Transport and Dispersion of Exciton-Polariton

Arkajit Mandal and David R. Reichman

Experiment: Ding Xu and Milan Dilor

Xu† Mandal† Baxter Cheng Lee Su Liu Reichman Delor**
Arxiv: 2205.01176 (2022) *† Equal*

Mandal Xu Mahajan Lee Delor Reichman**
Nano Lett. 2023

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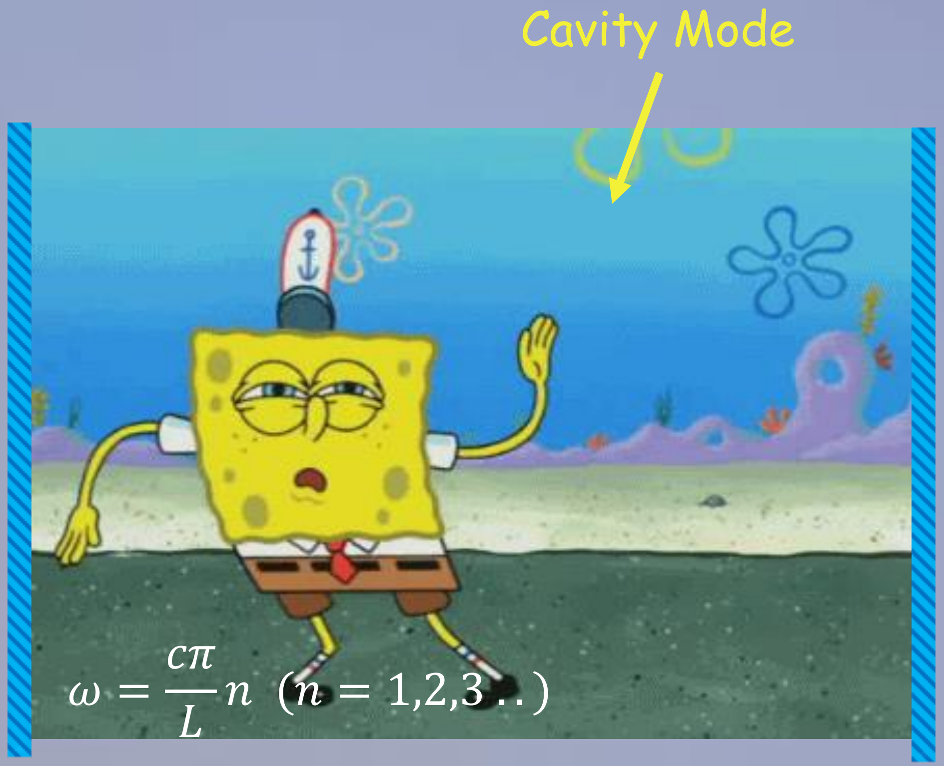
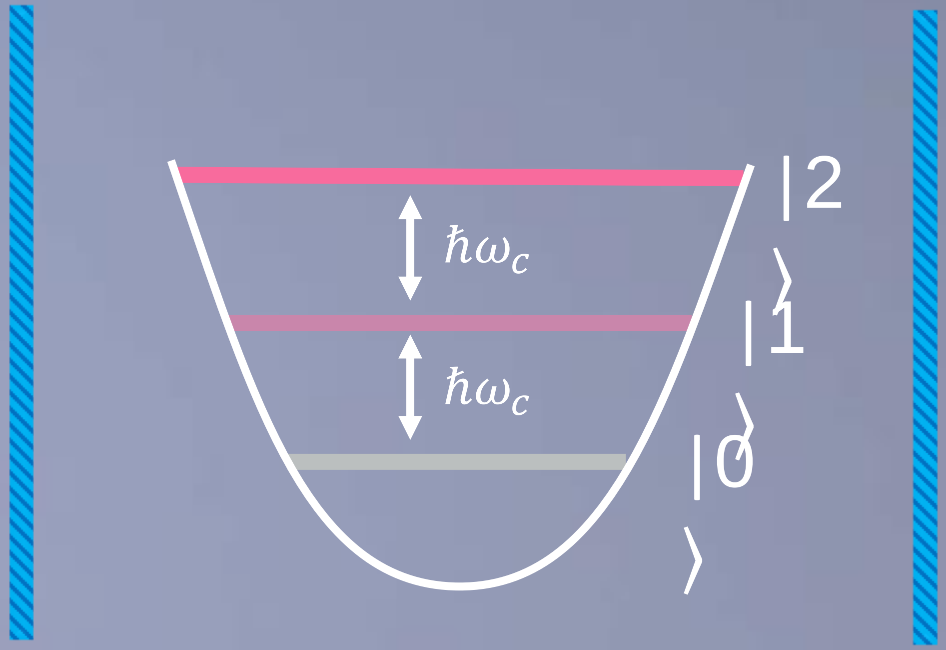
A Cavity

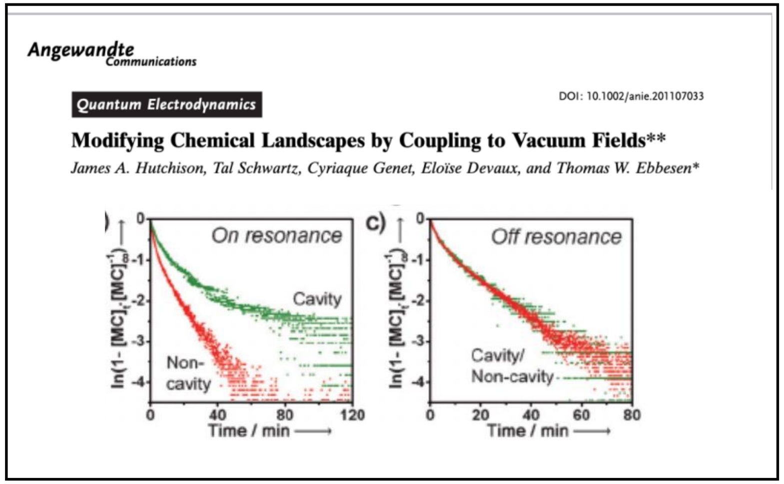


Quantization →

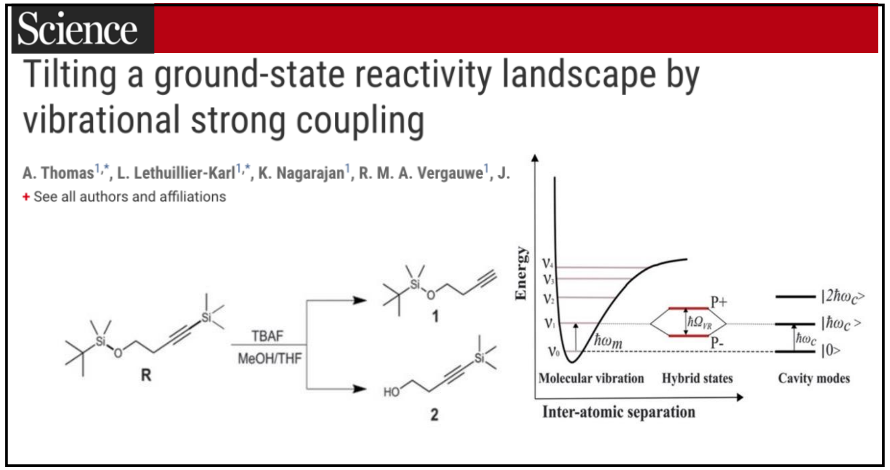


Polaritons

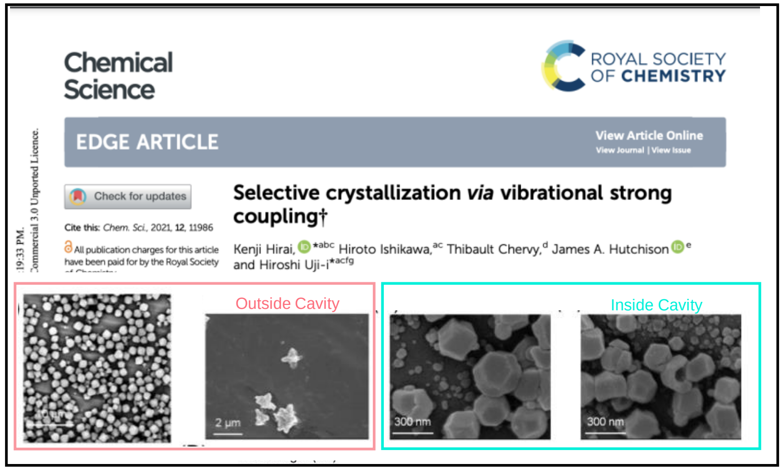
Photochemistry



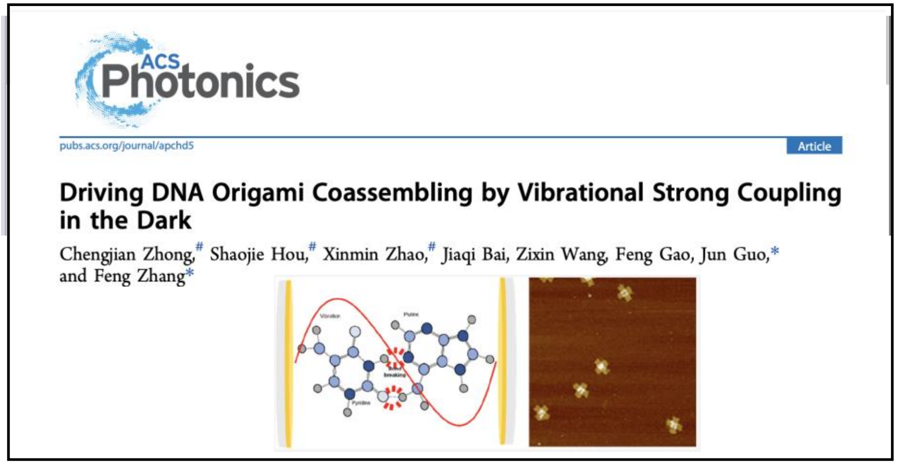
Chemical Kinetics



Crystallization



Biophysics



Theoretical Advances in Polariton Chemistry

Rubio (Max Planck), Mukamel (UCI), Huo (Rochester), Yuen-Zhou (UCSD), Herrera (U. Santiago), Groenhoff (U. Jyväskylä), Narang (UCLA), Keeling (St. Andrews), Rebeiro (Emory), Subotnik (UPenn), Feist (UAM), Nitzan (UPenn), Saalfrank (U. Potsdam), Vendrell (Heidelberg), Garcia-Vidal (UAM), Cederbaum (Heidelberg), Koch (NTNU), Kowalewski (Stockholm), Deprince (FSU), Vivok (U. Debrecen), Hammes-Schiffer (Yale), Scholes (Princeton), Foley (UNCC) and many more groups

Experiments in Polariton Chemistry/Physics

Ebbesen (Strasbourg), Xiong (UCSD), Menon (CCNY), Schwartz (Tel Aviv), Giebink (Penn), George (IISER-M), Simpkins (NRL) , Baumberg (St. Andrews), Vamivakas (Rochester), Krauss (Rochester), Haran (Weizmann), Witchman (Princeton), XYZ (Columbia), Delor (Columbia), Musser (Cornell) many more group…

Topics

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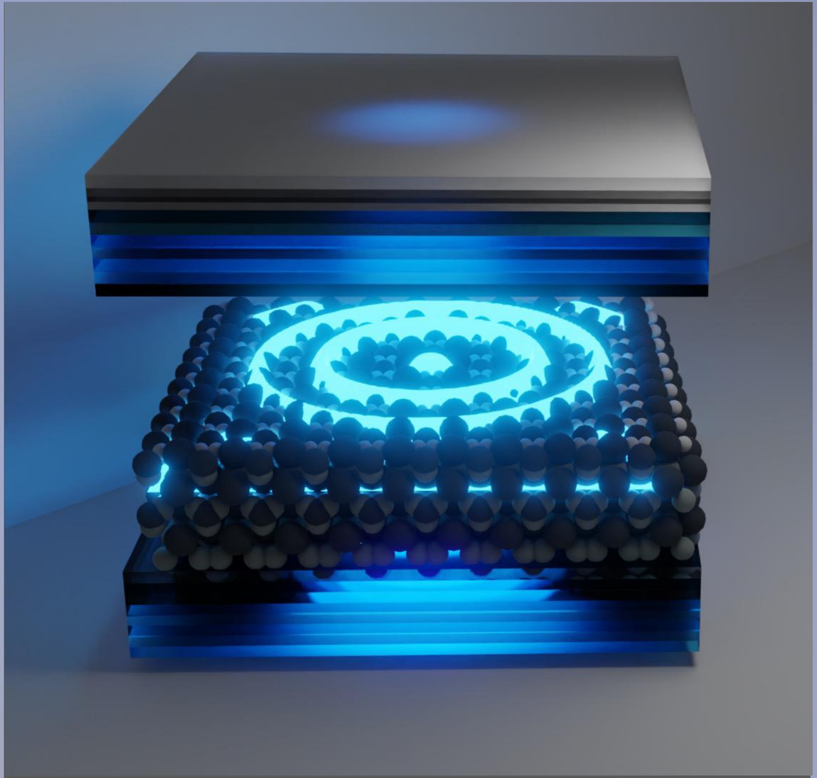
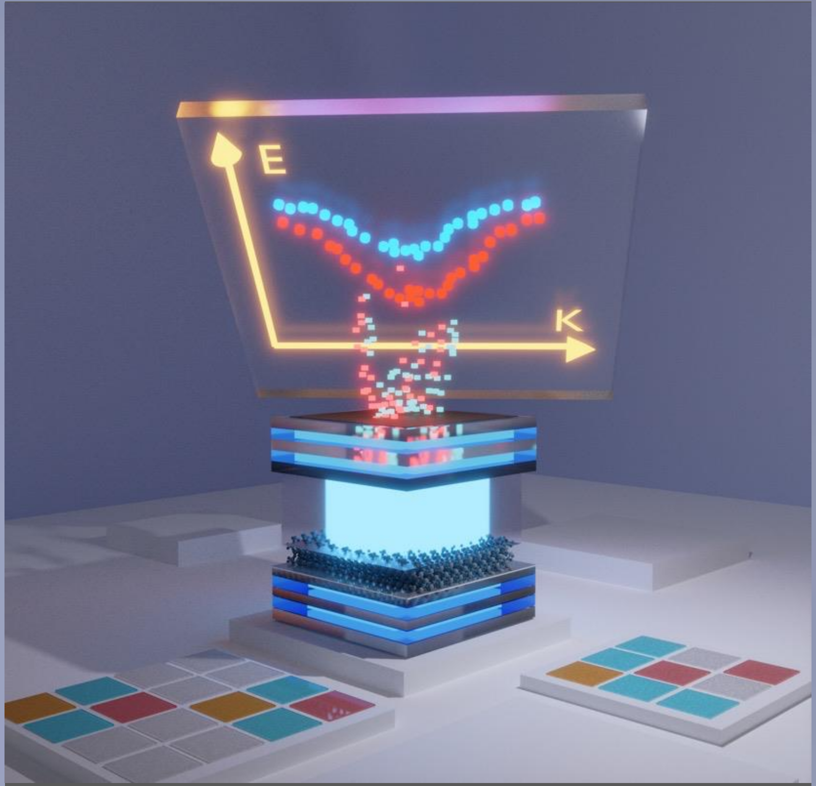


Exciton-Polariton
Transport

Xu† *Mandal*† *Baxter Cheng Lee Su Liu Reichman* Delor**
Arxiv: 2205.01176 (2022) *† Equal*

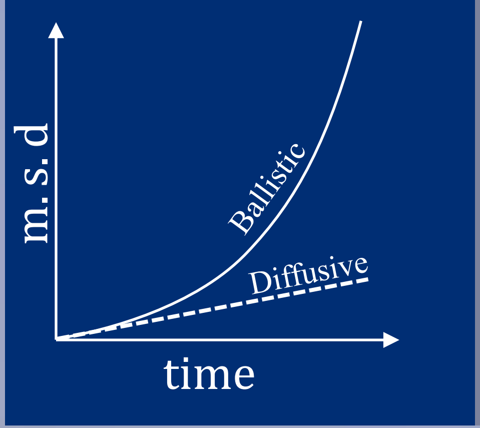


Exciton-Polariton
Dispersion

Mandal Xu Mahajan Lee Delor Reichman**
Nano Lett. 2023

Outline

1



Introduce the key concepts

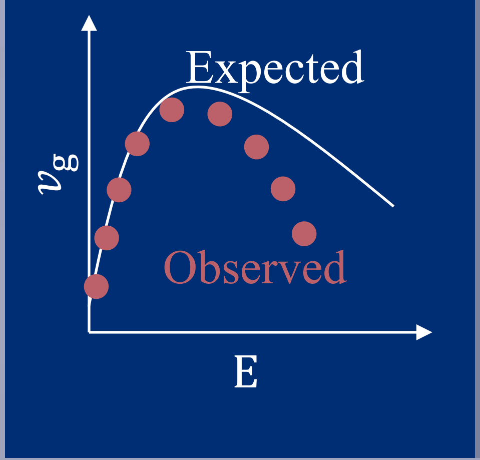
Coherent and Incoherent Transport

2

$$\hat{c}\hat{a}^{\dagger} + \hat{c}^{\dagger}\hat{a}$$

Light-Matter Interactions

3

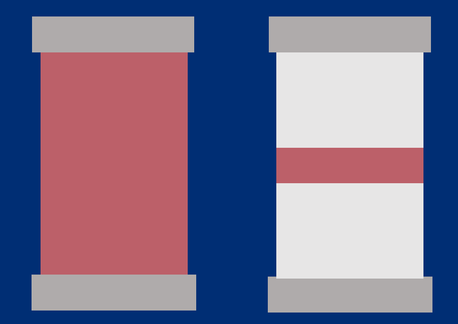


Simulation of Exciton Polariton Transport

Ballistic and Diffusive motion of Exciton-Polariton

Experiment + Theory

4

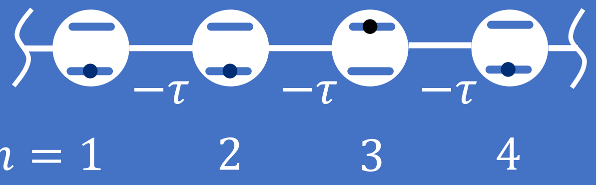


Microscopic theory of Dispersion

Experiment + Theory

1D Exciton

$$\hat{c}_n^\dagger = |n\rangle\langle G|$$



$$\hat{H}_{1\mathrm{D}} = \epsilon_0 \sum_n \hat{c}_n^\dagger \hat{c}_n - \tau \sum_n (\hat{c}_{n+1}^\dagger \hat{c}_n + \hat{c}_n^\dagger \hat{c}_{n+1})$$

$$|k\rangle = \frac{1}{\sqrt{N}} \sum_n e^{ikn} |n\rangle \qquad \hat{c}_k = \frac{1}{\sqrt{N}} \sum_n e^{ikn}\, \hat{c}_n$$

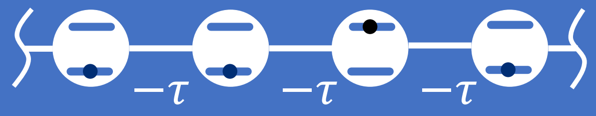
$$\hat{H}_{1\mathrm{D}}\, |k\rangle = (\epsilon_0 - 2\tau \cos k)|k\rangle$$

$$\hat{H}_{1\mathrm{D}} = \sum_k (\epsilon_0 - 2\tau \cos k)\hat{c}_k^\dagger \hat{c}_k = \sum_k \epsilon_k \hat{c}_k^\dagger \hat{c}_k$$

A set of two level systems with nearest neighbor coupling.

1D Exciton (Group Velocity)

$$\hat{c}_n^\dagger = |n\rangle\langle G|$$

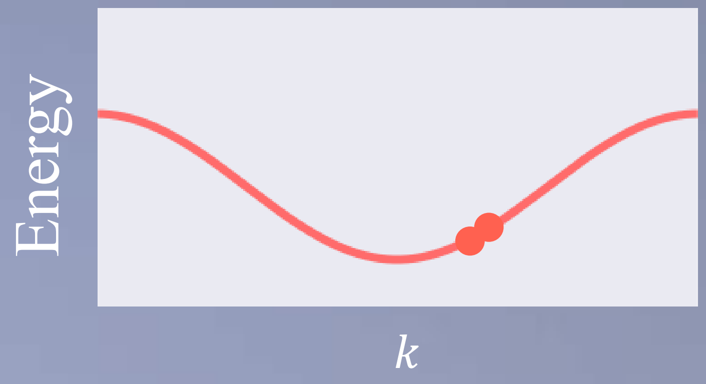


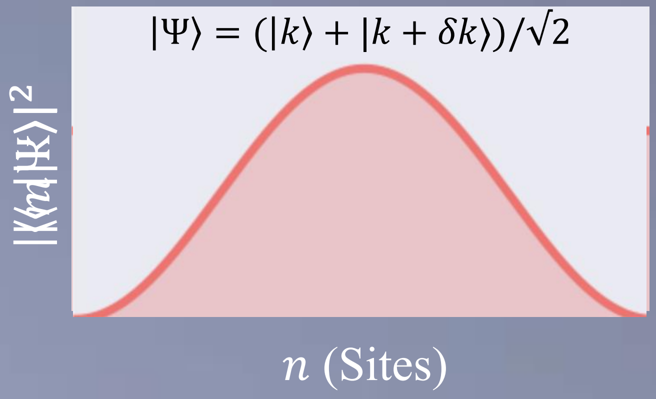
$$\hat{H}_{1D} = \epsilon_0 \sum_n \hat{c}_n^\dagger \hat{c}_n - \tau \sum_n (\hat{c}_{n+1}^\dagger \hat{c}_n + \hat{c}_n^\dagger \hat{c}_{n+1})$$

$$|k\rangle = \frac{1}{\sqrt{N}} \sum_n e^{ikn} |n\rangle \qquad \hat{c}_k = \frac{1}{\sqrt{N}} \sum_n e^{ikn} \hat{c}_n$$

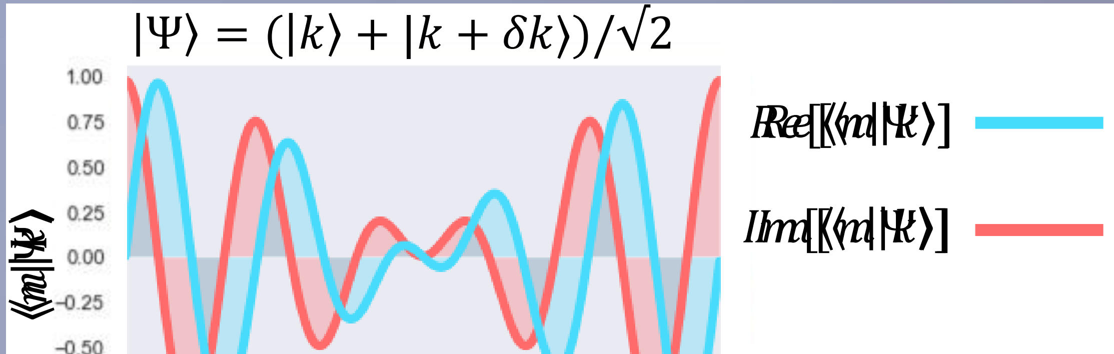
$$\hat{H}_{1D} |k\rangle = (\epsilon_0 - 2\tau \cos k)|k\rangle$$

$$\hat{H}_{1D} = \sum_k (\epsilon_0 - 2\tau \cos k) \hat{c}_k^\dagger \hat{c}_k = \sum_k \epsilon_k \hat{c}_k^\dagger \hat{c}_k$$





Eigenfunctions are delocalized in site space.
Localized in reciprocal space.

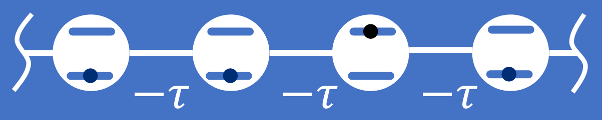


$$v_p = \epsilon_k / k$$

$$v_g = d\epsilon_k / dk$$

Ballistic (Coherent) Transport

$$\hat{c}_n^\dagger = |n\rangle\langle G|$$

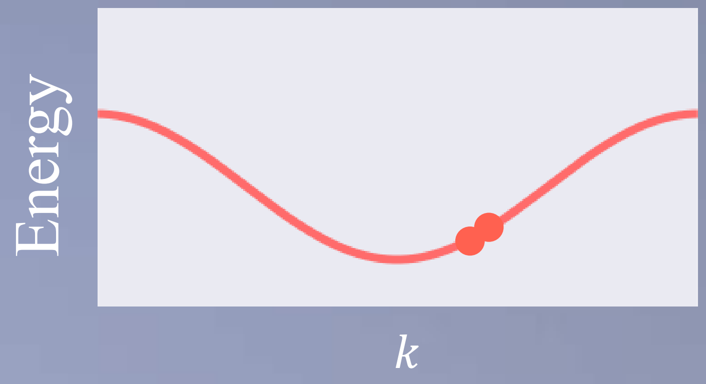


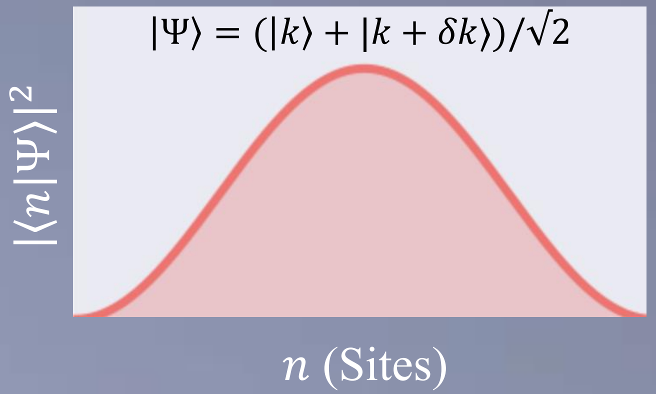
$$\hat{H}_{1\mathrm{D}} = \epsilon_0 \sum_n \hat{c}_n^\dagger \hat{c}_n - \tau \sum_n (\hat{c}_{n+1}^\dagger \hat{c}_n + \hat{c}_n^\dagger \hat{c}_{n+1})$$

$$|k\rangle = \frac{1}{\sqrt{N}} \sum_n e^{ikn} |n\rangle \qquad \hat{c}_k = \frac{1}{\sqrt{N}} \sum_n e^{ikn} \hat{c}_n$$

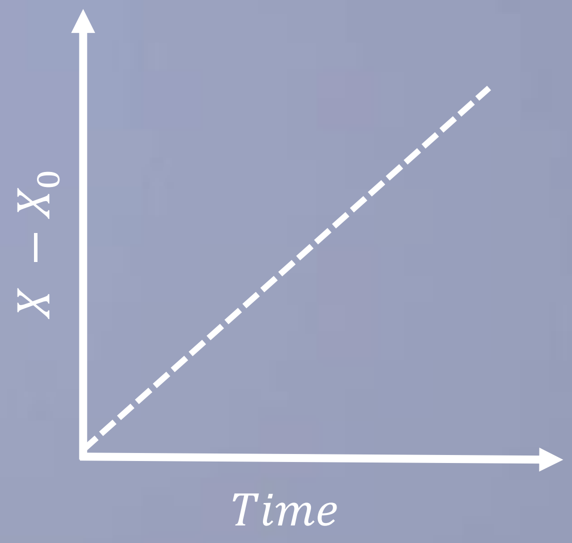
$$\hat{H}_{1\mathrm{D}} |k\rangle = (\epsilon_0 - 2\tau \cos k)|k\rangle$$

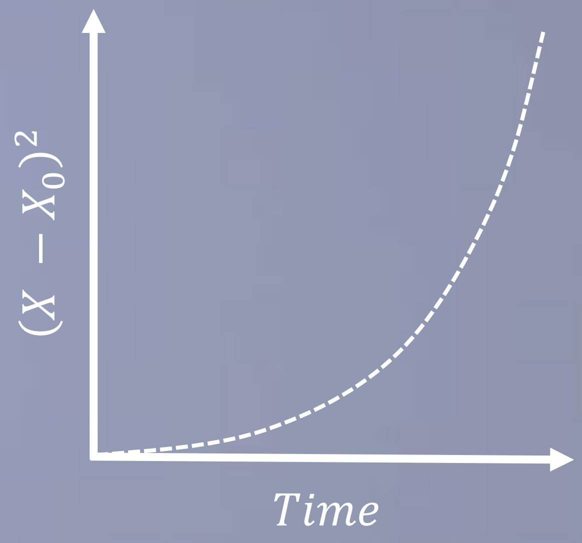
$$\hat{H}_{1\mathrm{D}} = \sum_k (\epsilon_0 - 2\tau \cos k)\hat{c}_k^\dagger \hat{c}_k = \sum_k \epsilon_k \hat{c}_k^\dagger \hat{c}_k$$



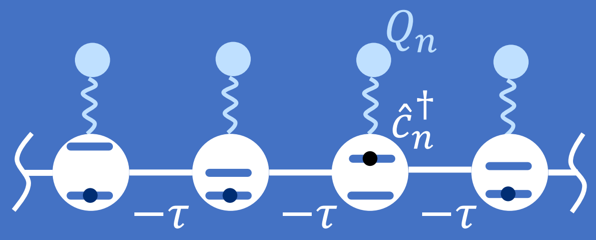


Ballistic (Coherent) Motion





1D Exciton + Phonon



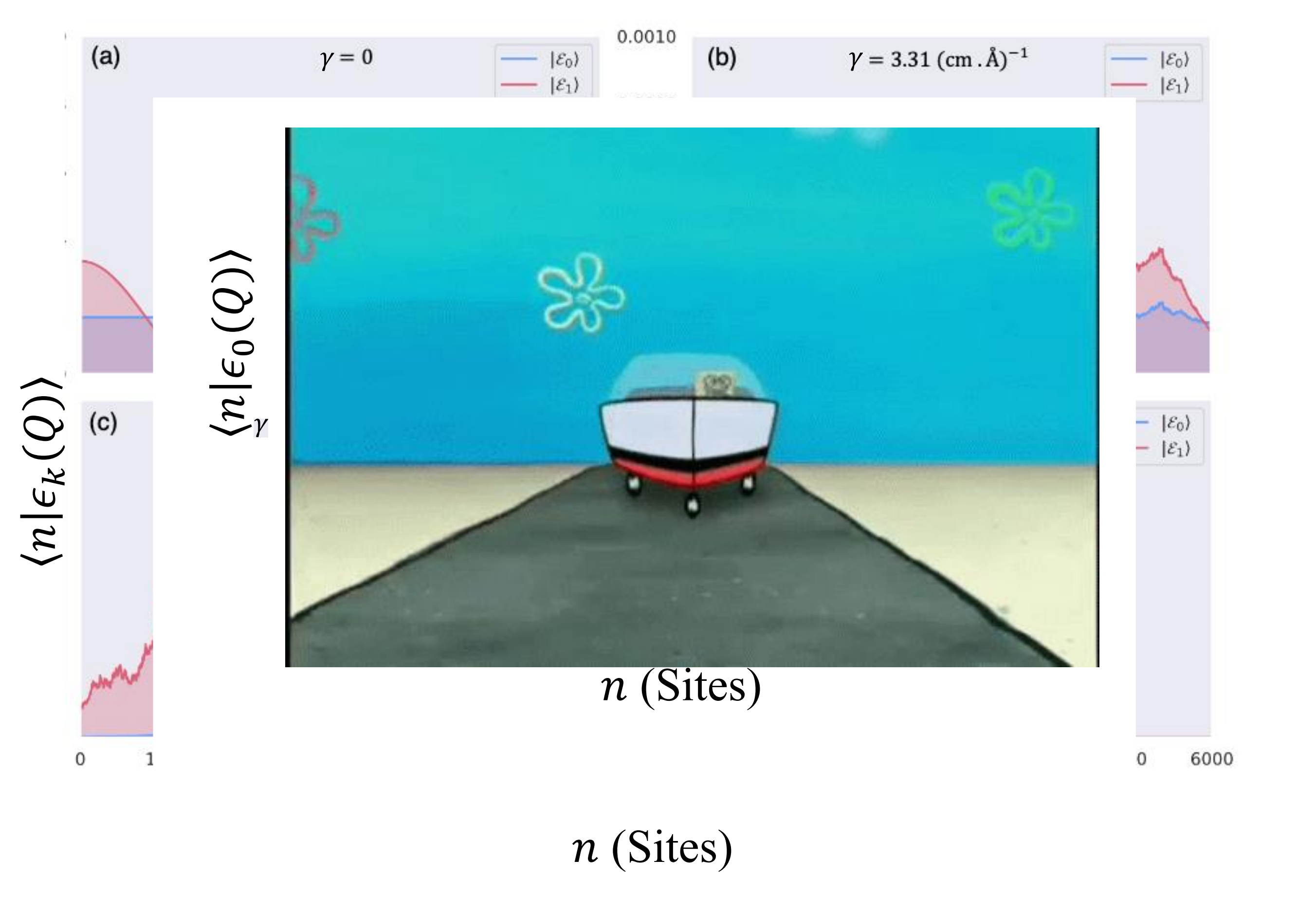
$$\hat{H}_{1D} = \epsilon_0 \sum_n \hat{c}_n^\dagger \hat{c}_n - \tau \sum_n \left(\hat{c}_{n+1}^\dagger \hat{c}_n + \hat{c}_n^\dagger \hat{c}_{n+1}\right)$$

$$+ \gamma \sum_n Q_n \hat{c}_n^\dagger \hat{c}_n + \frac{1}{2} \sum_n \left(P_n^2 + \omega_p^2 Q_n^2\right)$$

Electronic Hamiltonian

$$\hat{H}_{el} = \hat{H}_{1D} - \sum_n \frac{P_n^2}{2}$$

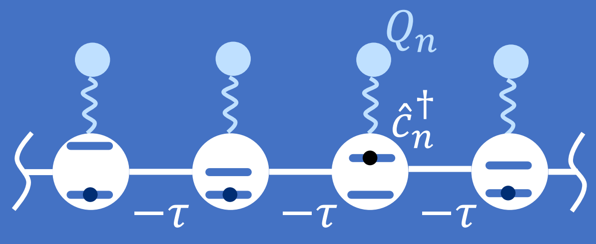
$$\hat{H}_{el} \, |\epsilon_k(\boldsymbol{Q})\rangle = \epsilon_k(\boldsymbol{Q}) |\epsilon_k(\boldsymbol{Q})\rangle$$



Phonon fluctuation leads to energetic disorder which leads to localization of eigenstates

This disorder is dynamic, i.e. time-dependent

Exciton Diffusion (Ehrenfest Method)

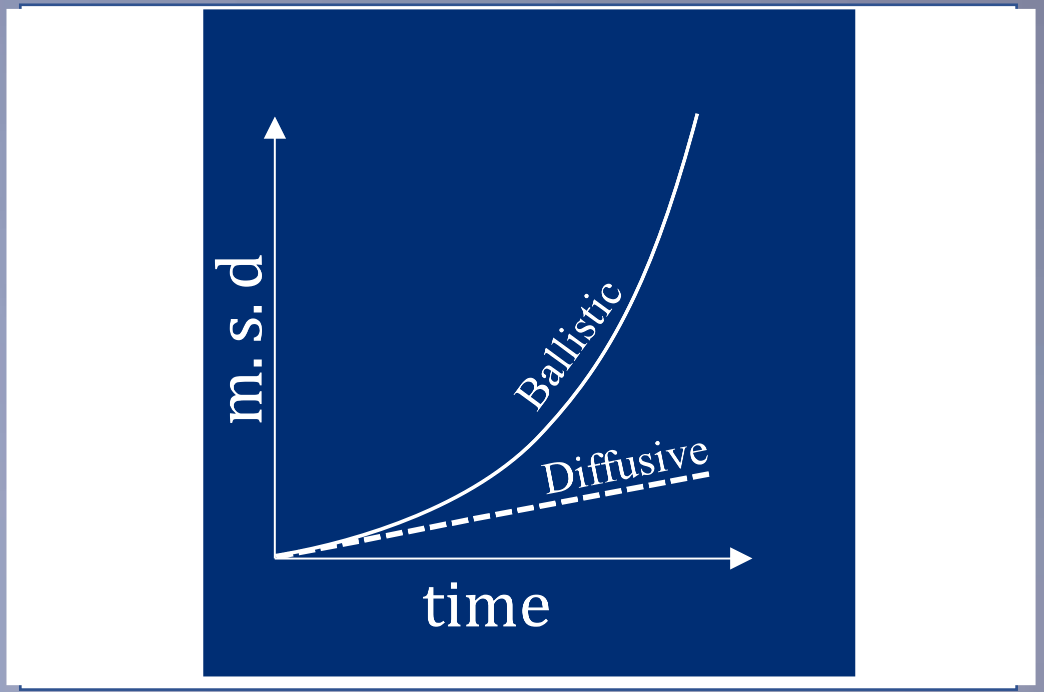


$$\hat{H}_{1\mathrm{D}} = \epsilon_0 \sum_n \hat{c}_n^\dagger \hat{c}_n - \tau \sum_n \left(\hat{c}_{n+1}^\dagger \hat{c}_n + \hat{c}_n^\dagger \hat{c}_{n+1}\right) + \gamma \sum_n Q_n \hat{c}_n^\dagger \hat{c}_n + \frac{1}{2} \sum_n \left(P_n^2 + \omega_p^2 Q_n^2\right)$$

Electronic Hamiltonian

$$\hat{H}_{el} = \hat{H}_{1\mathrm{D}} - \sum_n \frac{P_n^2}{2}$$

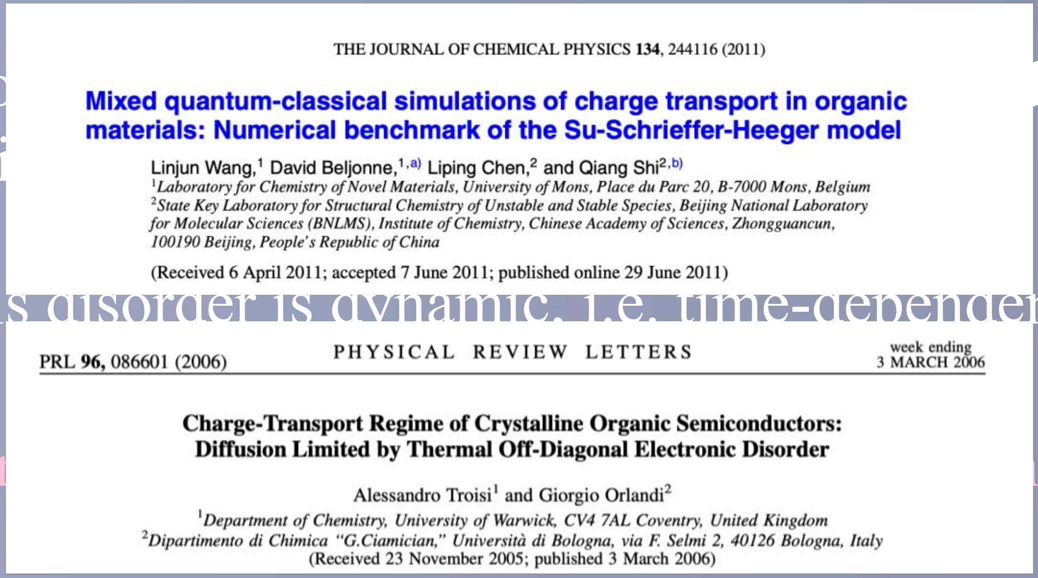
$$\hat{H}_{el} \, |\epsilon_k(\boldsymbol{Q})\rangle = \epsilon_k(\boldsymbol{Q}) |\epsilon_k(\boldsymbol{Q})\rangle$$



Pho… …ler
whi…

This disorder is dynamic, i.e. time-dependent

Dy…


THE JOURNAL OF CHEMICAL PHYSICS **134**, 244116 (2011)

Mixed quantum-classical simulations of charge transport in organic materials: Numerical benchmark of the Su-Schrieffer-Heeger model

Linjun Wang,[1] David Beljonne,[1,a)] Liping Chen,[2] and Qiang Shi[2,b)]
[1]Laboratory for Chemistry of Novel Materials, University of Mons, Place du Parc 20, B-7000 Mons, Belgium
[2]State Key Laboratory for Structural Chemistry of Unstable and Stable Species, Beijing National Laboratory for Molecular Sciences (BNLMS), Institute of Chemistry, Chinese Academy of Sciences, Zhongguancun, 100190 Beijing, People's Republic of China

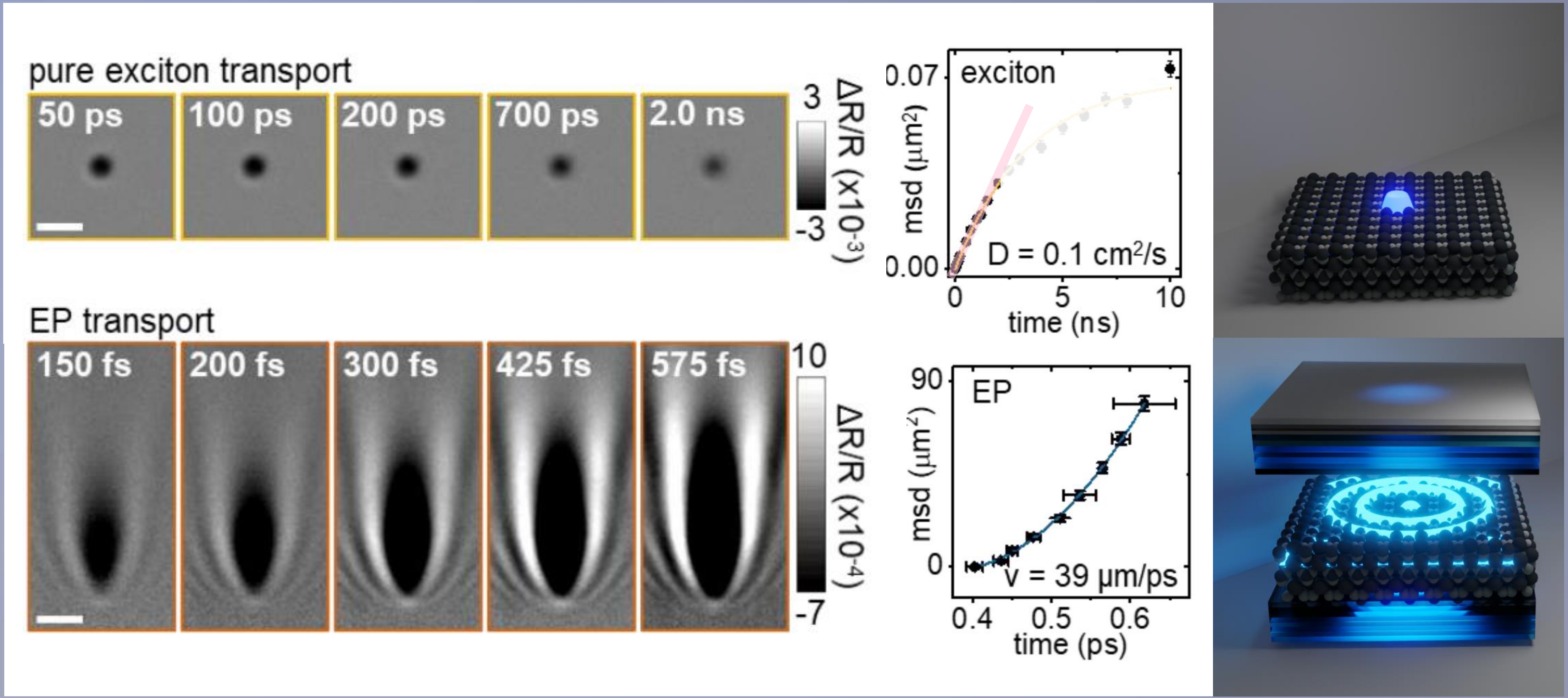
(Received 6 April 2011; accepted 7 June 2011; published online 29 June 2011)

PRL **96,** 086601 (2006) PHYSICAL REVIEW LETTERS week ending 3 MARCH 2006

Charge-Transport Regime of Crystalline Organic Semiconductors: Diffusion Limited by Thermal Off-Diagonal Electronic Disorder

Alessandro Troisi[1] and Giorgio Orlandi[2]
[1]Department of Chemistry, University of Warwick, CV4 7AL Coventry, United Kingdom
[2]Dipartimento di Chimica "G.Ciamician," Università di Bologna, via F. Selmi 2, 40126 Bologna, Italy
(Received 23 November 2005; published 3 March 2006)

Exciton-Polariton Transport



Xu†, Mandal†, …, Milan, Reichman (arXiv: 2205.01176) 2022

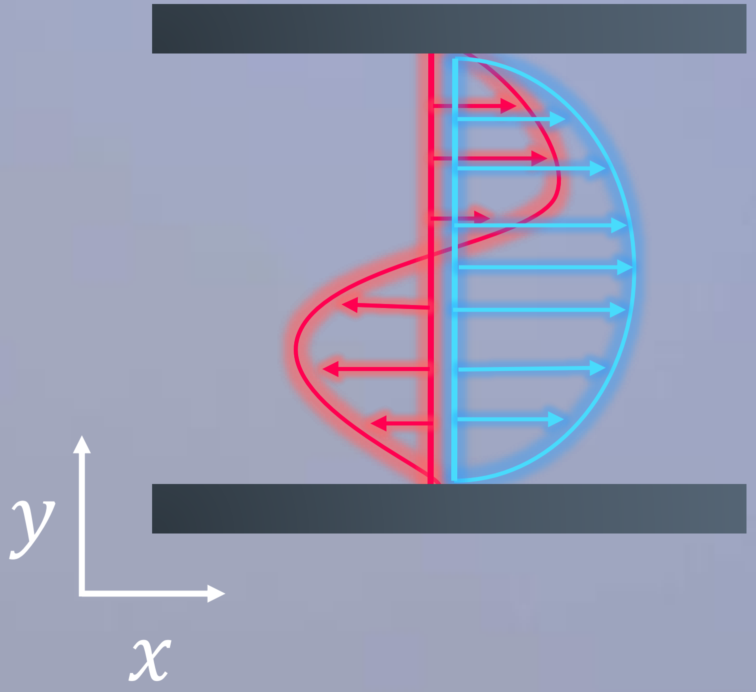
Also see:
Pandya et. Al. Adv. Sci. 9, 2105569 (2022)
Balasubrahmaniyam et. al. Nat. Matter (2023)

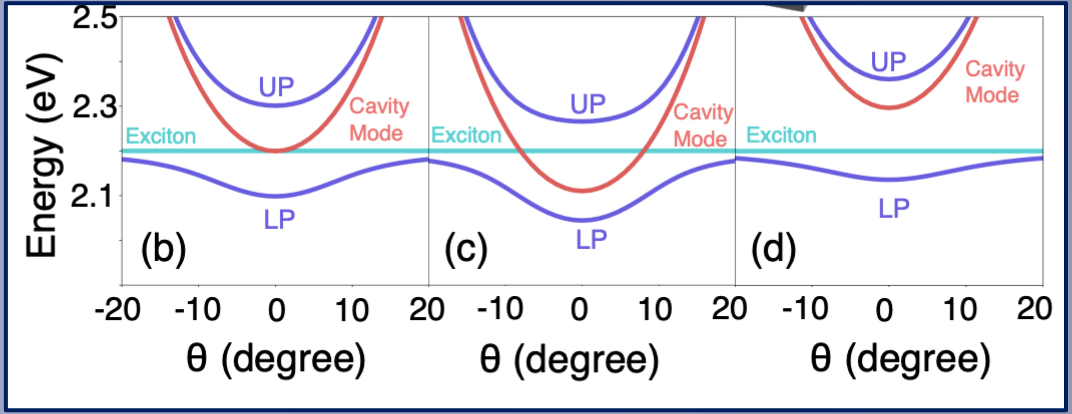
Light-Matter Hamiltonian

$$\hat{H}_{\mathrm{LM}} = \epsilon_0 \sum_n \hat{c}_n^\dagger \hat{c}_n - \tau \sum_n (\hat{c}_{n+1}^\dagger \hat{c}_n + \hat{c}_n^\dagger \hat{c}_{n+1}) + \gamma \sum_n Q_n \hat{c}_n^\dagger \hat{c}_n + \frac{1}{2} \sum_n (P_n^2 + \omega_p^2 Q_n^2)$$

$$+ \sum_{\mathbf{k}} \hat{a}_{\mathbf{k}}^\dagger \hat{a}_{\mathbf{k}}\, \omega_{\mathbf{k}} + g_c \sum_{n,\mathbf{k}} (\hat{c}_n^\dagger \hat{a}_{\mathbf{k}} e^{ik_x.\mathbf{R_n}} + \hat{a}_{\mathbf{k}}^\dagger \hat{c}_n e^{-ik_x.\mathbf{R_n}}) \sin(k_y.\mathbf{R_n})$$

Photon Modes

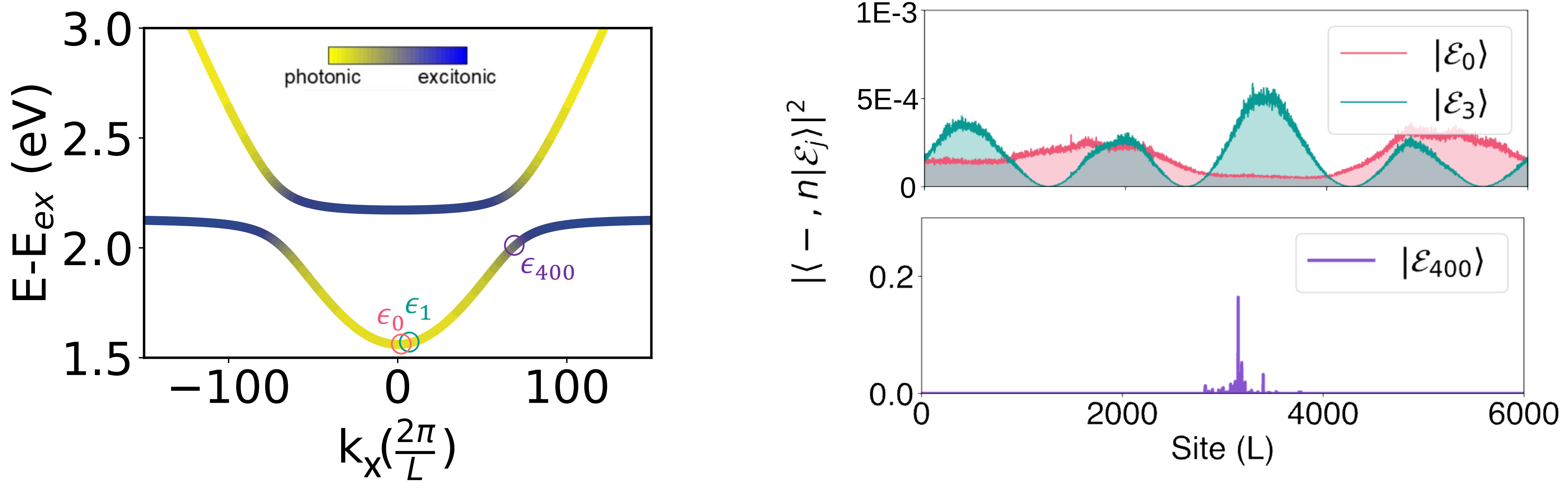




Cavity operators do not couple to phonons. Photon frequency is off-resonant to phonon frequency.

Polaritons, which are partially excitonic, have effectively smaller coupling to phonons.

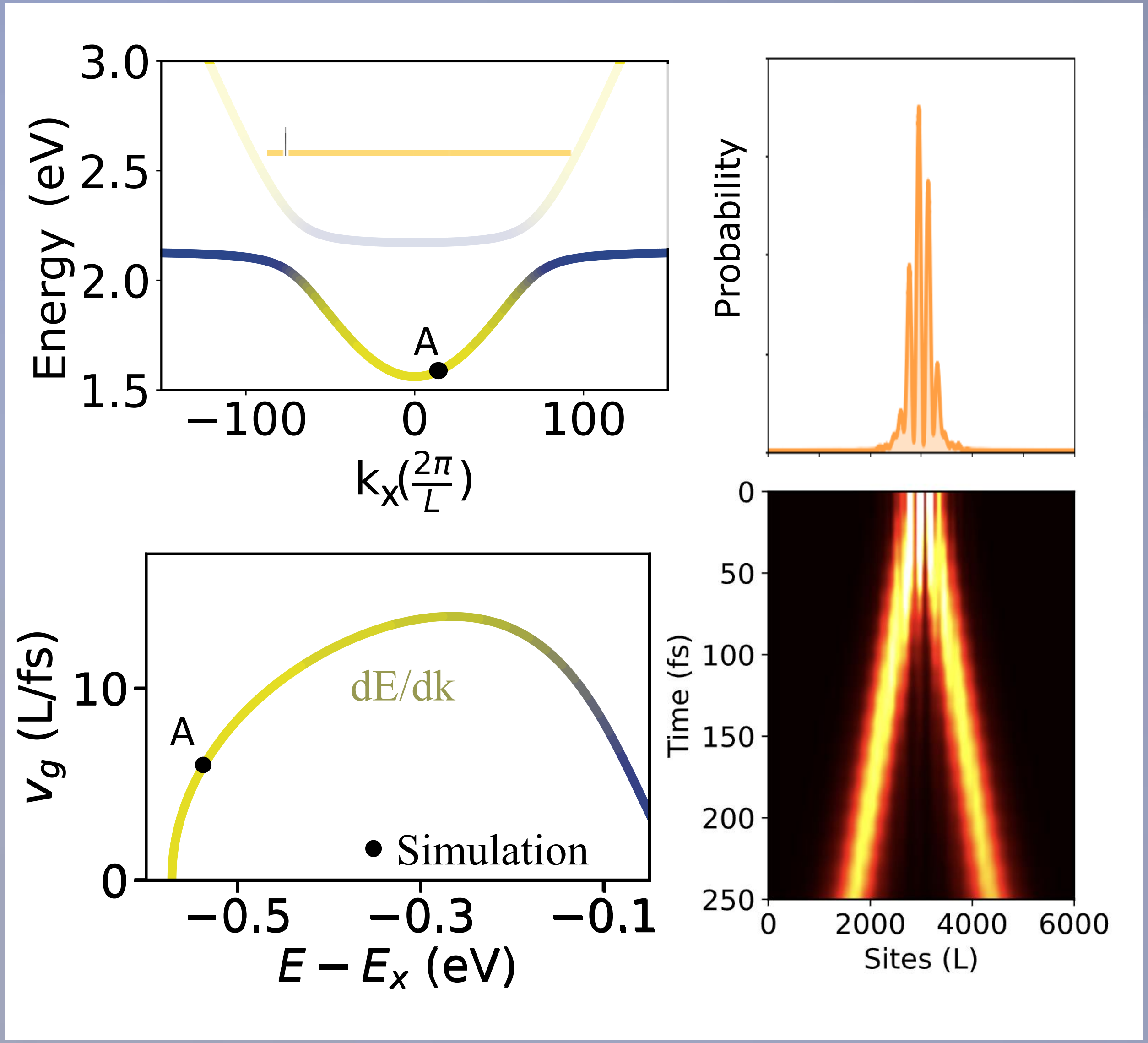
Polariton States at various Detuning



Higher Exciton character → Higher Phonon Coupling → Higher Localization

Xu[†], Mandal[†], Milan, Reichman (arXiv: 2205.01176) 2022

Group Velocity



At low exciton character (at large detuning) the exciton-polariton wavepacket propagates ballistically.

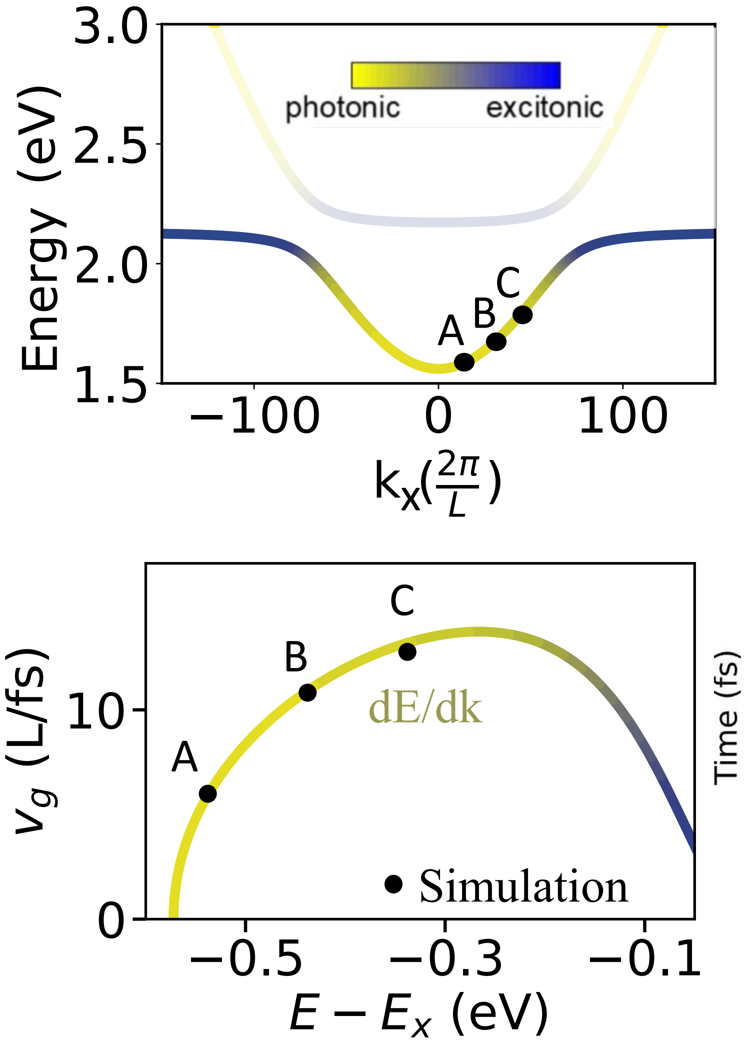
The wavefront velocity matches the expected group velocity.

Also see: Berghuis et. Al. ACS photonics 9 (7), 2263-2272

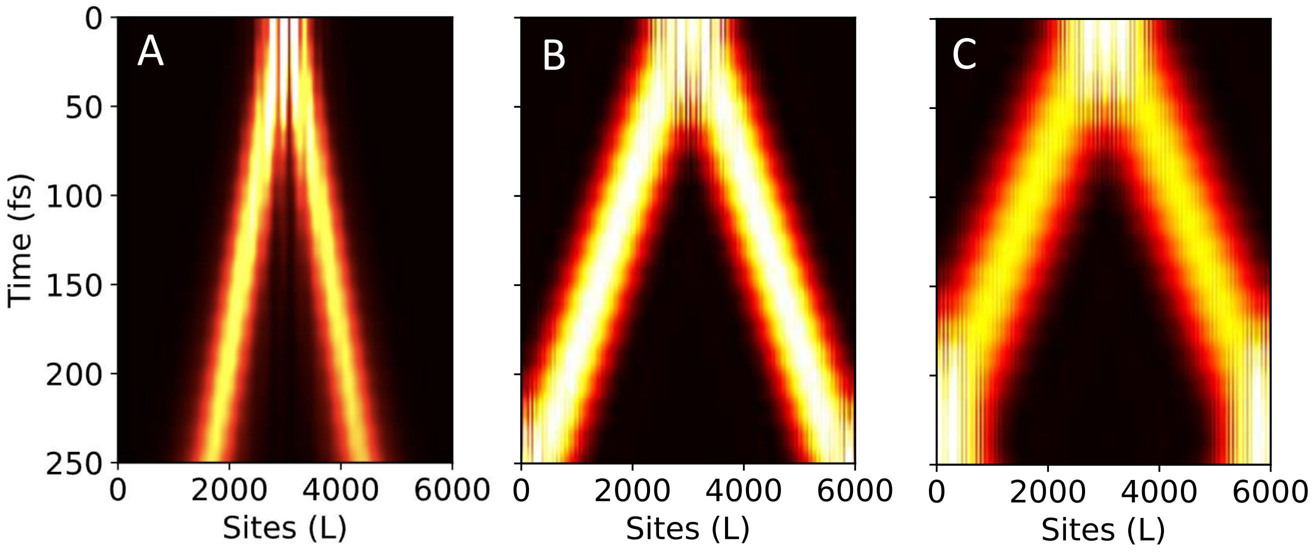
Sokolovskii et. Al. arXiv:2209.07309

Xu†, Mandal†, Milan, Reichman (arXiv: 2205.01176) 2022

Group Velocity vs Exciton Character

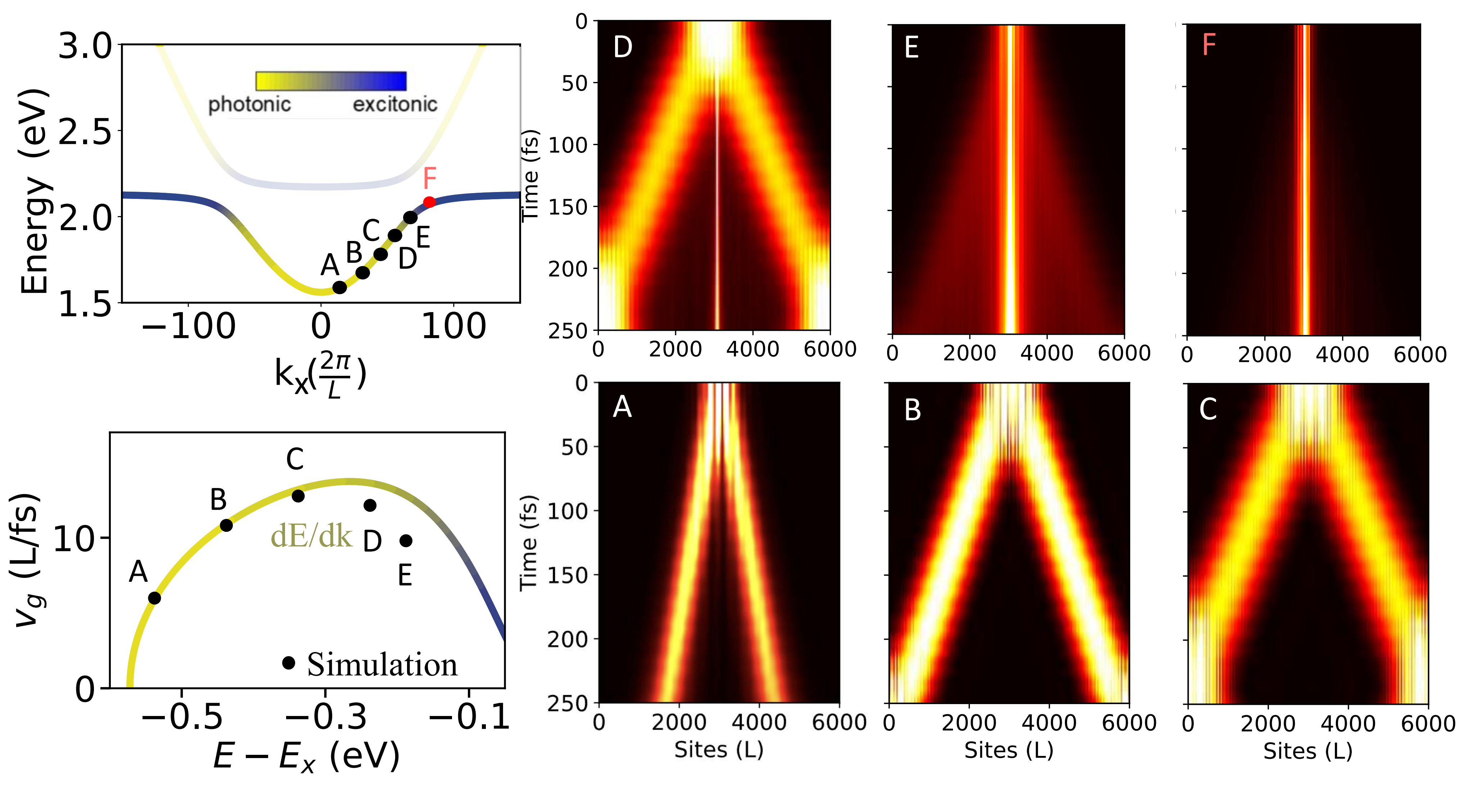


The wavefront velocity matches the expected group velocity for exciton character < 15%.



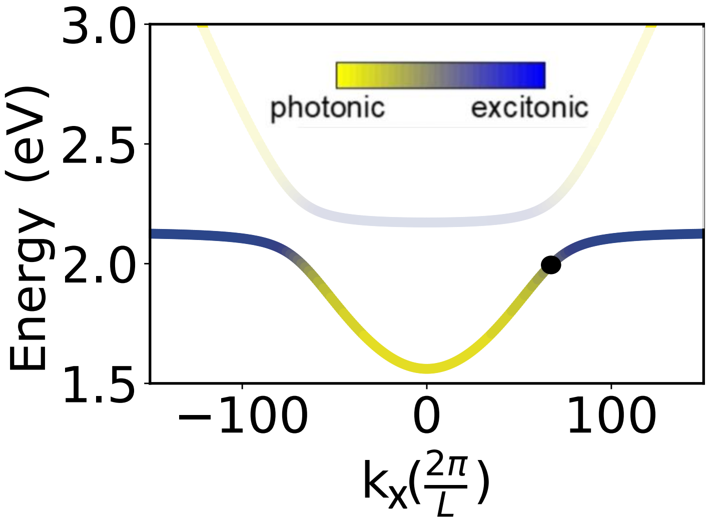
Xu†, Mandal†, Milan, Reichman (arXiv: 2205.01176) 2022

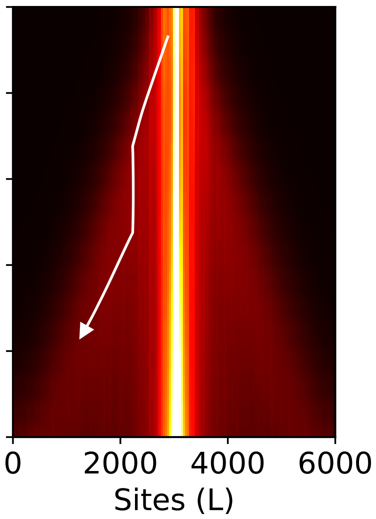
Group Velocity

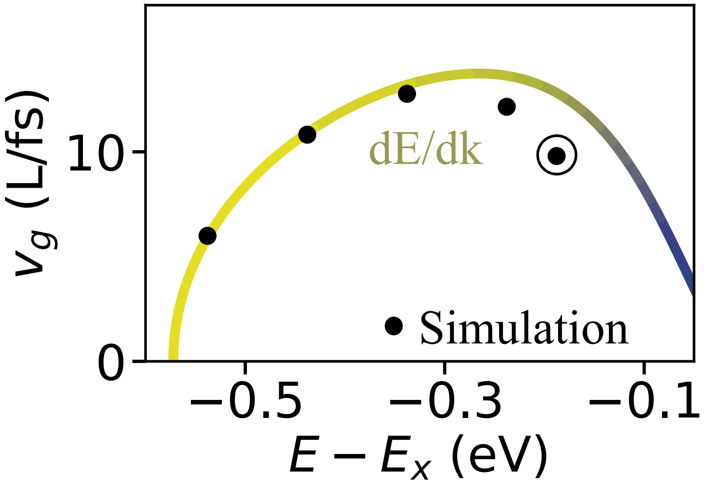


Xu†, Mandal†, Milan, Reichman (arXiv: 2205.01176) 2022

Velocity Renormalization





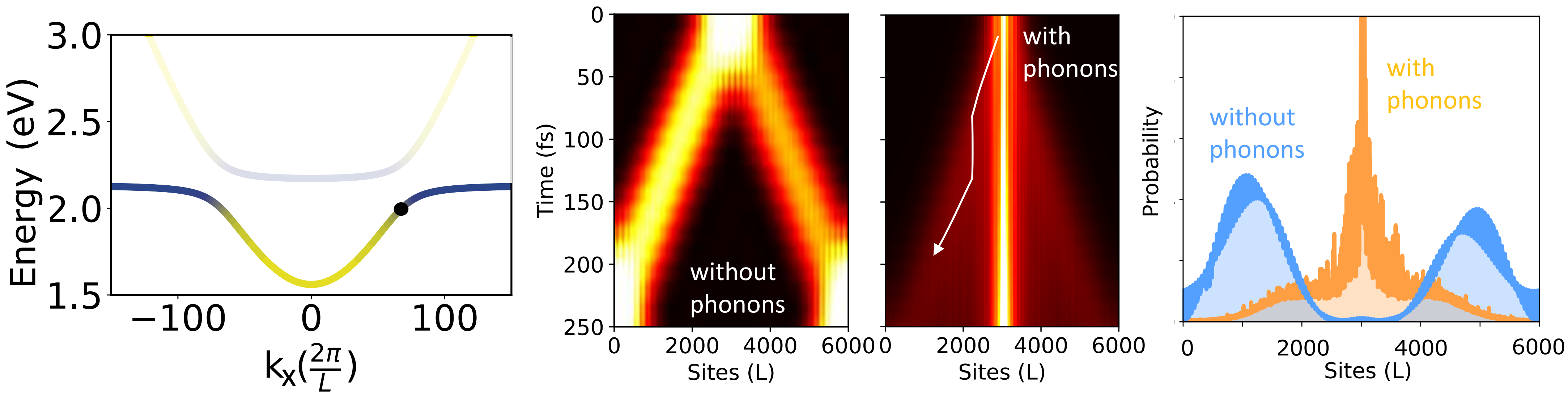


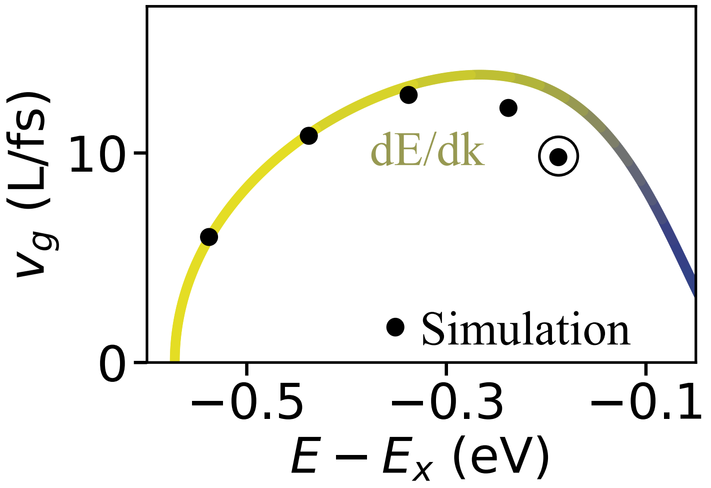
At exciton character 25-40% → Phonons **renormalizes** polariton group velocity

Phonon induced transient localization leads to reduced velocity

Xu†, Mandal†, Milan, Reichman (arXiv: 2205.01176) 2022

Group Velocity

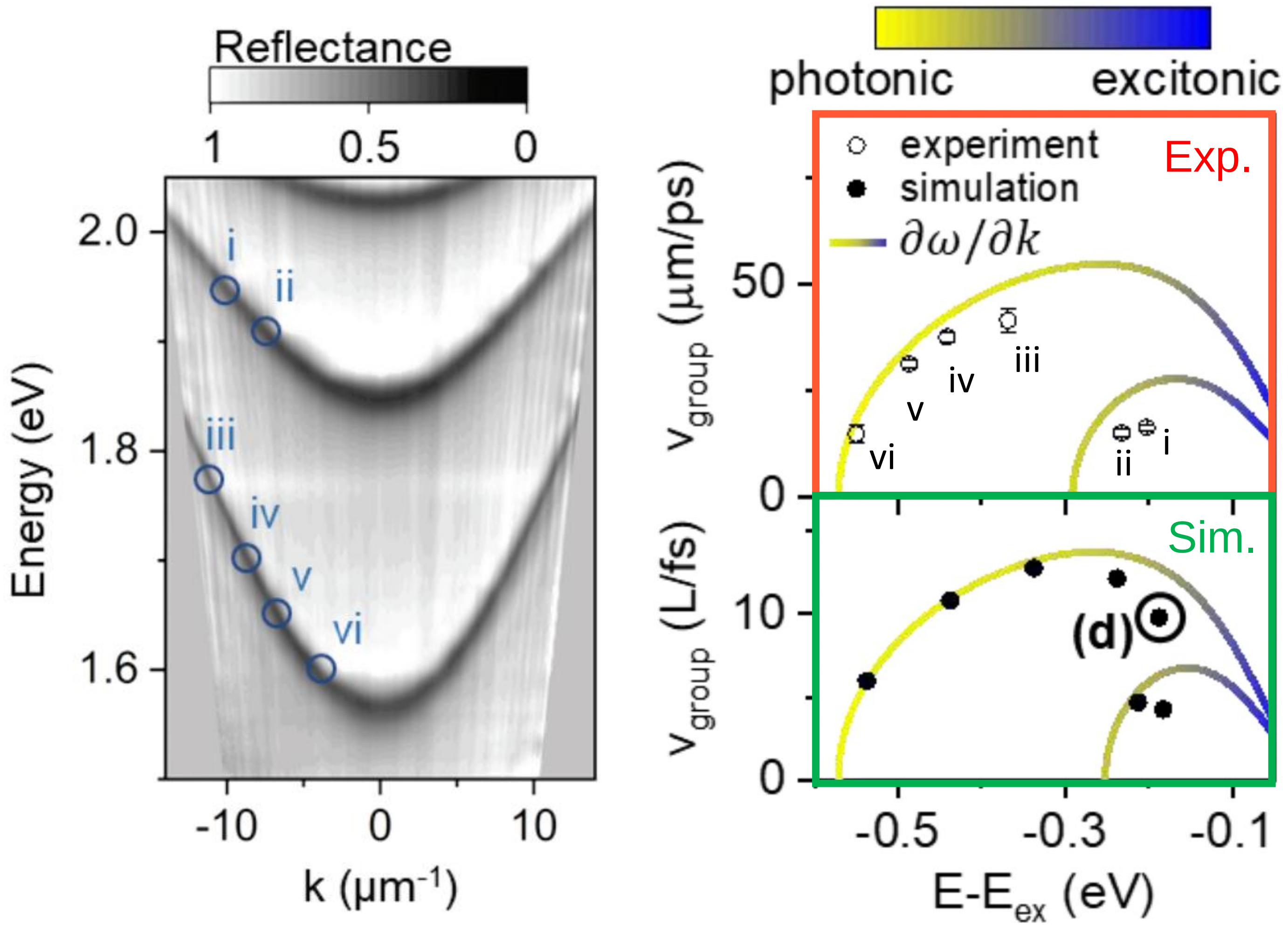




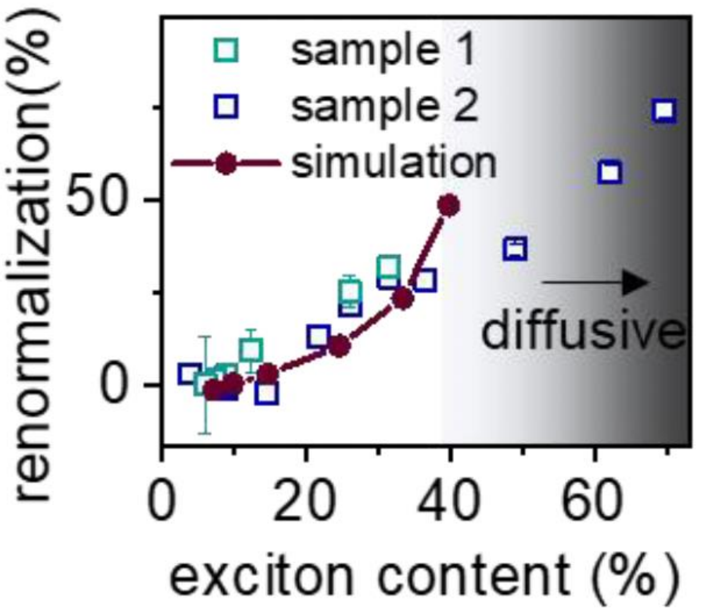
We can confirm the renormalization by comparing with a case without phonons.

Xu[†], Mandal[†], Milan, Reichman (arXiv: 2205.01176) 2022

Experiment & Theory: Group Velocity



We experimentally observe the same phenomena.



Xu†, Mandal†, Milan, Reichman (arXiv: 2205.01176) 2022

Non-Markovian Master Equation

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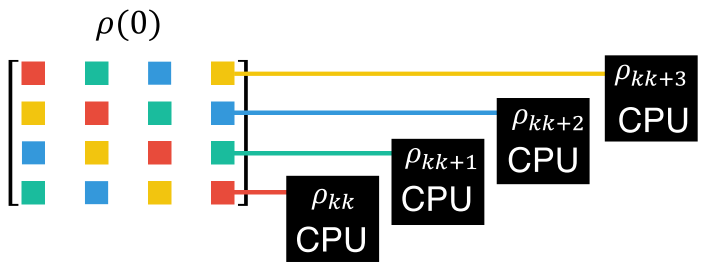
Aim: Simulate a matter system with ~10^4 sites and 10^2 cavity modes (~10000 states)

N^5 → N^4 → N^3 → N^2

$$\dot{\rho}_{k,k+\delta k}(t) = \int_0^t d\tau\, K_{k,k+\delta k,p,p+\delta k}(t-\tau)\rho_{p,p+\delta k,}(t)$$

$$K_{k,k',p,p'} = K_{k,k',p,p+(k'-k)}$$

Momentum Conservation makes K a 3-dimensional tensor instead of 4-dimentional

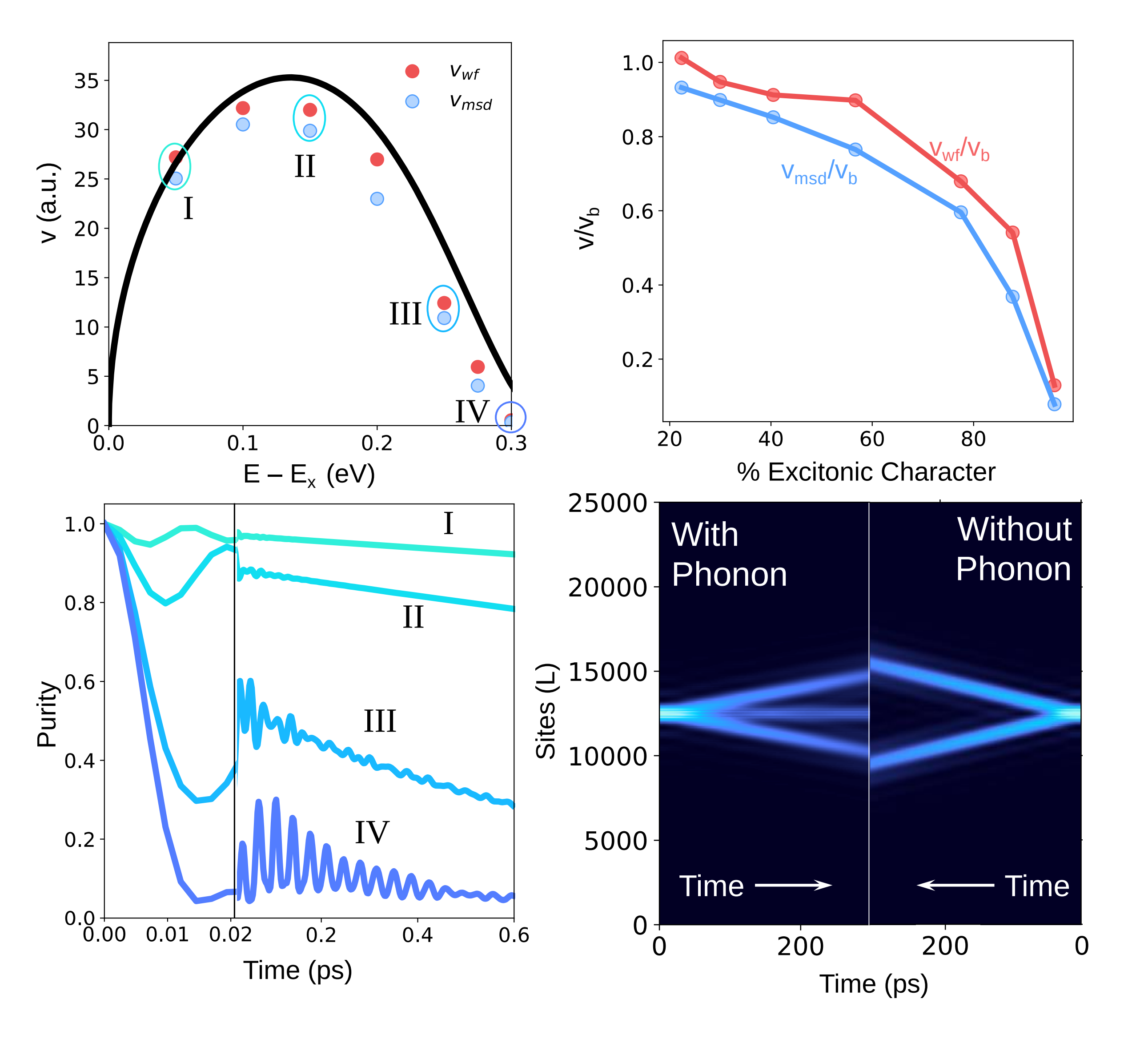


decoupled diagonals of density matrix in the reciprocal space – parallelized simulation

Prony decomposition of memory kernel + Coarse Graining Density Matrix.

Mandal, Reichman (in prep) 2023

Polariton Transport



Higher purity == Coherent.

We perform Non-Markovian Master Equation simulation with 25000 states!

Mandal, Reichman (in prep) 2023

Topics

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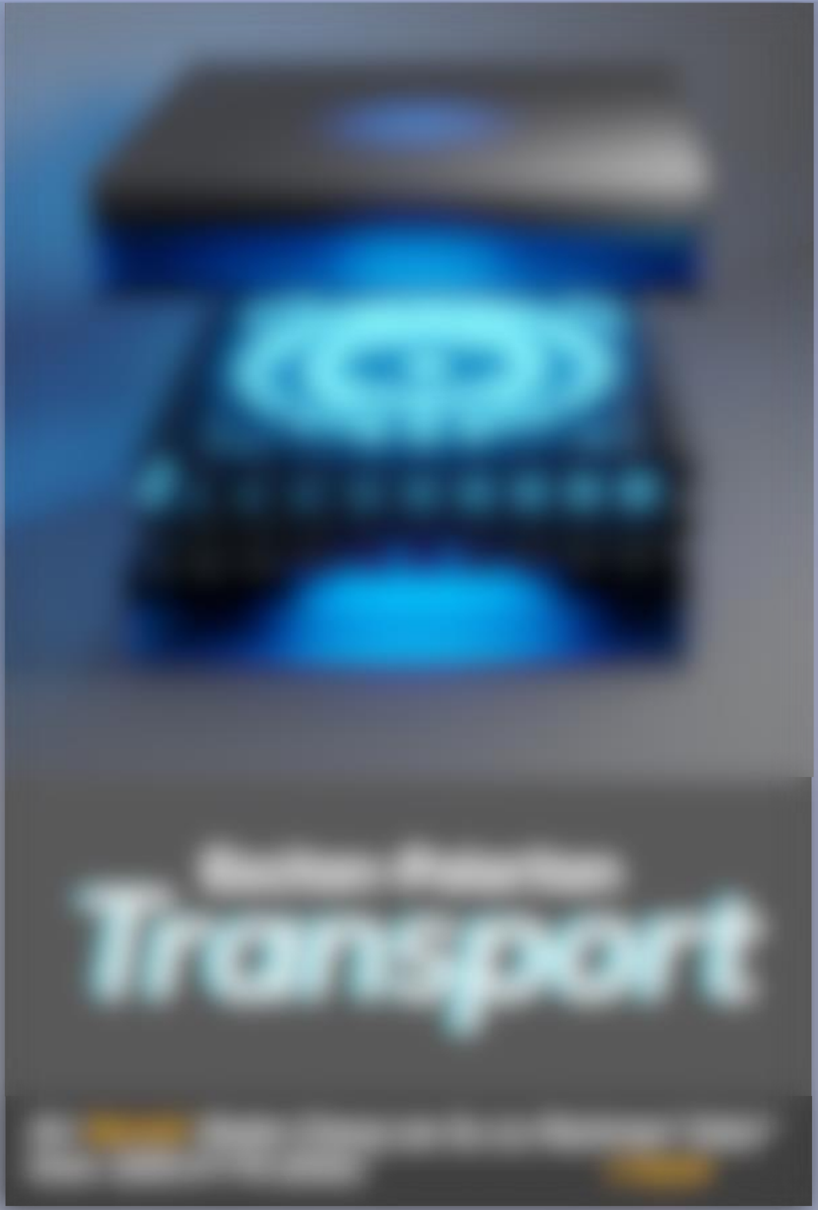
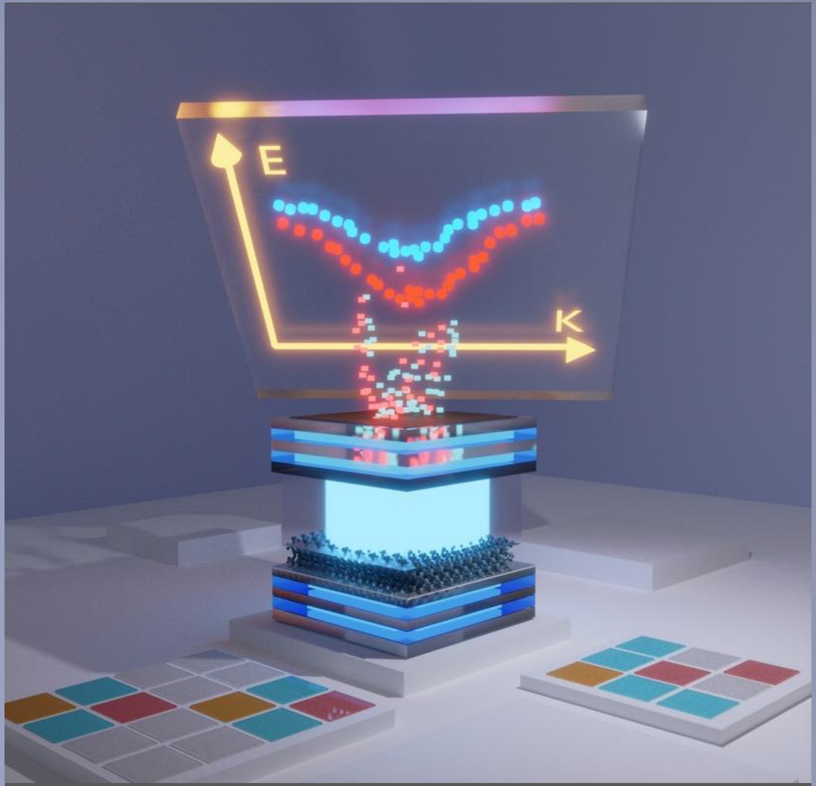




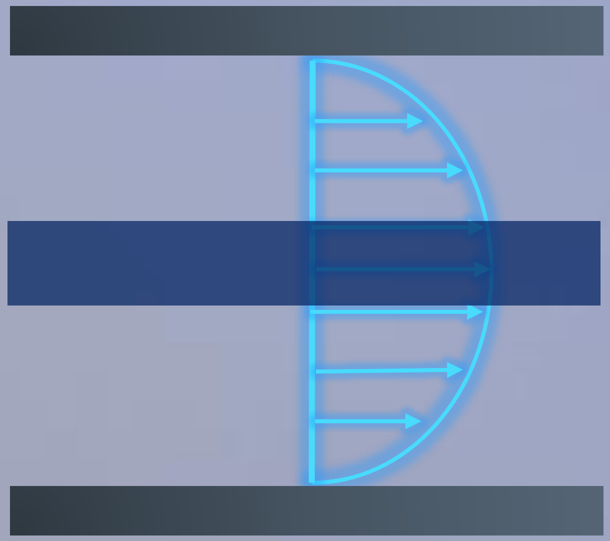
Exciton-Polariton

Dispersion

Mandal Xu Mahajan Lee Delor Reichman**
Nano Lett. 2023

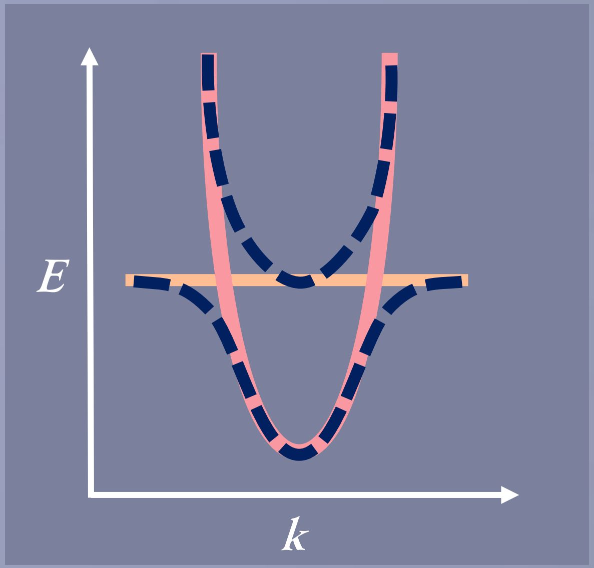
Light-Matter Hamiltonian

$$\hat{H}_{\mathrm{LM}} = \epsilon_0 \sum_n \hat{c}_n^\dagger \hat{c}_n - \tau \sum_n \left(\hat{c}_{n+1}^\dagger \hat{c}_n + \hat{c}_n^\dagger \hat{c}_{n+1}\right) + \sum_{\mathbf{k}} \hat{a}_{\mathbf{k}}^\dagger \hat{a}_{\mathbf{k}}\, \omega_{\mathbf{k}} + g_c \sum_{n,\mathbf{k}} \left(\hat{c}_n^\dagger \hat{a}_{\mathbf{k}} e^{ik_\parallel.\mathbf{R_n}} + \hat{a}_{\mathbf{k}}^\dagger \hat{c}_n e^{-ik_\parallel.\mathbf{R_n}}\right) \sin(k_\perp.\mathbf{R_n})$$



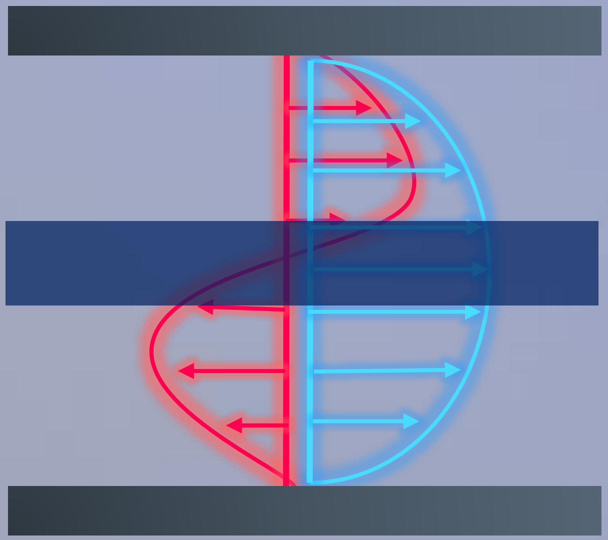
$$\hat{H}_{\mathrm{LM}} = \textstyle\sum_k \hat{c}_k^\dagger \hat{c}_k\, \epsilon_k + \hat{a}_k^\dagger \hat{a}_k \omega_k - g\left(\hat{a}_k^\dagger \hat{c}_k + \hat{c}_k^\dagger \hat{a}_k\right) = \sum H_k$$

$$\begin{pmatrix} \epsilon & \Omega_1 \\ \Omega_1 & \omega_{C_1} \end{pmatrix}$$



N+1 Model

Expectation: Exciton-Polariton Band Structure is obtained by a (N+1)x(N+1) Matrix.
N = number of cavity mode branches.



$$\hat{H}_{\mathrm{LM}} = \sum_k \hat{c}_k^\dagger \hat{c}_k \epsilon_k + \hat{a}_k^\dagger \hat{a}_k \omega_k - g\left(\hat{a}_k^\dagger \hat{c}_k + \hat{c}_k^\dagger \hat{a}_k\right) = \sum H_k$$

	Ω_1	Ω_2	Ω_3
Ω_1	c_1	0	0
Ω_2	0	c_2	0
Ω_3	0	0	c_3

Problem with N+1 Hamiltonian

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(N+1)x(N+1) model do not reproduce exciton polariton dispersion

2N x 2N model reproduce exciton polariton dispersion

N+1 Hamiltonian (cannot be used)

	Ω_1	Ω_2	Ω_3
Ω_1	c_1	0	0
Ω_2	0	c_2	0
Ω_3	0	0	c_3

2N Hamiltonian

	Ω_1	0	0	0	0
Ω_1	c_1	0	0	0	0
0	0		Ω_2	0	0
0	0	Ω_2	c_2	0	0
0	0	0	0		Ω_3
0	0	0	0	Ω_3	c_3

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Observation of photon-mode decoupling in a strongly coupled multimode microcavity

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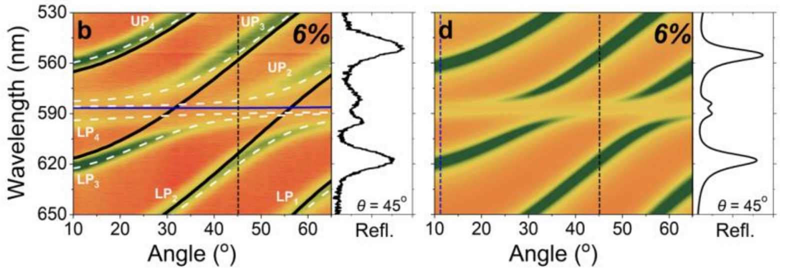
Kyriacos Georgiou,[1,2,a)] Kirsty E. McGhee,[1] Rahul Jayaprakash,[1] and David G. Lidzey[1,a)]



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Strong Coupling beyond the Light-Line

Kishan S. Menghrajani* and William L. Barnes*

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Ultralong-Range Polariton-Assisted Energy Transfer in Organic Microcavities

Kyriacos Georgiou,* Rahul Jayaprakash, Andreas Othonos and David G. Lidzey*

Theory

APPLIED PHYSICS LETTERS 107, 231104 (2015)

Maxwell consideration of polaritonic quasi-particle Hamiltonians in multi-level systems

Steffen Richter, Tom Michalsky, Lennart Fricke, Chris Sturm, Helena Franke, Marius Grundmann, and Rüdiger Schmidt-Grund
Institut für Experimentelle Physik II, Universität Leipzig, Linnéstr. 5, 04103 Leipzig, Germany

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PHYSICAL REVIEW B 103, L241407 (2021)
Letter

Coupling and decoupling of polaritonic states in multimode cavities

M. Balasubrahmaniyam,[1] Cyriaque Genet,[2] and Tal Schwartz[1,*]
[1]School of Chemistry, Raymond and Beverly Sackler Faculty of Exact Sciences and Tel Aviv University Center for Light-Matter Interaction, Tel Aviv University, Tel Aviv 6997801, Israel
[2]Université de Strasbourg, CNRS, Institut de Science et d'Ingénierie Supramoléculaires, UMR 7006, 67000 Strasbourg, France

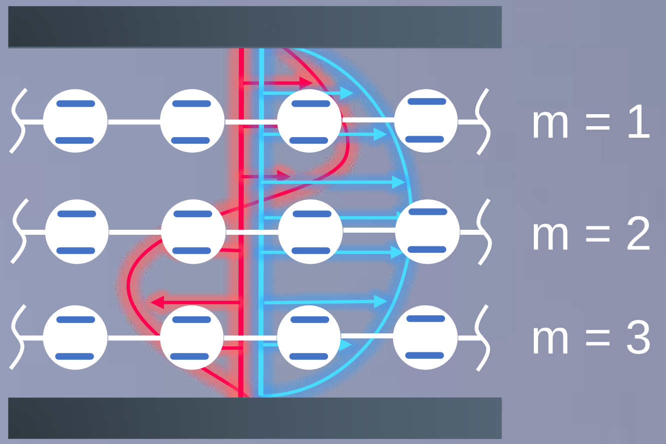
We have a simple microscopic understanding of this phenomena

Multilayer GTC Hamiltonian

$$\hat{H}_{\mathrm{LM}} = \sum_{k} \hat{c}^{\dagger}_{k_x,m}\hat{c}_{k_x,m}\,\epsilon_k + \hat{a}^{\dagger}_{k_x k_y}\hat{a}_{k_x k_y}\omega_{\boldsymbol{k}} + g\left(\hat{a}^{\dagger}_{k_x k_y}\hat{c}_{k_x,m} + \hat{c}^{\dagger}_{k_x,m}\hat{a}_{k_x k_y}\right)\sin k_y Y_m$$

$$= \sum_{k} \hat{H}_{k_x}$$

$$\begin{bmatrix}\hat{c}^{\dagger}_{k_x,1} & \cdots & \hat{c}^{\dagger}_{k_x,m}\end{bmatrix} \; Q \times R \; \begin{bmatrix}\hat{a}_{k_x,1} \\ \vdots \\ \hat{a}_{k_x,m}\end{bmatrix}$$



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New Excitonic Operators

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$$\hat{H}_{\mathrm{LM}} = \sum_{k} \hat{c}^{\dagger}_{k_x,m} \hat{c}_{k_x,m}\, \epsilon_k + \hat{a}^{\dagger}_{k_x k_y} \hat{a}_{k_x k_y} \omega_{\boldsymbol{k}} + g\left(\hat{a}^{\dagger}_{k_x k_y} \hat{c}_{k_x,m} + \hat{c}^{\dagger}_{k_x,m} \hat{a}_{k_x k_y}\right) \sin k_y Y_m$$

$$= \sum_{k} \hat{H}_{k_x}$$

$$\begin{bmatrix} \hat{c}^{\dagger}_{k_x,1} & \cdots & \hat{c}^{\dagger}_{k_x,m} \end{bmatrix} Q \times R \begin{bmatrix} \hat{a}_{k_x,1} \\ \vdots \\ \hat{a}_{k_x,m} \end{bmatrix}$$

$$\rightarrow \begin{bmatrix} \hat{c}^{\dagger}_{k_x,\kappa_1} & \cdots & \hat{c}^{\dagger}_{k_x,\kappa_{N_e}} \end{bmatrix}$$

For filled cavities $\hat{c}^{\dagger}_{k_x,k_y}$ operator only couple to $\hat{a}_{k_x k_y}$

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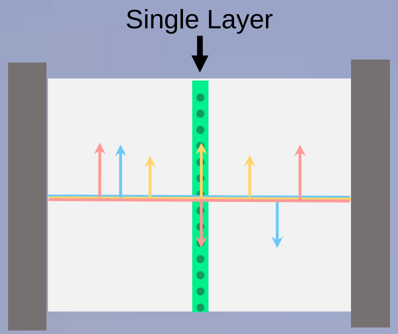
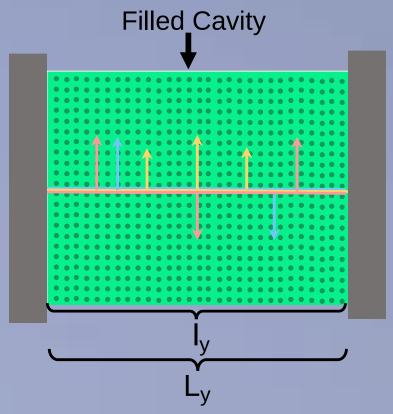
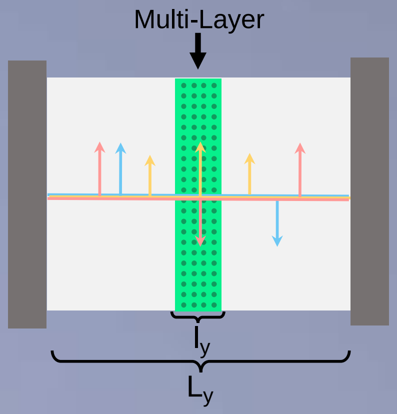
Simple Matrix Models



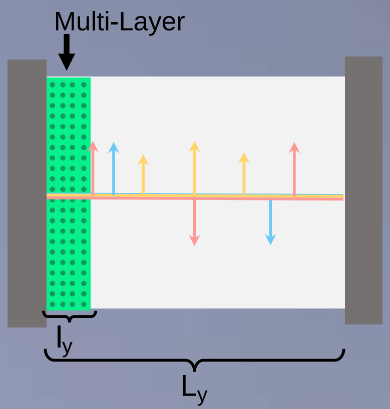
ϵ	Ω_1	Ω_2	Ω_3	Ω_4	Ω_5
Ω_1	ω_{c_1}	0	0	0	0
Ω_2	0	ω_{c_2}	0	0	0
Ω_3	0	0	ω_{c_3}	0	0
Ω_4	0	0	0	ω_{c_4}	0
Ω_5	0	0	0	0	ω_{c_5}



ϵ	Ω_1	0	0	0	0
Ω_1	ω_{c_1}	0	0	0	0
0	0	ϵ	Ω_2	0	0
0	0	Ω_2	ω_{c_2}	0	0
0	0	0	0	ϵ	Ω_3
0	0	0	0	Ω_3	ω_{c_3}



ϵ	Ω_1	Ω_3	0	0	0
Ω_1	ω_{c_1}	0	0	0	0
Ω_3	0	ω_{c_3}	0	0	0
0	0	0	ϵ	Ω_2	Ω_4
0	0	0	Ω_2	ω_{c_2}	0
0	0	0	Ω_4	0	ω_{c_4}



ϵ	Ω_1	Ω_2	Ω_3	Ω_4	Ω_5
Ω_1	ω_{c_1}	0	0	0	0
Ω_2	0	ω_{c_2}	0	0	0
Ω_3	0	0	ω_{c_3}	0	0
Ω_4	0	0	0	ω_{c_4}	0
Ω_5	0	0	0	0	ω_{c_5}

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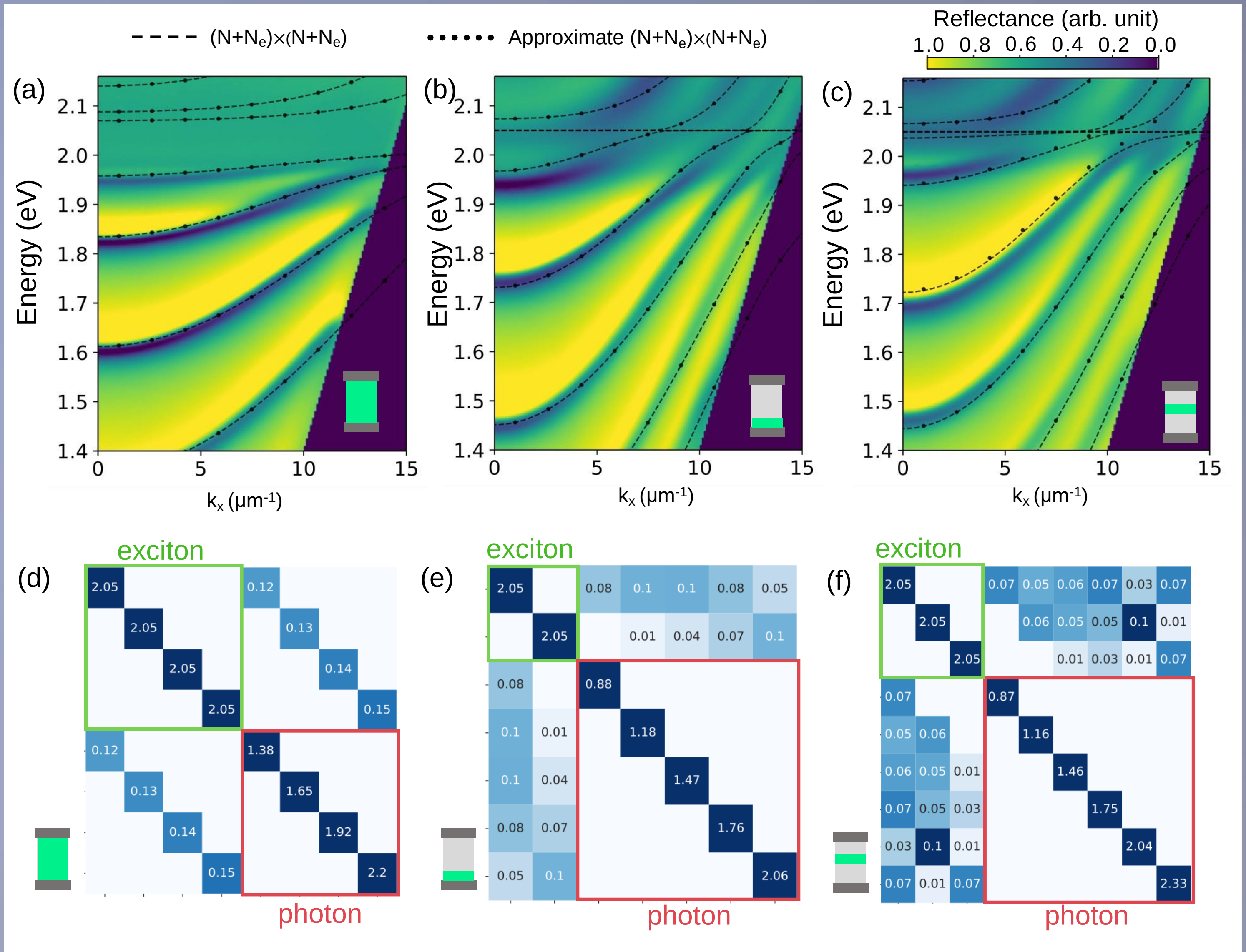
Comparing to Experiment

The theoretical models predicts the experiment



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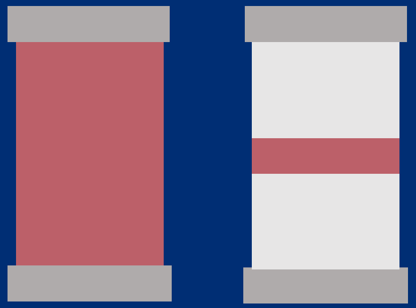
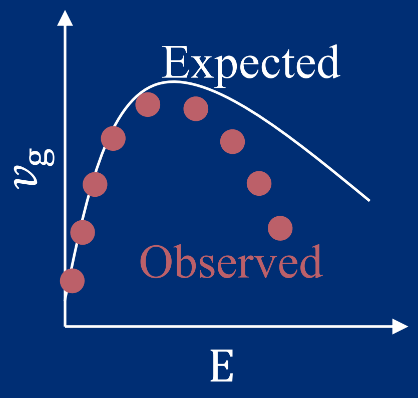
Conclusion



- For extended materials: 2N Hamiltonian (N = number of cavity mode branch) should be used.
- For arbitrary setup : $N+N_e$ Hamiltonian (N = number of cavity mode branch, $N_e < N$) should be used.



- We simulated exciton-polariton transport with Semi-classical approach.
- We find ballistic (Coherent) motion when exciton character is low. (< 40%)
- We find phonons can **rescale** group velocity through a transient localization mechanism.
- Coherence is long-lived (~ ps) for low exciton character.

Acknowledgements

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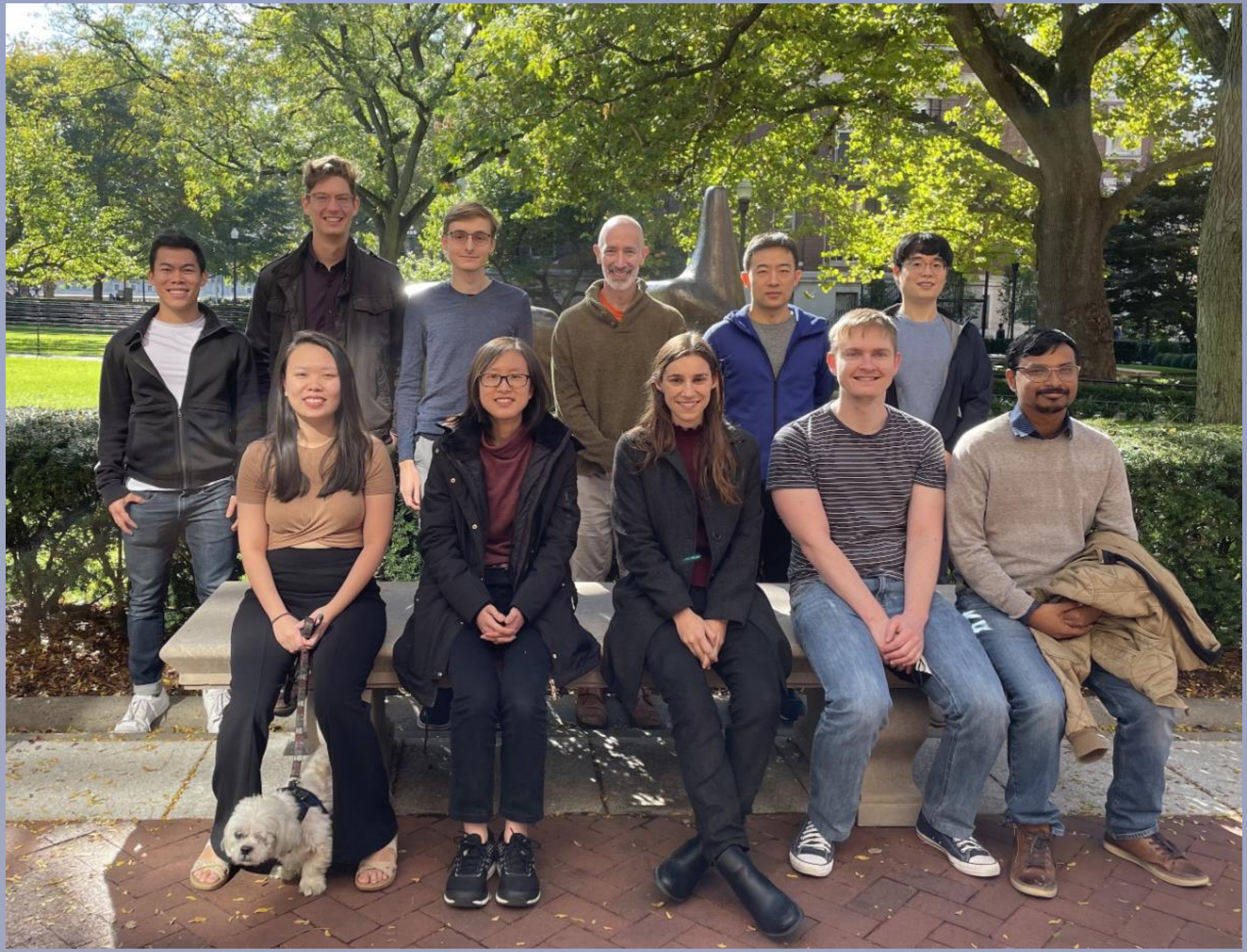

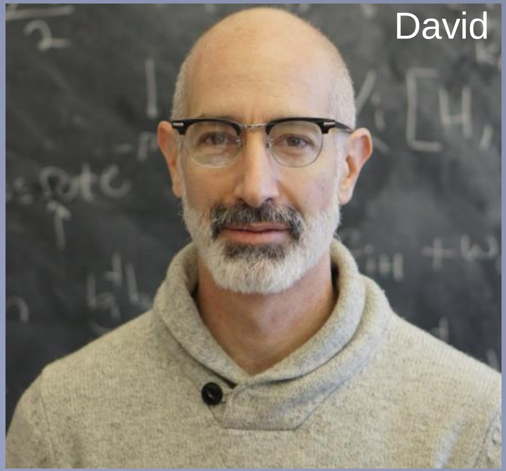

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