



# Exciton-phonon coupling from first principles

Claudio Attaccalite



CNRS concours 03/01 (2021)



# Short CV



**2001: Master in Physics,** La Sapienza University (Rome)



**2005: PhD in Physics,** SISSA, Trieste (Italie) *S. Sorella*



**2006-2007: PostDoc,** IEMN, Lille, *L. Wirtz*

**2008-2009: PostDoc,** Universidad del País Vasco, *A. Rubio*



**2009-2014: CNRS Researcher (CR2),** Institute Néel, France

**2014-2018: CNRS Researcher (CR1),** CINaM, France

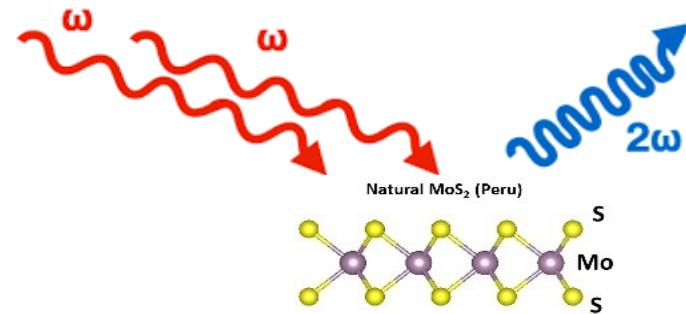
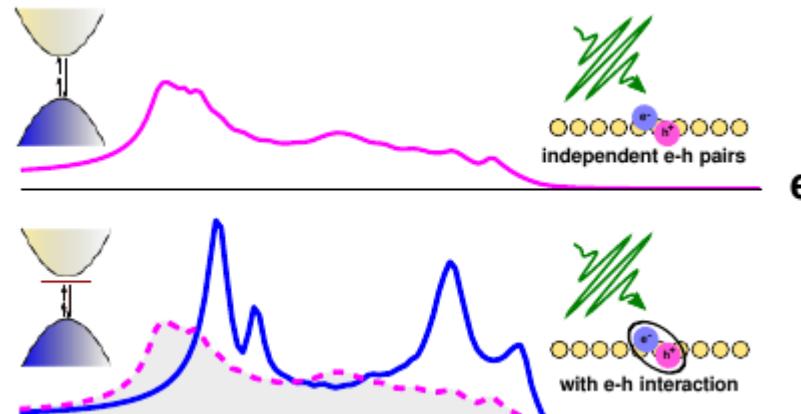
**2018-2019: Visiting Researcher,** Tor-Vergata Univ. (Rome)



# Research activity

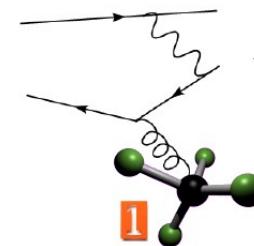
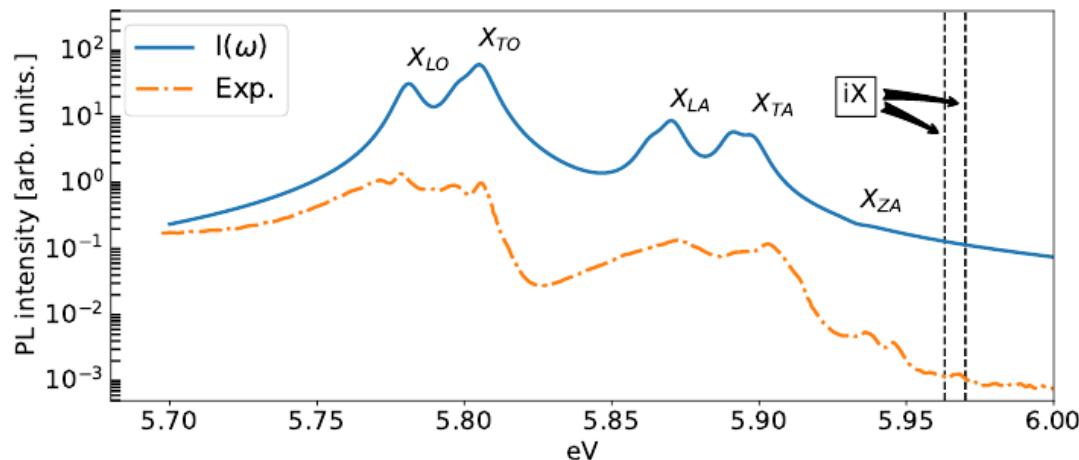


## Linear and non-linear optical response



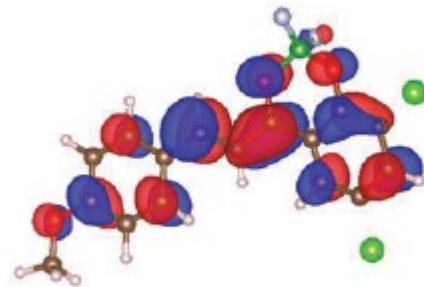
C. Attaccalite, E. Cannuccia, M. Grüning, PRB **95**, 125403 (2017)  
M. Grüning, C. Attaccalite, PRB-Rapid **88**, 081102 (2014)

## Excited states and electron-phonon coupling



C. Attaccalite et al., Nanoletters, **10**, 1172(2010)  
E. Cannuccia, B. Monserrat, C. Attaccalite, PRB B **99**, 125403 (2019)

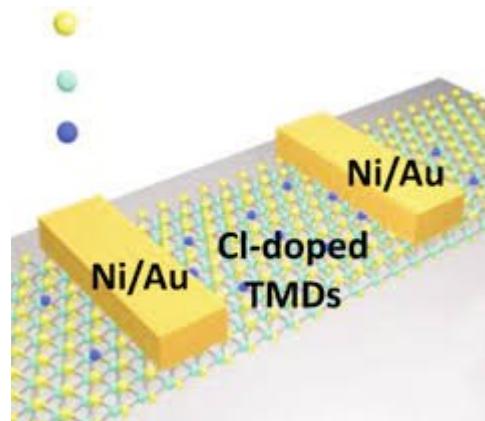
# Internal collaborations



## New light emitters in the infrared

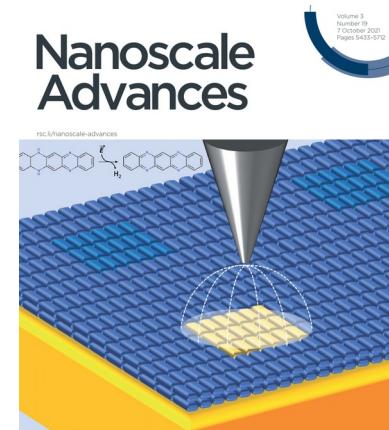
Mater. Chem. Font. 2019, 3, 16

A. D'Aleo, F. Fages



Opto-electronics with 2D  
materials  
R. Parret (2021-)

## Nanoscale Advances



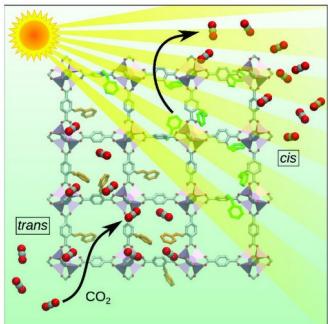
## Chemical reactions with a STM

Nanoscale Adv. 2021, 3, 5565

T. Leoni and C. Becker

PhD thesis founded by  
Aix-Marseille-Univ.

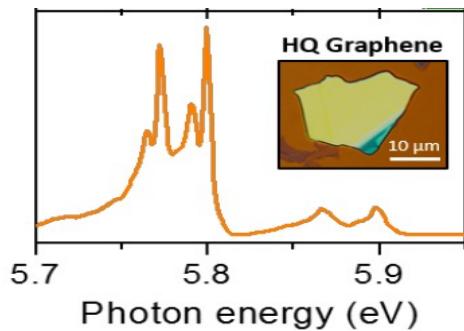
# External collaborations



**Photoactive MOF  
for Carbon Capture**  
**R.Poloni (2019–2021)**



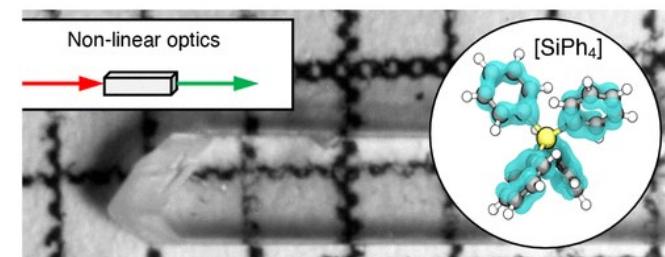
**Materials at high pressure**  
**L.Artus (2020–2021)**



**EELS and luminescence**  
**A.Loiseau, J.Barjon (2017–)**



**Graphene  
Flagship**



**New non-linear crystals**  
**S.Sanna, S.Chatterjee (2020–)**

Invited Prof. at  UNIVERSITÄT  
PADERBORN

# Scientific implication

## Scientific Production →

57 papers (6 PRL, 1 NanoLetter), 1 patent,  
more than **4000** citations, h-index **31**  
**16** invited talks



## Supervision → 3 PhD, 2 postdocs, 3 master, 2 internship



## Teaching →

Statistical Mechanics (18/19), Density Func. Theory(17/22)  
**9** International Schools(2008-)



## Scientific research projects:

Region Rhone-Alpes  
Graphene-Flagship  
Emergence CNRS  
AMU PhD program  
COST actions  
HPC-Europe  
ETSF interships  
PRACE



## Conference Organization:

3 Symposium  
8 Conferences/Workshop  
2 Schools  
2 Year Weekly seminars at CINaM



# Service to the community

Editor fellow of **SciPost**



Membre du bureau du **GDR REST**

Membre du bureau du **COST ACTION TUMIEE**

Membre du Comité scientifique du **GDR HOWDI et GDR-IRN HOWDI**

Membre du bureau de la Division Matière Condensée de la **SFP**

Correspondant science ouverte du **CINaM**



## Code development

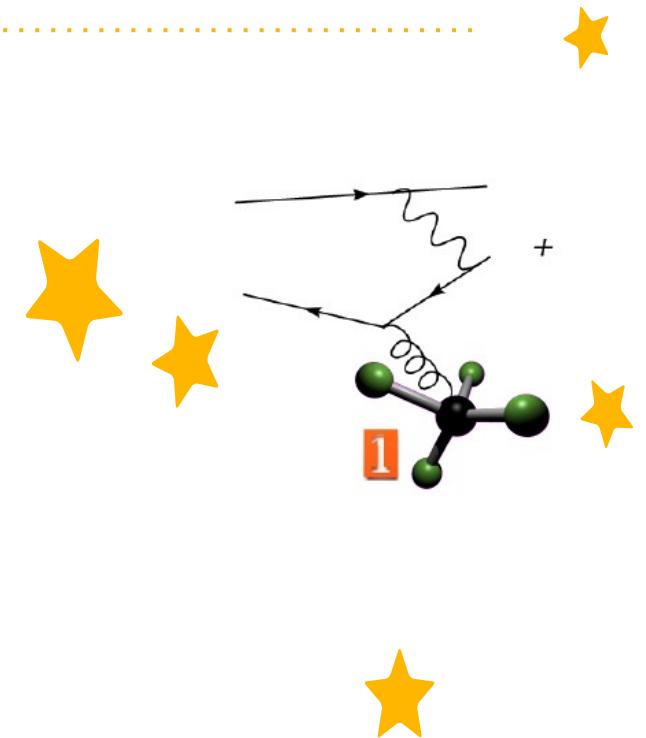


D. Sangalli, et al. J. Phys. Cond. Matt. 31, 325902(2020)  
K. Nakano, C. Attaccalite et al. J. Chem. Phys. 152,  
204121 (2020)

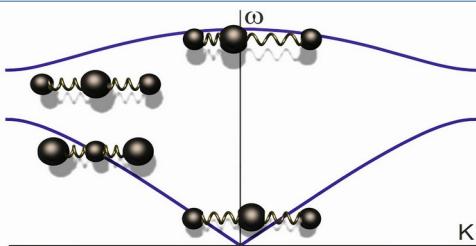


**Yambo school**  
2021/2022  
100 participants

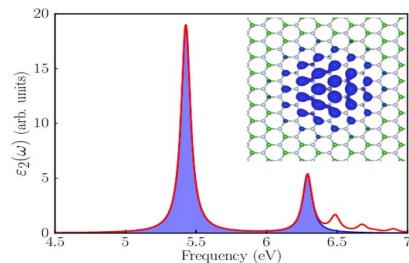
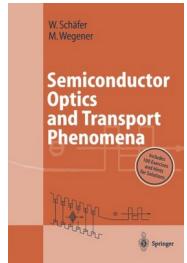
# Exciton-phonon coupling from first principles



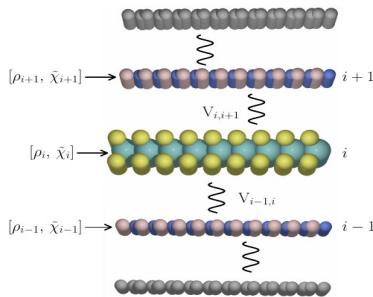
# Exciton phonon coupling



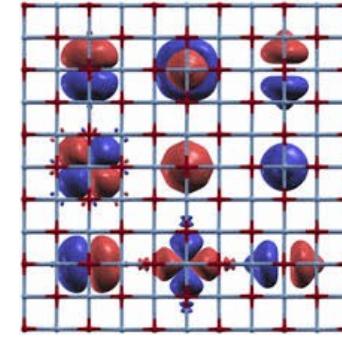
Phonons  
Density-Functional  
Perturbation-Theory



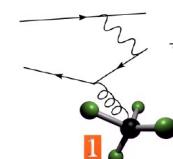
Electronic excitations  
Many-body theory



Environment effects  
Modeling



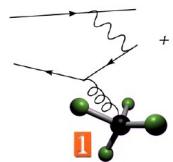
Wannier interpolation



Exciton-phonon Hamiltonian

$$\mathcal{H}_{\text{el-ph}} = \sum_{kq\mu} M_{kq}^\mu (v_{k+q}^\dagger v_k - u_{k+q}^\dagger u_k)(a_{q\mu} + a_{-q\mu}^\dagger)$$

# Expected results

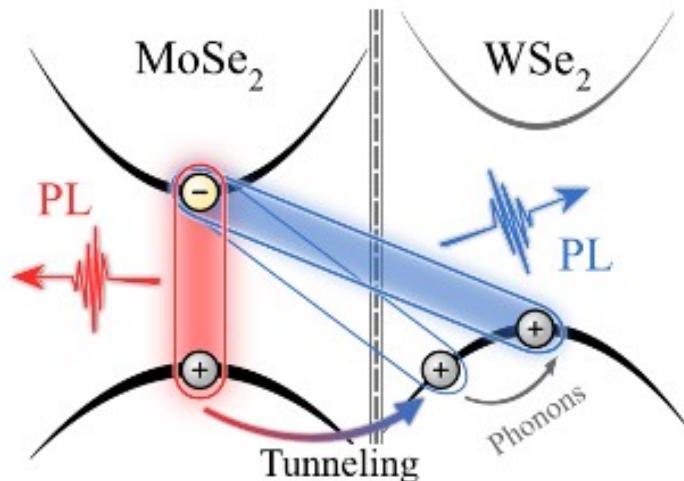


## Exciton-phonon Hamiltonian

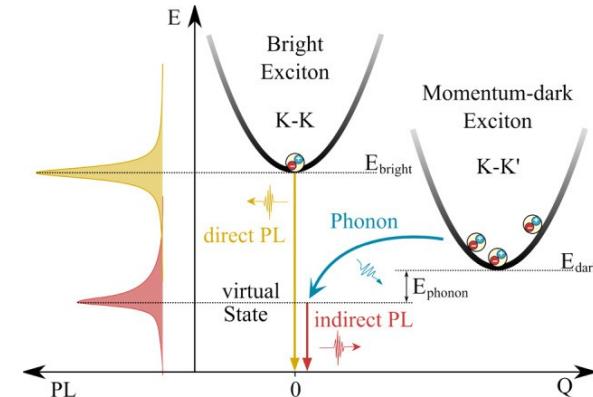
$$\mathcal{H}_{\text{el-ph}} = \sum_{kq\mu} M_{kq}^\mu (v_{k+q}^\dagger v_k - u_{k+q}^\dagger u_k) (a_{q\mu} + a_{-q\mu}^\dagger)$$



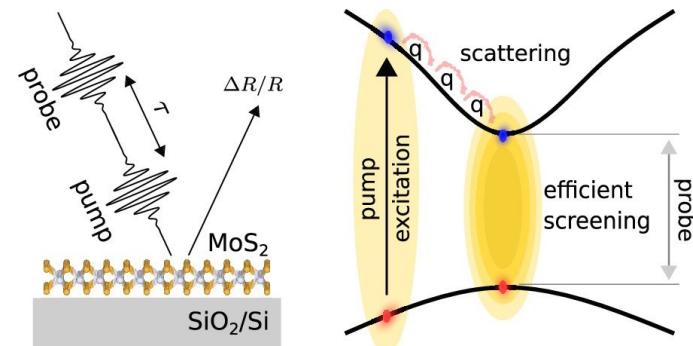
## Exciton dynamics



## Phonon-assisted luminescence



## Pump-probe



# CV

Non-linear-optics

Electron-phonon

Code development

# Project

Exciton-phonon

from first principles

Exciton-dynamics

Time-resolved

spectroscopy

# Thanks!

Any questions?





# Exciton phonons

Exciton-phonon matrix elements

$$D_{S,S'}^{\lambda\mathbf{q}} = \sum_{\mathbf{k},c,v} \left[ \sum_{c'} \left( A_{c'\mathbf{k}+\mathbf{q},v\mathbf{k}}^{S'(\mathbf{q})} \right)^* g_{c'\mathbf{k}+\mathbf{q},c\mathbf{k}}^{\lambda\mathbf{q}} A_{c\mathbf{k},v\mathbf{k}}^S - \sum_{v'} \left( A_{c\mathbf{k},v'\mathbf{k}-\mathbf{q}}^{S'(\mathbf{q})} \right)^* g_{v\mathbf{k},v'\mathbf{k}-\mathbf{q}}^{\lambda\mathbf{q}} A_{c\mathbf{k},v\mathbf{k}}^S \right]$$

Exciton-phonon Hamiltonian

$$H^{exc-ph} = \sum_{S,\mathbf{Q}} E_{\mathbf{Q}}^S a_{S\mathbf{Q}}^\dagger a_{S\mathbf{Q}} + \sum_{\lambda\mathbf{q}} \omega_{\lambda\mathbf{q}} b_{\lambda\mathbf{q}}^\dagger b_{\lambda\mathbf{q}} + \sum_{S,S',\mathbf{Q},\mathbf{q}} D_{S,S'}^{\lambda\mathbf{q}}(\mathbf{Q}) a_{S\mathbf{Q}+\mathbf{q}}^\dagger a_{S'\mathbf{Q}} (b_{\lambda\mathbf{q}} + b_{\lambda,-\mathbf{q}}^\dagger)$$

Equations of motion

$$\frac{d}{dt} n_{\mathbf{k}} = \frac{2}{\hbar} \text{Im}\{M_{\mathbf{k}} S_{\mathbf{k}}\}, \quad \text{Phonon occupation}$$

$$i\hbar \frac{d}{dt} S_{\mathbf{k}} = (E_0 - \omega_{\mathbf{k}}) S_{\mathbf{k}} - M_{\mathbf{k}}^* N_0 + \sum_{\mathbf{q},\zeta=\pm} D_{\mathbf{q}} \mathcal{U}_{\mathbf{kq}}^\zeta + \sum_{\mathbf{k}'} M_{\mathbf{k}'}^* n_{\mathbf{k}\mathbf{k}'} \quad \text{Polarization}$$

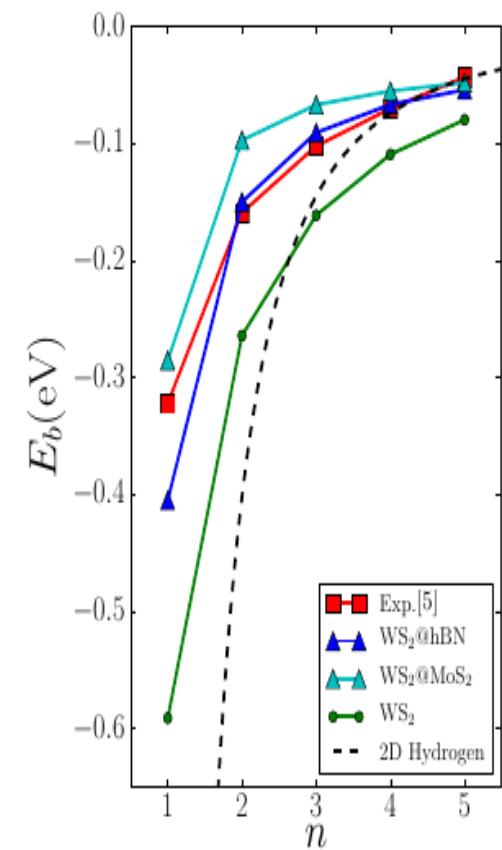
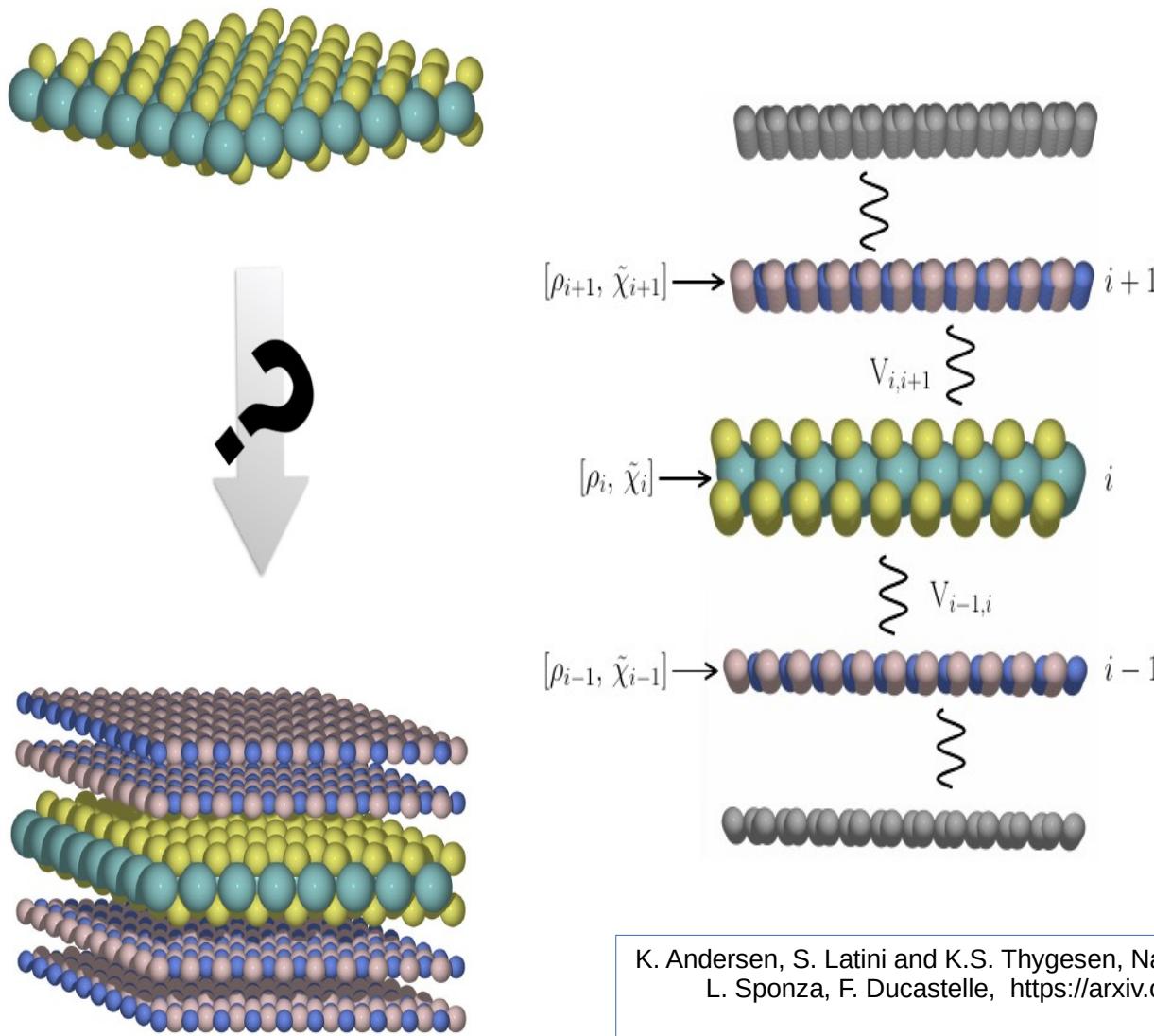
$$i\hbar \frac{d}{dt} \mathcal{U}_{\mathbf{kq}}^\zeta = (E_{-\mathbf{q}} - \zeta \Omega_{\mathbf{q}} - \omega_{\mathbf{k}}) \mathcal{U}_{\mathbf{kq}}^\zeta - M_{\mathbf{k}}^* \mathcal{C}_{\mathbf{0q}}^\zeta + D_{\mathbf{q}}^* \eta_{\mathbf{q}}^\zeta S_{\mathbf{k}} + \sum_{\mathbf{q}',\zeta'=\pm} D_{\mathbf{q}'} X_{\mathbf{kq}\mathbf{q}'}^{\zeta\zeta'}, \quad \text{PH-assist. pol.}$$

$$i\hbar \frac{d}{dt} \mathcal{C}_{\mathbf{Qq}}^\zeta = (E_{\mathbf{Q}-\mathbf{q}} - \zeta \Omega_{\mathbf{q}} - E_{\mathbf{Q}}) \mathcal{C}_{\mathbf{Qq}}^\zeta - D_{\mathbf{q}}^* Q_{\mathbf{Qq}}^\zeta \quad \text{Exciton-phonon}$$

$$- \sum_{\mathbf{k}} (M_{\mathbf{k}} \mathcal{U}_{\mathbf{kq}}^\zeta \delta_{\mathbf{Q},0} - M_{\mathbf{k}}^* \mathcal{U}_{\mathbf{k},-\mathbf{q}}^{-\zeta} \delta_{\mathbf{Q},-\mathbf{q}}) + \sum_{\mathbf{q}',\zeta'=\pm} (D_{\mathbf{q}'} Y_{\mathbf{Qq}\mathbf{q}'}^{\zeta\zeta'} - D_{\mathbf{q}'}^* Y_{\mathbf{Q}+\mathbf{q}',\mathbf{q},-\mathbf{q}'}^{\zeta\zeta'}),$$

$$\frac{d}{dt} N_{\mathbf{Q}} = -\frac{2}{\hbar} \sum_{\mathbf{k}} \text{Im}\{M_{\mathbf{k}} S_{\mathbf{k}}\} \delta_{\mathbf{Q},0} - \frac{i}{\hbar} \sum_{\mathbf{q},\zeta=\pm} D_{\mathbf{q}} (\mathcal{C}_{\mathbf{Qq}}^\zeta - \mathcal{C}_{\mathbf{Q}+\mathbf{q},-\mathbf{q}}^\zeta) \quad \text{Exciton-density}$$

# Quantum Electrostatic Heterostructure model and its relatives



K. Andersen, S. Latini and K.S. Thygesen, Nanoletter, **15**, 4616 (2015)  
L. Sponza, F. Ducastelle, <https://arxiv.org/abs/2011.07811>

# Environment: phonons

