TEXAS A&M UNIVERSITY

Microscopic Theory of Polaron-Polariton Dispersion & Propagation

Logan Blackham

Mandal Group

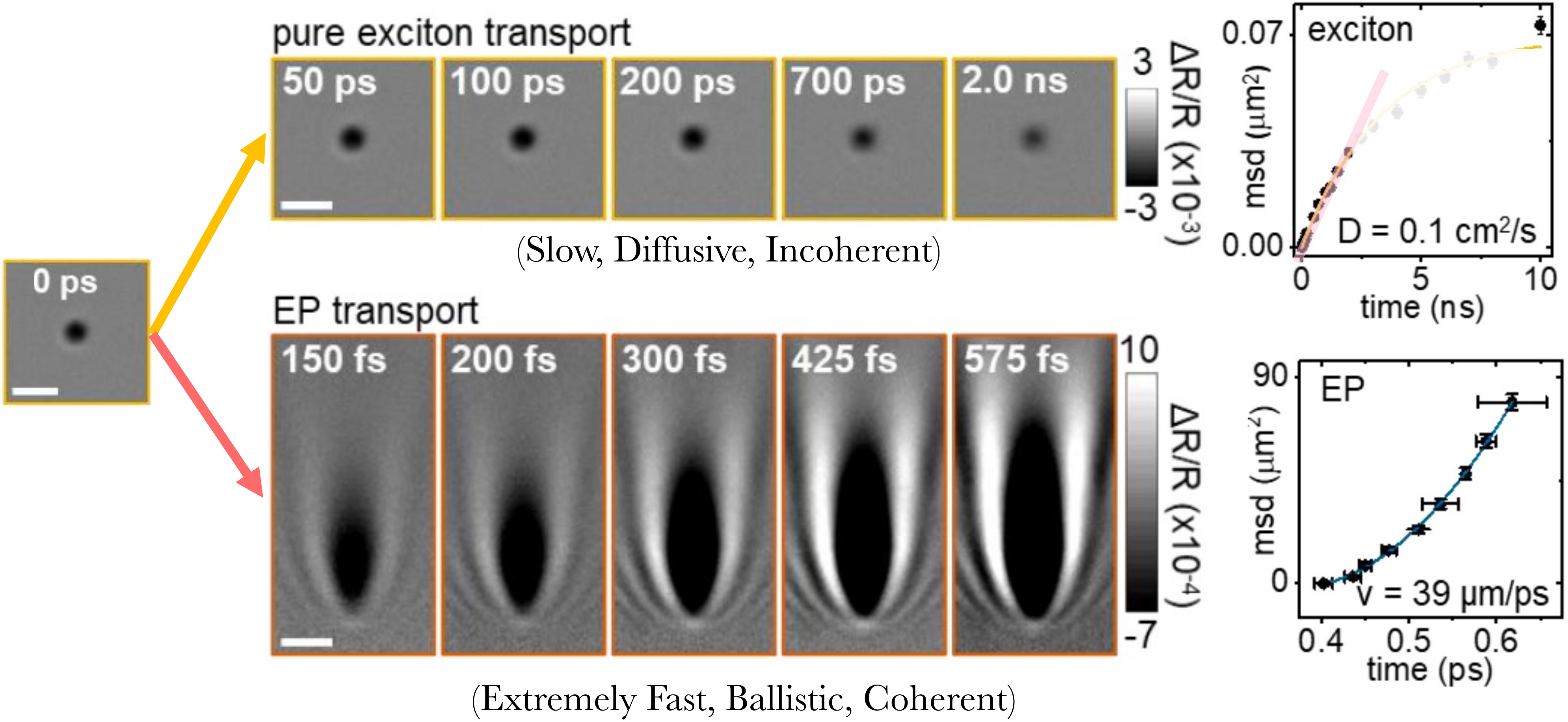
Blackham †, Manjalingal †, Rahmanian, Mandal, *Nano Letters* (2025)

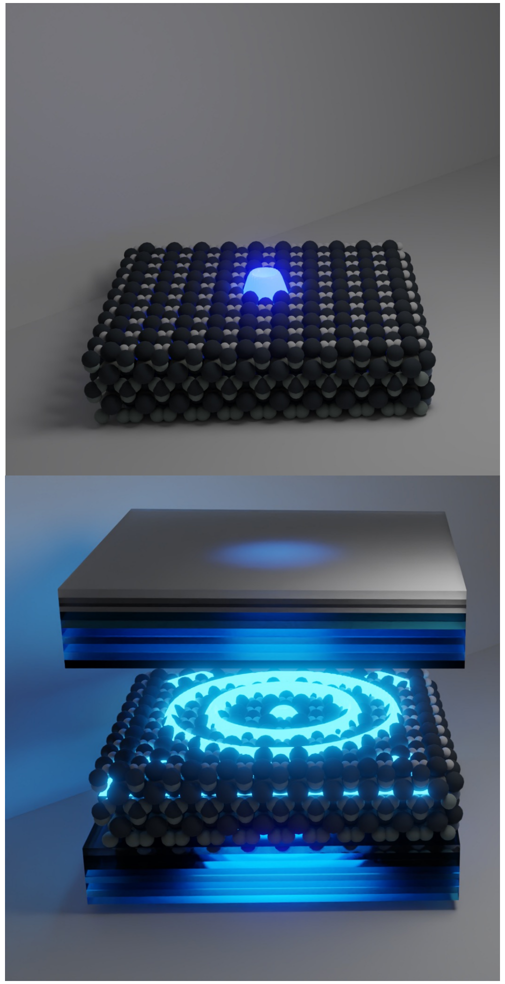
Manjalingal †, Rahmanian †, **Blackham**, Mandal, *ACS Photonics* (2025)

Rahmanian, Manjalingal, **Blackham**, Mandal, *arXiv* (2025)

†equal contribution

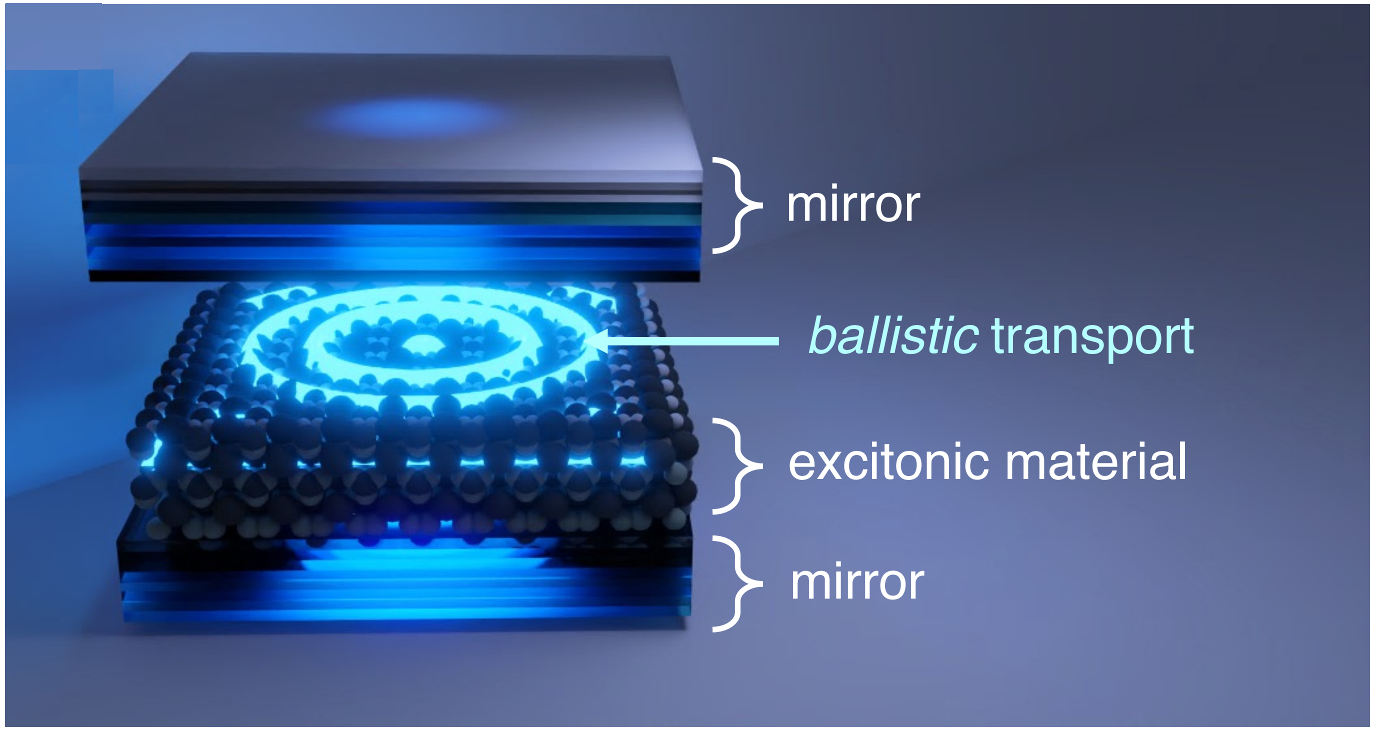
Exciton-Polariton Transport

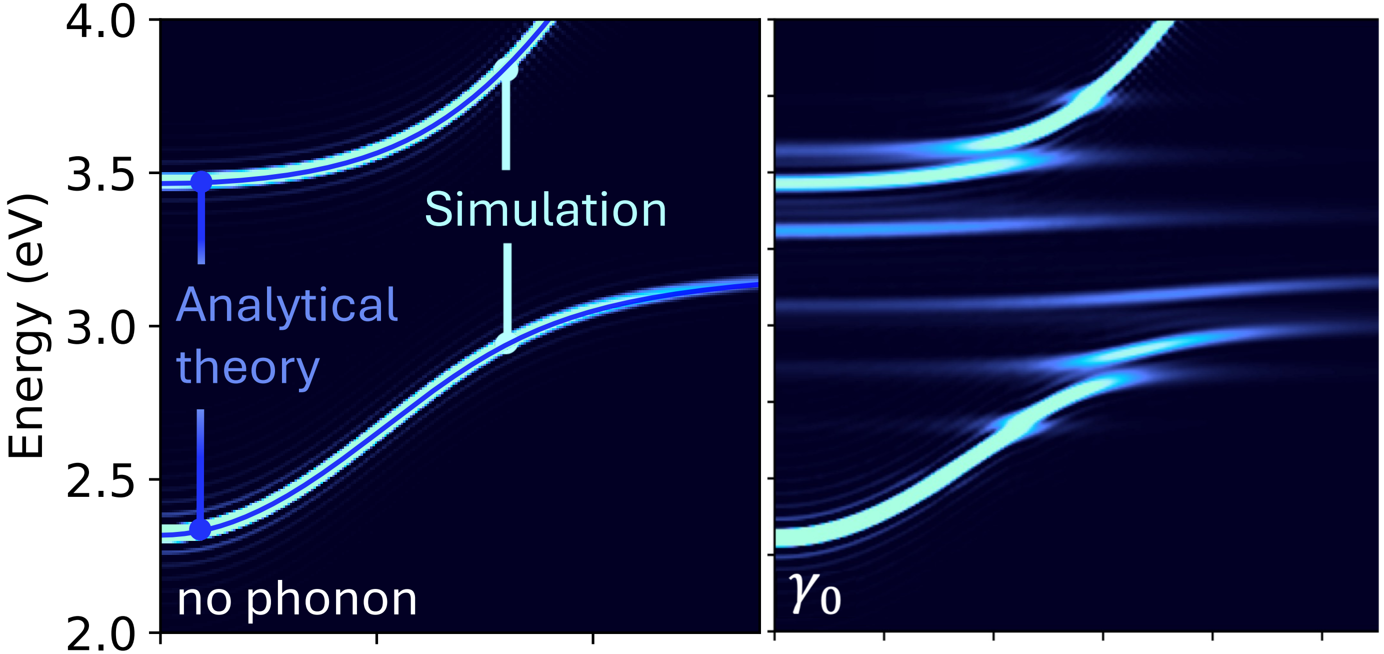


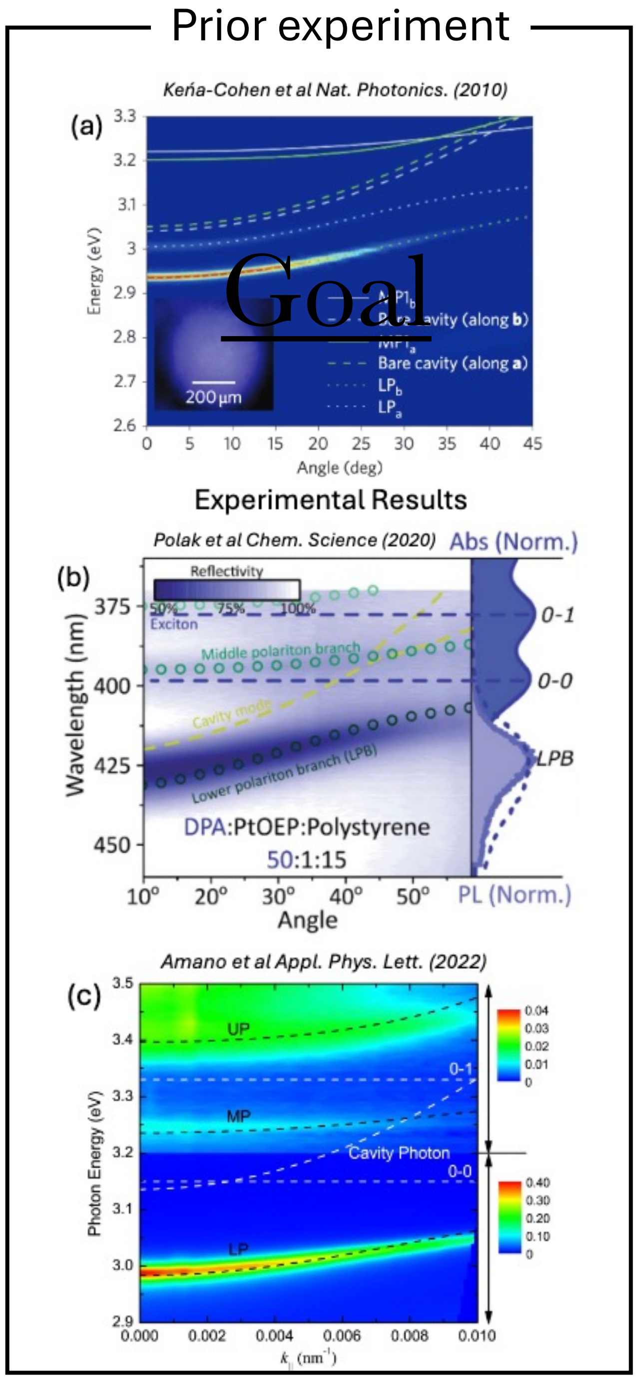


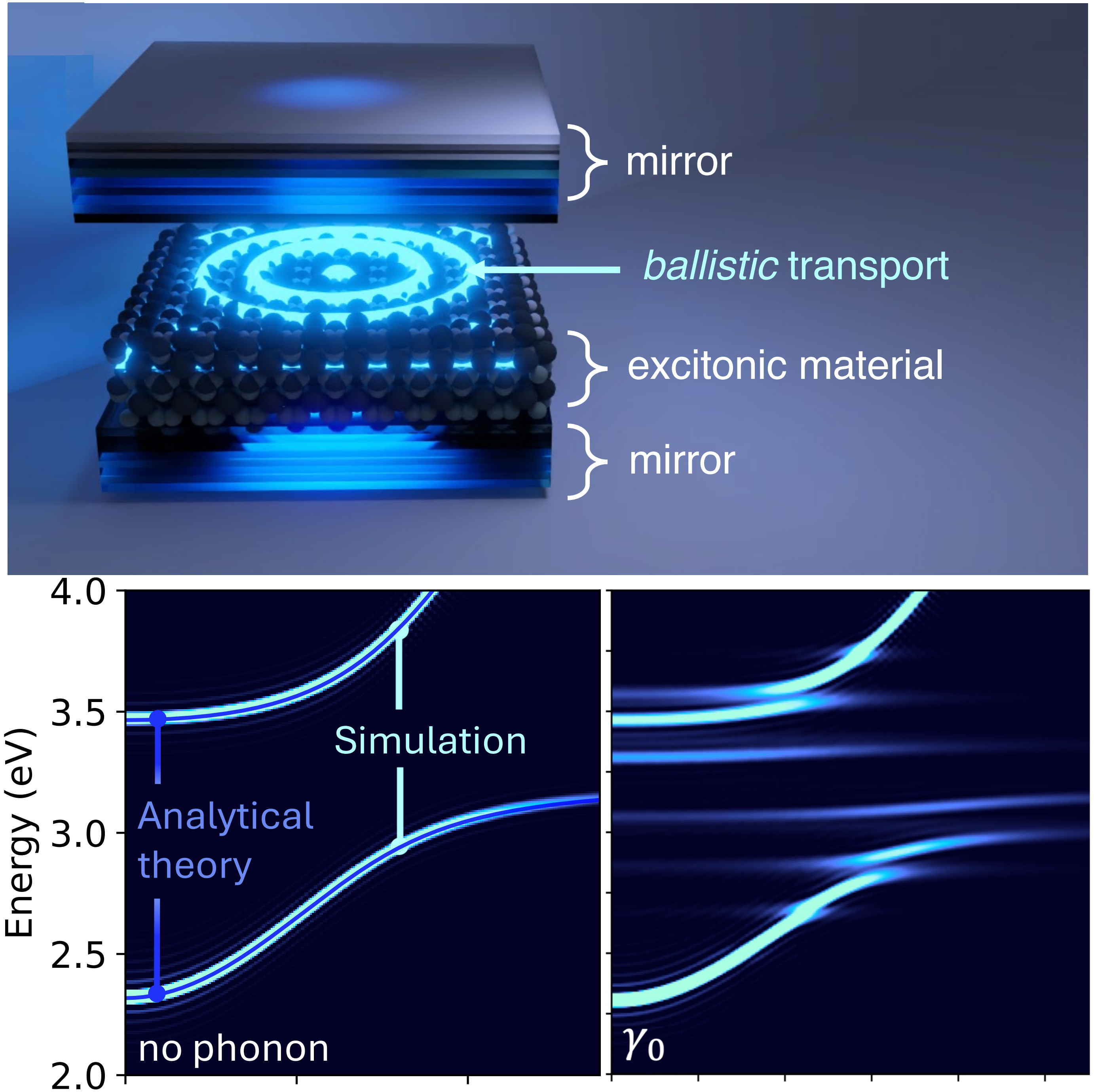
Xu et. al. Nat. Comm. (2023)

Also see:
Pandya et. al., Adv Sci (2022)
Balasubrahmaniyam et. al., Nat. Mater. (2023)


mirror
ballistic transport
excitonic material
mirror


Energy (eV)
4.0
3.5
3.0
2.5
2.0
Simulation
Analytical theory
no phonon
γ_0


Prior experiment
Keña-Cohen et al Nat. Photonics. (2010)
(a)
Energy (eV)
Angle (deg)
Goal
200 μm
Bare cavity (along b)
Bare cavity (along a)
Experimental Results
Polak et al Chem. Science (2020)
Abs (Norm.)
(b)
Reflectivity
Exciton
Middle polariton branch
Cavity mode
Lower polariton branch (LPB)
Wavelength (nm)
0-1
0-0
LPB
DPA:PtOEP:Polystyrene
50:1:15
Angle
PL (Norm.)
Amano et al Appl. Phys. Lett. (2022)
(c)
UP
MP
LP
0-1
0-0
Cavity Photon
Photon Energy (eV)



Goal

- Develop a theory to capture the vibronic structure in exciton-polariton dispersion

Prior Theoretical Approaches

Tichauer, Sokolovskii, Groenhof, Adv. Sci. (2023)
Englehardt et. al, Phys. Rev. Lett. (2023)
Ying et. al. Nat. Commun. (2025)

$$\hat{H}_{\text{LM}} = \sum_n \hat{X}_n^\dagger \hat{X}_n \varepsilon_0 + \tau \sum_n (\hat{X}_n^\dagger \hat{X}_{n+1} + \hat{X}_{n+1}^\dagger \hat{X}_n)$$
$$+ \sum_k \hat{a}_k^\dagger \hat{a}_k \omega_c(k) + \sum_{n,k} \frac{\Omega_k}{\sqrt{N}} \left[\hat{a}_k^\dagger \hat{X}_n e^{-ik\cdot r_n} + \hat{a}_k \hat{X}_n^\dagger e^{ik\cdot r_n} \right]$$
$$+ \sum_n \frac{P_n^2}{2} + \frac{1}{2}\omega^2 R_n^2 + \sum_n \gamma \hat{X}_n^\dagger \hat{X}_n R_n$$

Mixed Quantum Classical Dynamics

$$\dot{P}_n(t) = -\left\langle \Psi(t) \middle| \frac{\mathrm{d}\hat{H}_{\text{LM}}}{\mathrm{d}R_n} \middle| \Psi(t) \right\rangle, \quad \dot{R}_n(t) = P_n(t)$$
$$i|\dot{\Psi}(t)\rangle = \left[\hat{H}_{\text{LM}} - \frac{1}{2} \sum_n \frac{P_n^2}{2} \right] |\Psi(t)\rangle$$

Classical Path Approximation

$$R_n(t) \approx R_n(0)\cos\omega t + \frac{1}{\omega} P_n(0) \sin\omega t$$
$$\hat{H}_{\text{LM}} \to \hat{H}_{\text{LM}}(t) = \hat{H}_{\text{EP}} + \hat{P} e^{i\omega t} + \hat{P}^\dagger e^{-i\omega t}$$
$$\hat{P} = \sum_n \gamma \hat{X}_n^\dagger \hat{X}_n \Big(R_n(0)/2 + P_n(0)/2i\omega \Big)$$

J. Phys. Chem. B **1999,** *103,* 10978−10991

On the Adequacy of Mixed Quantum-Classical Dynamics in Condensed Phase Systems

S. A. Egorov†
Theoretical Chemistry Institute and Department of Chemistry, University of Wisconsin, Madison, Wisconsin 53706

Eran Rabani‡ **and B. J. Berne***
Department of Chemistry, Columbia University, 3000 Broadway, New York, New York 10027

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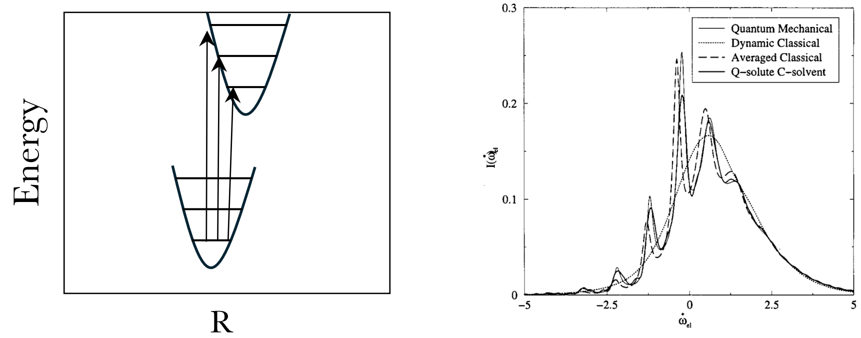
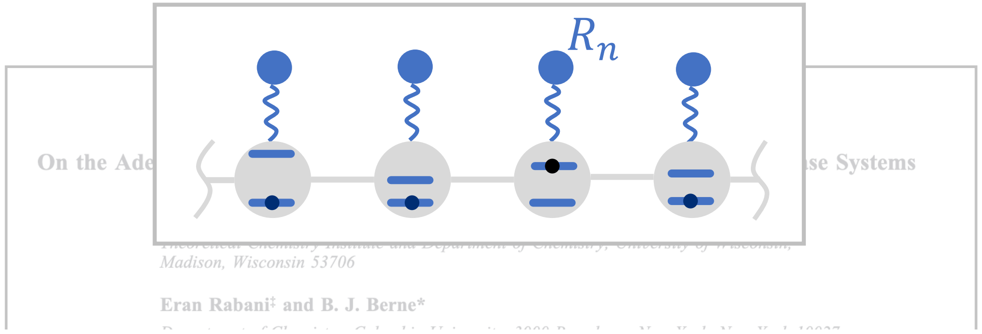


$$\hat{H}_{\mathrm{LM}} = \sum_n \hat{X}_n^\dagger \hat{X}_n \varepsilon_0 + \tau \sum_n (\hat{X}_n^\dagger \hat{X}_{n+1} + \hat{X}_{n+1}^\dagger \hat{X}_n)$$
$$+ \sum_k \hat{a}_k^\dagger \hat{a}_k \omega_c(k) + \sum_{n,k} \frac{\Omega_k}{\sqrt{N}} \left[\hat{a}_k^\dagger \hat{X}_n e^{-ik\cdot r_n} + \hat{a}_k \hat{X}_n^\dagger e^{ik\cdot r_n} \right]$$
$$+ \sum_n \frac{P_n^2}{2} + \frac{1}{2}\omega^2 R_n^2 + \sum_n \gamma \hat{X}_n^\dagger \hat{X}_n R_n$$



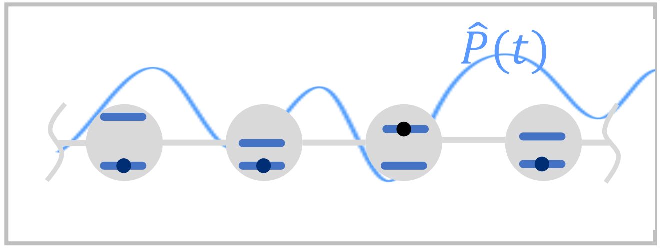
Mixed Quantum Classical Dynamics

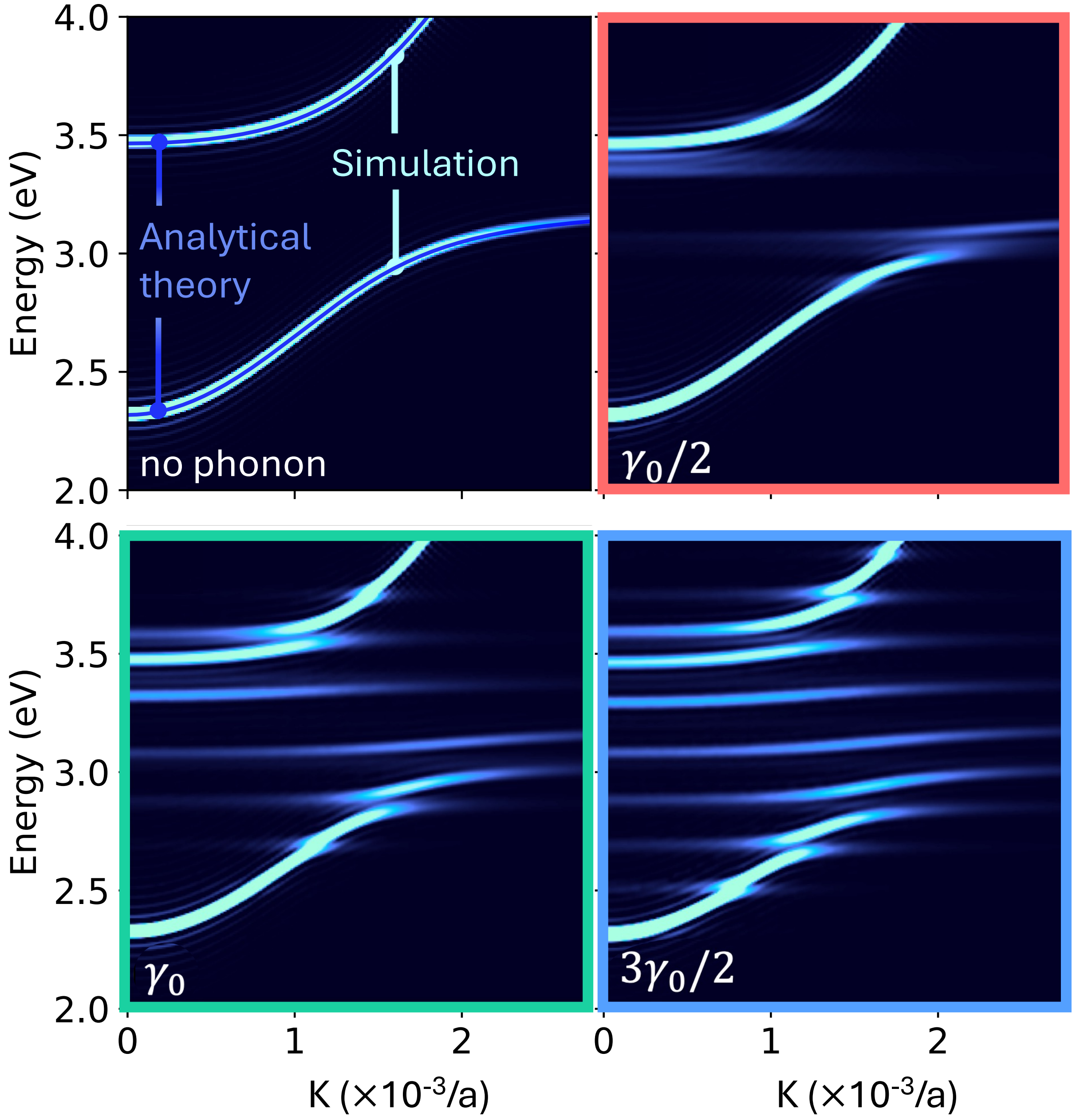
$$\dot{P}_n(t) = -\left\langle \Psi(t) \middle| \frac{\mathrm{d}\hat{H}_{\mathrm{LM}}}{\mathrm{d}R_n} \middle| \Psi(t) \right\rangle, \quad \dot{R}_n(t) = P_n(t)$$
$$i|\dot{\Psi}(t)\rangle = \left[\hat{H}_{\mathrm{LM}} - \frac{1}{2}\sum_n \frac{P_n^2}{2} \right] |\Psi(t)\rangle$$

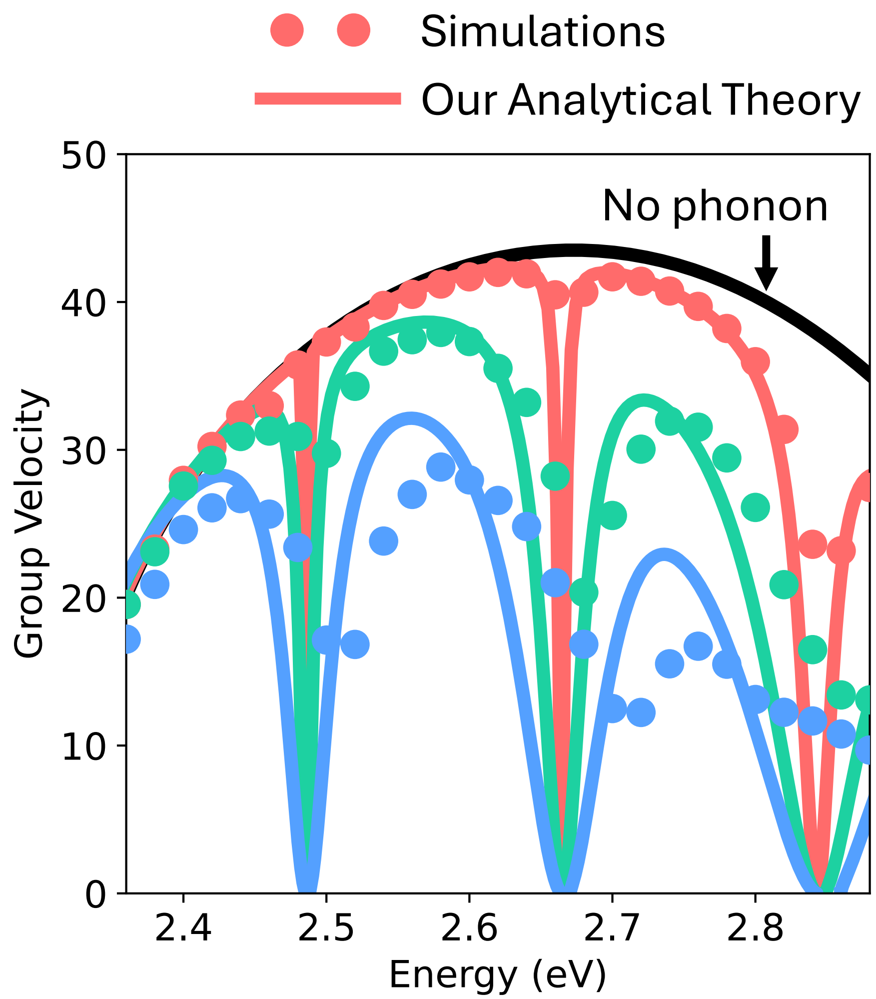
Exciton-Polariton is interacting
with a
time-dependent **phonon field**

Classical Path Approximation

$$R_n(t) \approx R_n(0)\cos\omega t + \frac{1}{\omega} P_n(0) \sin\omega t$$
$$\hat{H}_{\mathrm{LM}} \to \hat{H}_{\mathrm{LM}}(t) = \hat{H}_{\mathrm{EP}} + \hat{P} e^{i\omega t} + \hat{P}^\dagger e^{-i\omega t}$$
$$\hat{P} = \sum_n \gamma \hat{X}_n^\dagger \hat{X}_n \left(R_n(0)/2 + P_n(0)/2i\omega \right)$$

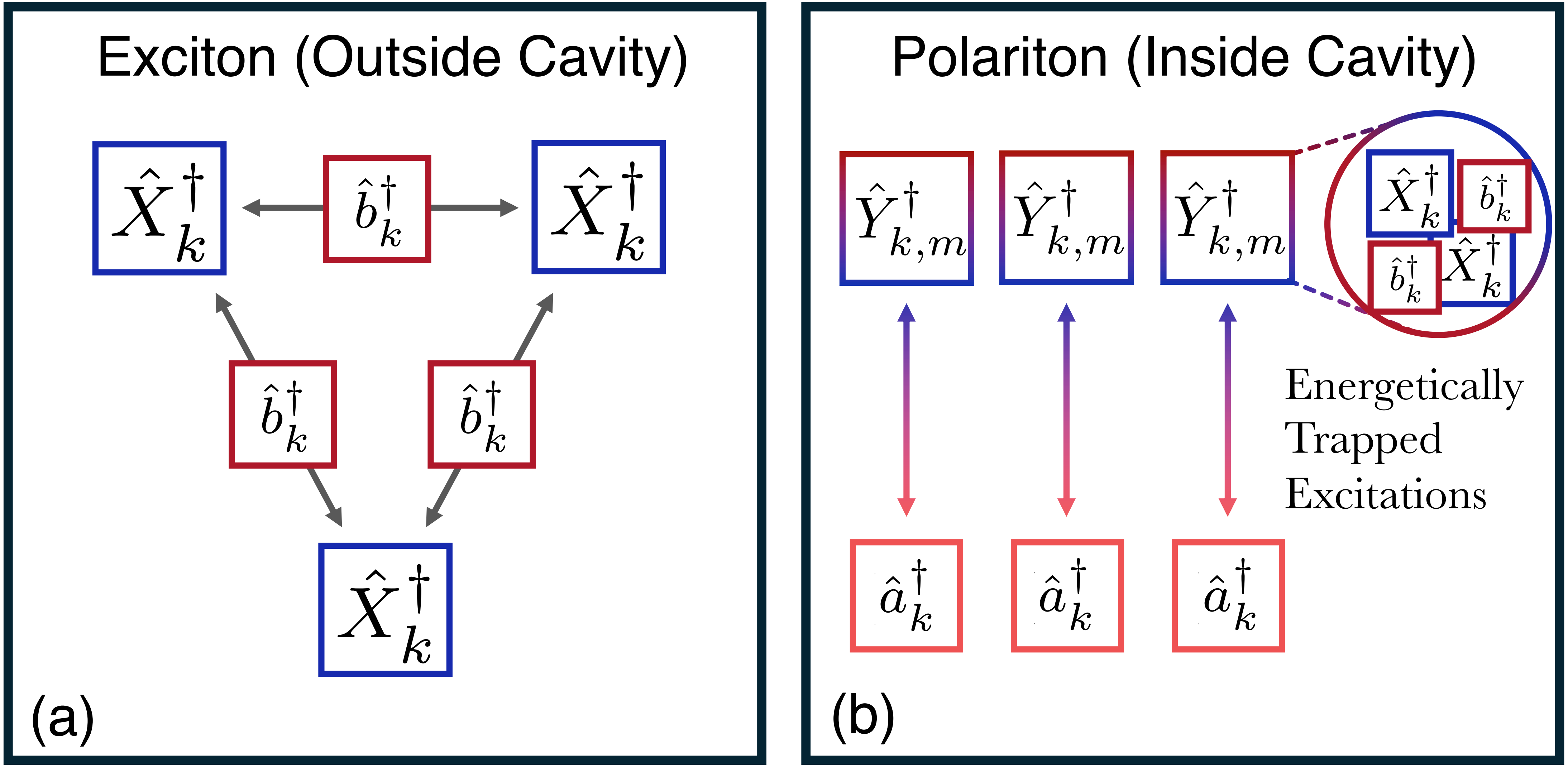



Energy (eV)
4.0
3.5
3.0
2.5
2.0
Simulation
Analytical theory
no phonon
$\gamma_0/2$
γ_0
$3\gamma_0/2$
0
1
2
K (×10⁻³/a)

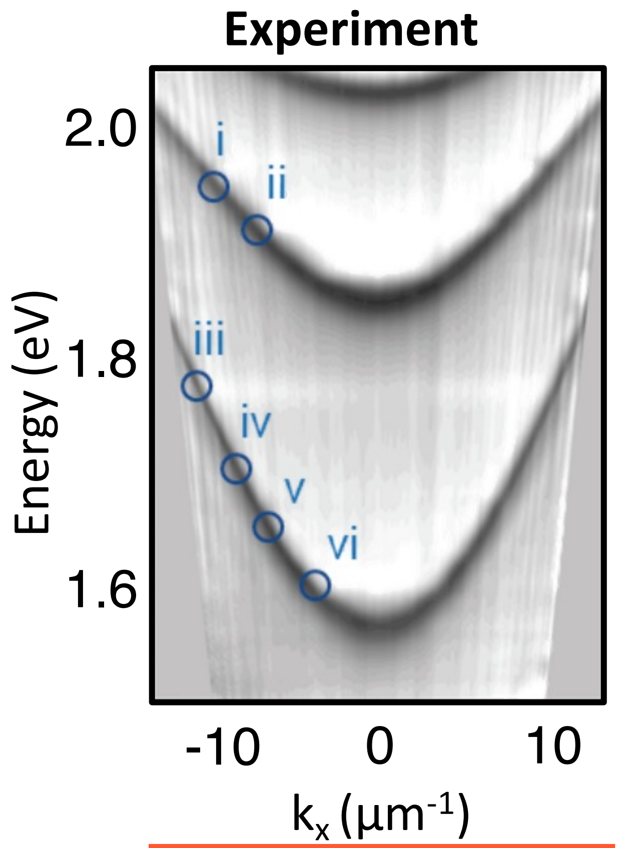

Simulations
Our Analytical Theory
No phonon
Group Velocity
0
10
20
30
40
50
2.4
2.5
2.6
2.7
2.8
Energy (eV)

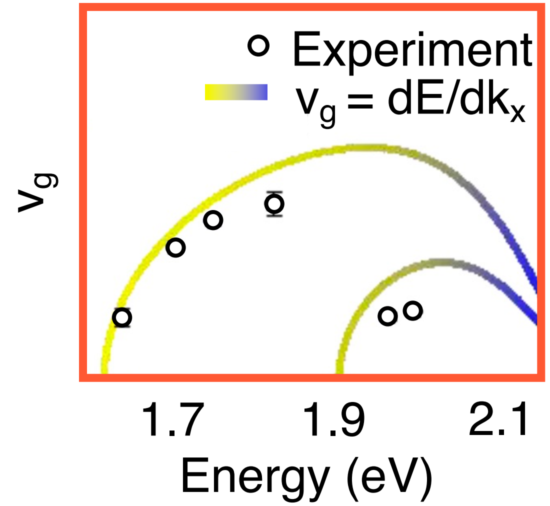
$$\hat{Y}^\dagger_{k,m} = \sum_n \frac{Q_{m0}(Z_n)}{\sqrt{\mathcal{S}_m}} e^{-ik\cdot r_n} \hat{X}^\dagger_n \frac{(\hat{B}^\dagger)^m}{m!}$$

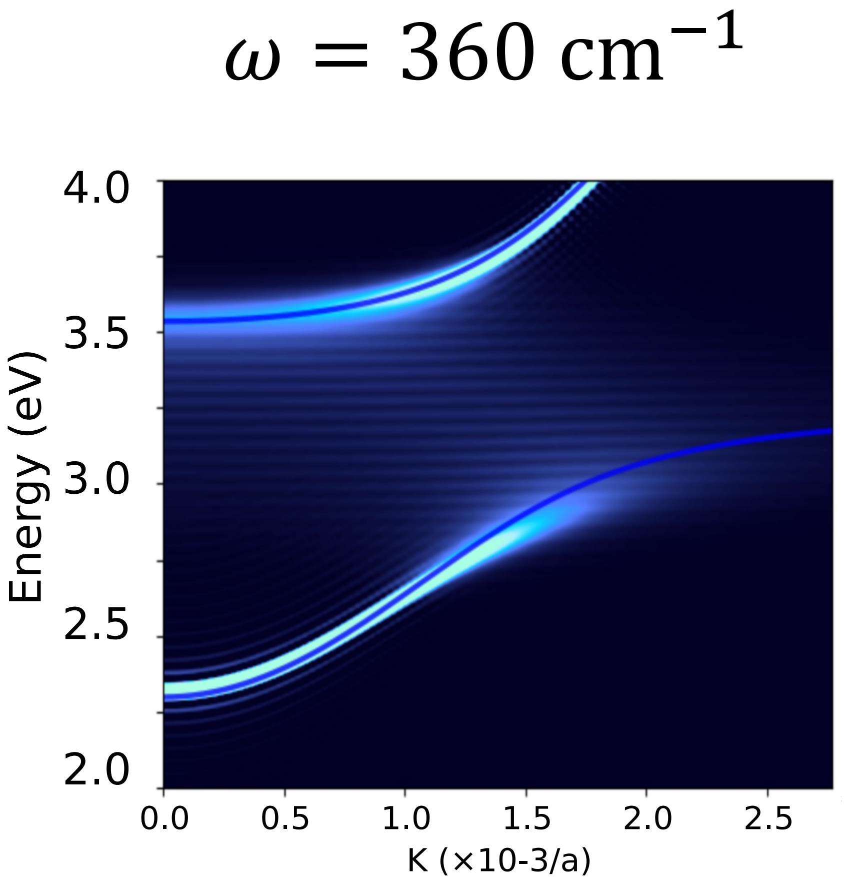
We introduce a "Polaron Operator"

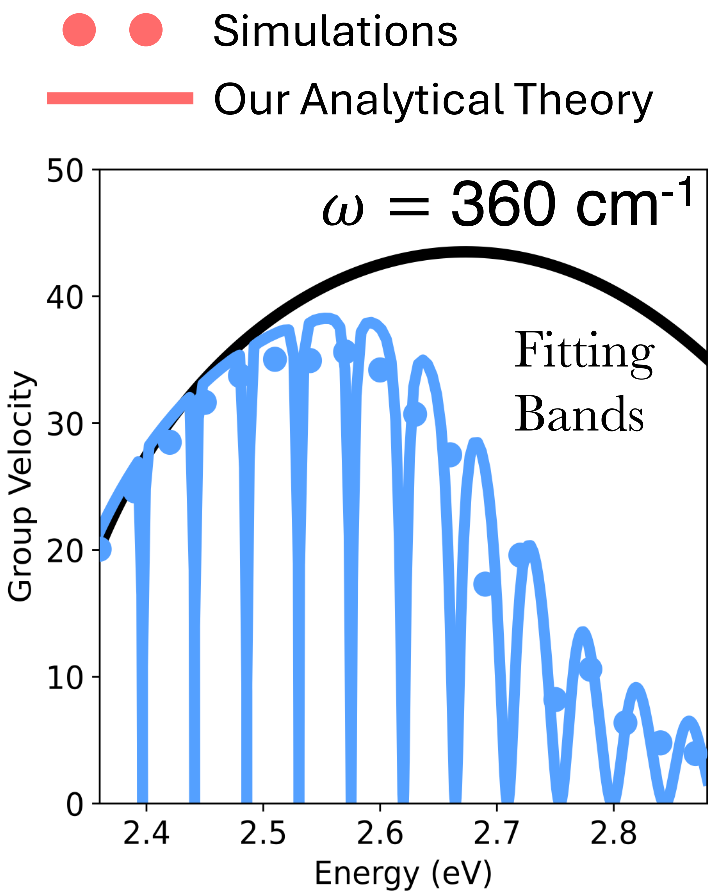


Xu†, Mandal†… Delor, Reichman Nat. Comm. (2023)

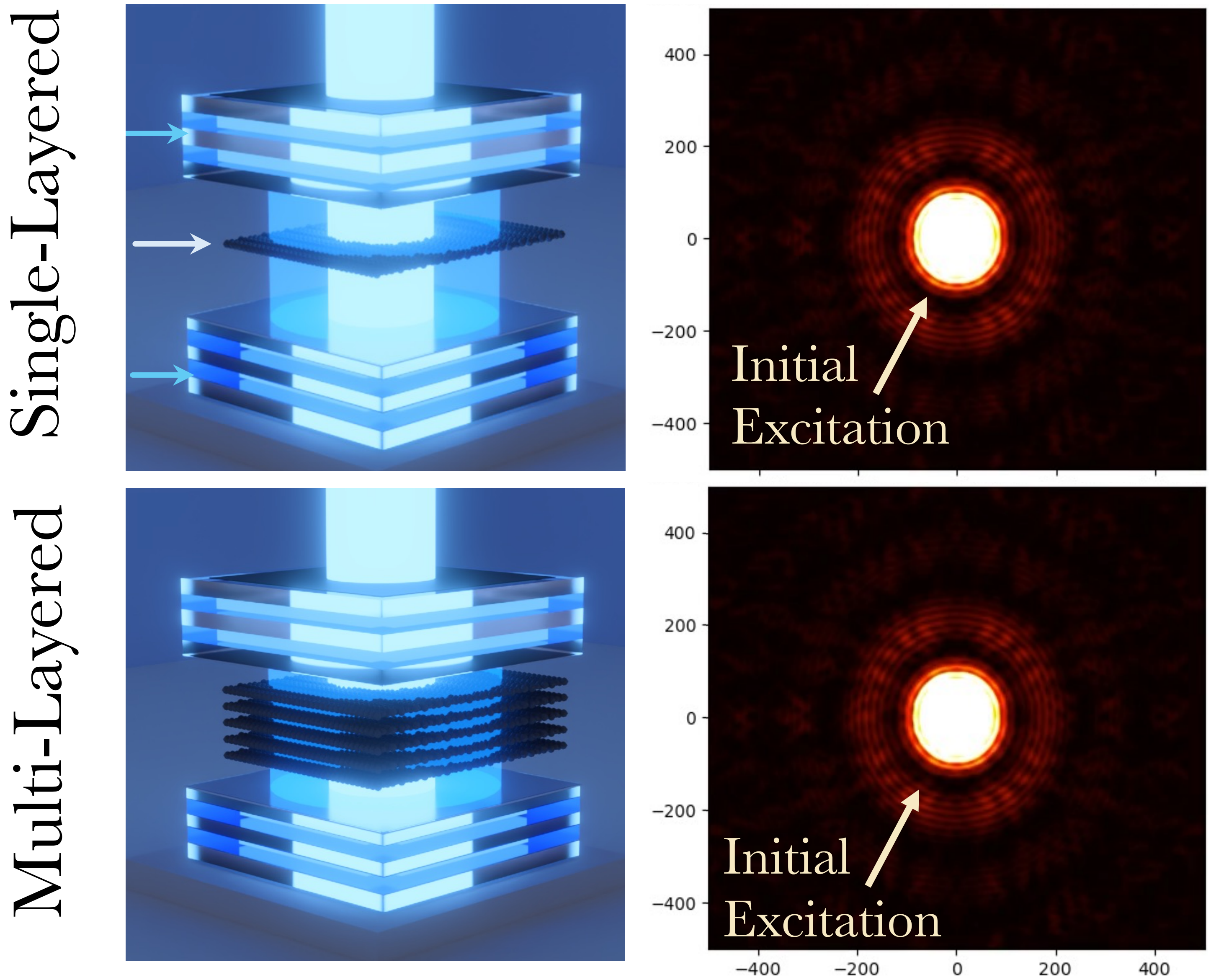




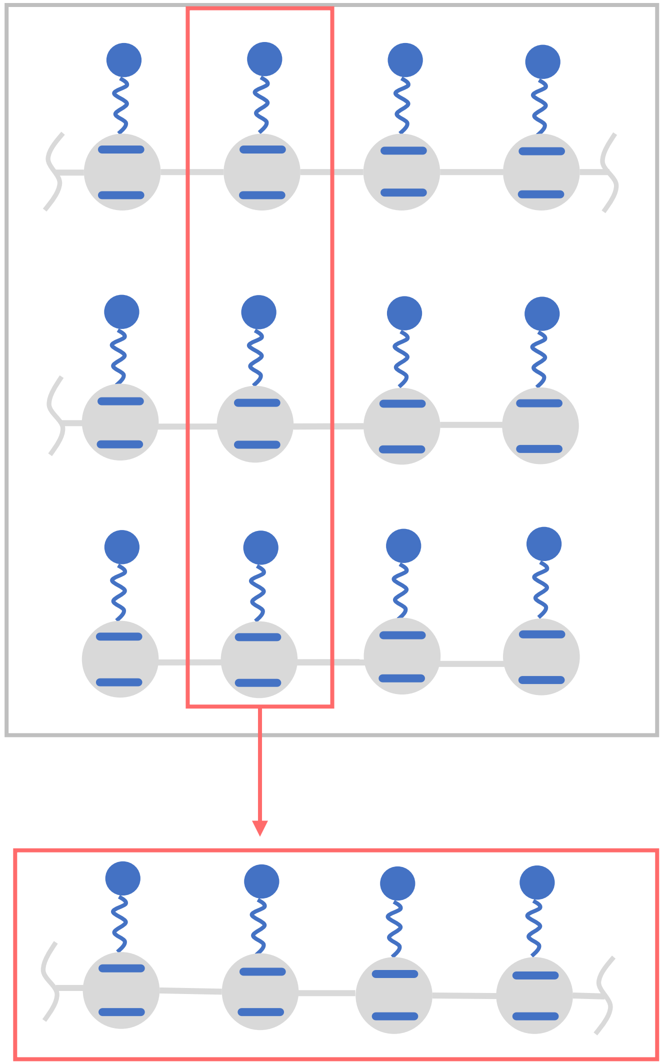




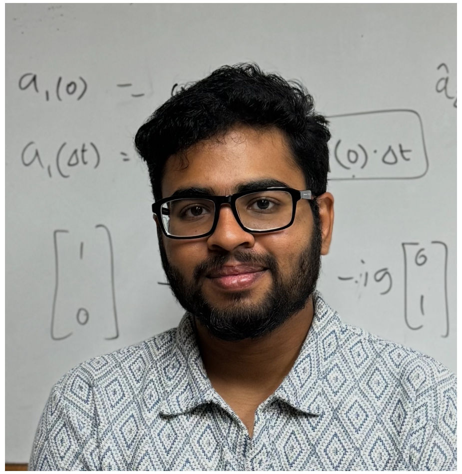
- The polaron-polariton band may not show up in spectra
- The slopes of the polaron-polariton band dictates the group velocity
- This explains what has been observed in experiments

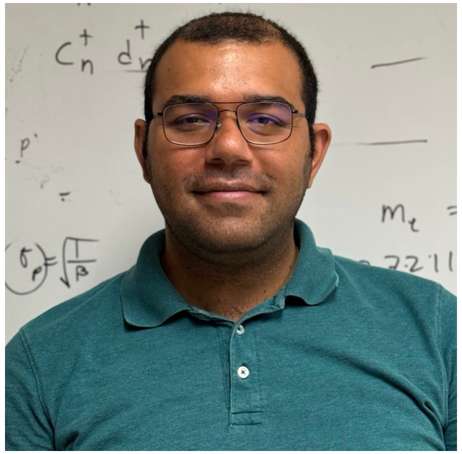


Phonon Averaging Effect



Rahmanian, Manjanlingal, Blackham, Mandal arXiv [quant-ph] (2025)





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