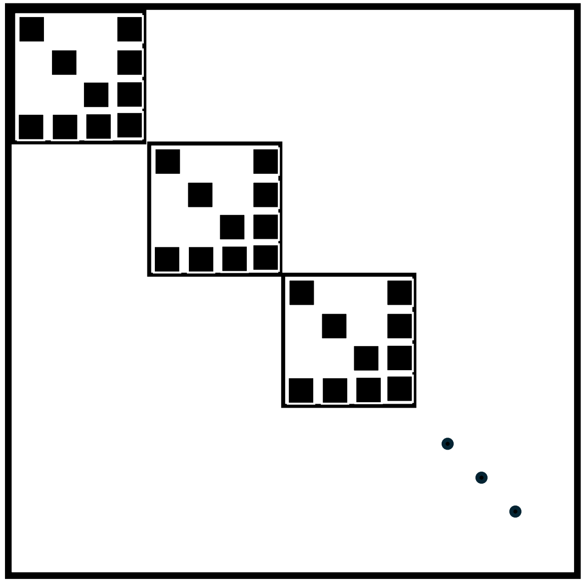
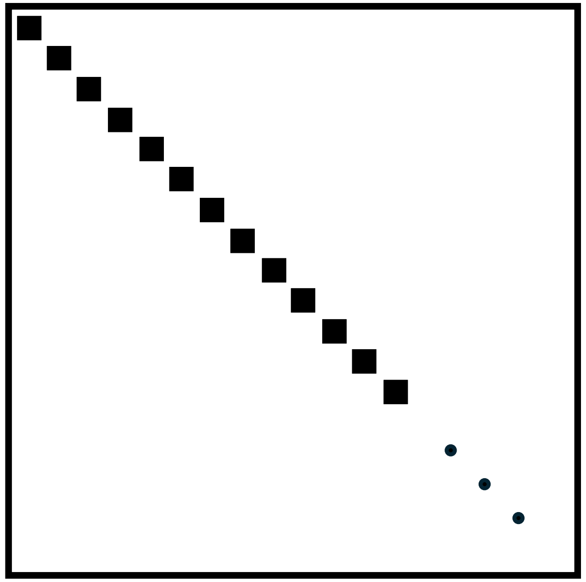
- Contains bare exciton, bare photon, and exciton-photon interaction terms (photon in ***k*-space**)
- It is block diagonal if exciton in ***k*-space**

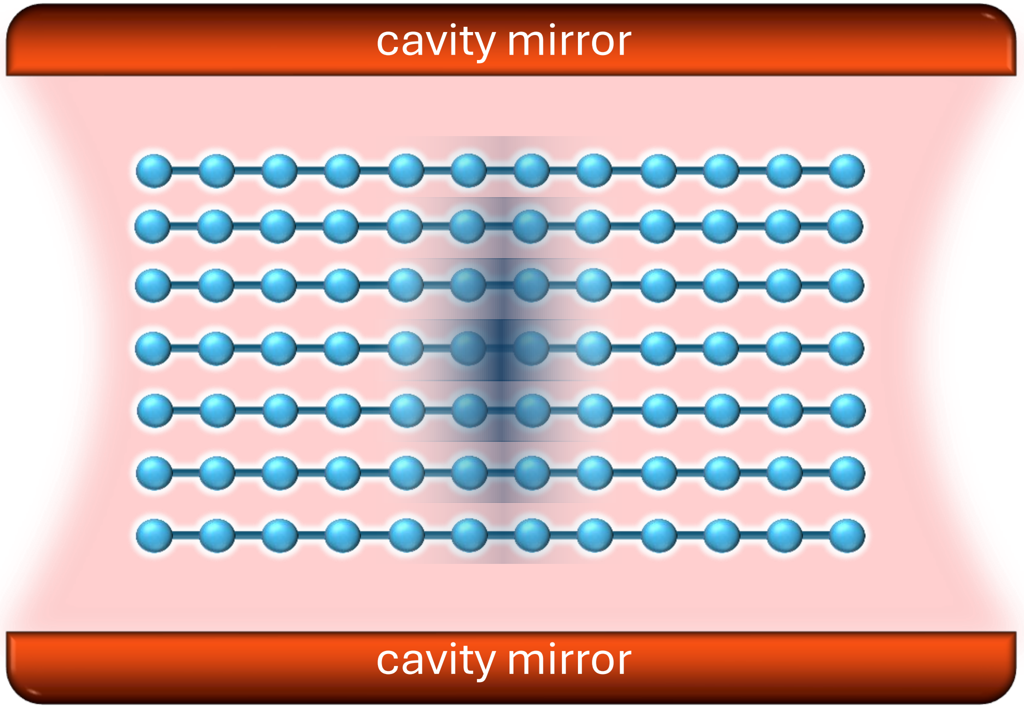


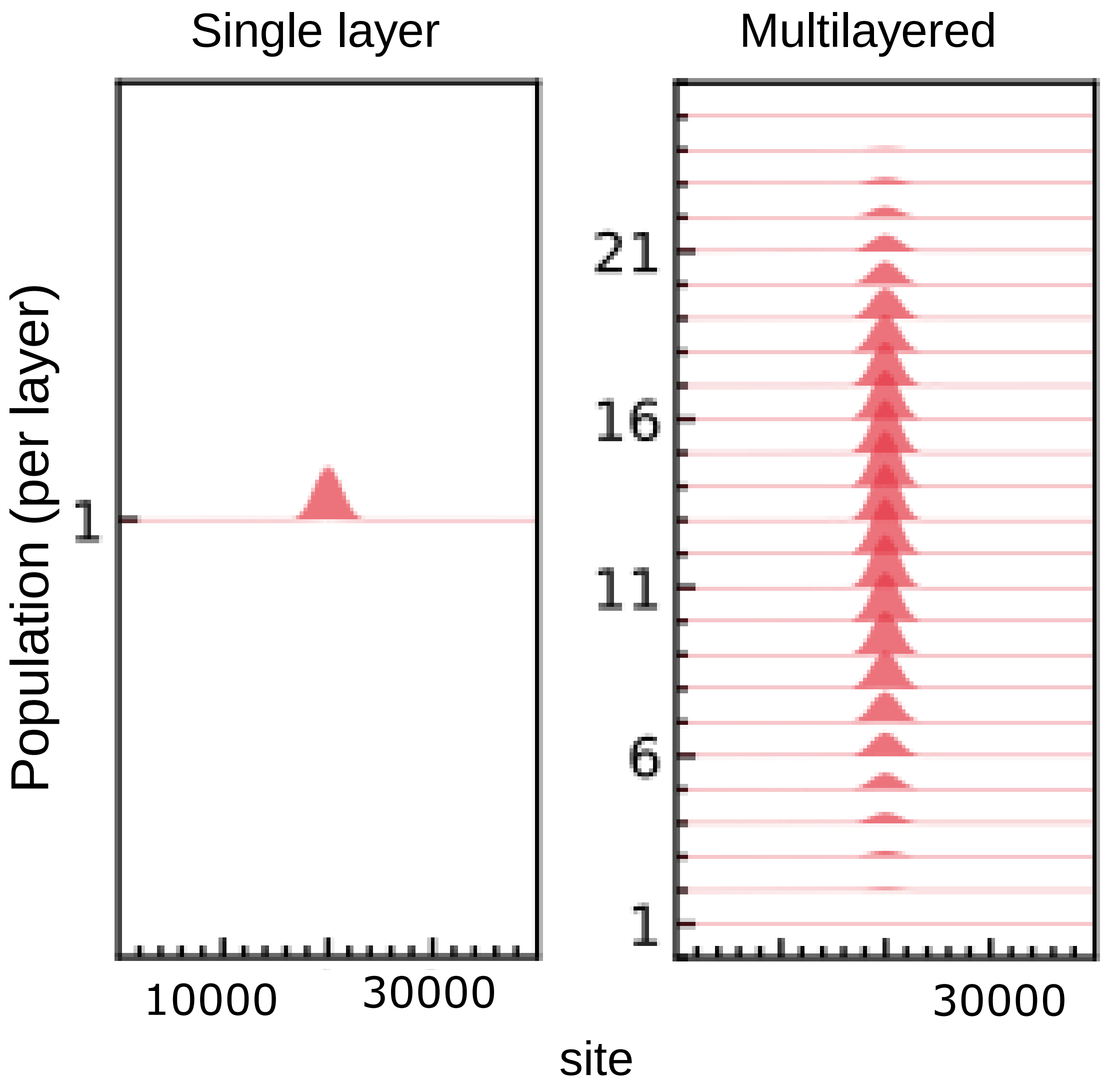
- Contains cavity photon loss and exciton-phonon interaction terms (photon in ***k*-space**)
- It is diagonal if exciton in **real space**



# Results

Generating initial state

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$$\approx e^{-i\hat{H}_{\mathrm{env}}\delta t/2} \hat{U}_{\mathrm{ft}}\hat{U}_{\mathrm{B}} \cdot e^{-i\boxed{(\hat{U}_{\mathrm{B}}^{\dagger}\hat{U}_{\mathrm{ft}}^{\dagger}\hat{H}_{\mathrm{EP}}\hat{U}_{\mathrm{ft}}\hat{U}_{\mathrm{B}})}\delta t} \hat{U}_{\mathrm{B}}^{\dagger}\hat{U}_{\mathrm{ft}}^{\dagger} e^{-i\hat{H}_{\mathrm{env}}\delta t/2} \, |\Psi(t)\rangle$$

$$\boxed{\sum_k \left[\epsilon_k \hat{X}^{\dagger}_{k,b}\hat{X}_{k,b} + \sqrt{S}\Omega_k \left(\hat{X}^{\dagger}_{k,b}\hat{a}_k + \mathrm{h.c.}\right) + \omega_k \hat{a}^{\dagger}_k \hat{a}_k\right] + \sum_{k,d} \epsilon_k \hat{X}^{\dagger}_{k,d}\hat{X}_{k,d}}$$

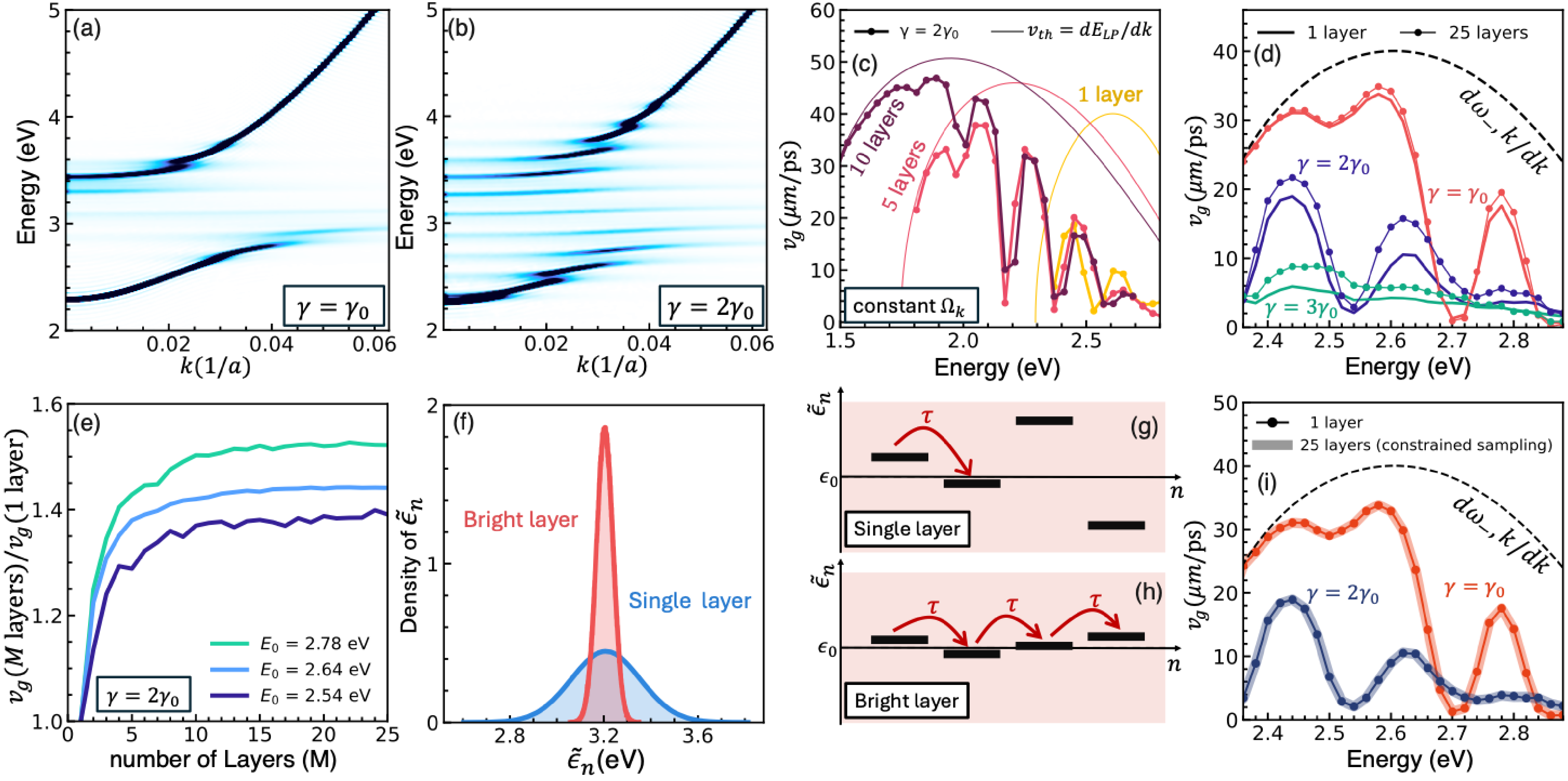
Saeed R. K., et al. arXiv:2502.12933 (2025).

# Results

we model the materials part using a system based on a multilayered perovskite material [2] and weak Huang–Rhys factor [3] in an optical cavity

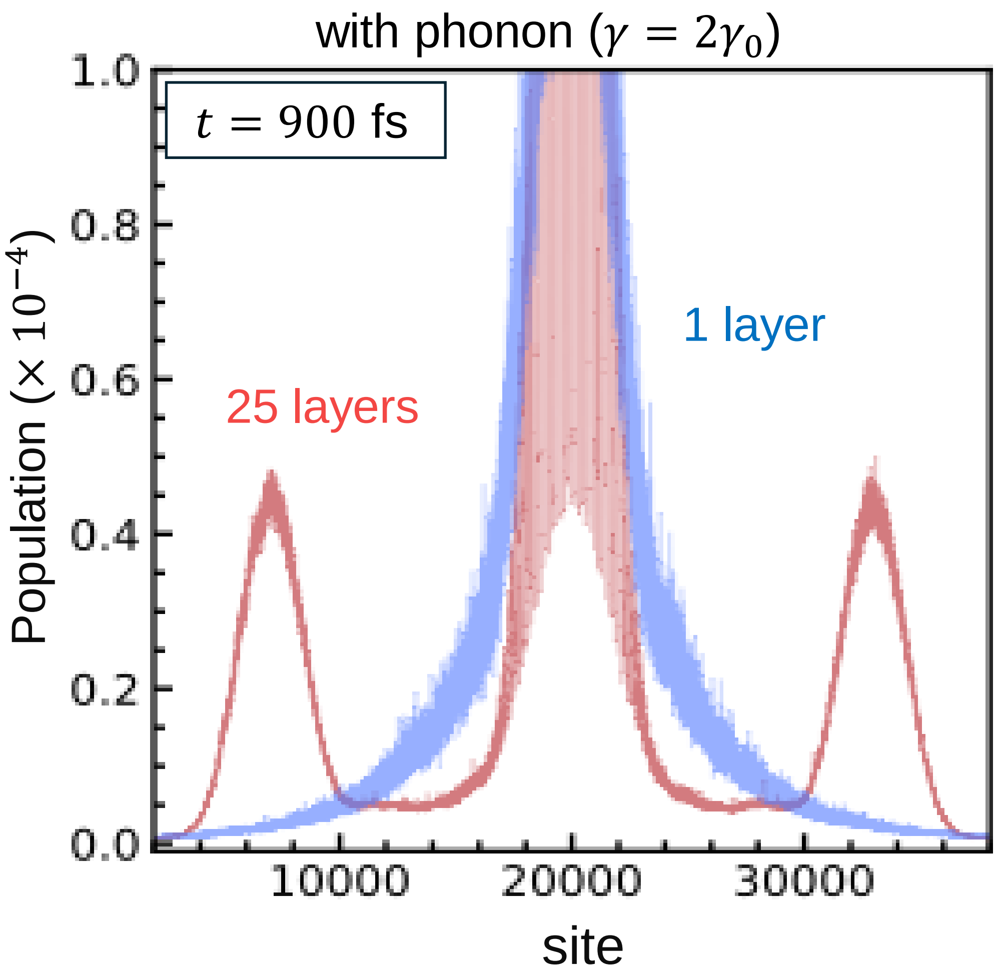
| | |
|---|---|
| Refractive index | $\eta = 2.4$ |
| Exciton hopping integral | $\tau = 400 \text{ cm}^{-1}$ |
| Exciton on-site energy | $\epsilon_0 = 3.2$ eV |
| Lattice spacing | $a = 1.2$ nm |
| Number of sites per layer | $N = 40001$ |
| Nuclear time step | $dtN \approx 00.48$ fs |
| Exciton time steps | $dtE \approx 0.012$ fs |
| Phonon mode frequency | $\omega = 1440 \text{ cm}^{-1}$ |
| $\boxed{\text{Exciton-phonon coupling constant}}$ | $\boxed{\gamma_0 = 1.1\sqrt{\omega^3} = 5.8 \times 10^{-4} \text{ a.u.}}$ |
| Exciton-photon coupling | $\Omega_0 = 480$ meV |
| Distance between two reflective mirrors of cavity | $L = 100\ nm$ |
| Interlayer spacing of multilayered material | $a_y = 4$ nm |
| $\boxed{\text{Rabi splitting}}$ | $\boxed{\approx 874 \text{ meV}}$ |


1. Saeed R. K., et al. arXiv:2502.12933 (2025).
2. Janke, Svenja M., et al. *The Journal of Chemical Physics* 152.14 (2020).
3. Whalley, Lucy D., et al. *Journal of the American Chemical Society* 143.24 (2021): 9123-9128.



(a)
Energy (eV)
$k(1/a)$
$\gamma = \gamma_0$
(b)
Energy (eV)
$k(1/a)$
$\gamma = 2\gamma_0$
(c)
$\gamma = 2\gamma_0$
$v_{th} = dE_{LP}/dk$
10 layers
5 layers
1 layer
constant $\Omega_k$
$v_g(\mu m/ps)$
Energy (eV)
(d)
1 layer
25 layers
$d\omega_{-,k}/dk$
$\gamma = 2\gamma_0$
$\gamma = \gamma_0$
$\gamma = 3\gamma_0$
$v_g(\mu m/ps)$
Energy (eV)
(e)
$v_g(M\text{ layers})/v_g(1\text{ layer})$
number of Layers (M)
$\gamma = 2\gamma_0$
$E_0$ = 2.78 eV
$E_0$ = 2.64 eV
$E_0$ = 2.54 eV
(f)
Density of $\tilde{\epsilon}_n$
Bright layer
Single layer
$\tilde{\epsilon}_n$(eV)
(g)
$\tilde{\epsilon}_n$
$\epsilon_0$
$n$
$\tau$
Single layer
(h)
$\tilde{\epsilon}_n$
$\epsilon_0$
$n$
$\tau$
Bright layer
(i)
1 layer
25 layers (constrained sampling)
$d\omega_{-,k}/dk$
$\gamma = 2\gamma_0$
$\gamma = \gamma_0$
$v_g(\mu m/ps)$
Energy (eV)

# Exciton population transport over time

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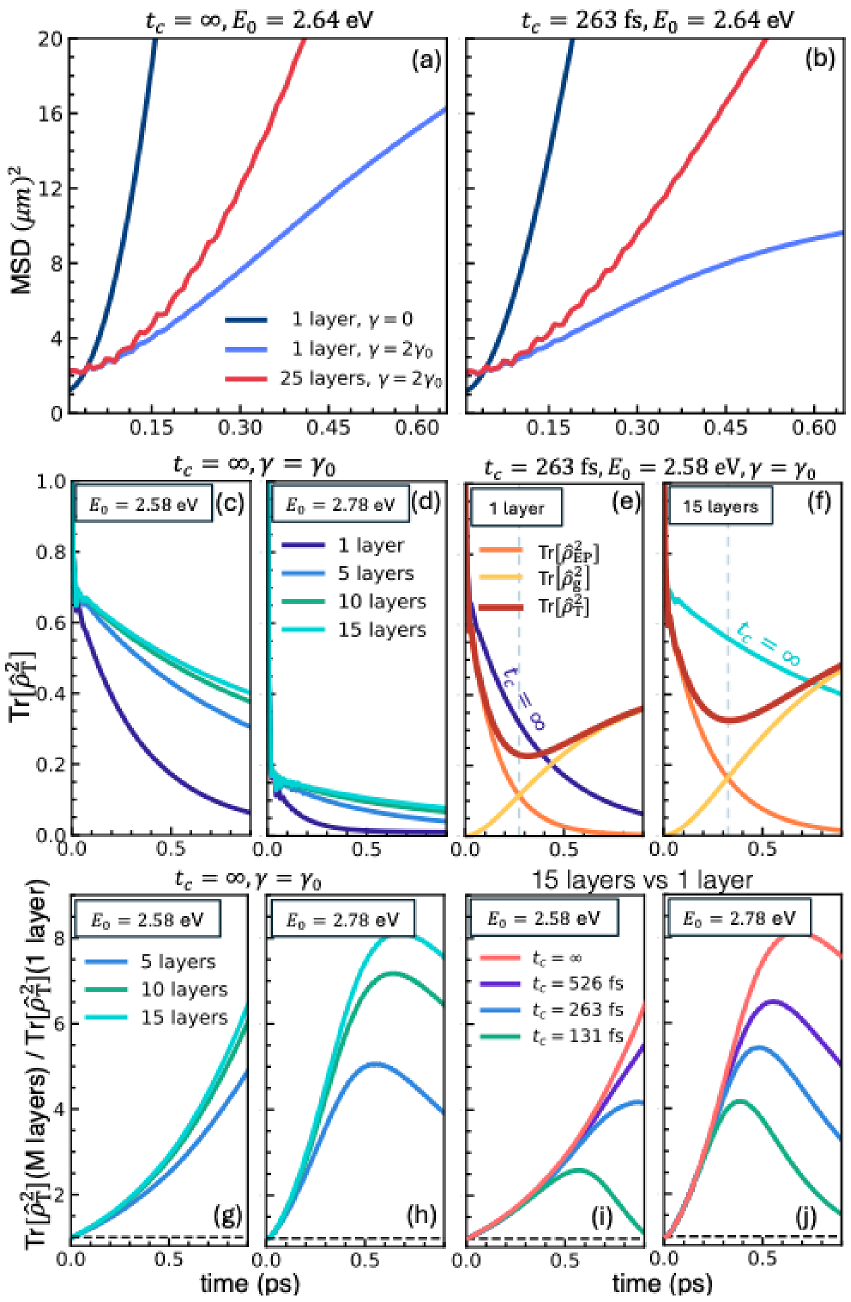
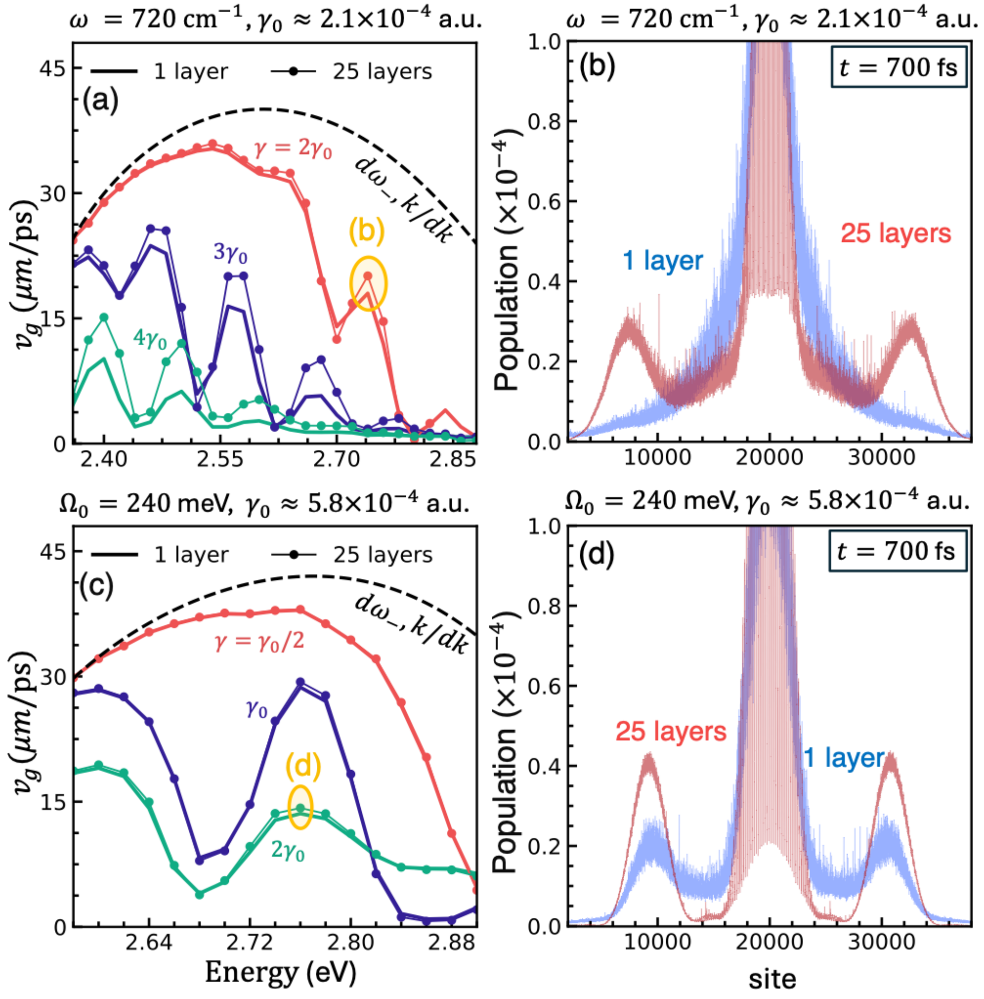


$t_c = \infty, E_0 = 2.64$ eV
$t_c = 263$ fs, $E_0 = 2.64$ eV
(a)
(b)
MSD $(\mu m)^2$
1 layer, $\gamma = 0$
1 layer, $\gamma = 2\gamma_0$
25 layers, $\gamma = 2\gamma_0$
$t_c = \infty, \gamma = \gamma_0$
$t_c = 263$ fs, $E_0 = 2.58$ eV, $\gamma = \gamma_0$
$E_0 = 2.58$ eV (c)
$E_0 = 2.78$ eV (d)
1 layer (e)
15 layers (f)
1 layer
5 layers
10 layers
15 layers
$\mathrm{Tr}[\hat{\rho}^2_{\mathrm{EP}}]$
$\mathrm{Tr}[\hat{\rho}^2_{\mathrm{g}}]$
$\mathrm{Tr}[\hat{\rho}^2_{\mathrm{T}}]$
$t_c = \infty$
$\mathrm{Tr}[\hat{\rho}^2_{\mathrm{T}}]$
$t_c = \infty, \gamma = \gamma_0$
15 layers vs 1 layer
$E_0 = 2.58$ eV
$E_0 = 2.78$ eV
5 layers
10 layers
15 layers
$t_c = \infty$
$t_c = 526$ fs
$t_c = 263$ fs
$t_c = 131$ fs
(g)
(h)
(i)
(j)
$\mathrm{Tr}[\hat{\rho}^2_{\mathrm{T}}]$(M layers) / $\mathrm{Tr}[\hat{\rho}^2_{\mathrm{T}}]$(1 layer)
time (ps)
time (ps)

$\omega = 720$ cm$^{-1}$, $\gamma_0 \approx 2.1\times10^{-4}$ a.u.
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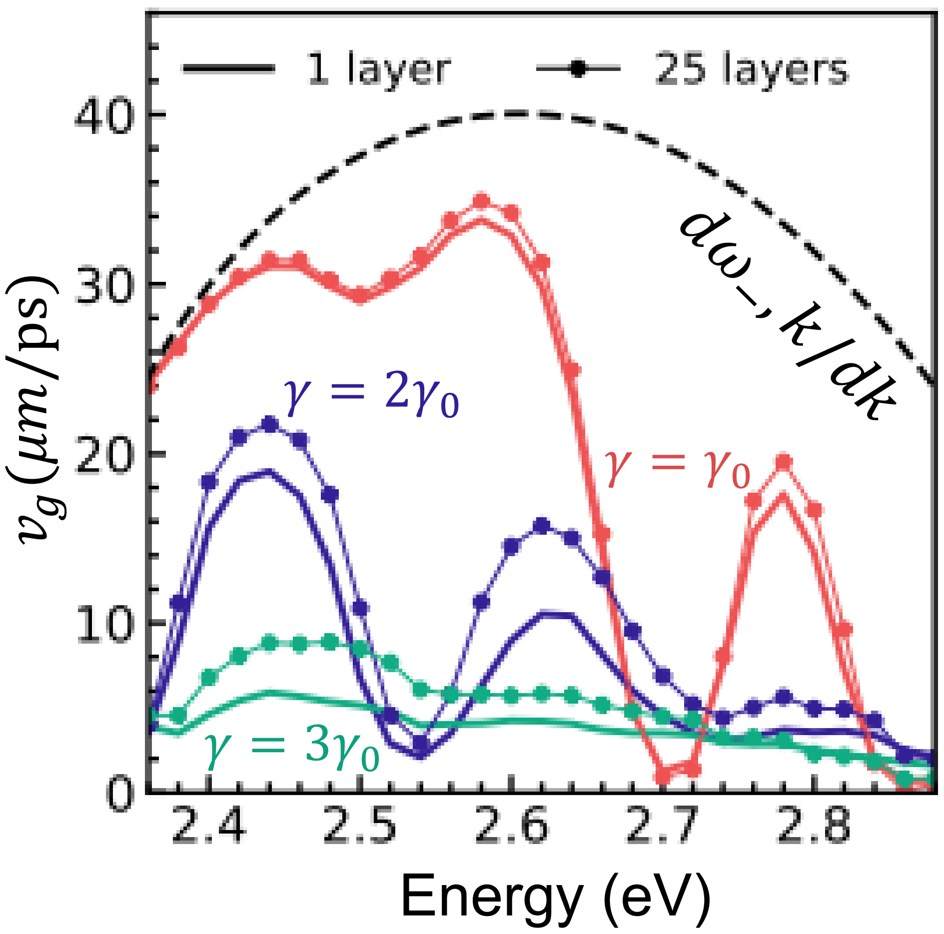
1 layer
25 layers
(a)
$\gamma = 2\gamma_0$
$d\omega_{-,k}/dk$
(b)
$3\gamma_0$
$4\gamma_0$
$v_g (\mu m/\mathrm{ps})$
(b)
$t = 700$ fs
1 layer
25 layers
Population $(\times 10^{-4})$
$\Omega_0 = 240$ meV, $\gamma_0 \approx 5.8\times10^{-4}$ a.u.
$\Omega_0 = 240$ meV, $\gamma_0 \approx 5.8\times10^{-4}$ a.u.
1 layer
25 layers
(c)
$d\omega_{-,k}/dk$
$\gamma = \gamma_0/2$
$\gamma_0$
(d)
$2\gamma_0$
$v_g (\mu m/\mathrm{ps})$
Energy (eV)
(d)
$t = 700$ fs
25 layers
1 layer
Population $(\times 10^{-4})$
site

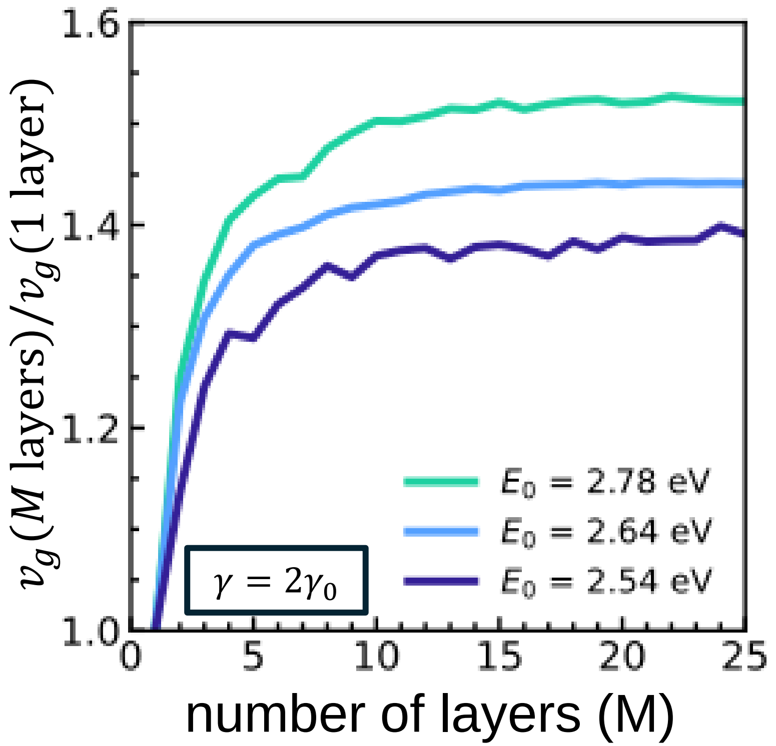
# Results

Exciton group velocity




SRK, et al. arXiv:2502.12933 (2025).

$\gamma$: Exciton-phonon coupling constant
$v_g$: Group velocity
$\omega_-$: Lower polariton energy dispersion
$E_0$: Initial State energy

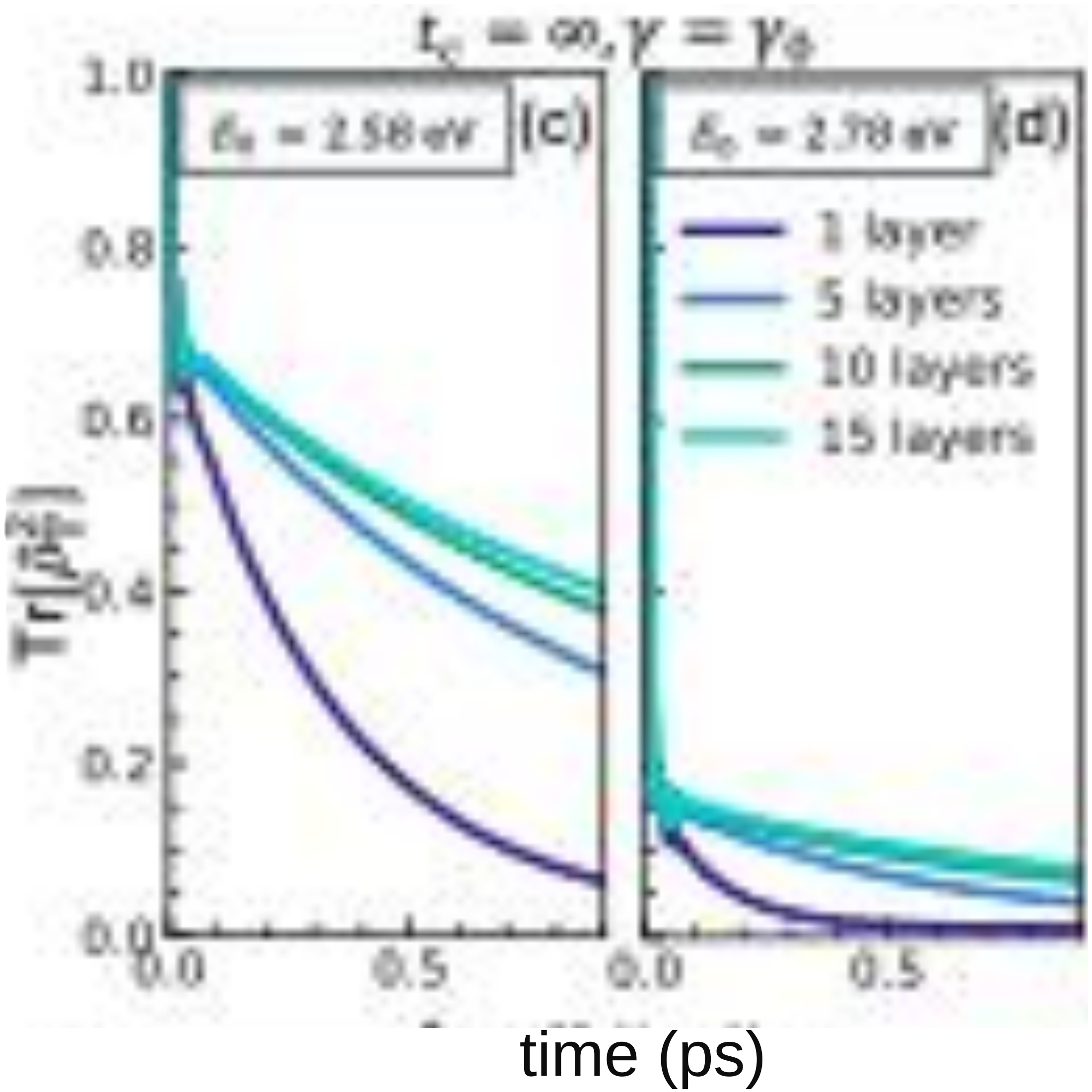


# Purity

We measure coherence using purity

- **<u>Purity of 1</u>** means pure state
- **<u>Purity less than 1</u>** means mixed state
- The larger purity, the more coherent
- decoherence means lower purity

Multi-layered materials extends coherence time by 10x time!
(via the phonon fluctuation Synchronization effects! )




Saeed R. K., et al. arXiv:2502.12933 (2025).