Electron-phonon coupling in light-driven solids: bridging ab-initio theory and pump-probe experiments

Fabio Caruso
University of Kiel, Germany

July 16th, 2024

Frontiers in many-body excited-state dynamics from first principles

CECAM-HQ, Lausanne

https://cs2t.de

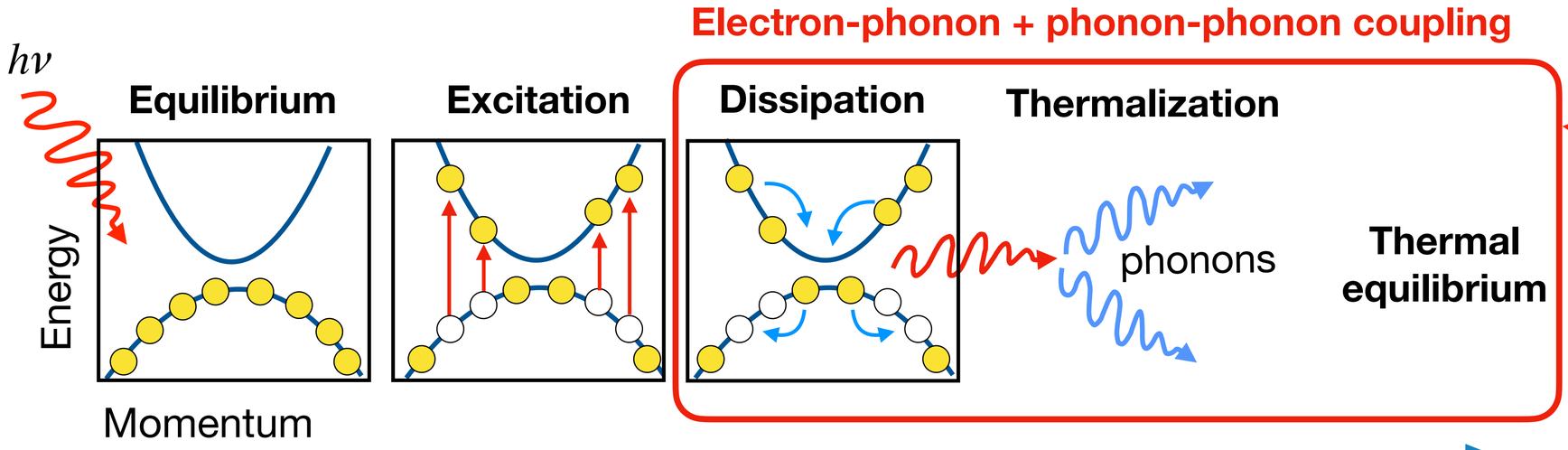
C | S² | T Computational Solid-State Theory group

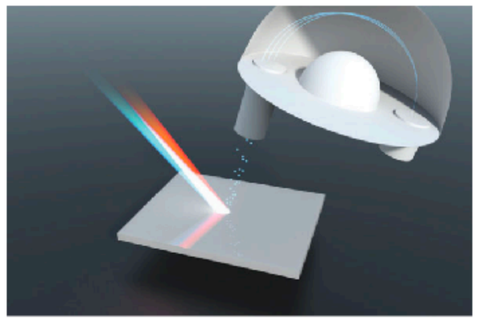
C | A | U Christian-Albrechts-Universität zu Kiel

Funded by DFG Deutsche Forschungsgemeinschaft German Research Foundation

MARIE CURIE ACTIONS

Research in the Computational Solid-State Theory group @ Uni. Kiel





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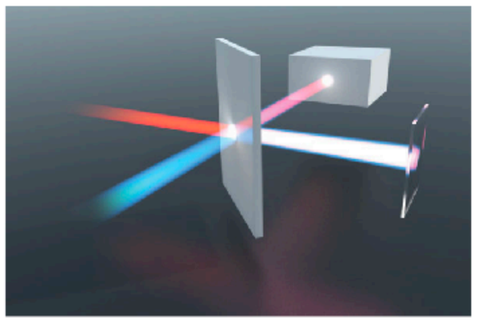
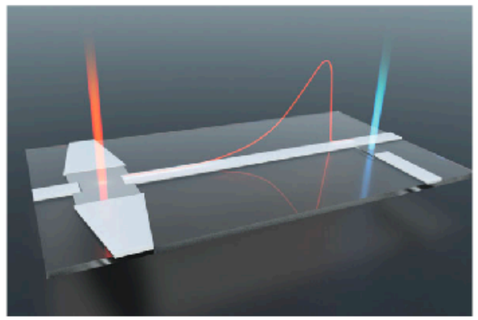
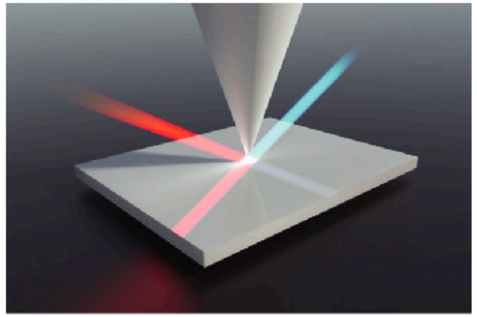


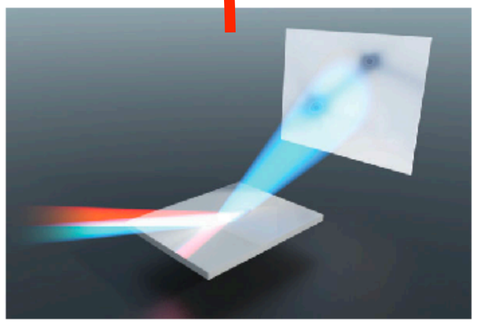
Optical probes



Transport



Scanning probes

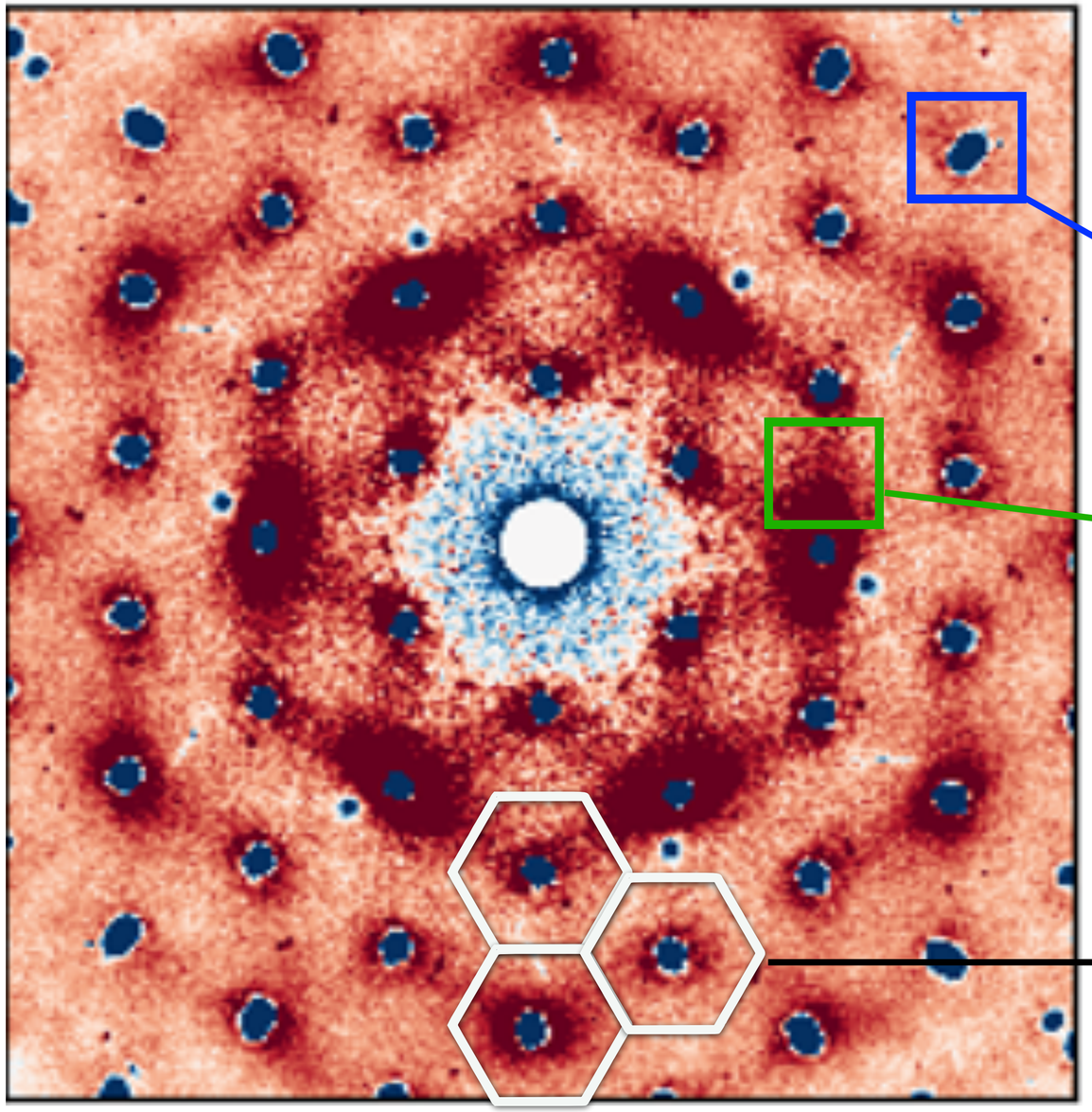


Scattering probes

de la Torre, et al. Rev Mod. Phys. (2021)

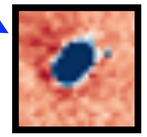
direct imaging of nuclear degrees of freedom

$$\Delta I(t) = I(t) - I(t = 0)$$

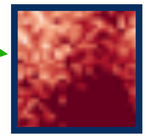


measurements: **Helene Seiler**

ultrafast electron diffuse scattering (UEDS) for bulk MoS_2



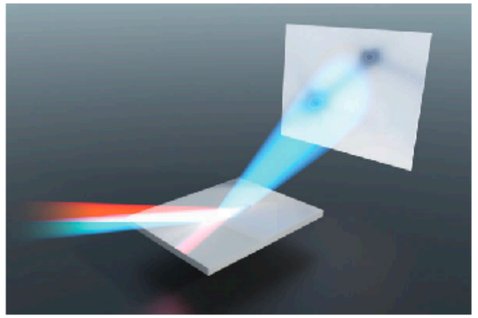
elastic scattering (Bragg peaks)



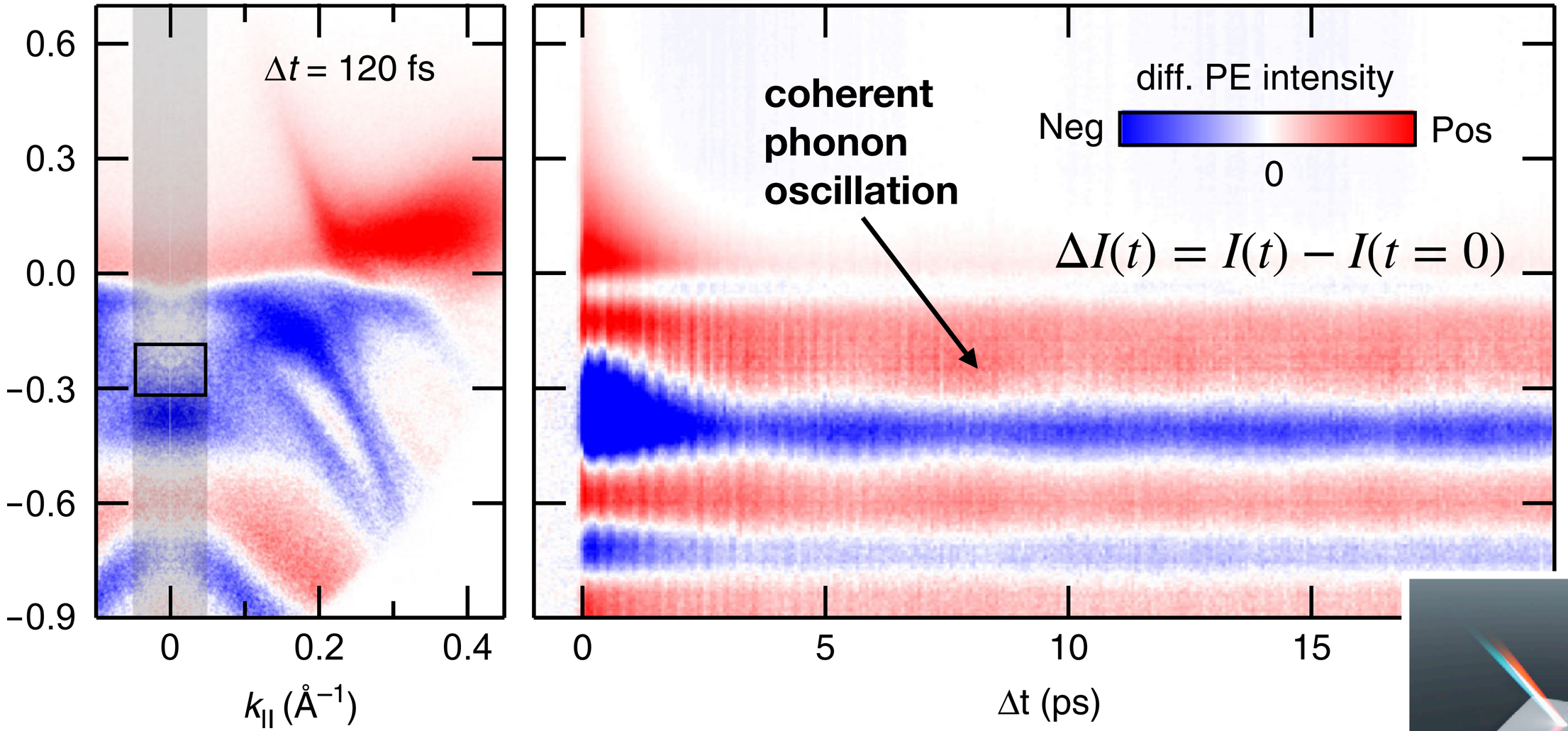
diffuse scattering (phonon-assisted electron scattering

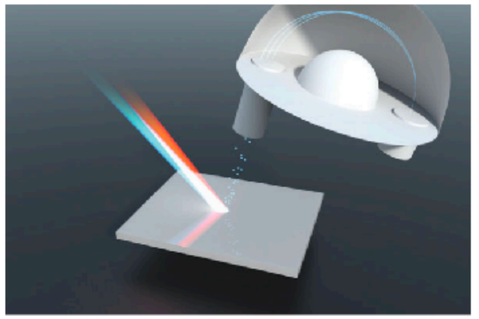
- thermal (temperature increase)
- non-thermal (phonons out of equilibrium)

Brillouin zone



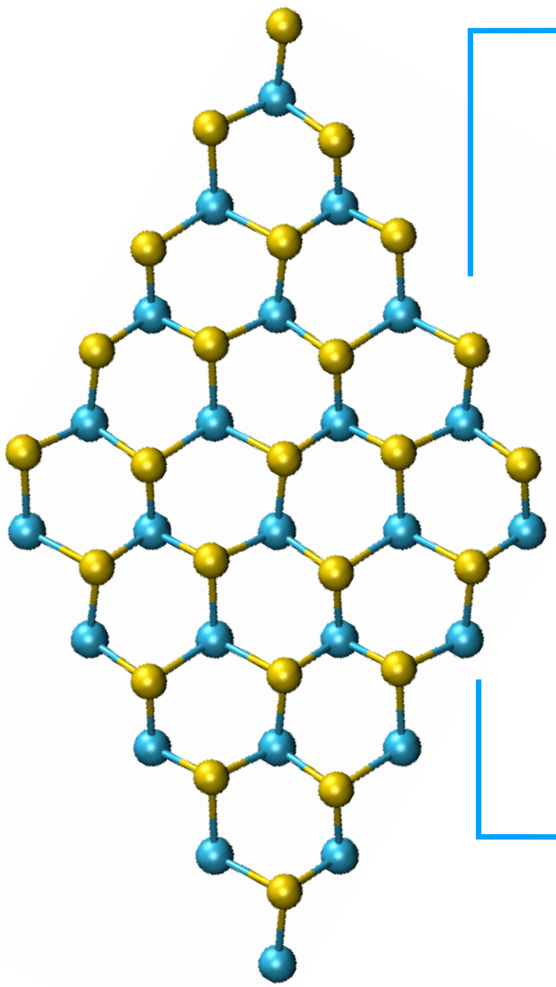
time- and angle-resolved photoemission spectroscopy (trARPES) for bulk WTe_2





measurements: P. Hein, M. Bauer, et al., Nature Comm. (2020)

In this talk



Part 1

Ab-initio simulations of ultrafast phonon dynamics: what can we learn?

Part 2

Non-equilibrium lattice dynamics "a la carte”: opportunities for engineering phonons out of equilibrium

Part 3

Transient screening of the electron-phonon coupling in bulk MoS_2 ?

Part 1

Ab-initio simulations of the non-equilibrium phonon dynamics: what can we learn?

Non-equilibrium phonon dynamics from the time-dependent Boltzmann equation (TDBE)

The TDBE:

$$\frac{\partial f_{n\mathbf{k}}}{\partial t} = \Gamma^{\mathrm{e-ph}}_{n\mathbf{k}} + \Gamma^{\mathrm{e-e}}_{n\mathbf{k}} \rightarrow \text{electron dynamics}$$

$$\frac{\partial n_{\mathbf{q}\nu}}{\partial t} = \Gamma^{\mathrm{e-ph}}_{\mathbf{q}\nu} + \Gamma^{\mathrm{ph-ph}}_{\mathbf{q}\nu} \rightarrow \text{phonon dynamics}$$

electron-phonon coupling

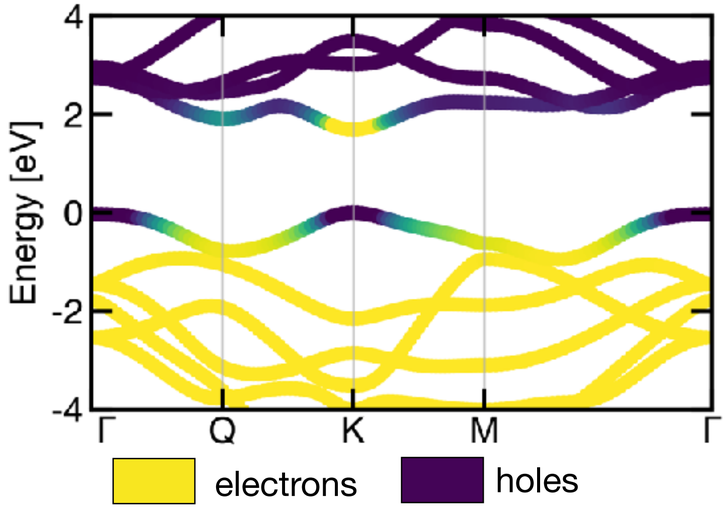
electron and phonon occupations: $n_{\mathbf{q}\nu}$ and $f_{n\mathbf{k}}$
collision integrals: $\Gamma_{n\mathbf{k}}$

Bonitz, Quantum Kinetic Theory, Springer (2016)
Bernardi, Louie et al., PNAS (2015)
FC, J. Phys. Chem. Lett. (2021)
Tong, Bernardi, Phys. Rev. Res. (2021)
Review: FC, Novko, Adv. Phys. X (2022)

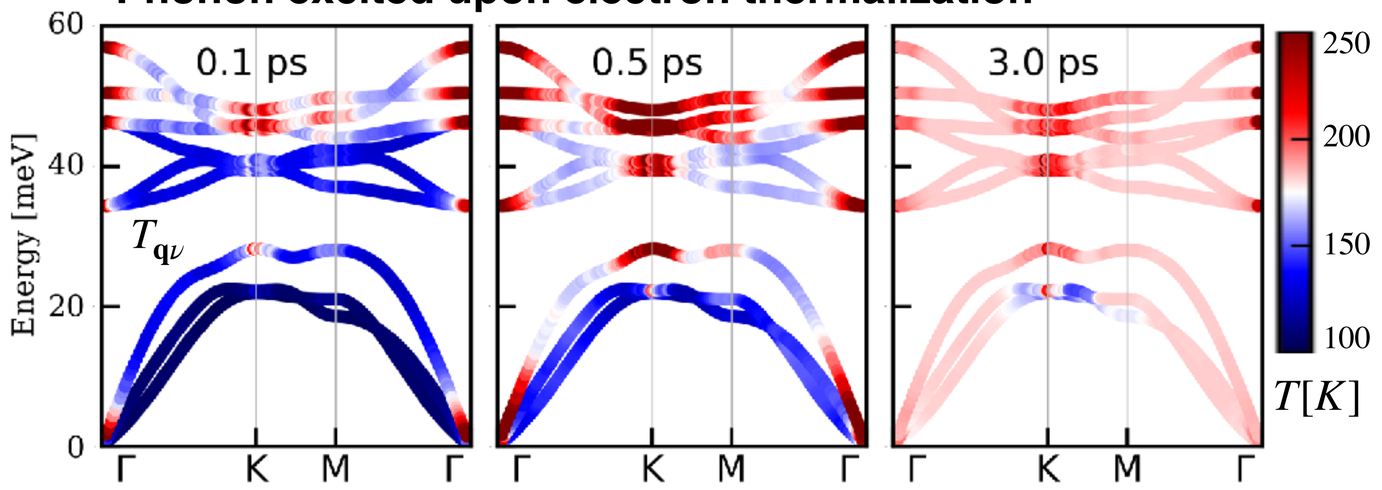
EPW®

Coupled electron-phonon dynamics in monolayer MoS_2

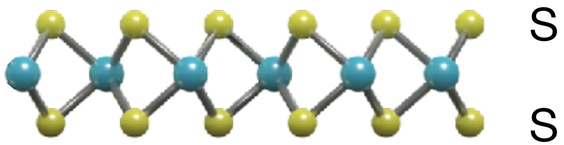
Electronic excitation

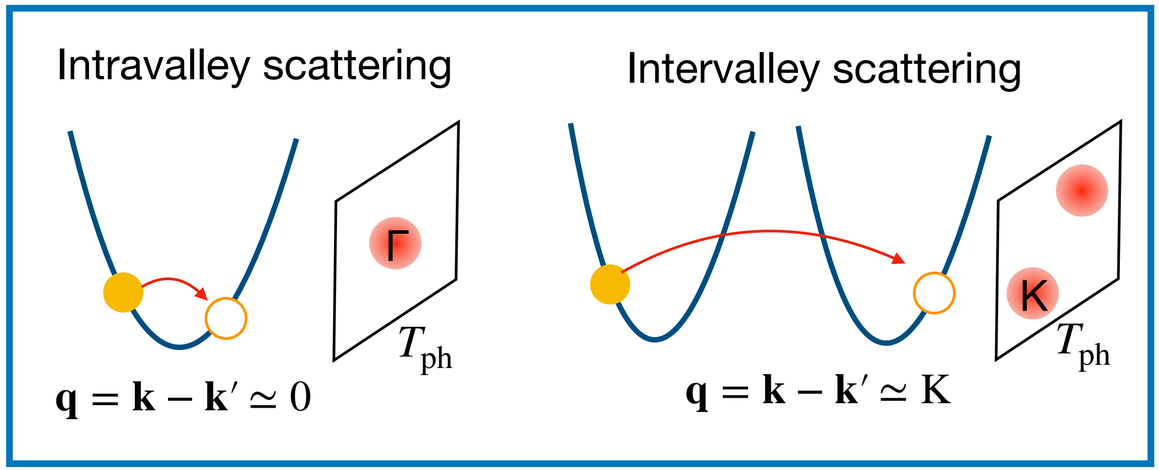


Phonon excited upon electron thermalization

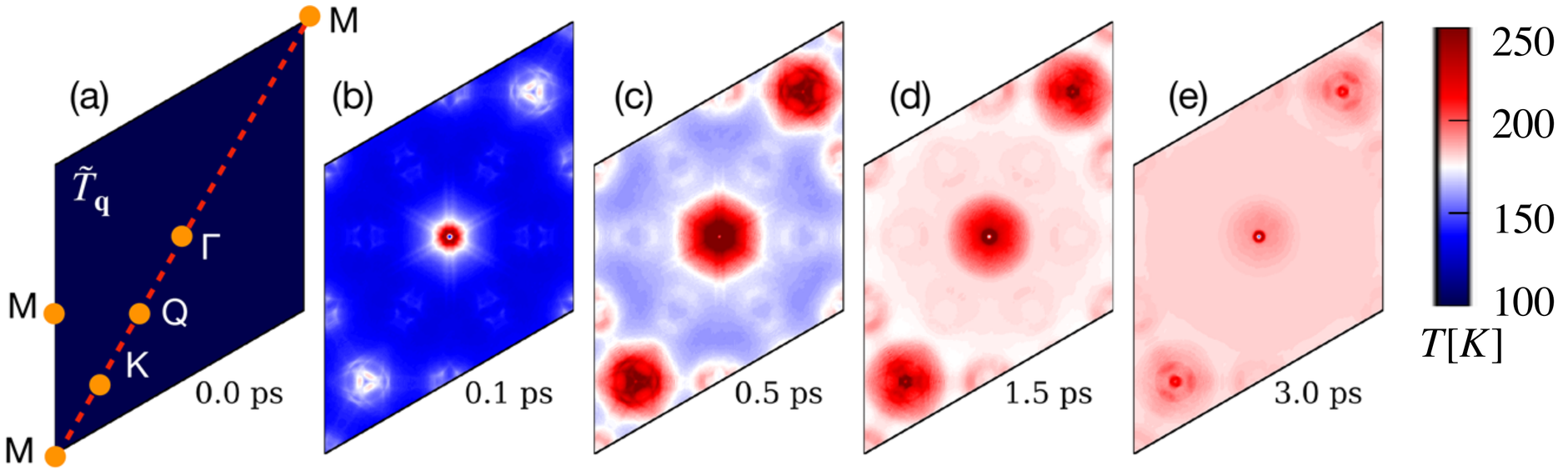


Momentum selectivity and non-equilibrium phonon dynamics





Non-equilibrium phonon population in 2D MoS_2



FC, J. Phys. Chem. Lett. (2021)

NANO LETTERS

pubs.acs.org/NanoLett

Letter

Accessing the Anisotropic Nonthermal Phonon Populations in Black Phosphorus

Hélène Seiler,* Daniela Zahn, Marios Zacharias, Patrick-Nigel Hildebrandt, Thomas Vasileiadis, Yoav William Windsor, Yingpeng Qi, Christian Carbogno, Claudia Draxl, Ralph Ernstorfer, and Fabio Caruso*

Cite This: *Nano Lett.* 2021, 21, 6171–6178

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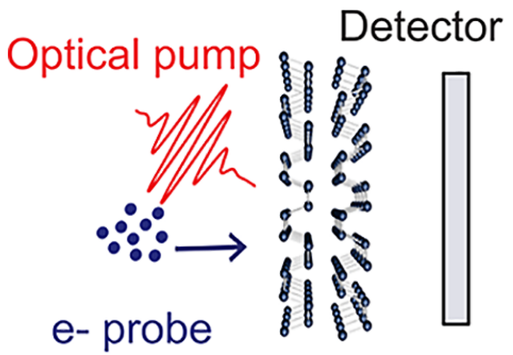
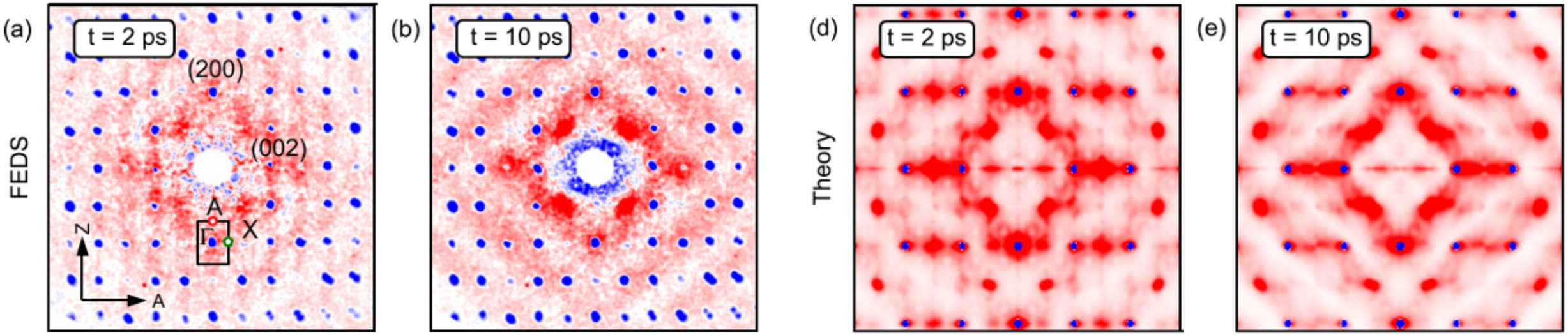
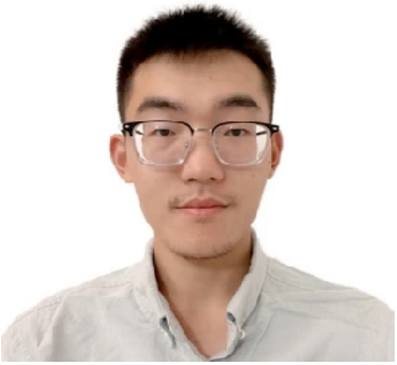




Part 2

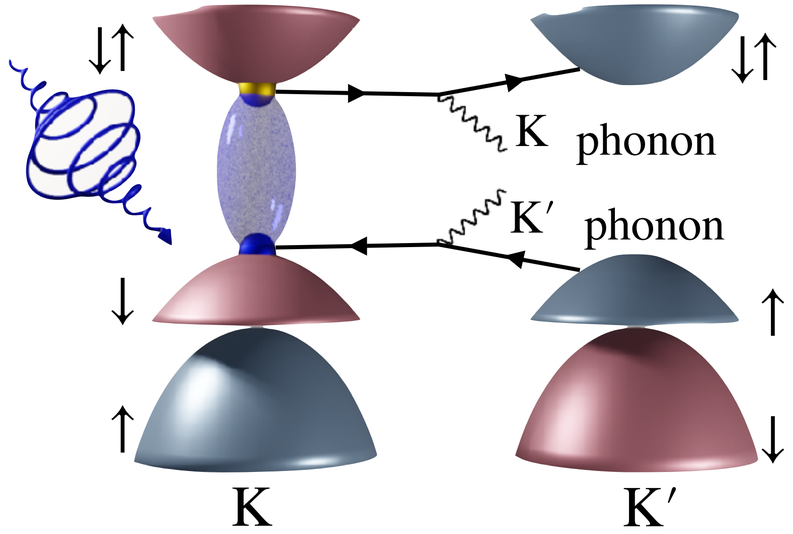
Non-equilibrium lattice dynamics "a la carte”: opportunities for engineering phonons out of equilibrium

How to control non-equilibrium dynamical states of the lattice?



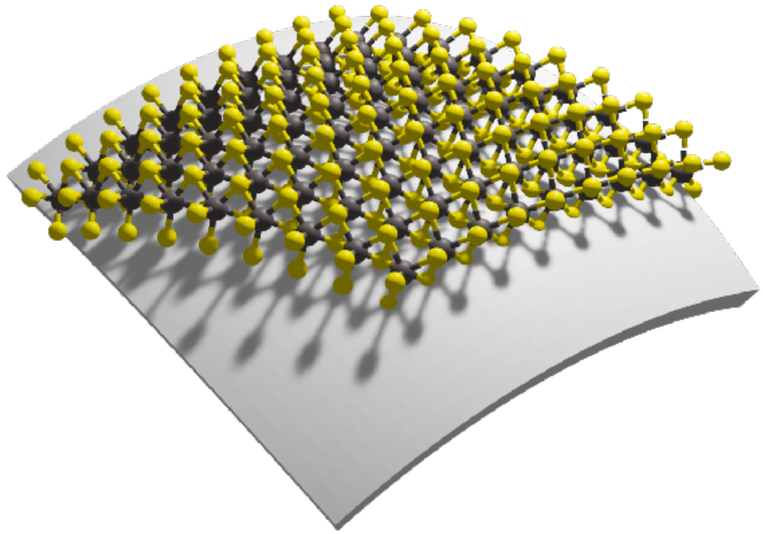
Yiming Pan

Option 1: tailored electronic excitations



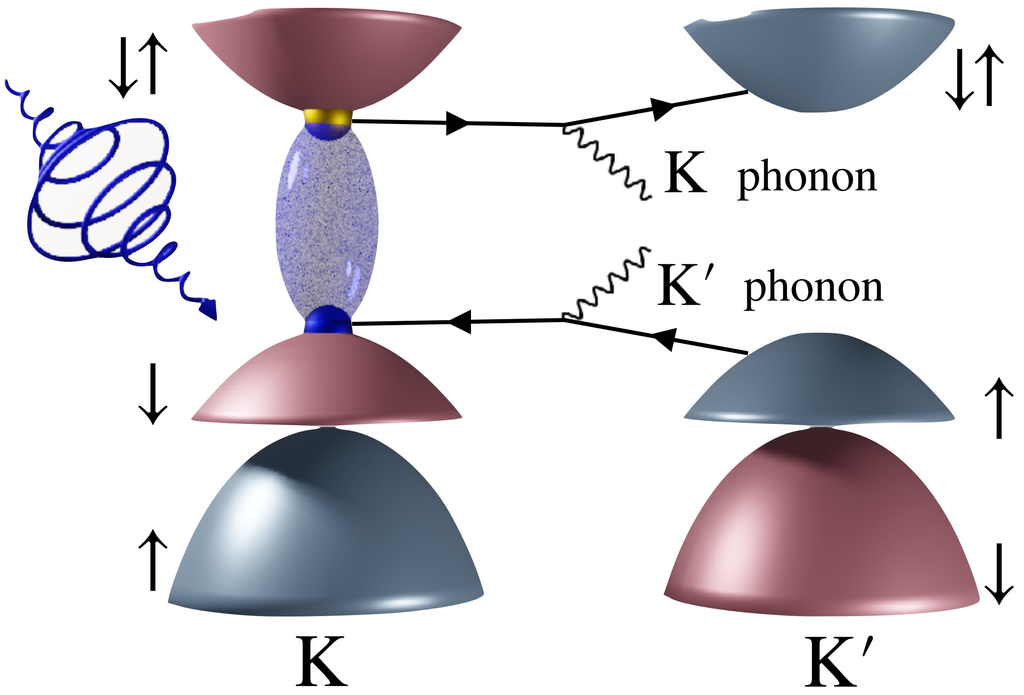
Pan, FC, Nano Lett. (2023)

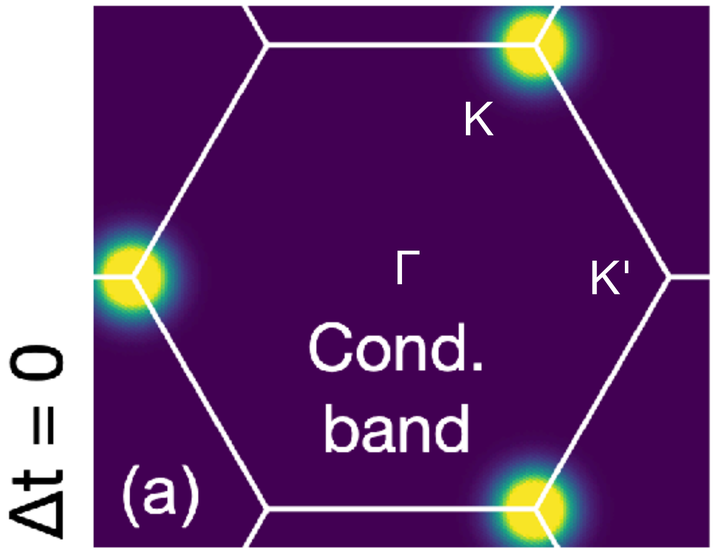
Option 2: control relaxation pathways



Pan, FC, npj 2D Mater. Appl. (2024)

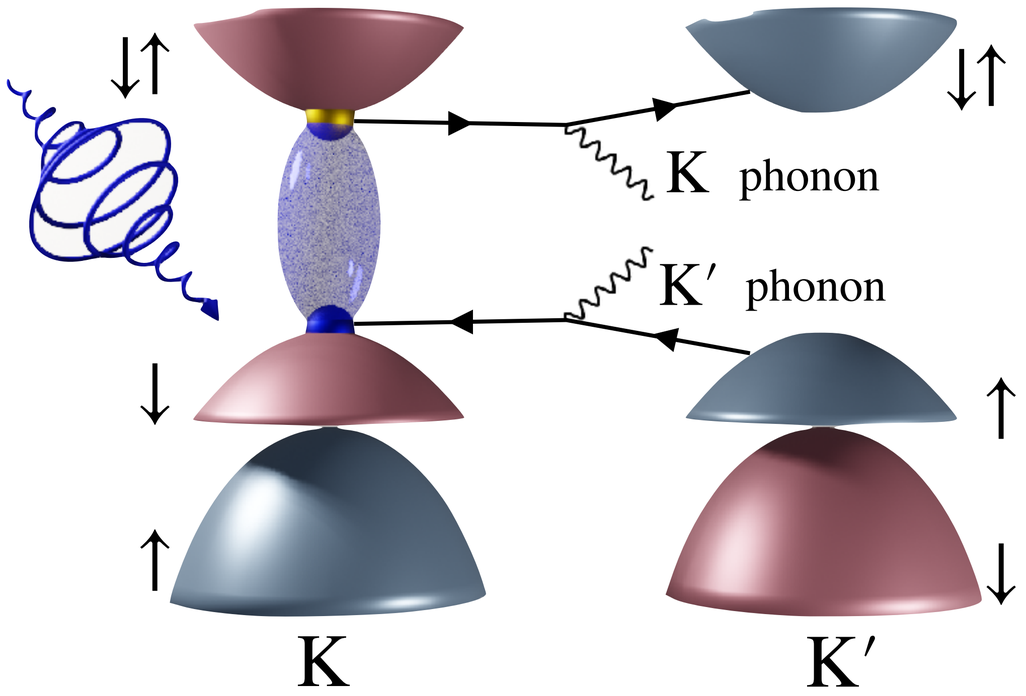
Valley selective optical excitation in MoS_2

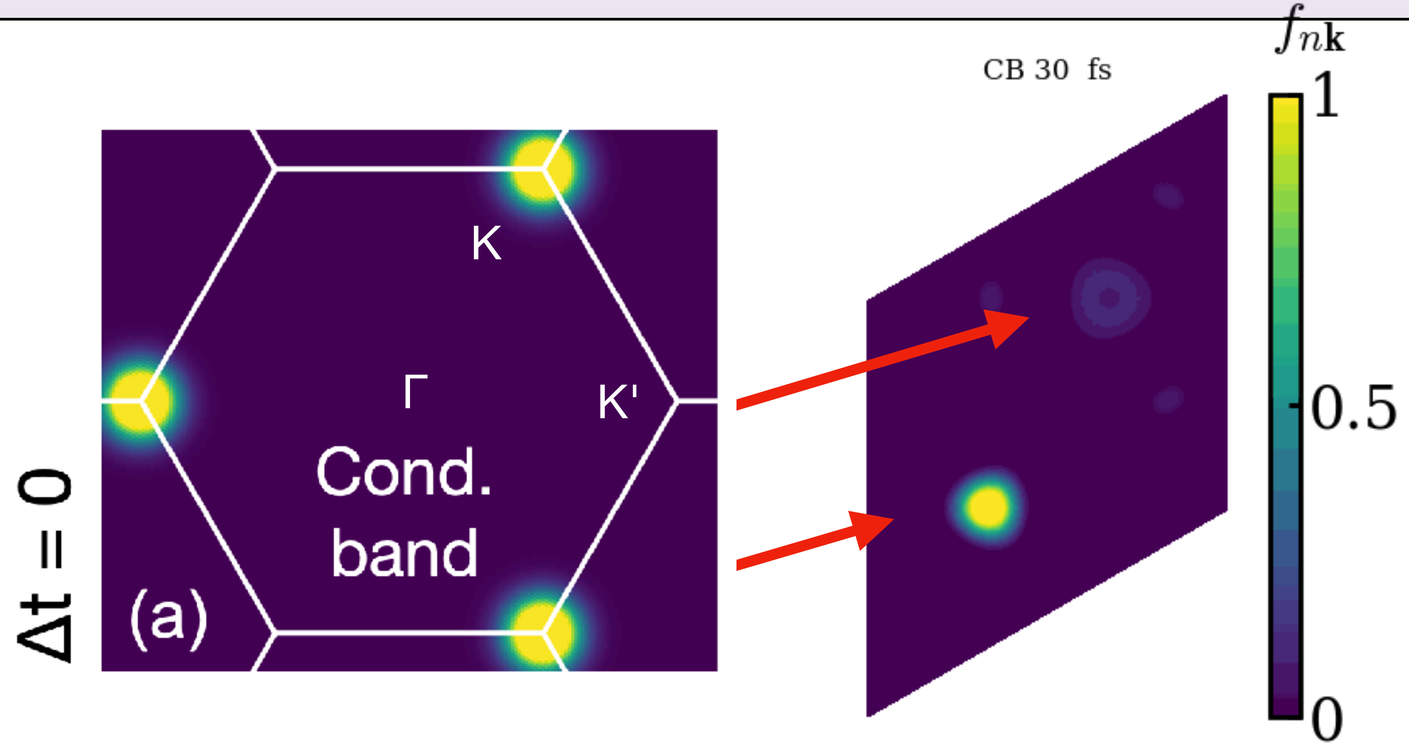




- three-fold rotational invariance
- non-centrosymmetryc crystal structure

Valley selective optical excitation in MoS_2



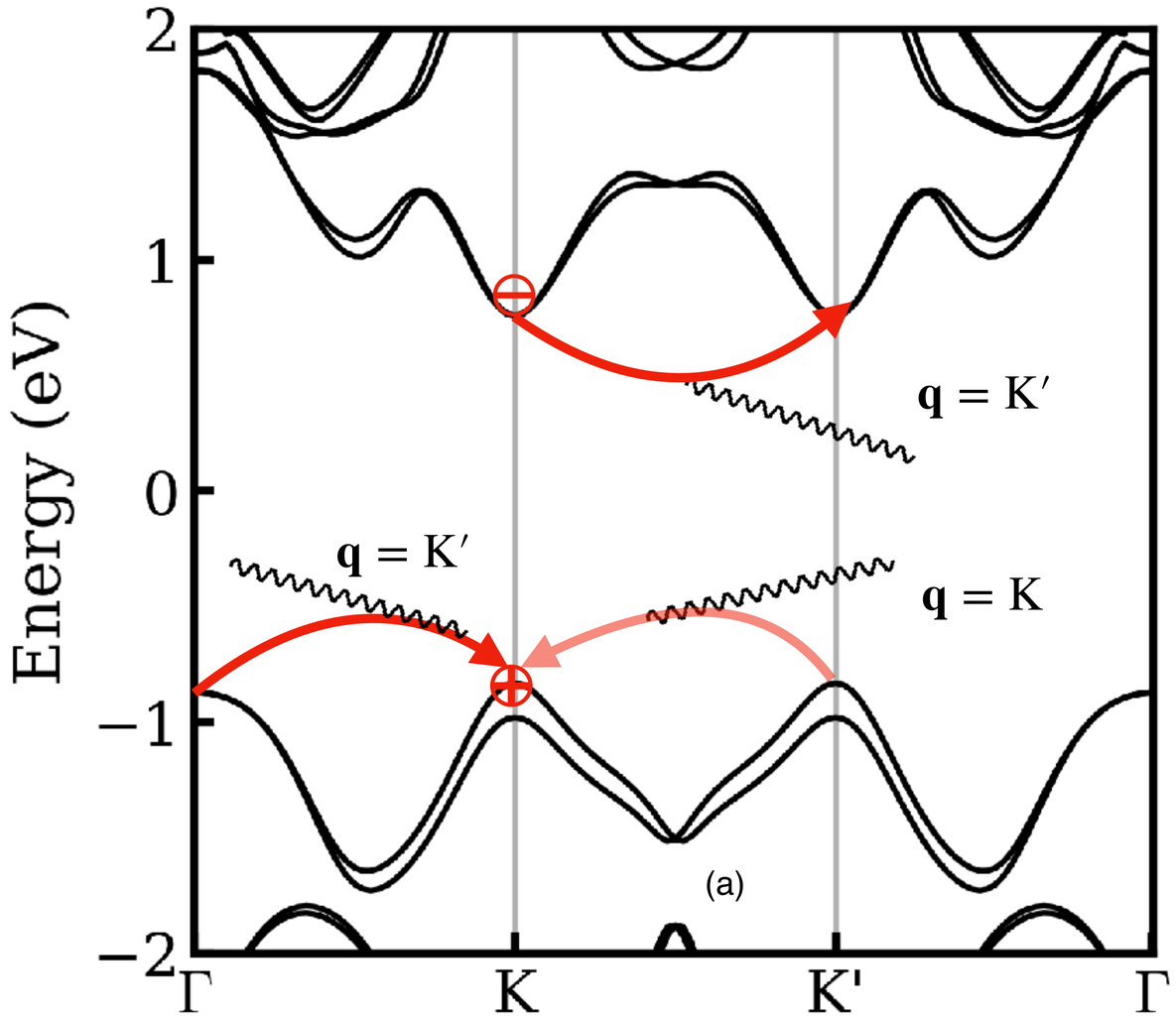


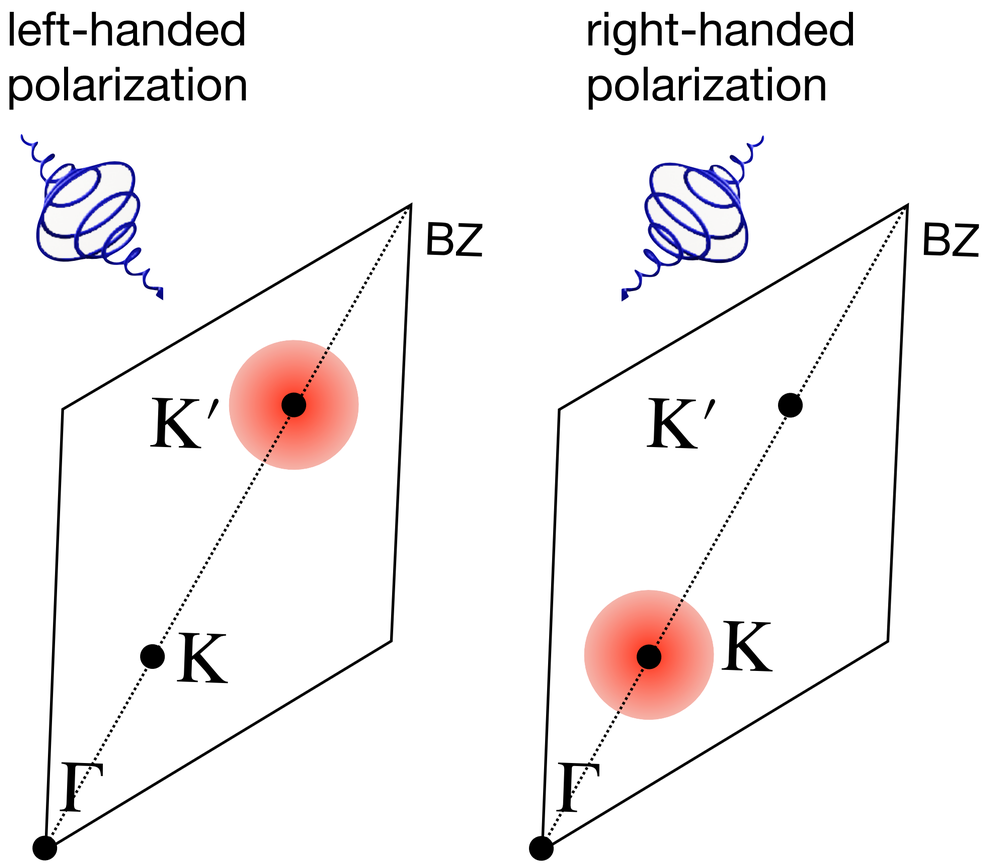
- three-fold rotational invariance
- non-centrosymmetryc crystal structure

- **Ultrafast valley depolarization dynamics**
- **Different timescales for valence and conduction band**

Yao, Niu, et al., Phys. Rev. B (2008)
Mak, Heinz, et al., Nature Nanotec. (2012)
Molina-Sánchez, et al., Nano Lett. 17, 4549 (2017)
Dal Conte, et al., Phys. Rev B 92, 235425 (2015)
Beyer et al., Phys. Rev. Lett. 123, 236802 (2019)
Xu, Duan, et al., Nano Lett. (2021)
Lin, Montserrat, et al., Phys. Rev. Lett. (2022)

The decay path of valley-polarized carriers in MoS_2



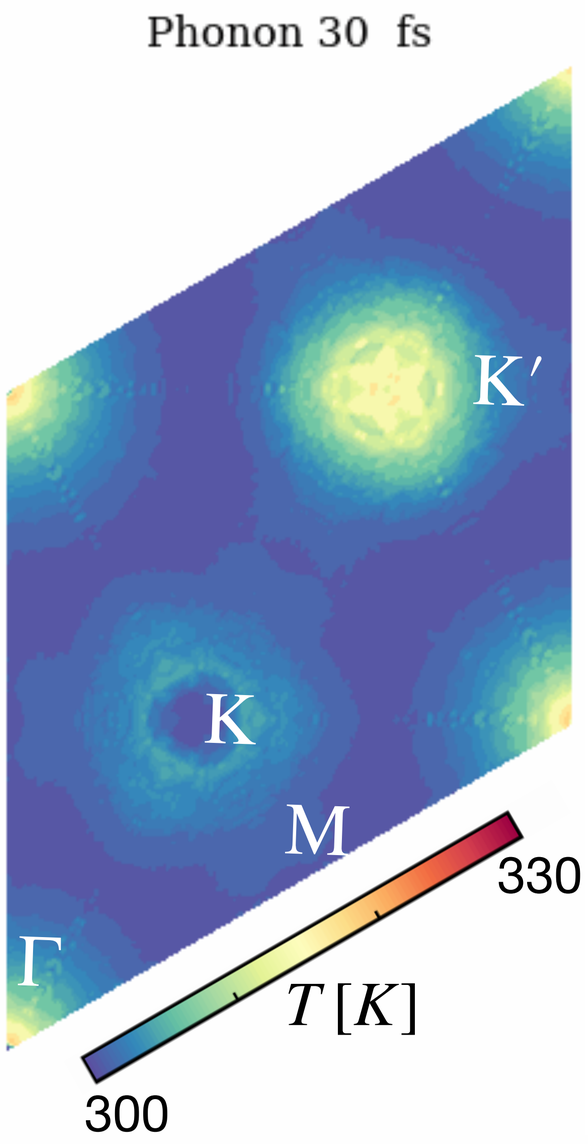


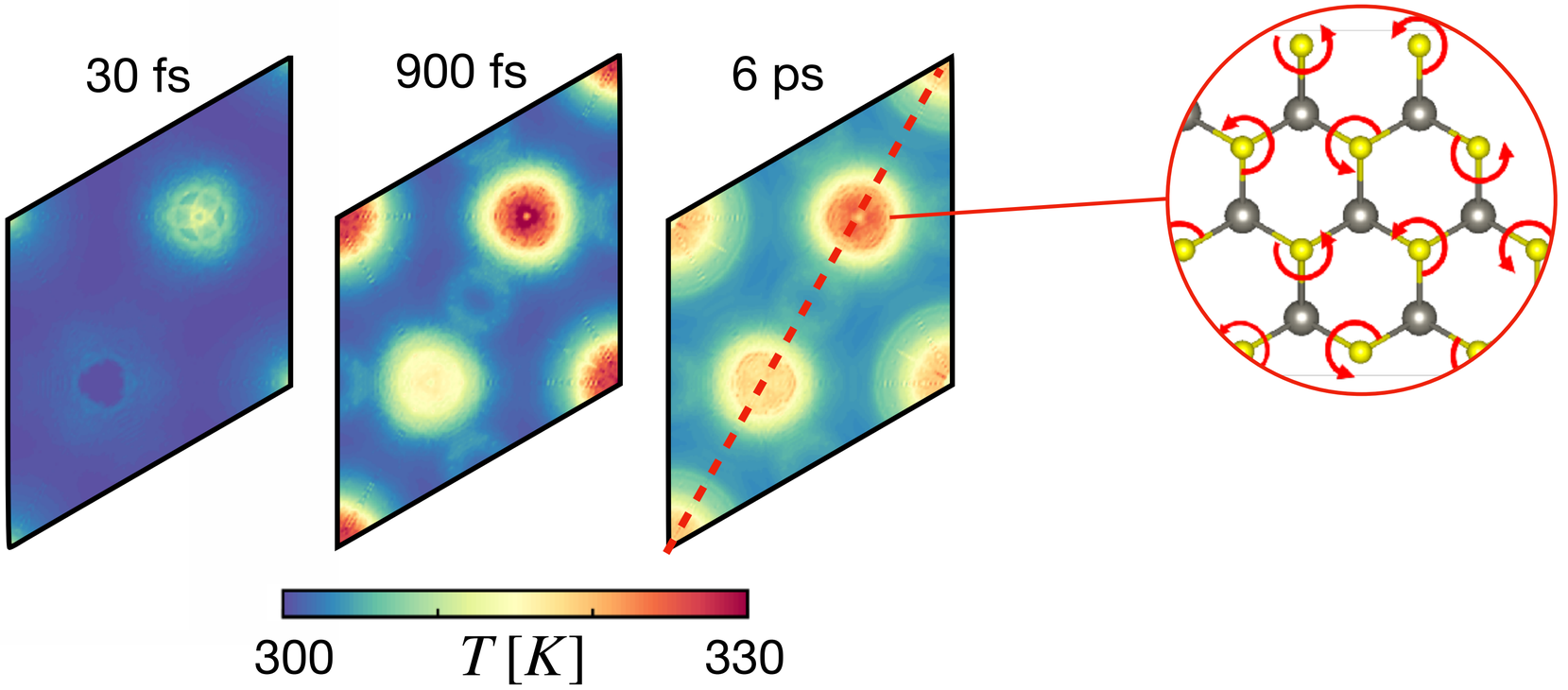
valley-polarized phonons
at the K and -K high-symmetry points

Pan, FC, Nano Lett. (2023)

Valley-polarized non-equilibrium phonon populations in MoS_2

Ab-initio simulation of non-equilibrium phonon population established upon valley depolarization

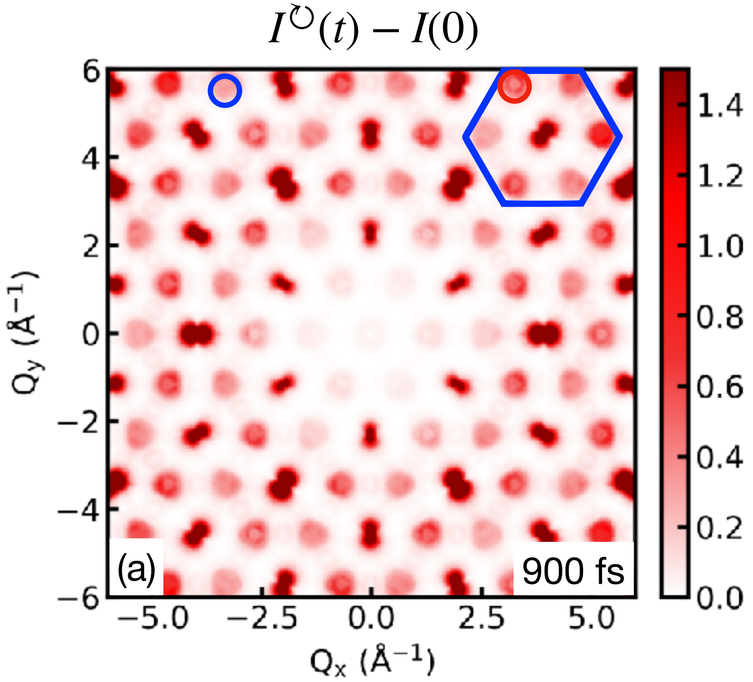


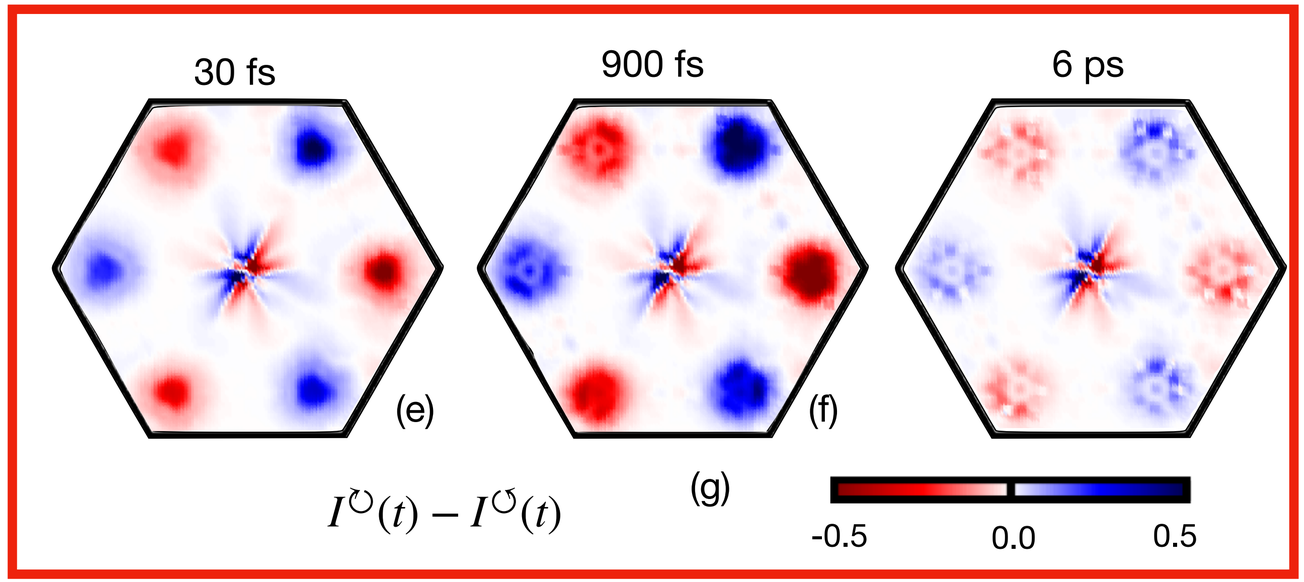


Pan, FC, Nano Lett. (2023)

Valley-polarized non-equilibrium phonon populations in MoS_2

Ultrafast electron diffuse scattering signal (simulations)



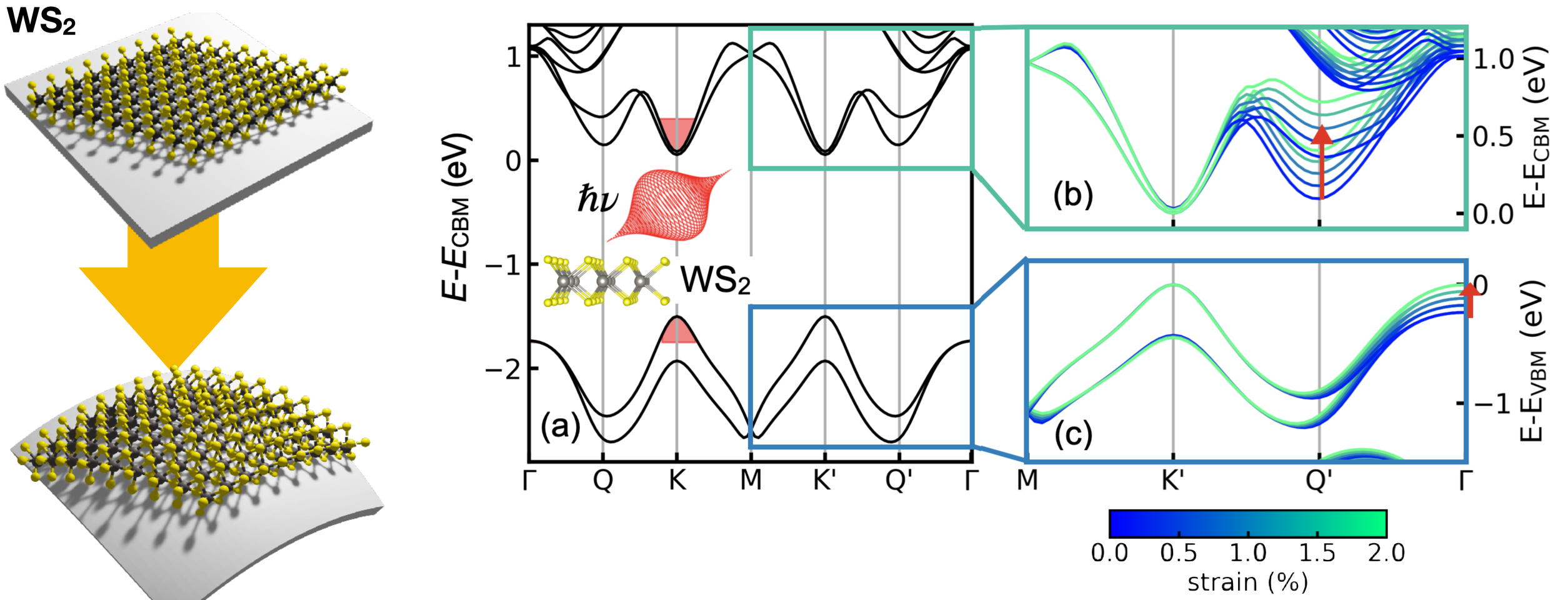


vibrational circular dichroism

$I^{\circlearrowright}(t) - I^{\circlearrowleft}(t)$ **: dichroic diffraction intensity (changes by switching polarization)**

Pan, FC, Nano Lett. (2023)

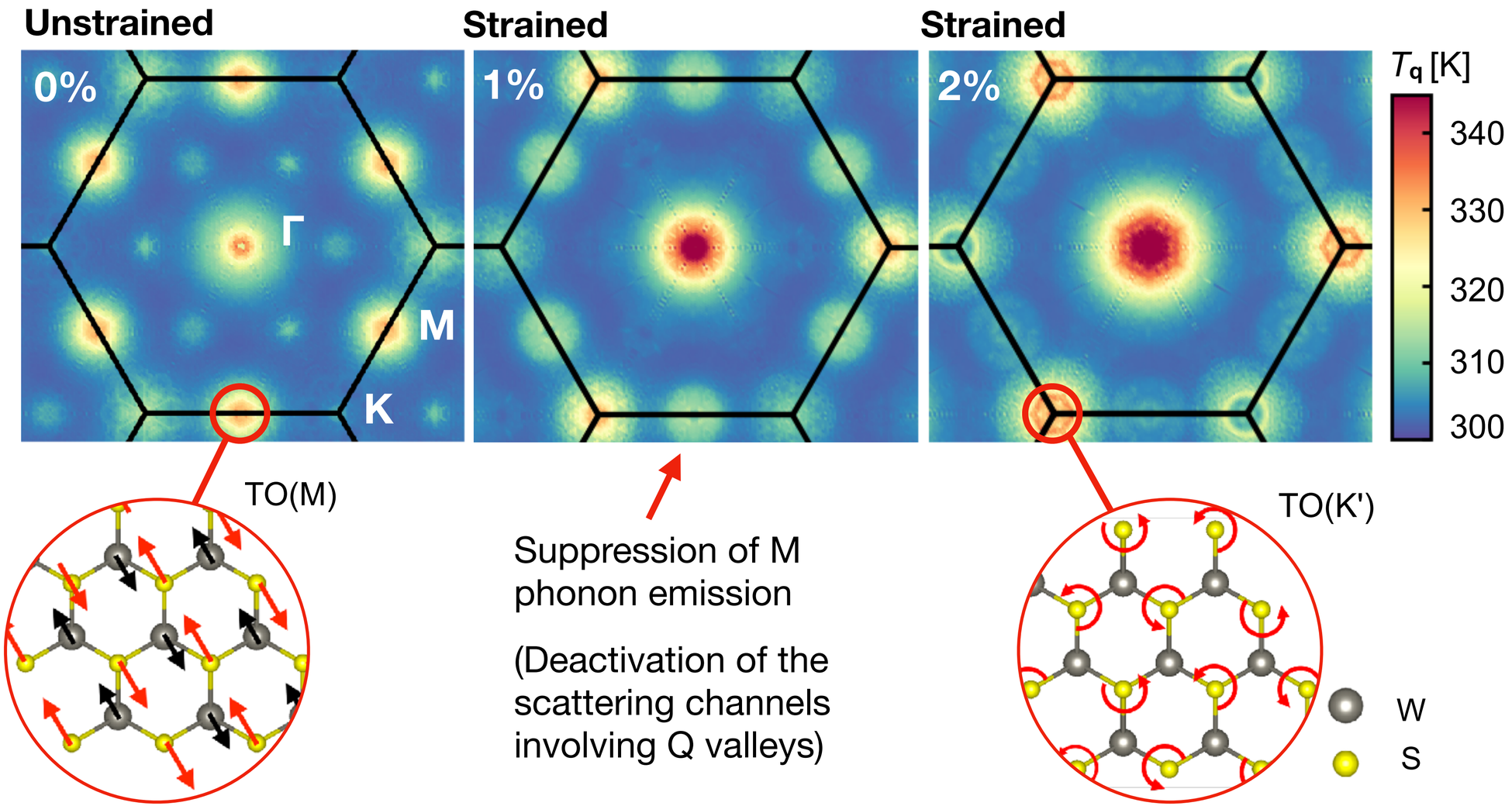
Strain-engineering band structures and relaxation channels: monolayer WS_2



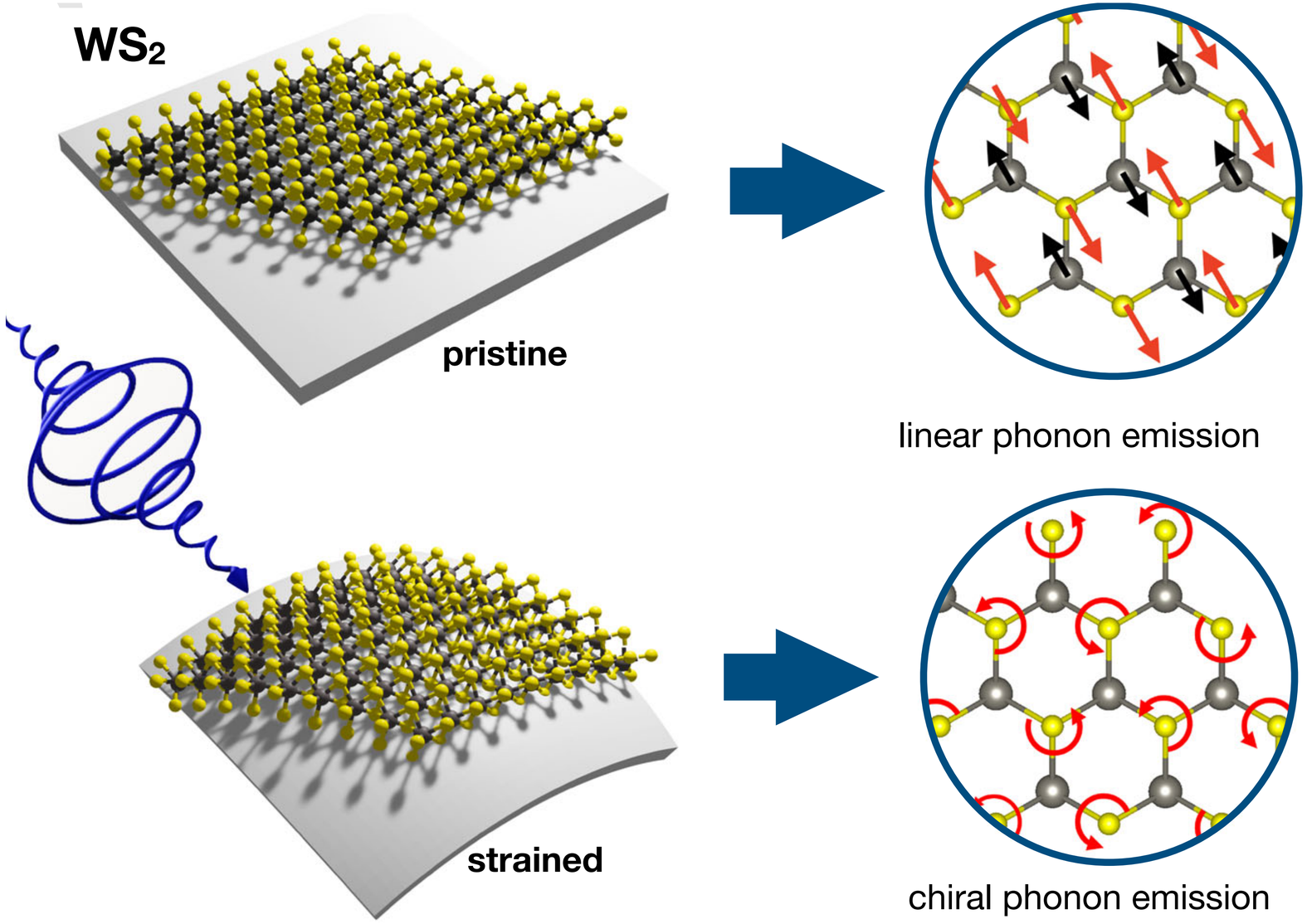
Pan, FC, npj 2D Mater. Appl. (2024)

Strain-induced activation of chiral phonon emission

Phonon population (1ps after valley-polarized excitation):



Pan, FC, npj 2D Mater. Appl. (2024)



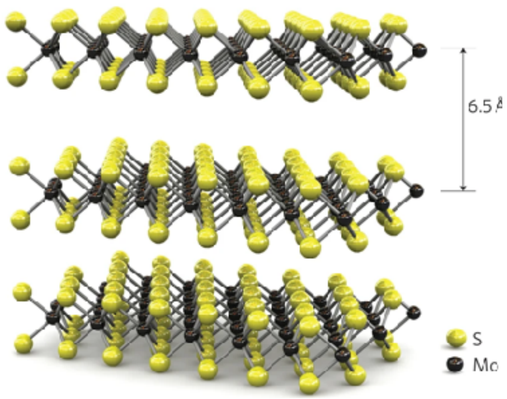
Pan, FC, npj 2D Mater. Appl. (2024)

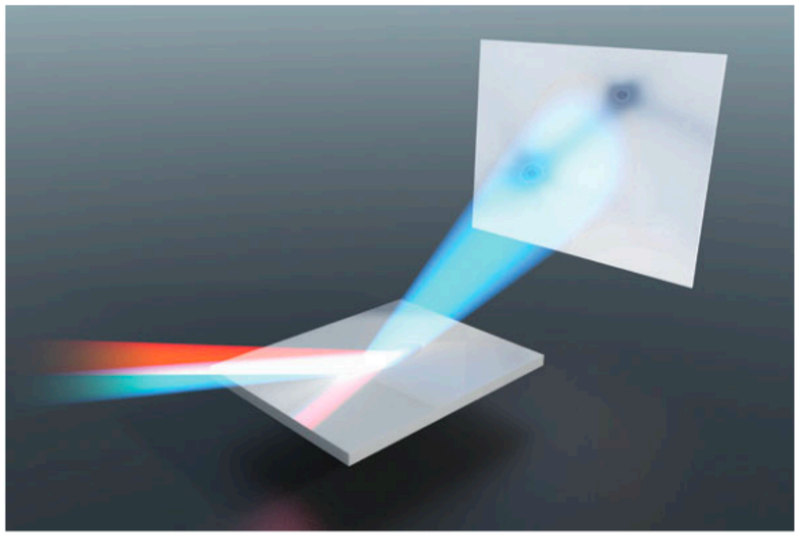
Part 3

Transient screening of the electron-phonon coupling in bulk MoS_2 ?

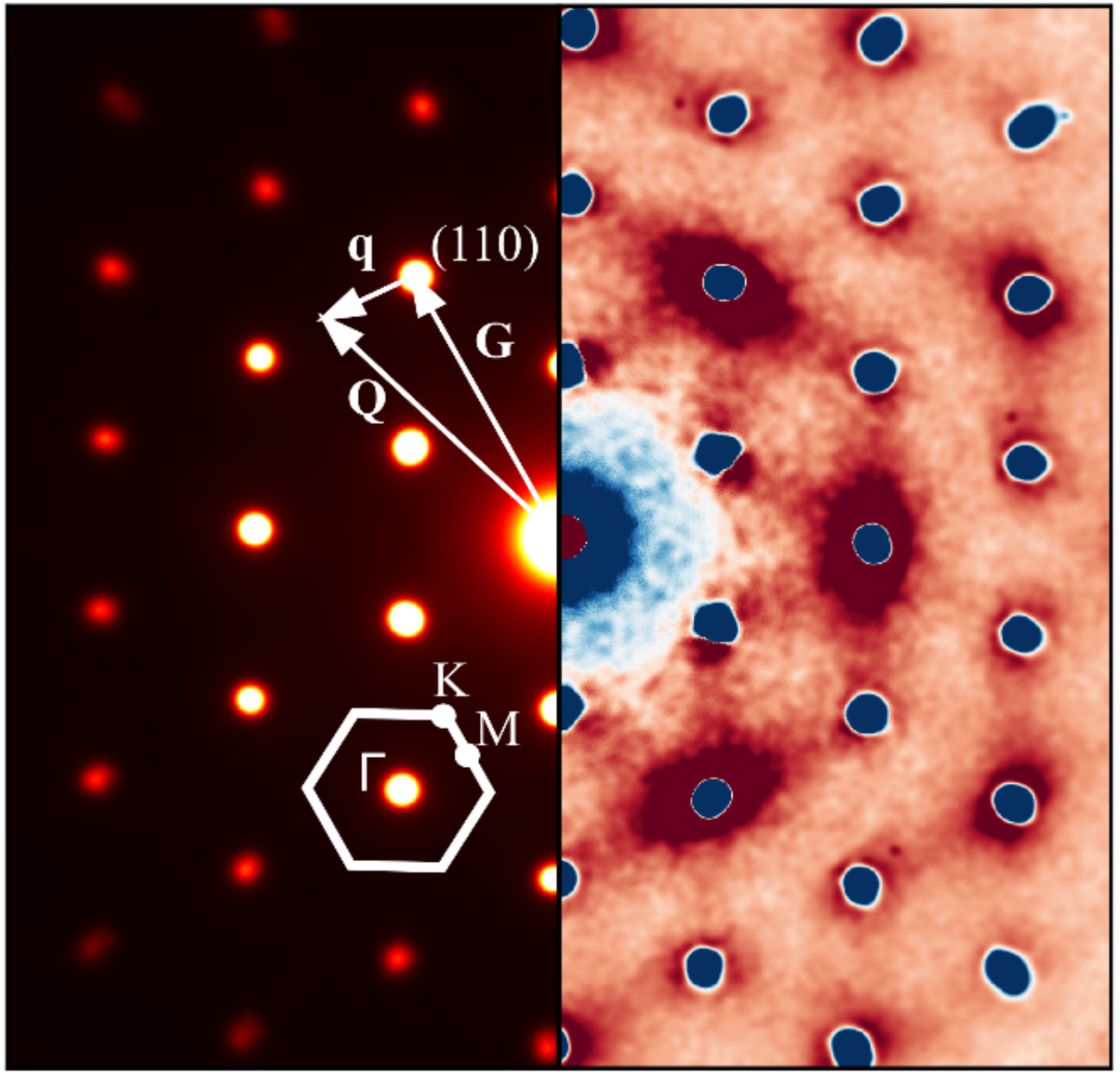
Ultrafast electron diffuse scattering: the case of bulk MoS_2

H. Seiler (FU Berlin)

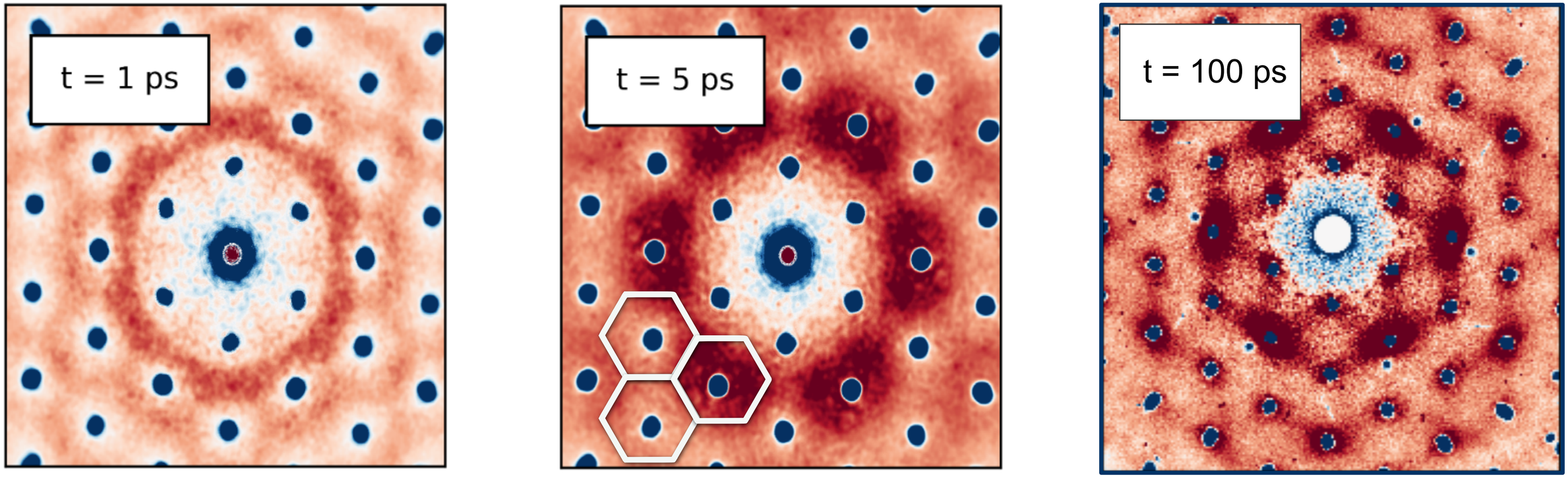




$I(t=0)$ $\Delta I(t) = I(t) - I(t=0)$



Two distinct non-thermal phonon populations (experiments)



non-thermal phonon populations

thermal equilibrium (at larger T)

Intensity of scattered radiation by a vibrating lattice

$$I(\mathbf{Q},\tau) = I_0(\mathbf{Q},\tau) + I_1(\mathbf{Q},\tau) + \cdots$$

Zero-phonon (von Laue condition)

Phonon contribution to diffraction

$$I_1(\mathbf{Q}) \propto \sum_{\nu} \frac{n_{\mathbf{q}\nu} + 1/2}{\omega_{\mathbf{q}\nu}} \left| \mathfrak{F}_{1\nu}(\mathbf{Q}) \right|^2$$

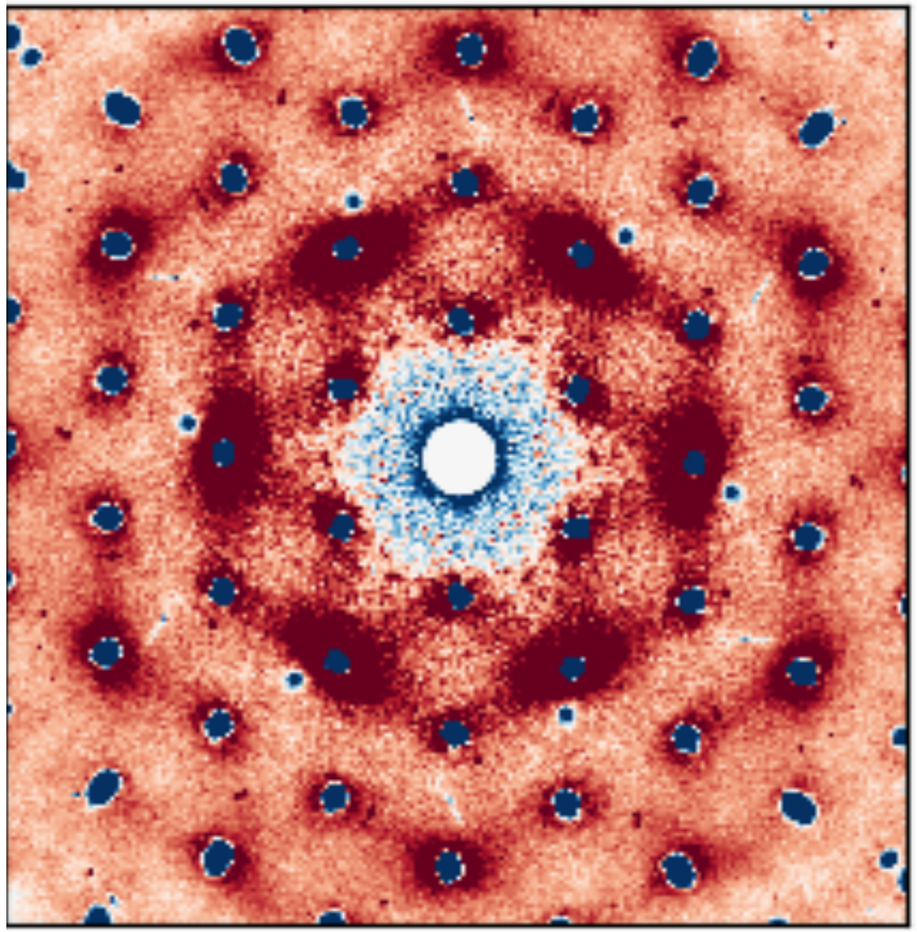
1-phonon contribution

$$\mathfrak{F}_{1\nu}(\mathbf{Q}) = \sum_{\kappa} e^{-W_\kappa(\mathbf{Q})} \frac{f_\kappa(\mathbf{Q})}{\sqrt{M_\kappa}} \left(\mathbf{Q} \cdot \mathbf{e}_{\mathbf{q}\nu\kappa} \right)$$

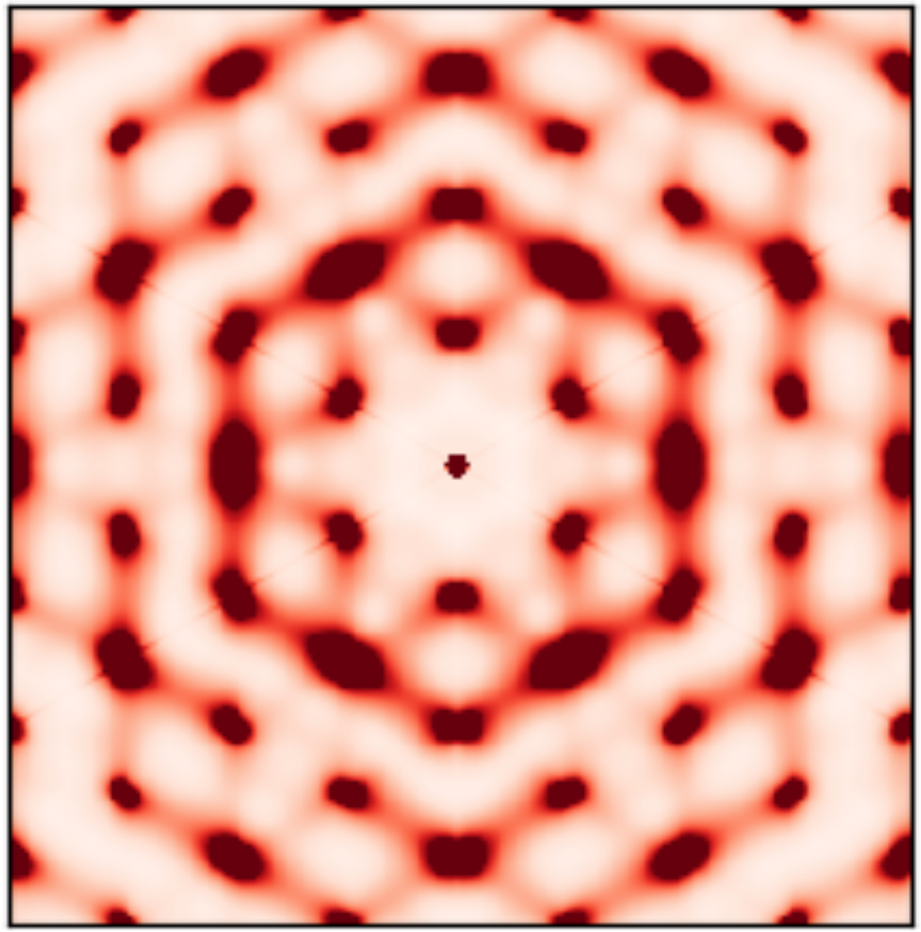
1-phonon structure factor

Diffuse scattering at thermalization: theory vs experiment

Experiment: delay 100 ps



Theory: multi-phonon SF, ΔT = 80 K

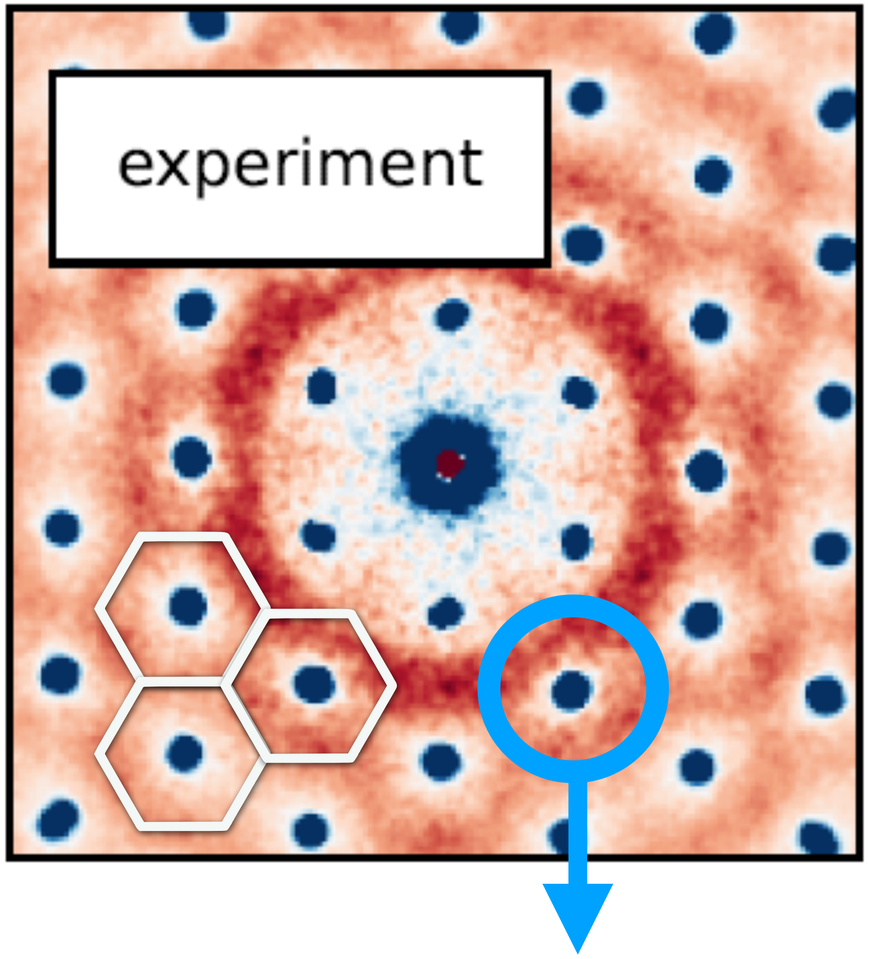


Marios Zacharias

Zacharias, Seiler, FC, et al. Phys. Rev. Lett. (2021)
Zacharias, Seiler, FC, et al. Phys. Rev. B (2021)

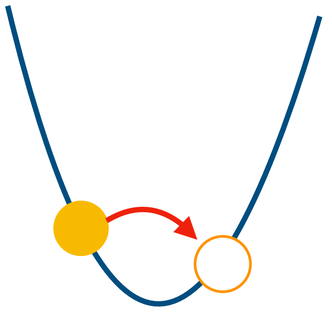
Diffuse scattering from non-equilibrium phonon populations

Experiment: delay 1 ps



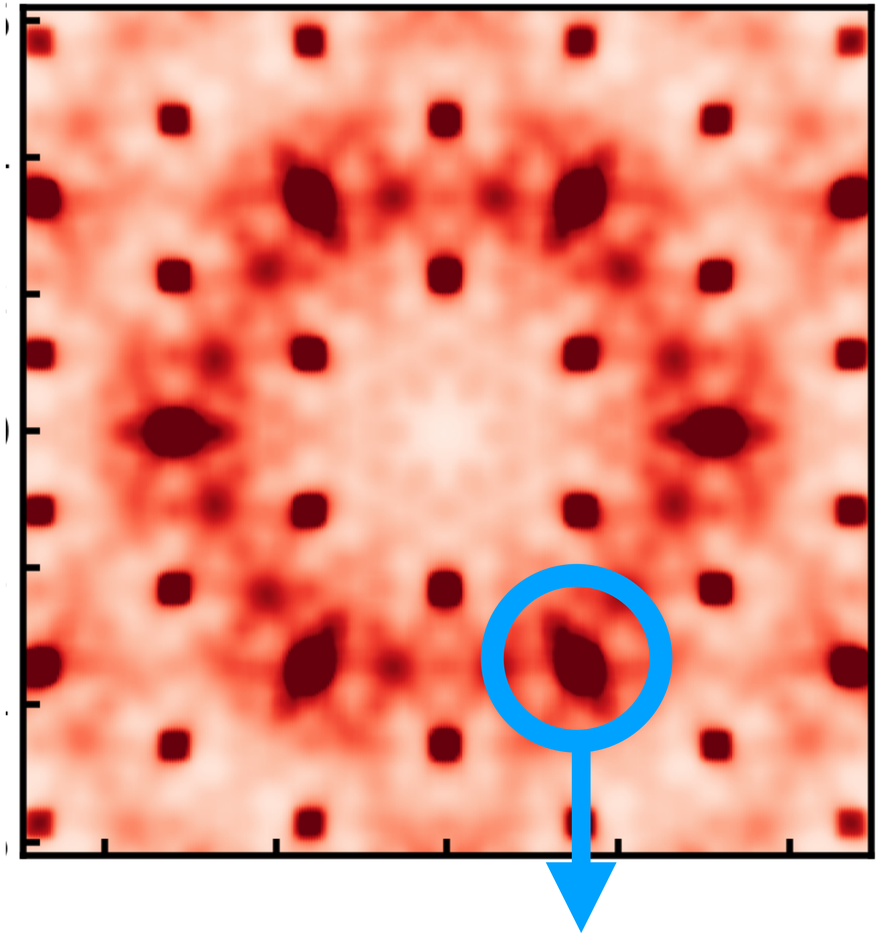
Suppression of phonon emission around Γ

Suppression of Intravalley electrong scattering due to EPI



$$\mathbf{q} = \mathbf{k} - \mathbf{k}' \simeq 0$$

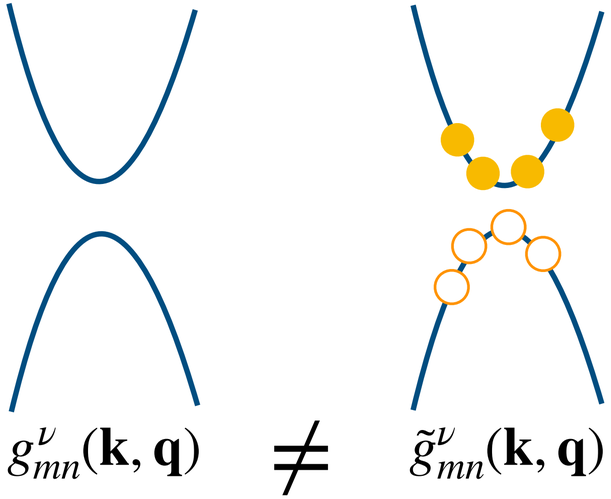
Theory: delay 1 ps



Strong phonon emission around Γ

Photo-screening of the electron-phonon interaction

$$g^{\nu}_{mn}(\mathbf{k},\mathbf{q}) = \langle m\mathbf{k}+\mathbf{q} \,|\, \Delta_{\mathbf{q}\nu} V_{\mathrm{KS}} \,|\, n\mathbf{k} \rangle$$



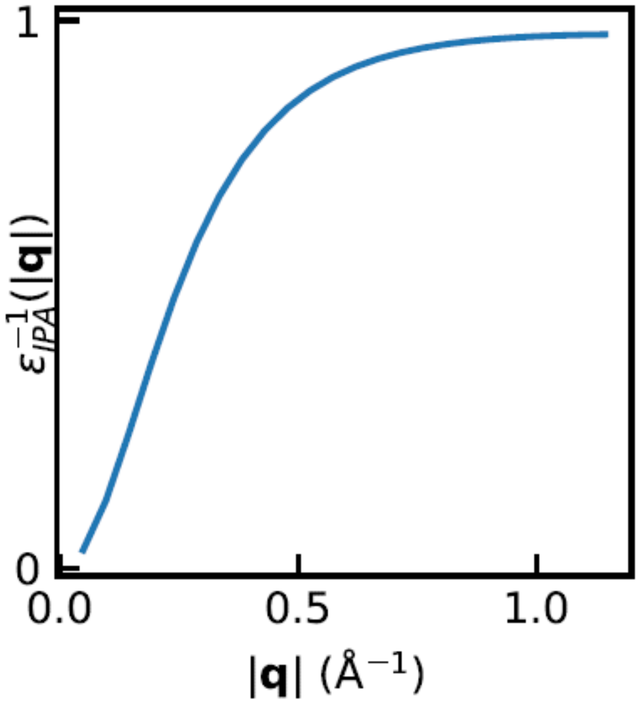
$g^{\nu}_{mn}(\mathbf{k},\mathbf{q}) \neq \tilde{g}^{\nu}_{mn}(\mathbf{k},\mathbf{q})$

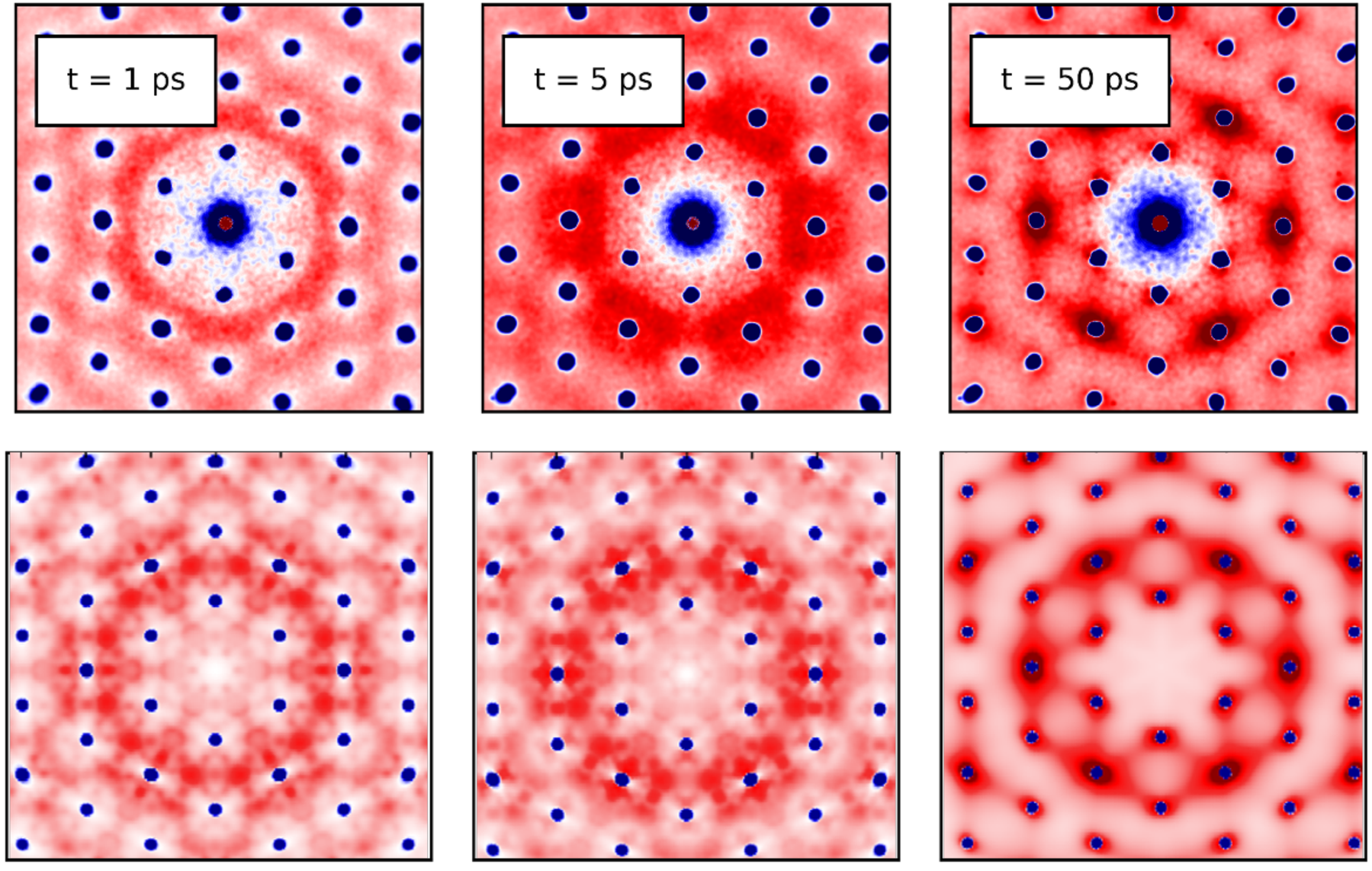
Renormalization of screening due to photoexcited carriers (independent particles):

$$\delta\chi_0(\mathbf{q}) = \sum_{mm'\mathbf{k}} \frac{\delta f_{m\mathbf{k}}(t) - \delta f_{m'\mathbf{k}+\mathbf{q}}(t)}{\varepsilon_{m\mathbf{k}} - \varepsilon_{m'\mathbf{k}+\mathbf{q}}} \,|\, \langle u_{m'\mathbf{k}+\mathbf{q}} \,|\, u_{m\mathbf{k}} \rangle \,|^2$$

Electron-phonon matrix elements for photo-doping:

$$\tilde{g}^{\nu}_{mn}(\mathbf{k},\mathbf{q}) = \frac{g^{\nu}_{mn}(\mathbf{k},\mathbf{q})}{1 - \frac{4\pi e^2}{|\mathbf{q}|^2 \epsilon_{\mathrm{undoped}}} \delta\chi_0(\mathbf{q})}$$

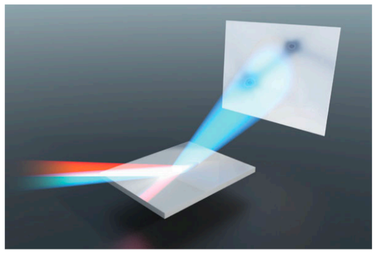


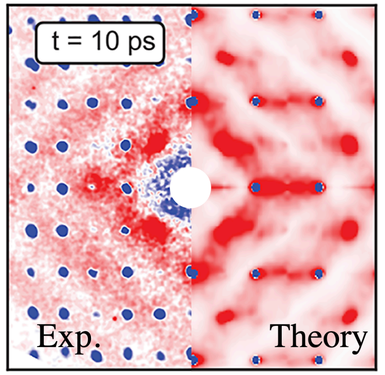

t = 1 ps
t = 5 ps
t = 50 ps

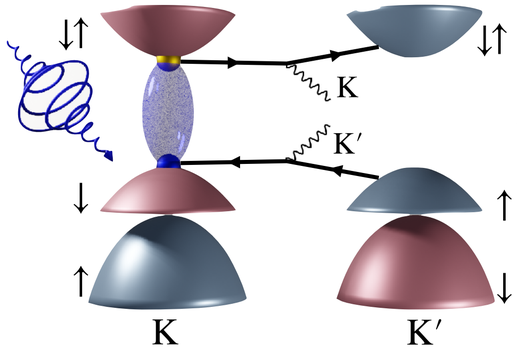
Final remarks

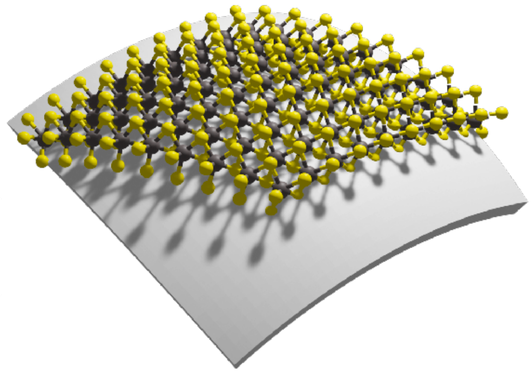
Part 1

Ab-initio simulations of ultrafast phonon dynamics: what can we learn?







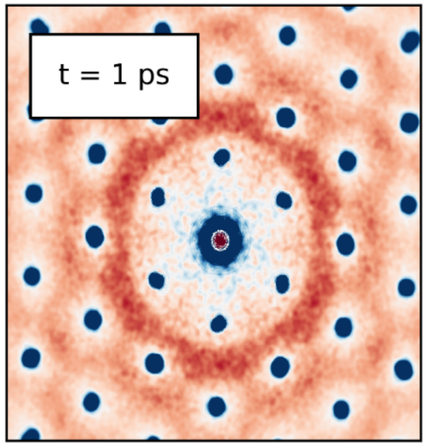


Part 2

Non-equilibrium lattice dynamics "a la carte": opportunities for engineering phonons out of equilibrium

Part 3

Transient screening of the electron-phonon coupling in bulk MoS_2 ?



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