Strongly Interacting Polaritons in Moiré Materials

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Work in collaboration with Arturo Camacho-Guardian

[Arturo Camacho-Guardian & NRC, arXiv:2108.06177]

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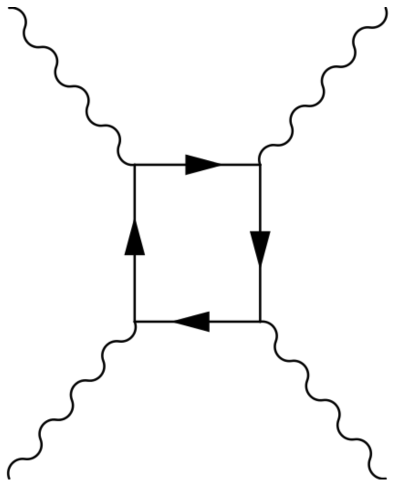
SIMONS FOUNDATION

Photon-Photon Interactions

Free space

photon-photon scattering via background polarisation

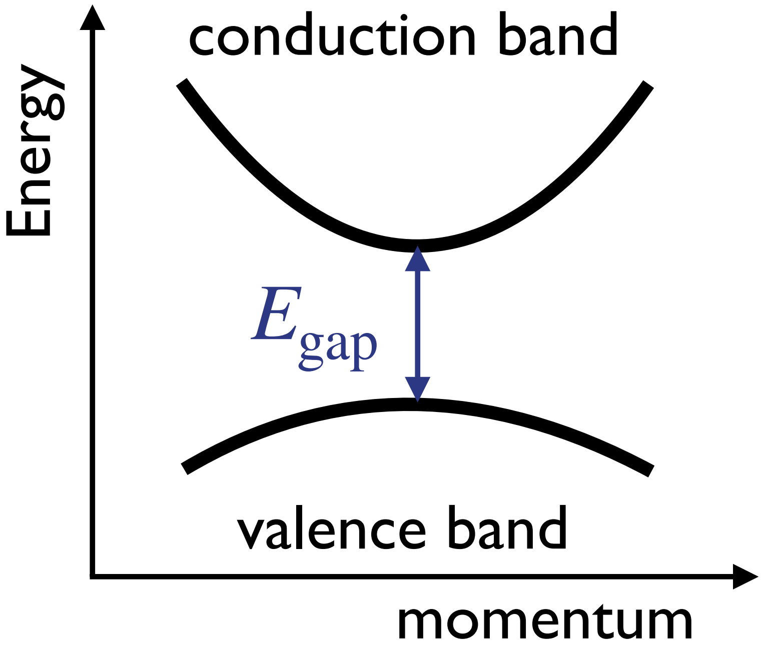
strong scattering when electron-positron creation permitted



[source: Wikipedia]

Insulating medium (semiconductor)

electron-hole creation for $E_{\text{photon}} \geq E_{\text{gap}}$

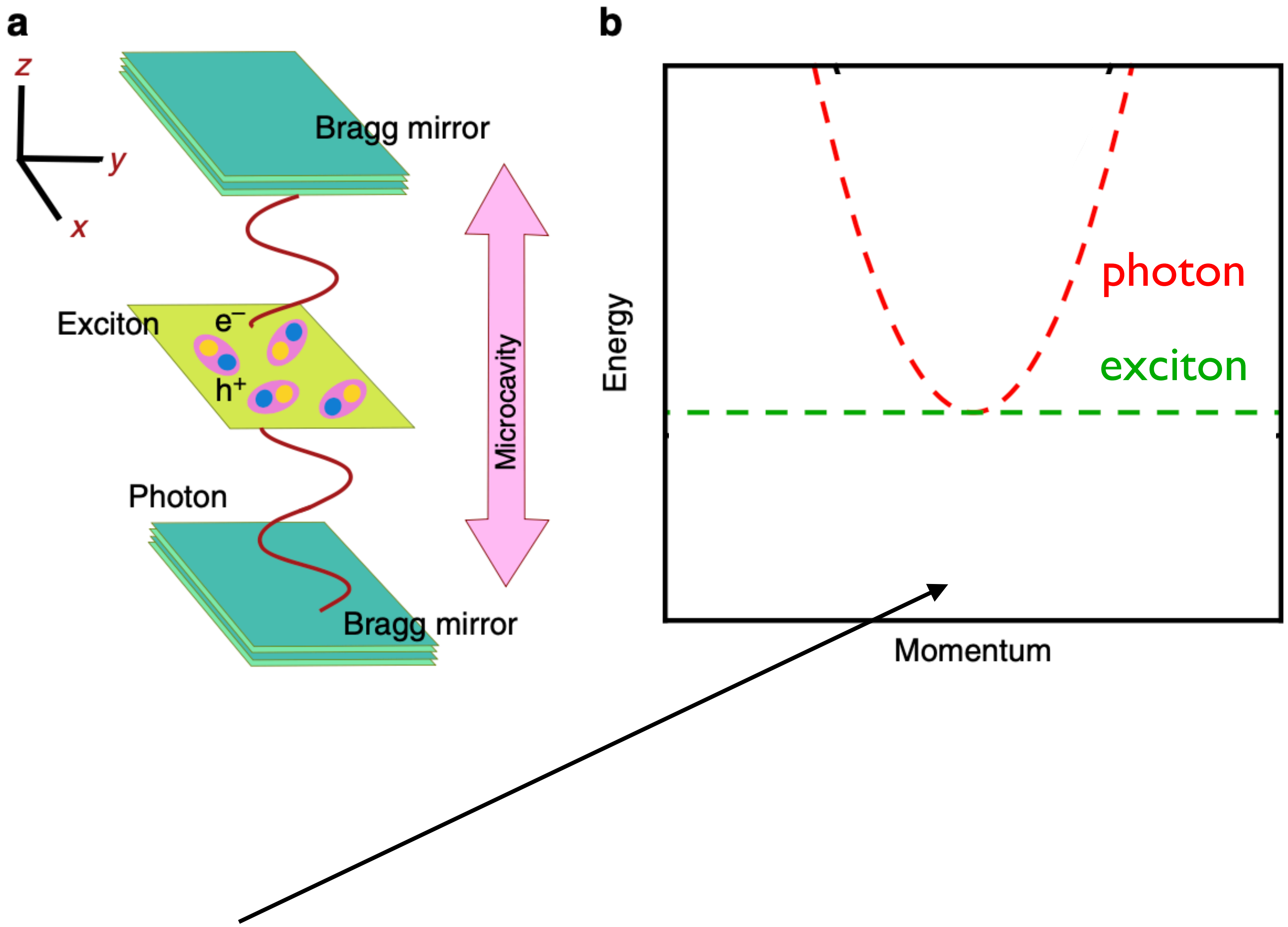


excitons — bound e-h pairs — at $E_X \lesssim E_{\text{gap}}$

Exciton-Polaritons

Two-dimensional semiconductor + optical microcavity

[figure: R.T. Juggins, J. Keeling & M.H. Szymańska, Nat. Commun. **9**, 4062 (2018)]



⇒ interacting polaritons (part photon, part exciton)

Interacting Polaritons

- Collisional thermalisation

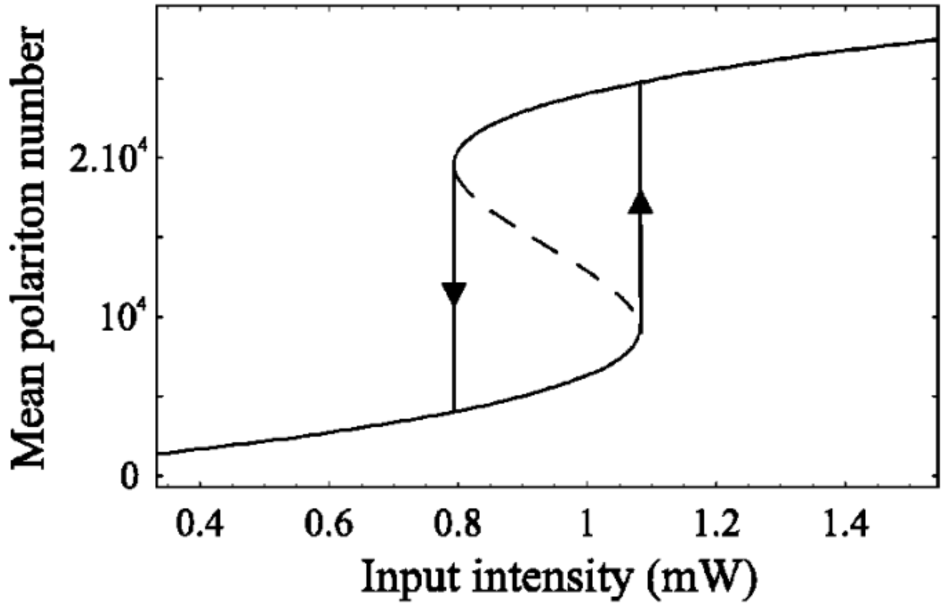
⇒ Bose-Einstein condensation, superfluidity, quantum vortices…

[I. Carusotto, RMP **85**, 299 (2013)]

- Non-equilibirium: optical bistability

$$E_{\mathrm{pol}}^{\mathrm{eff}} = E_{\mathrm{pol}} + \alpha \langle n_{\mathrm{pol}} \rangle$$

[A. Baas, J. Ph. Kaar, H. Eleuch, E. Giacobino, PRA **69**, 023809 (2004)]



Typically weakly interacting: Gross-Pitaevskii theory

- Strong interactions and correlations…

0D [T. Fink, A. Schade, S. Höfling, C. Schneider & A. Imamoglu, Nat. Phys. **14**, 365 (2018)]

1D [D.E. Chang, V. Gritsev, G. Morigi, V. Vuletić, M.D. Lukin & E.A. Demler, Nat. Phys. **4**, 884 (2008);
O. Firstenberg, T. Peyronel, Q.-Y. Liang, A.V. Gorshkov, M.D. Lukin & V. Vuletić, Nature **502**, 71 (2013)]

quantum chemistry [D. Sidler, M. Ruggenthaler, C. Schäfer, E. Ronca, A. Rubio, arXiv:2108.12244]

Here: strong interactions in Moiré materials

Outline

- Exciton-polaritons in Moiré materials
- 1) Coherent optical drive: Bistabilities with strong correlations
- 2) Incoherent pumping: multi-photon resonances
- Summary & Outlook

Moiré Materials

- Large-scale periodic structures of mis-aligned atomically-thin sheets

- High tunability: materials, twist, gating, …

- Diverse electronic states

- Superconductors, correlated & topological insulators, ferroelectrics, orbital magnetism, …

- Rich *bosonic* physics, with excitons and polaritons

[Photo credit: Benjamin Remez]

- "Dark" exciton condensates are "leaky" [B. Remez & NRC, arXiv:2110.07628]

⇒ Strongly interacting polaritons [A. Camacho-Guardian & NRC, arXiv:2108.06177]

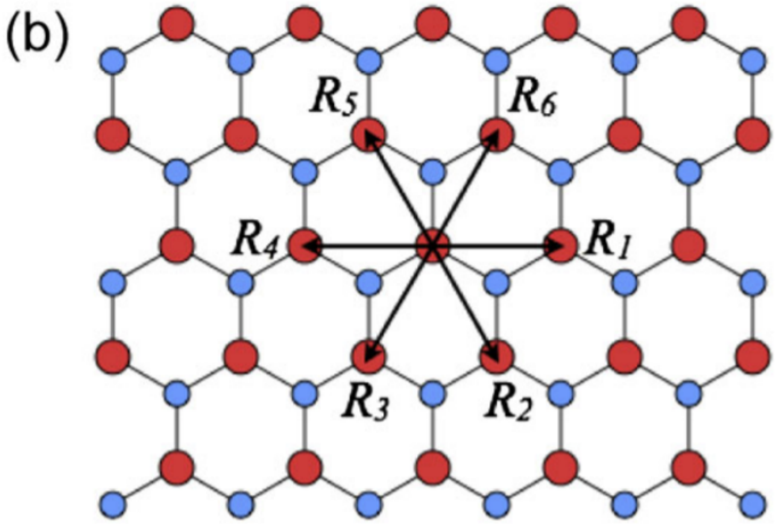
Transition metal dichalcogenide (TMD) materials

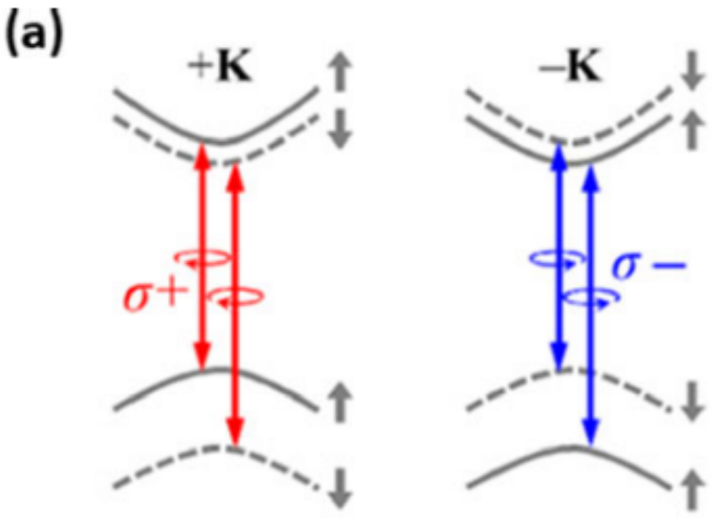
- 2D sheets of “MX_2”: MoS_2, WSe_2, $TiTe_2$,…

- Honeycomb lattice, broken inversion symmetry
  - Gap minima at K and K’ points
  - Strong SOC → spin-valley locking

- Here: one valley (e.g. circularly polarized pump)

[Xiao et al, PRL **108**, 196802 (2012)]



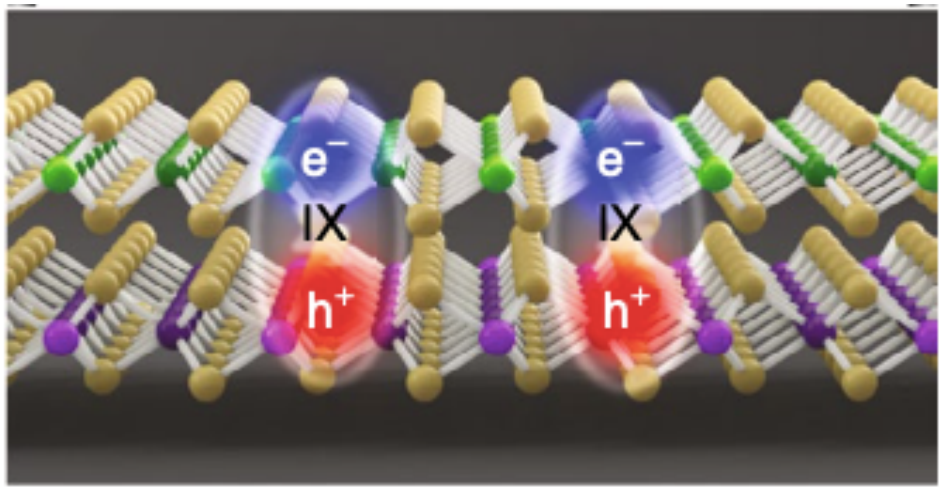


[Yu, et al, 2D Mater. **5**, 035021 (2018)]

TMD Moiré excitons

Bilayer

- Excitons can have hybrid character:
  - Inter-layer excitons: Strong interactions
  - Intra-layer excitons: Strong light-matter coupling

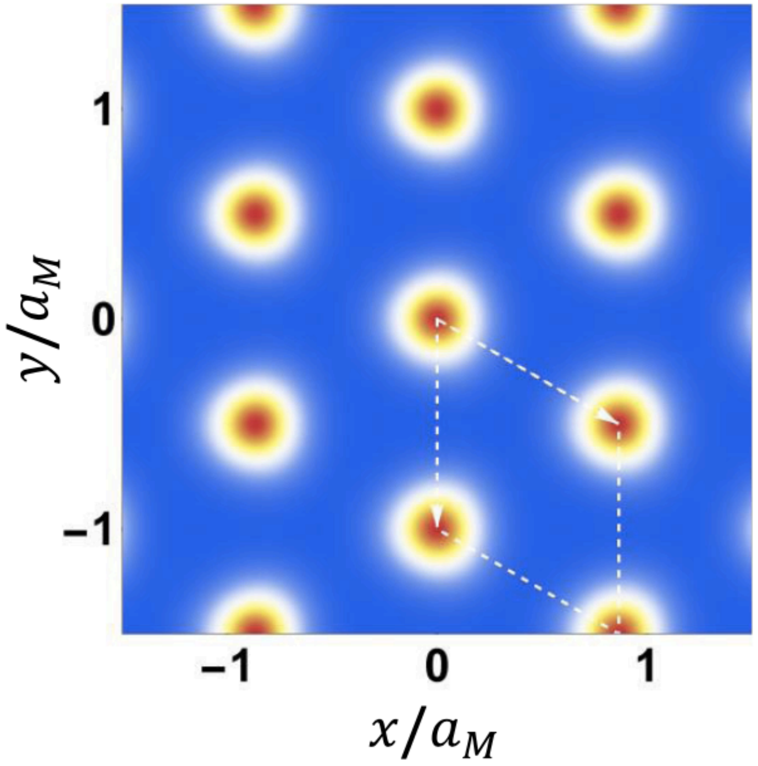


[Pasqual Rivera, Hongyi Yu, Kyle L. Seyler, Nathan P. Wilson, Wang Yao and Xiaodong Xu, Nat. Nanotech. **13**, 1004 (2018)]

Twisted bilayer: Moiré potential

- Spatially modulated band gap
- Excitons strongly localised to sites of a triangular lattice

⇒ very strong local interactions



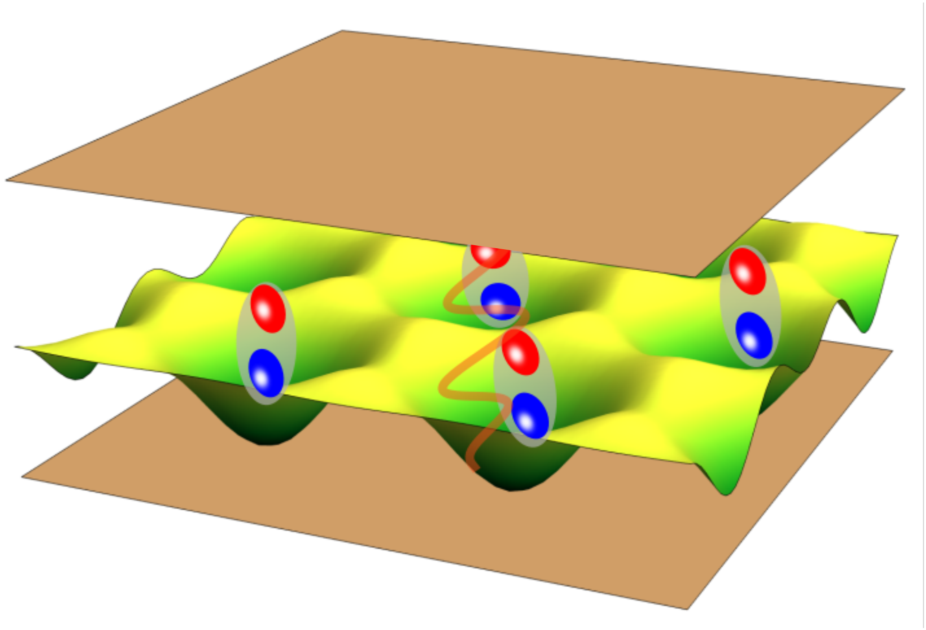
[Wu, Lovorn, MacDonald, PRB (2018)]

System: Moiré excitons + Cavity photons

[Arturo Camacho-Guardian & NRC, arXiv:2108.06177]

$$\hat{H}_X = \sum_{i=1}^{N_s} \omega_X \hat{x}_i^\dagger \hat{x}_i + \frac{U_X}{2}\sum_{i=1}^{N_s} \hat{x}_i^\dagger \hat{x}_i^\dagger \hat{x}_i \hat{x}_i - \cancel{\sum_{\langle i,j\rangle} t_{ij} \hat{x}_i^\dagger \hat{x}_j}$$

Strongly interacting excitons



$$\hat{H}_c = \sum_{\mathbf{k}} \omega_{\mathbf{k}} \hat{a}_{\mathbf{k}}^\dagger \hat{a}_{\mathbf{k}}$$

Cavity

$$\hat{H}_{l-m} = \Omega \sum_{\mathbf{k}}^{N_s} \hat{a}_{\mathbf{k}} \hat{x}_{\mathbf{k}}^\dagger + \hat{a}_{\mathbf{k}}^\dagger \hat{x}_{\mathbf{k}}$$

Light-matter coupling

Master equation

$$\frac{d\hat{\rho}}{dt} = -\mathrm{i}[\hat{H}, \hat{\rho}] + \mathscr{D}_{\mathrm{loss}}[\hat{\rho}]$$

$$\mathscr{D}_{\mathrm{loss}}[\hat{\rho}] = \frac{\gamma_c}{2}\left(2\hat{a}^\dagger \rho \hat{a}^\dagger - \{\hat{a}\hat{a}^\dagger, \hat{\rho}\}\right) + \frac{\gamma_x}{2}\sum_{i=1}^{N_s} 2\hat{x}_i \rho \hat{x}_i^\dagger - \{\hat{x}_i \hat{x}_i^\dagger, \hat{\rho}\}$$

Lossy photons and excitons

Energy scales:

[MoSe$_2$-WS$_2$: Long Zhang *et al.* (Michigan), Nature **591**, 61 (2021)]

$\Omega \sim 15\,\mathrm{meV}$ ⇒ strong coupling

$\gamma_c \sim 3\,\mathrm{meV}$ $\gamma_x \sim 8\,\mathrm{meV}$

$U_X \sim 30 - 40\,\mathrm{meV}$ ⇒ exciton blockade

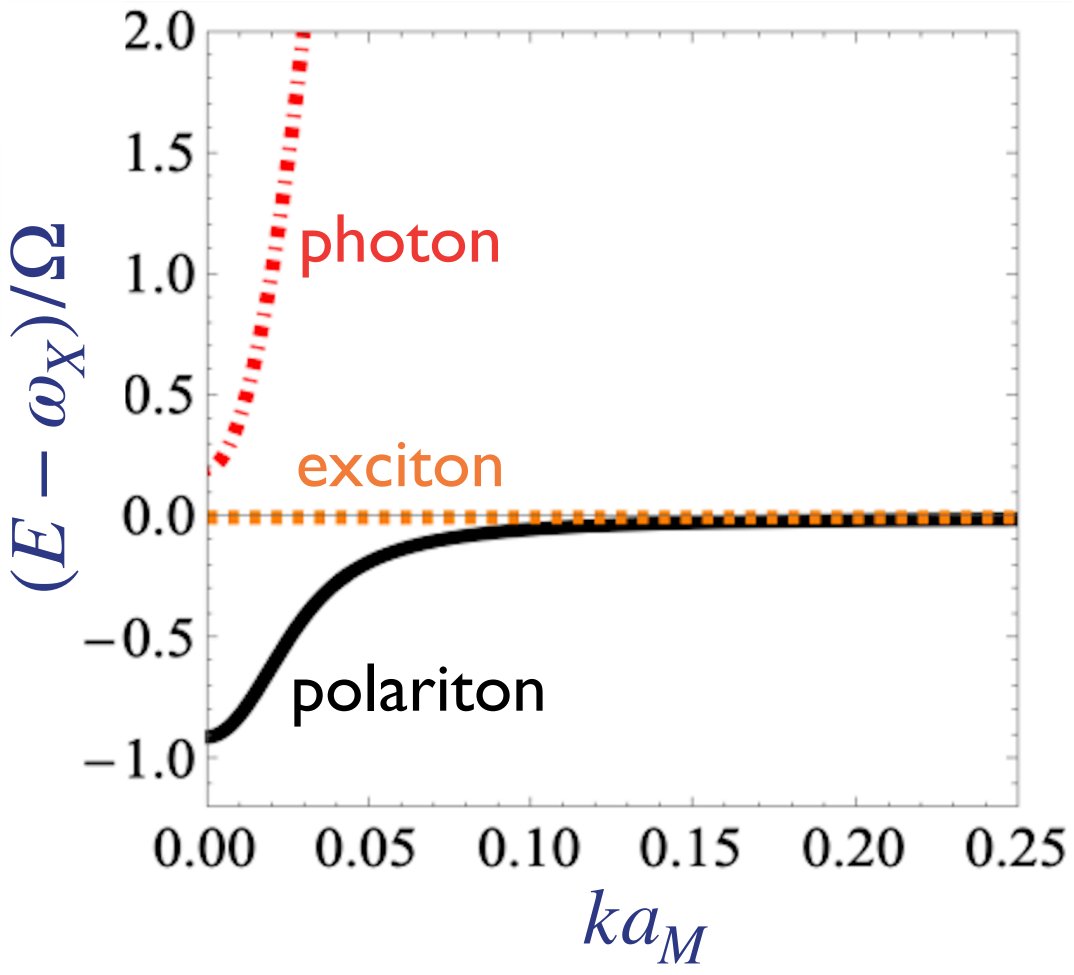
Theoretical Model & Approximations

Cavity mode $\omega_k \simeq \omega_c + \frac{1}{2}\frac{c^2k^2}{\omega_c}$

$$\hat{H}_{l-m} = \Omega \sum_{\mathbf{k}}^{N_s} \hat{a}_{\mathbf{k}}\hat{x}_{\mathbf{k}}^\dagger + \hat{a}_{\mathbf{k}}^\dagger \hat{x}_{\mathbf{k}}$$

photon decouples for $k^2 \gtrsim 2\omega_c\Omega/c^2$

Typically $\Delta k a_M \sim 0.02 - 0.1 \ll 1$



long-range coupling of excitons $(\lambda/a_M)^2 \sim (60-300)^2 \gg 1$

⇒ single cavity mode [photon acts as a mean field]

$$\hat{H}_{l-m} \to \frac{\Omega}{\sqrt{N_s}} \sum_{i=1}^{N_s} (\hat{a}\hat{x}_i^\dagger + \hat{a}^\dagger \hat{x}_i)$$

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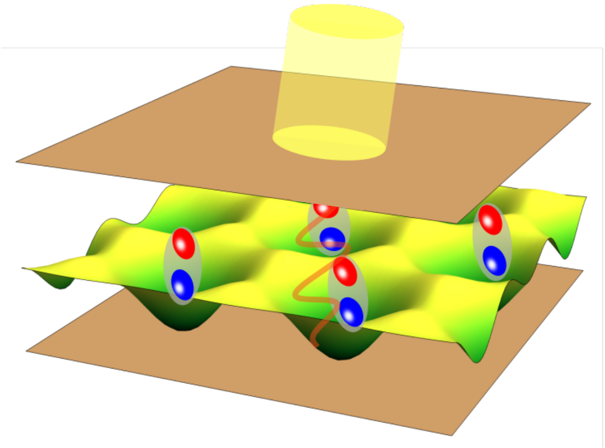
1) Coherent optical drive

$$\hat{H}_{\text{drive}} = F\hat{a}^{\dagger}\mathrm{e}^{-\mathrm{i}\omega_p t} + F^{*}\hat{a}\mathrm{e}^{\mathrm{i}\omega_p t}$$



- classical light field $\hat{a} \to \langle\hat{a}\rangle = \sqrt{N_s}\alpha$ (large photon occupation)

$$\alpha = \frac{1}{\Delta_c}(f + \Omega\langle\hat{x}\rangle) \qquad \Delta_c = \omega_p - \omega_c + \mathrm{i}\gamma_c/2 \qquad f = F/\sqrt{N_s}$$

- dynamics of (lossy) excitons from *local* self-consistent theory

$$\hat{H}_{\text{local}} = \sum_{i=1}^{N_s} \omega_X \hat{x}_i^{\dagger}\hat{x}_i + \frac{U_X}{2}\sum_{i=1}^{N_s} \hat{x}_i^{\dagger}\hat{x}_i^{\dagger}\hat{x}_i\hat{x}_i + \sum_{i=1}^{N_s}(f_X\hat{x}_i^{\dagger} + f_X^{*}\hat{x}_i)$$

$$f_X = \frac{\Omega}{\Delta_c}f + \left(\frac{\Omega^2}{\Delta_c}\right)\langle\hat{x}\rangle \qquad \mathscr{D}_{\text{loss}}[\hat{\rho}] = \frac{\gamma_x}{2}\sum_{i=1}^{N_s} 2\hat{x}_i\rho\hat{x}_i^{\dagger} - \{\hat{x}_i\hat{x}_i^{\dagger}, \hat{\rho}\}$$

1) Coherent optical drive: Methods

- semiclassical (Gross-Pitaevskii) approximation

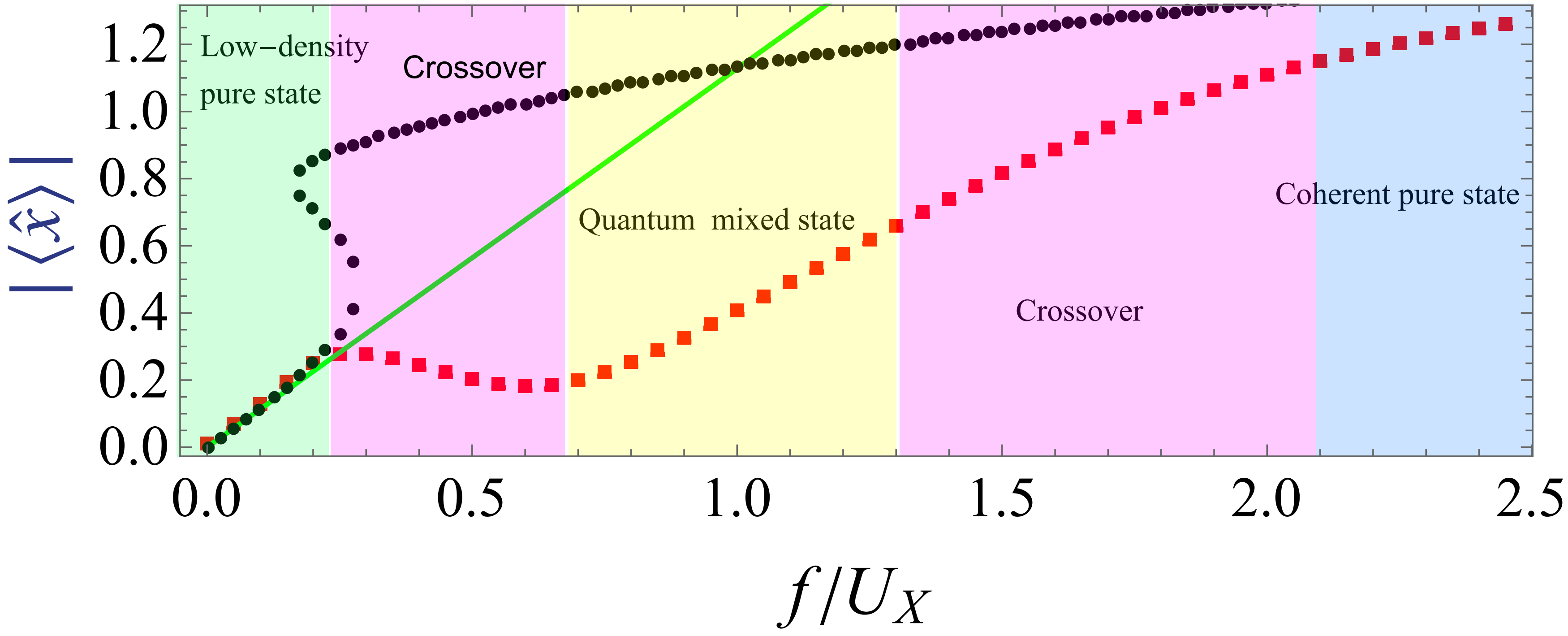
$$\frac{U_X}{2}\sum_i \hat{x}_i^\dagger \hat{x}_i^\dagger \hat{x}_i \hat{x}_i \rightarrow \frac{U_X}{2}\sum_i \left(\langle \hat{x}_i^\dagger \rangle \langle \hat{x}_i^\dagger \rangle \hat{x}_i \hat{x}_i + \hat{x}_i^\dagger \langle \hat{x}_i^\dagger \rangle \langle \hat{x}_i \rangle \hat{x}_i + \ldots \right)$$

⇒ classical nonlinear optical response (bi-stability etc.)

[A. Baas, J. Ph. Kaar, H. Eleuch, E. Giacobino, PRA **69**, 023809 (2004)]

- *exact* quantum dynamics shows strong deviations

[Arturo Camacho-Guardian & NRC, arXiv:2108.06177]

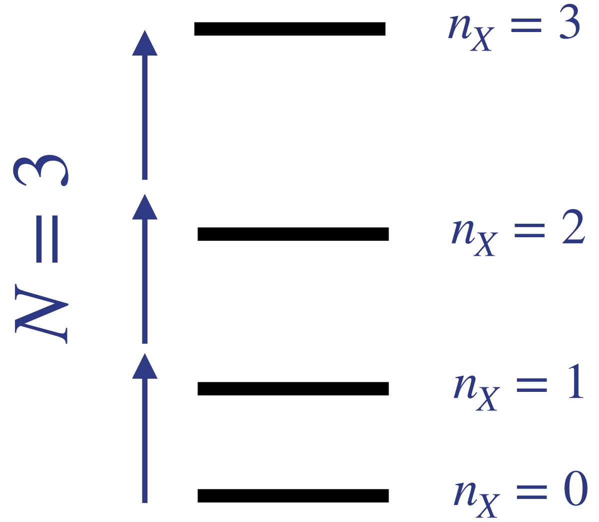


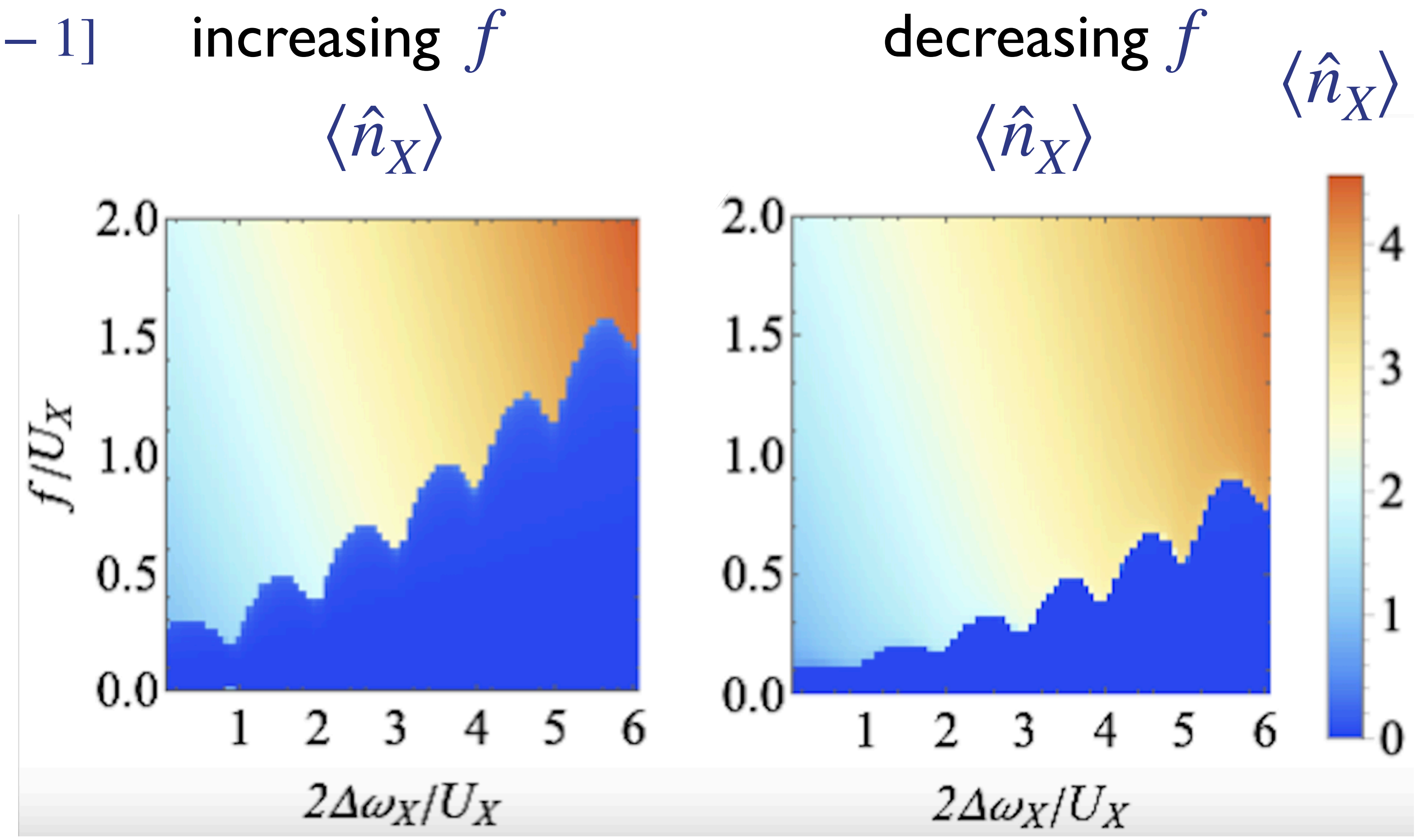
$$(\omega_p - \omega_X)/U_X = 1.2$$

$$(\omega_p - \omega_c)/U_X = 1$$

1) Coherent optical drive: Exact results

$[(\omega_p - \omega_c)/U_X = -1]$



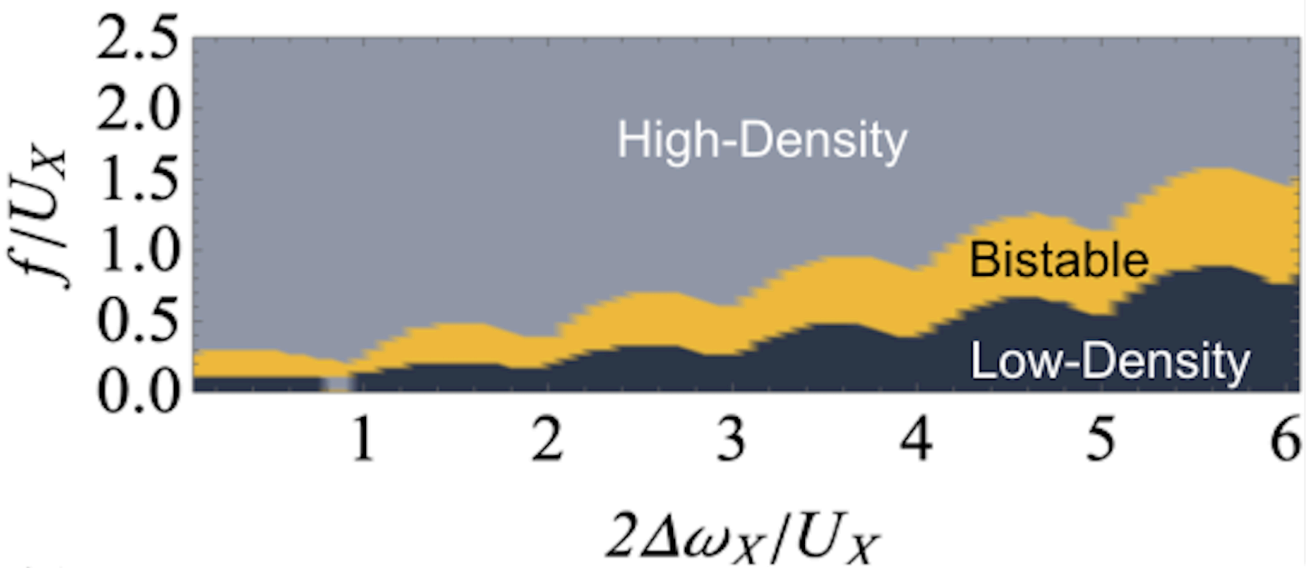

increasing f

decreasing f

N-photon resonance condition

$$N\omega_p = N\omega_X + \frac{U_X}{2}N(N-1)$$

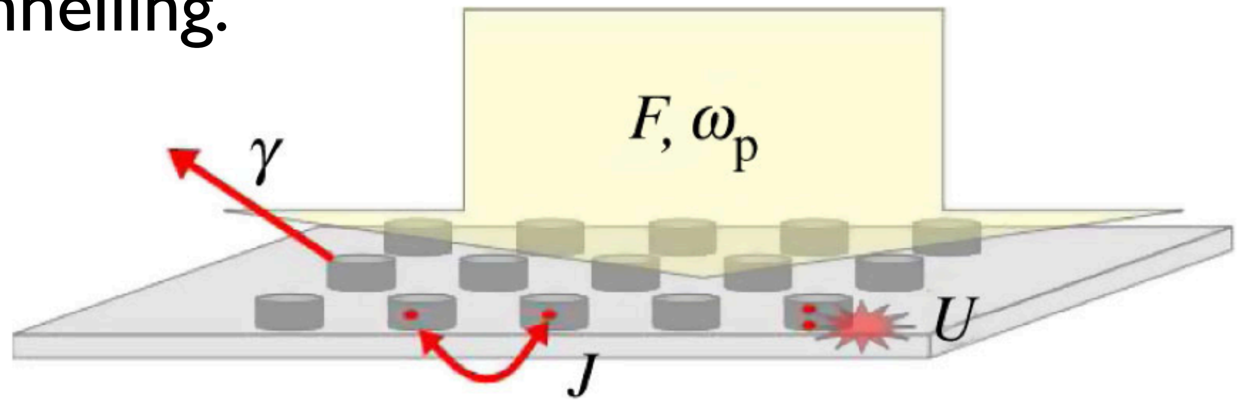
$$\Delta\omega_X = \omega_p - \omega_X = \frac{U_X}{2}(N-1)$$


Hysteresis

Signatures of beyond-GP physics in the bistability thresholds

1) Coherent optical drive: Relation to Cavity Arrays

Arrays of non-linear cavities are predicated to show similar bistability patterns, promoted by inter-cavity tunnelling.



[A. Le Boité, G. Orso & C. Ciuti, PRL **110**, 233601 (2013);
M. Biondi, G. Blatter, H. E. Türeci, and S. Schmidt, PRA **96**, 043809 (2017)]

Here: array of cavities → lattice of localized excitons

Additional "cavity photon" plays important roles:

- inter-cavity (exciton) tunnelling set by cavity photon field $J \sim \dfrac{\Omega^2}{\omega_X - \omega_c}$
- parameter $\Delta k a_M \ll 1$ justifies collective mean-field treatment

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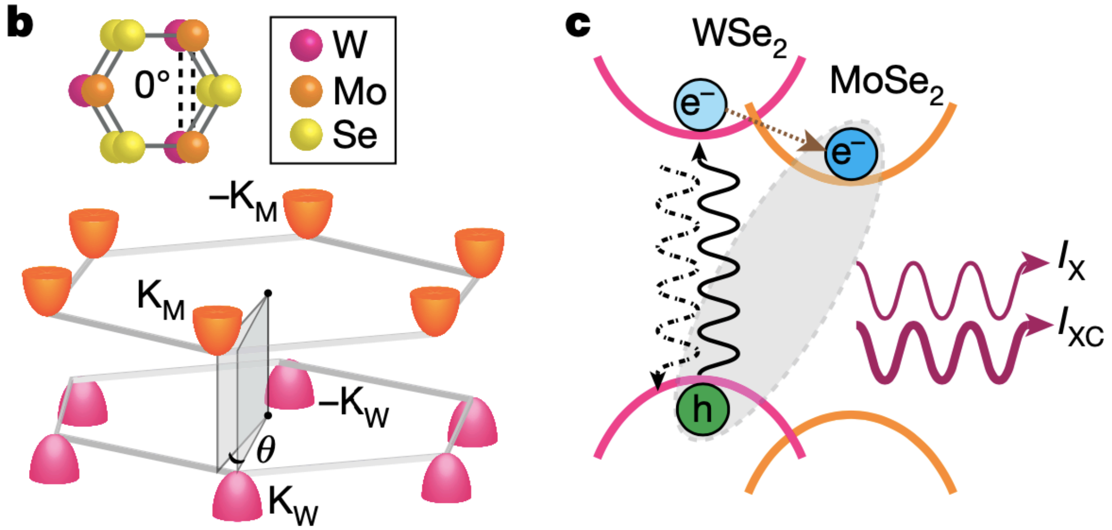
2) Incoherent pumping of excitons

Lasing of interlayer excitons in WSe₂-MoSe₂ bilayer

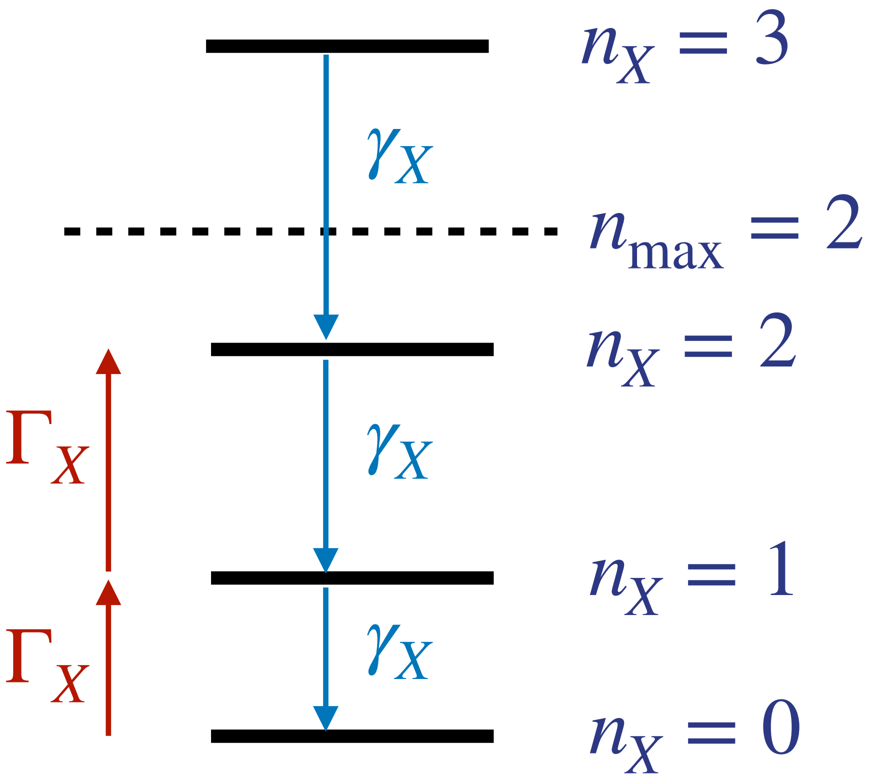


[Eunice Y. Palik, Long Zhang, G. William Burg, Rahul Gogna, Emanuel Tutuc & Hui Deng, Nature **576**, 80 (2019)]

$$\frac{d\hat{\rho}}{dt} = -\mathrm{i}[\hat{H}, \hat{\rho}] + \mathscr{D}_{\text{loss}}[\hat{\rho}] + \mathscr{D}_{\text{gain}}[\hat{\rho}]$$

[coherent drive $F = 0$]

$$\mathscr{D}_{\text{gain}}[\hat{\rho}] = \frac{\Gamma_x}{2} \sum_{i=1}^{N} 2\hat{y}_i^\dagger \rho \hat{y}_i - \{\hat{y}_i^\dagger \hat{y}_i, \hat{\rho}\}$$



$\hat{y}_i^\dagger = \hat{x}_i^\dagger \hat{P}_i$ where $\hat{P}_i$ projects onto states in the range $\hat{n}_i = 0,1,2\ldots n_{\text{max}} - 1$

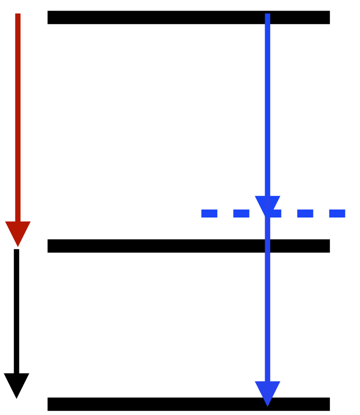
[Arturo Camacho-Guardian & NRC, arXiv:2108.06177]

2) Incoherent pumping of excitons

e.g. $n_{\mathrm{max}} = 2$

Single-photon transitions

Multi-photon transition



$n_X = 2$

$n_X = 1$

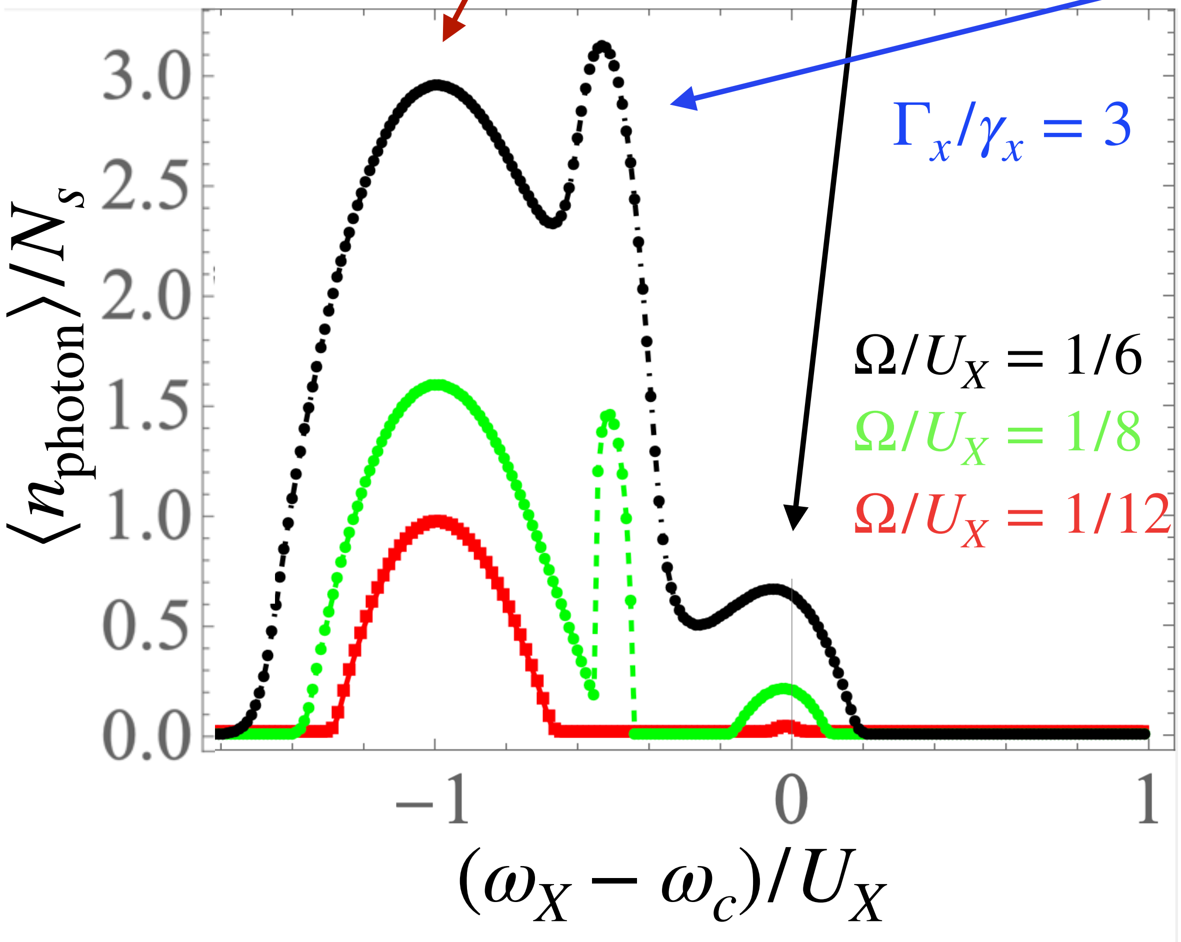
$n_X = 0$

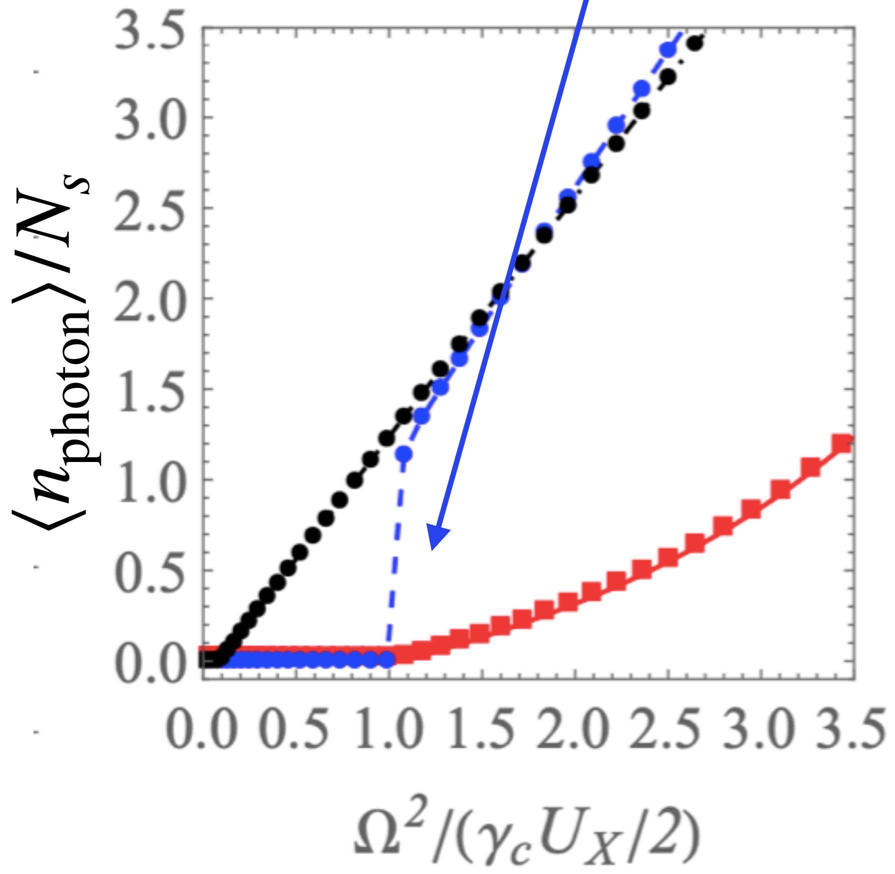
$\omega_c \simeq \omega_X + U_X$

$\omega_c \simeq \omega_X$

$2\omega_c \simeq 2\omega_X + U_X$

discontinuous





2) Incoherent pumping of excitons

cf. two-photon micromaser (Rydberg atom)

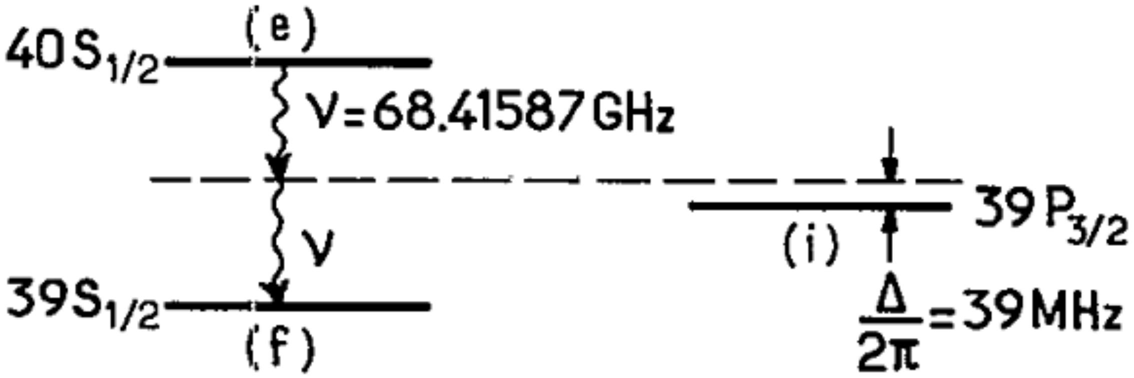


FIG. 1. Level scheme relevant to the Rb two-photon maser.

[M. Brune, J. M. Raimond, P. Goy, L. Davidovich, and S. Haroche, Phys. Rev. Lett. **59**, 1899 (1987)]

Coherent two-photon emission for $\Delta\ (\simeq U_X/2)\ \gtrsim \gamma_x$

Strong interactions permit two-photon cavity resonance

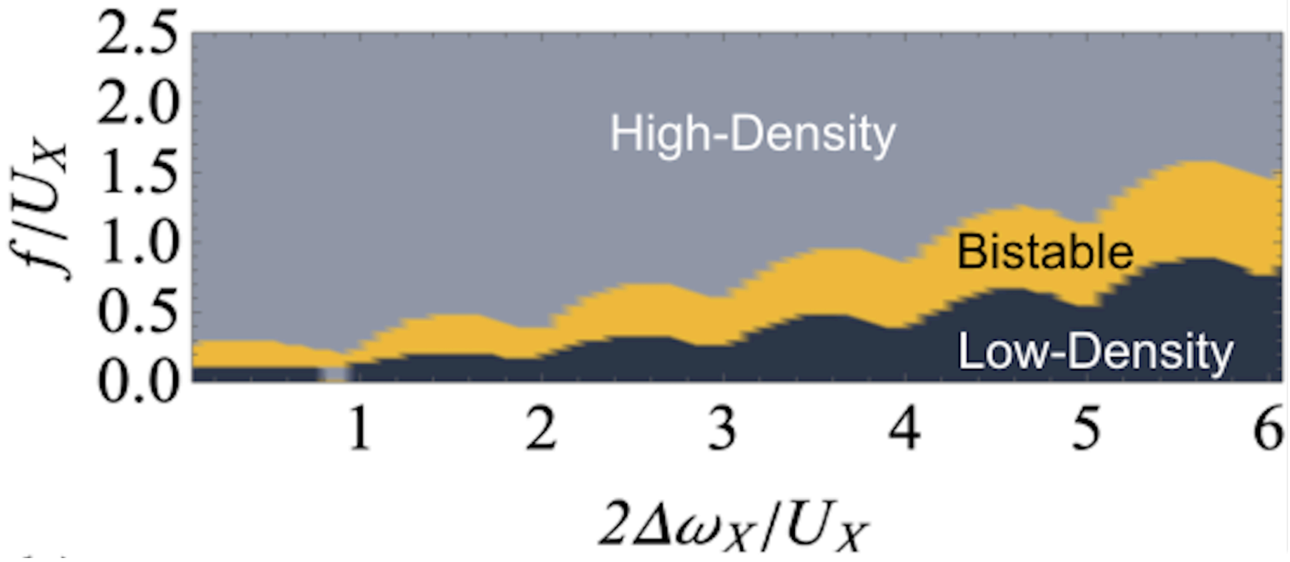
Summary

Polaritons in Moiré materials access regimes of very strong interactions.

Show qualitative features beyond Gross-Pitaevskii:

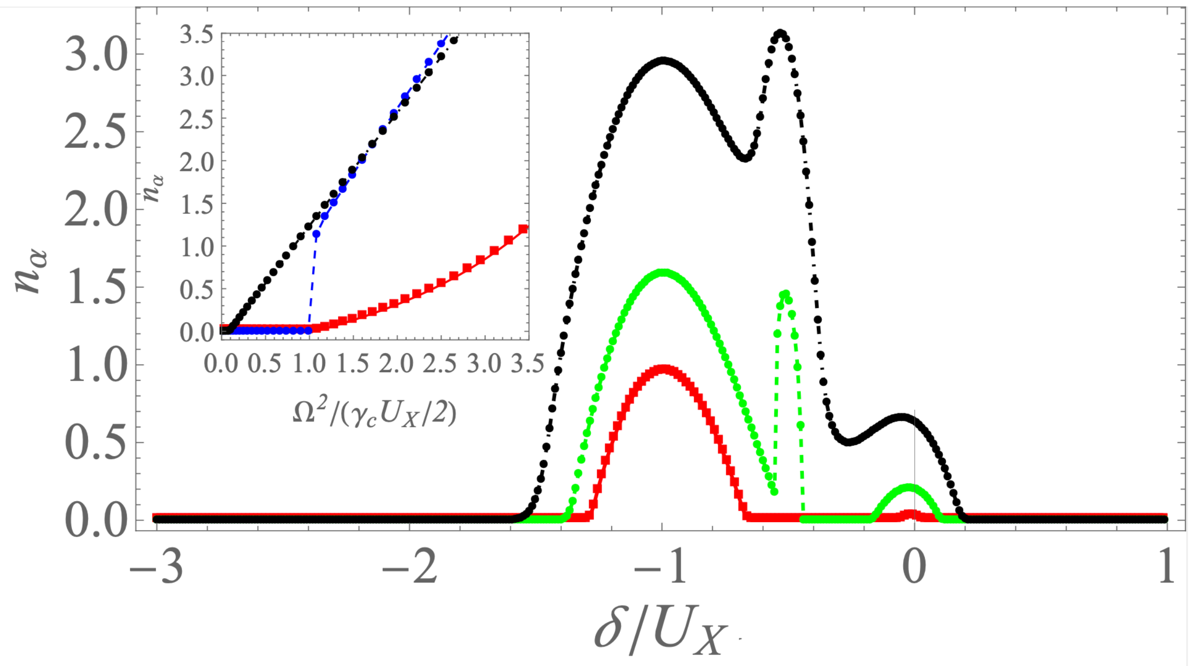
- Bi-stabilities controlled by N-exciton resonances

  Beyond-GP physics evident in the bistability thresholds



- Lasing on multi-photon transition

  Strong interactions permit coherent two-photon cavity resonance

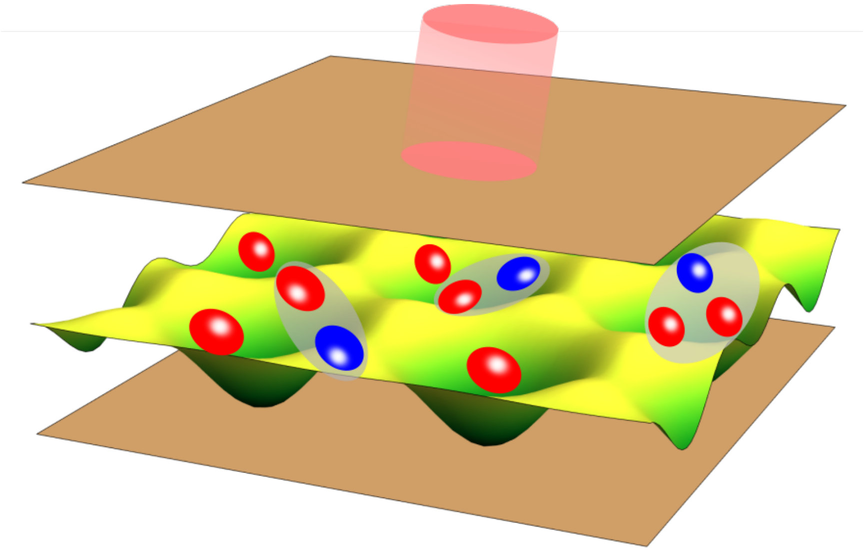


Outlook

- Two-component systems (unpolarised pump)
- Fluctuations of cavity mode (non-zero $\Delta k a_M$)

[T. Fink, A. Schade, S. Höfling, C. Schneider & A. Imamoglu, Nat. Phys. **14**, 365 (2018)]

- Electron doping: correlated phases & trions



- Strong coupling limit $\Omega \gtrsim E_X$ for reduced gaps

[Y. Ashida, A. Imamoglu & E. Demler, PRL **126**, 153603 (2021)]

$\Rightarrow$ Prospects for nonlinear optical devices at low photon density

$\Rightarrow$ Towards electronic control of entangled states of light