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

Fabio Caruso
University of Kiel, Germany

September 18th, 2024

SFB1242 International conference

Nonequilibrium Dynamics of Condensed Matter in the Time Domain

Schluchsee, Germany

https://cs2t.de

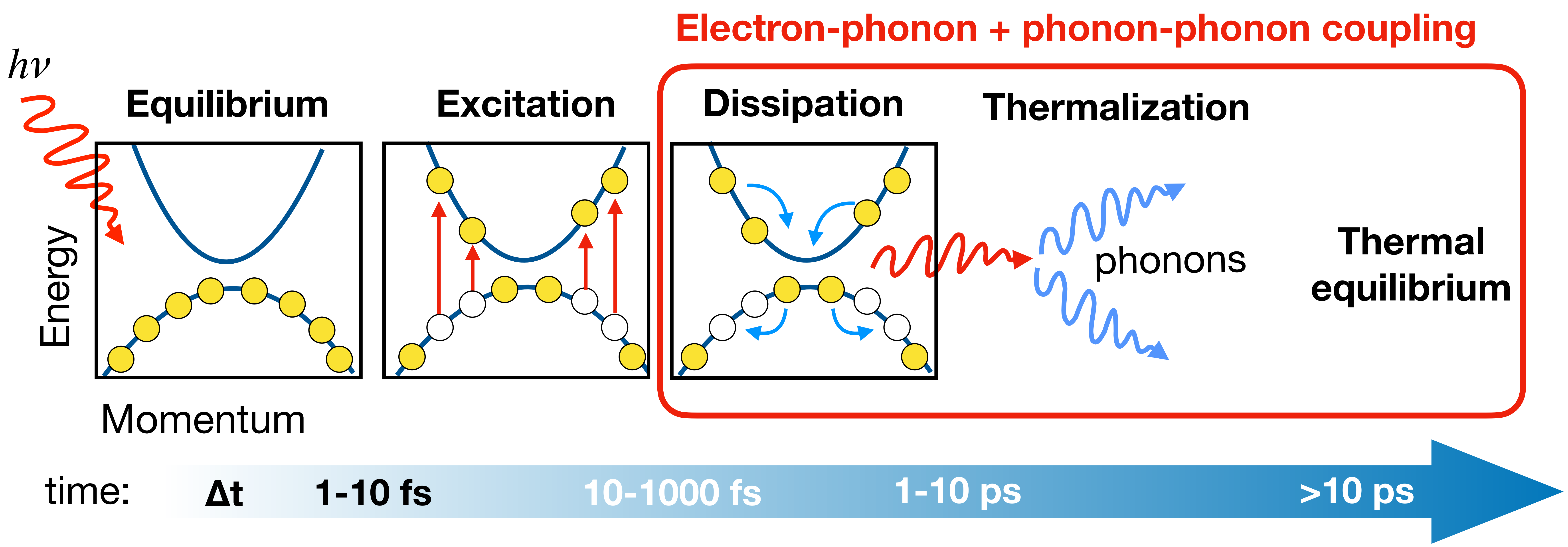
C S^2 T Computational Solid-State Theory group

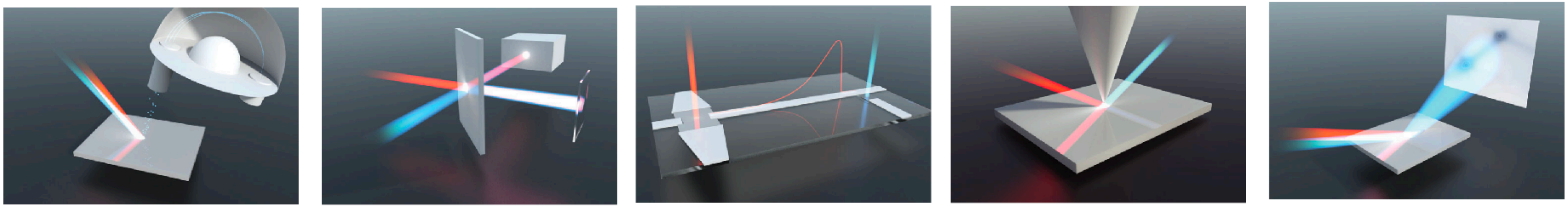
C A U Christian-Albrechts-Universität zu Kiel

Funded by DFG Deutsche Forschungsgemeinschaft German Research Foundation

MARIE CURIE ACTIONS

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



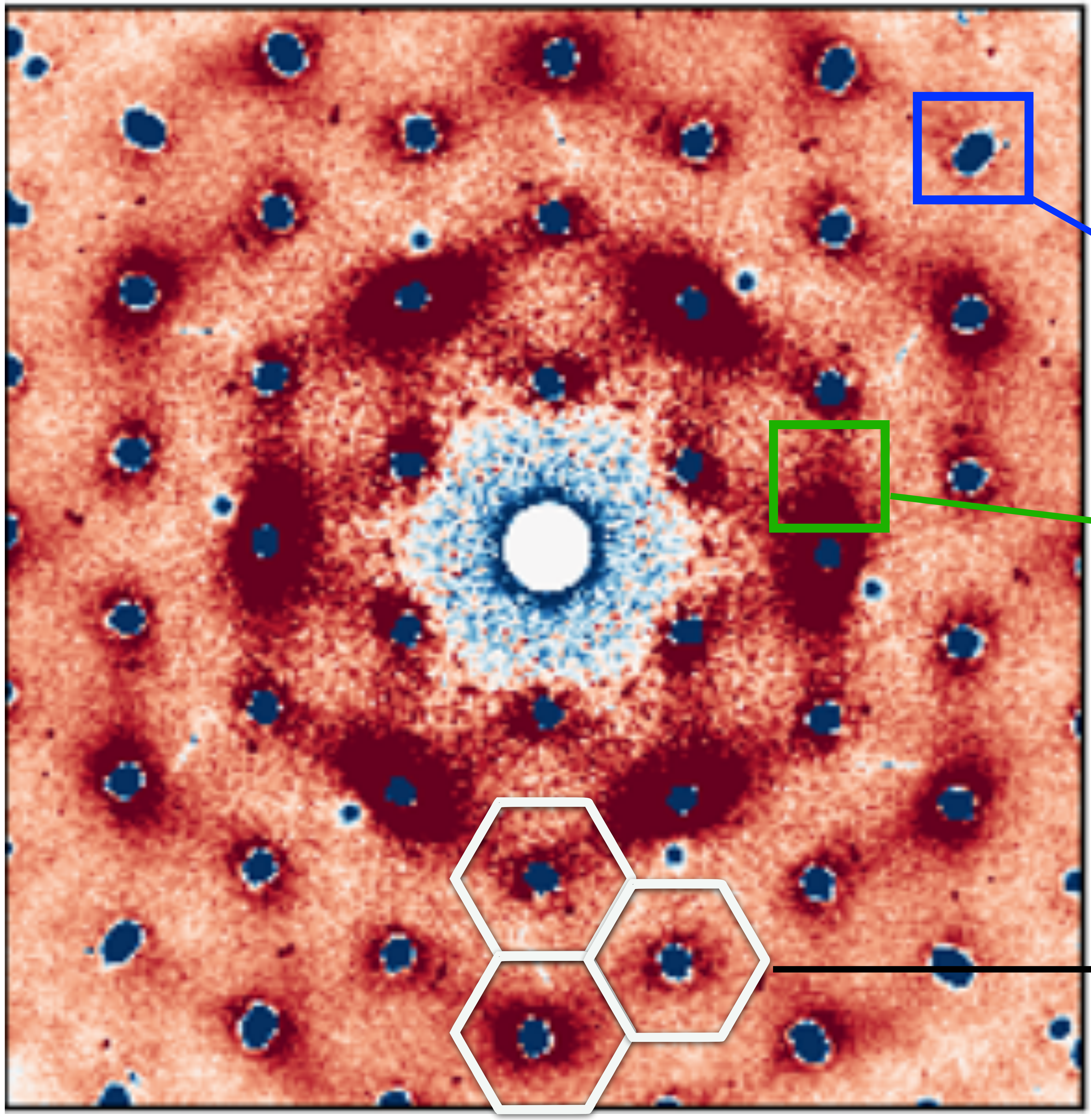


Photoemission | Optical probes | Transport | Scanning probes | Scattering probes

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

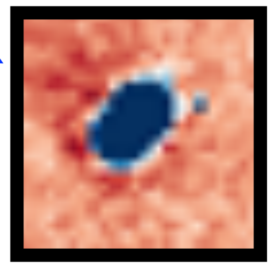
direct imaging of nuclear degrees of freedom

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



measurements: **Helene Seiler**

ultrafast electron diffuse scattering (UEDS) for bulk MoS_2

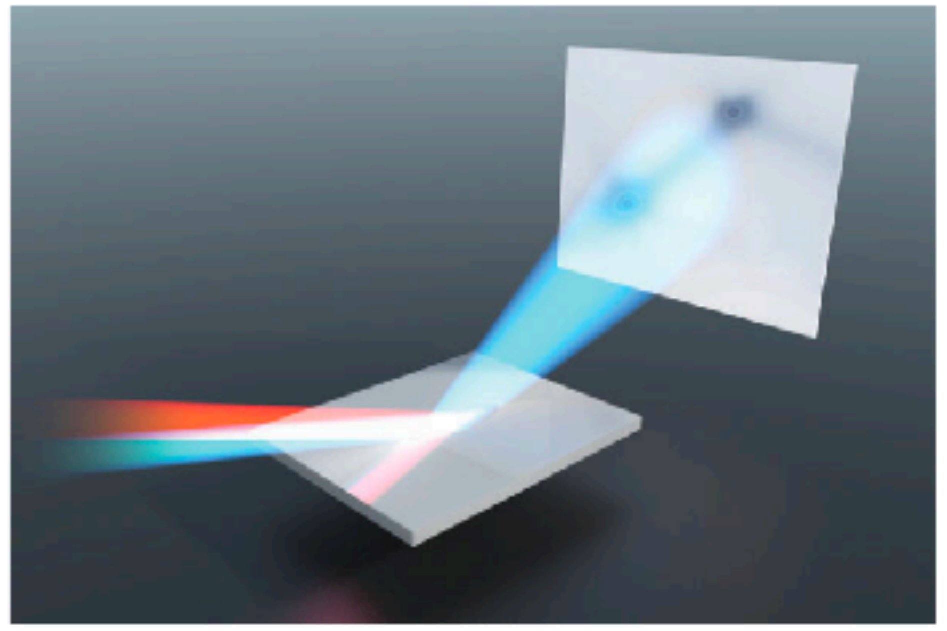


elastic scattering (Bragg peaks)

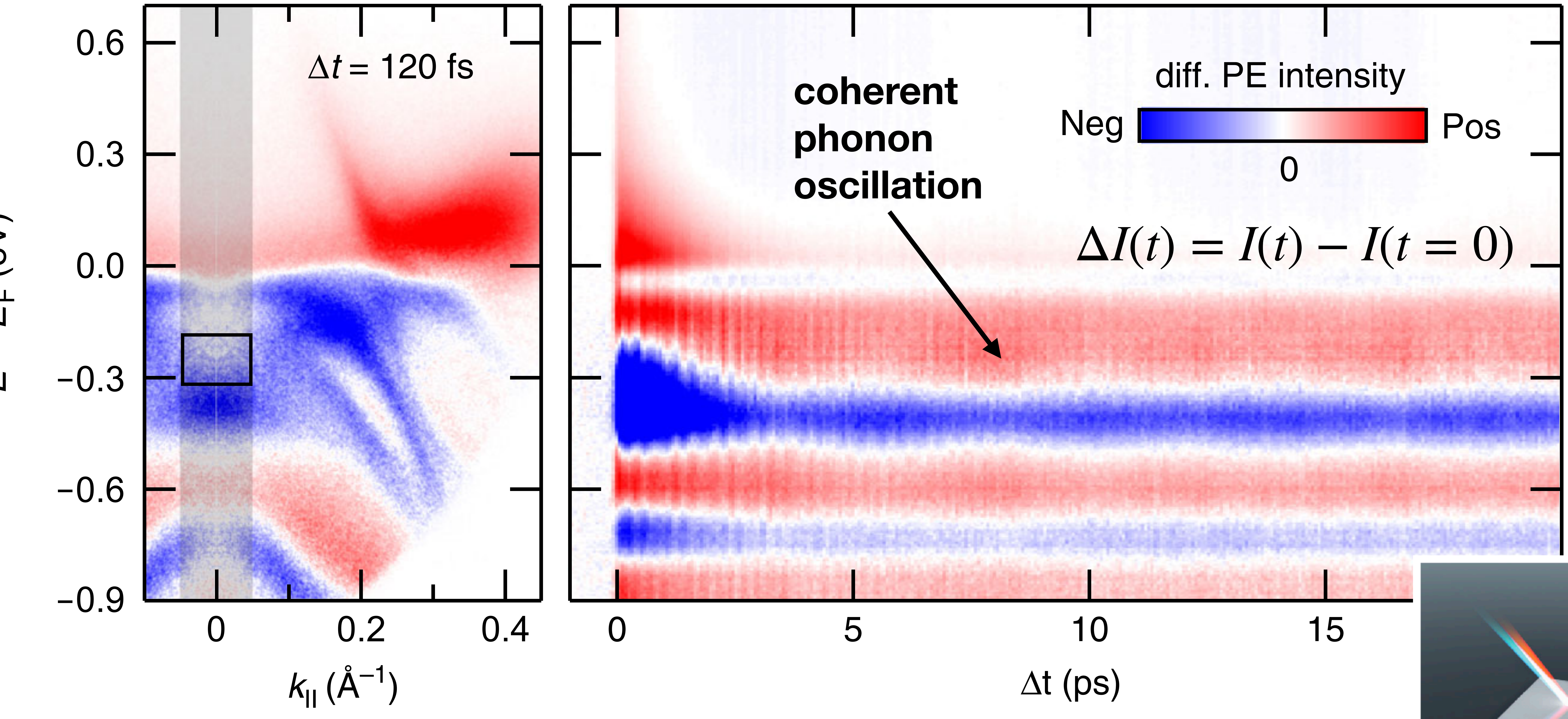
diffuse scattering (phonon-assisted electron scattering

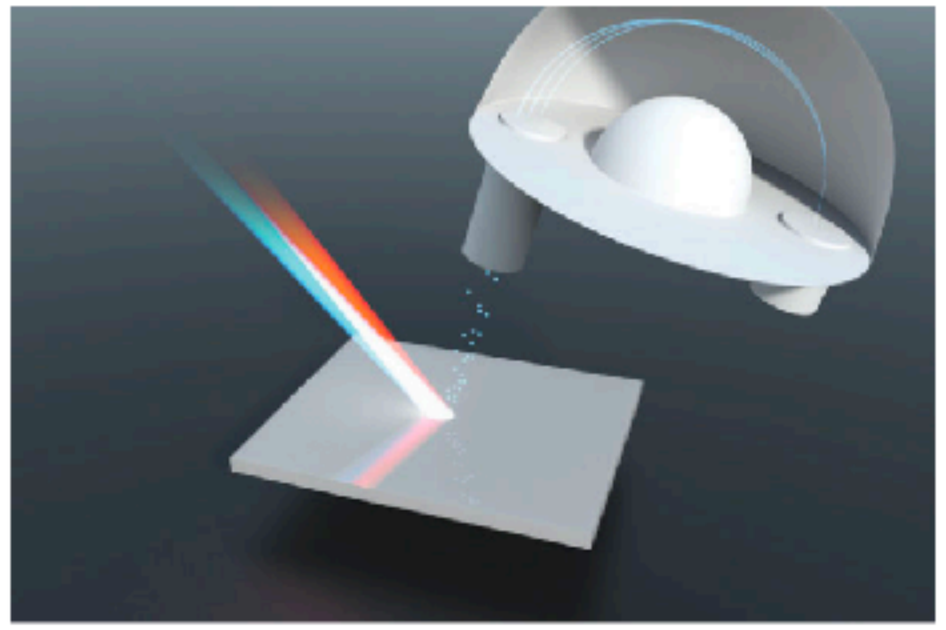
- thermal (temperature increase)
- non-thermal (phonons out of equilibrium)

Brillouin zone



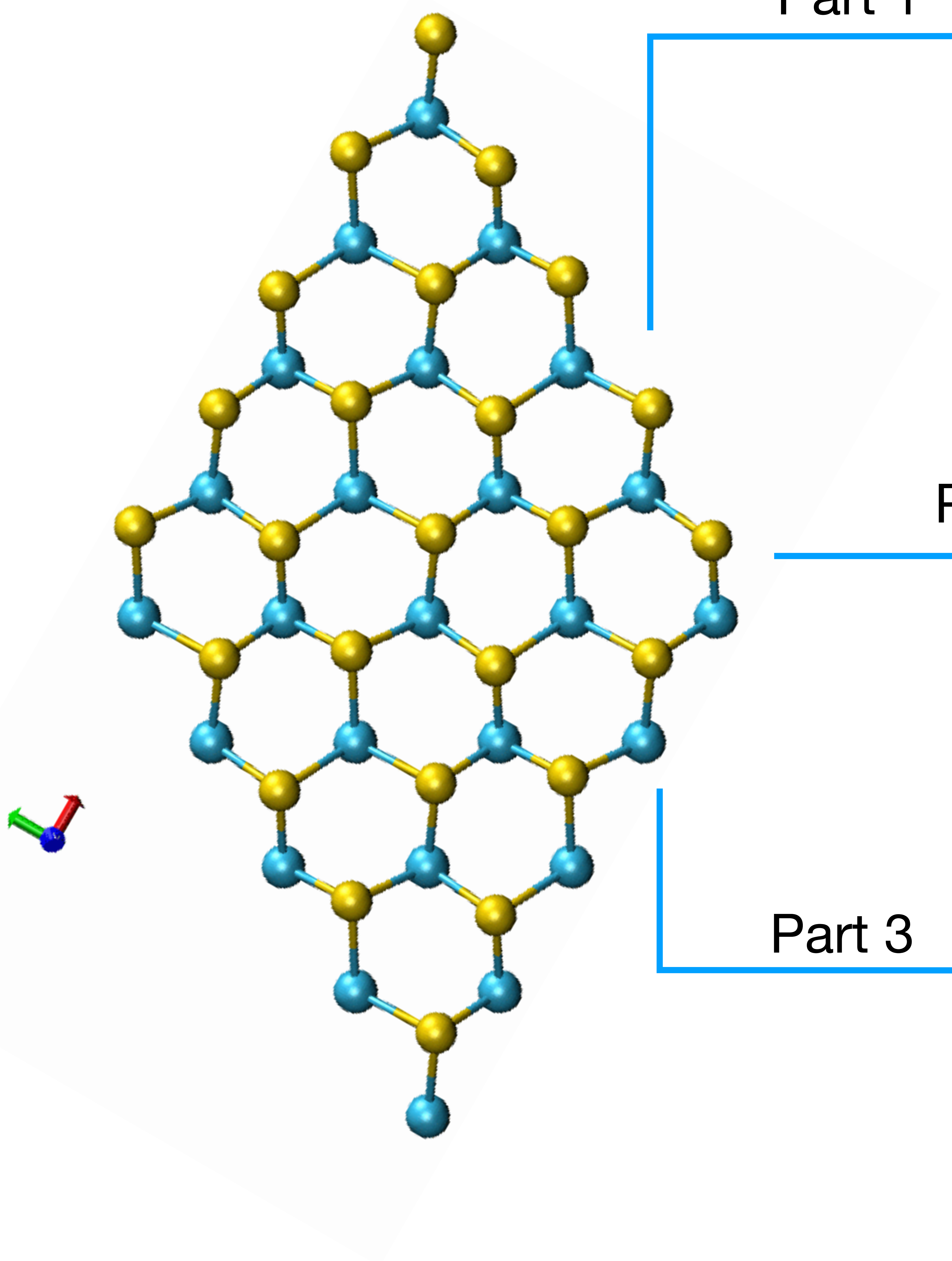
time- and angle-resolved photoemission spectroscopy (trARPES) for bulk WTe_2





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

In this talk



Part 1

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

Part 2

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

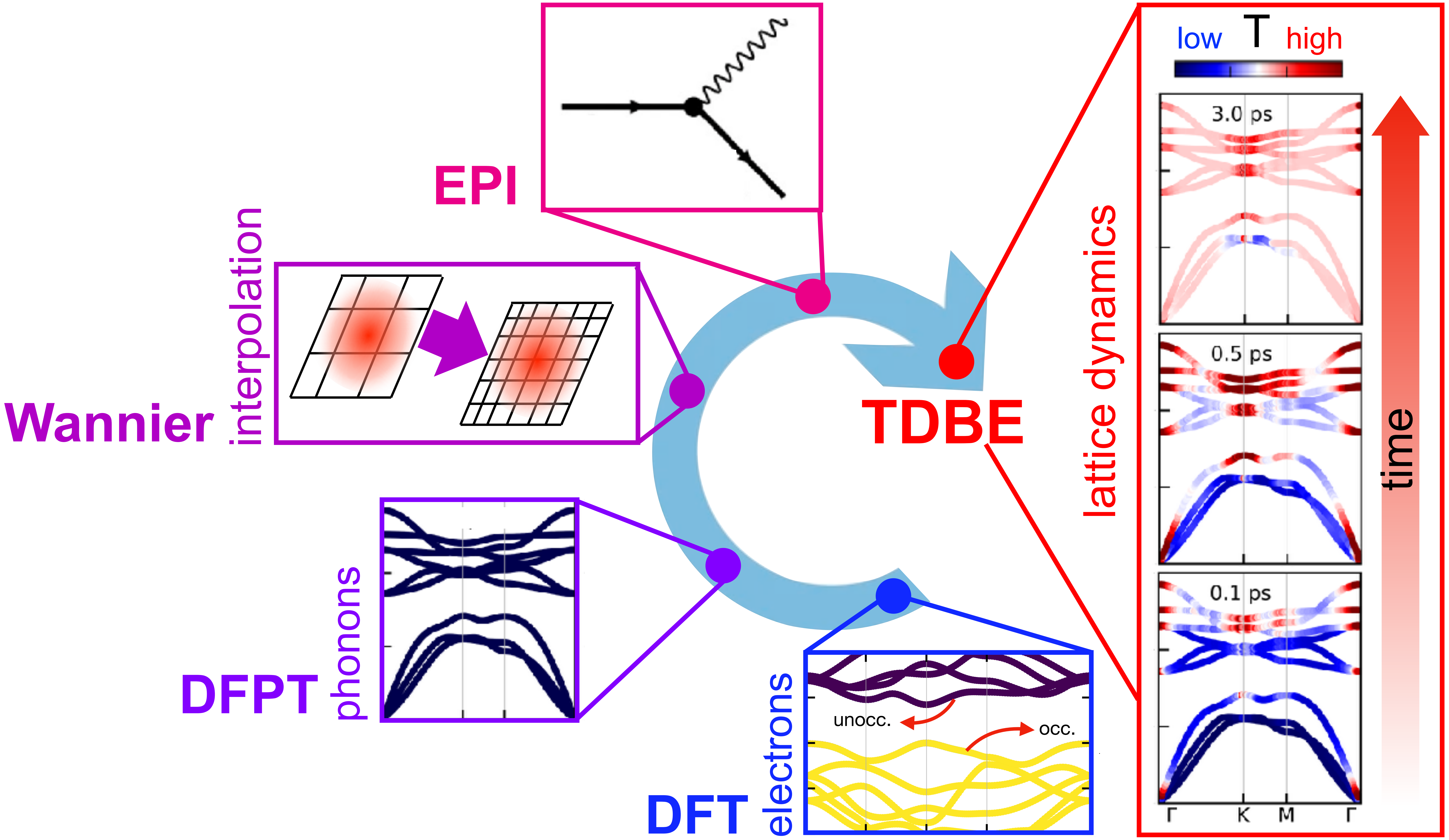
Part 3

Coherent phonons and quasiparticle renormalization in semimetals from first principles

Part 1

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

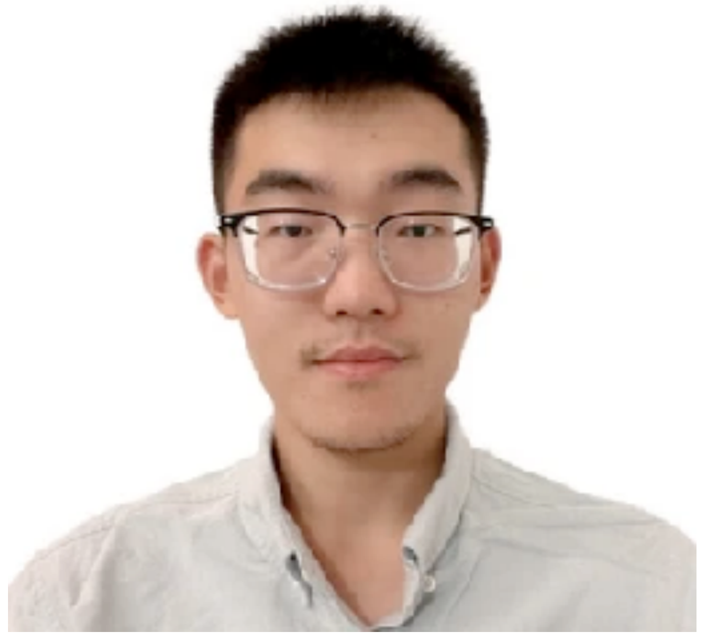
Ultrafast dynamics simulations from first principles



TDBE: Time-dependent Boltzmann equation

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

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



Y. Pan

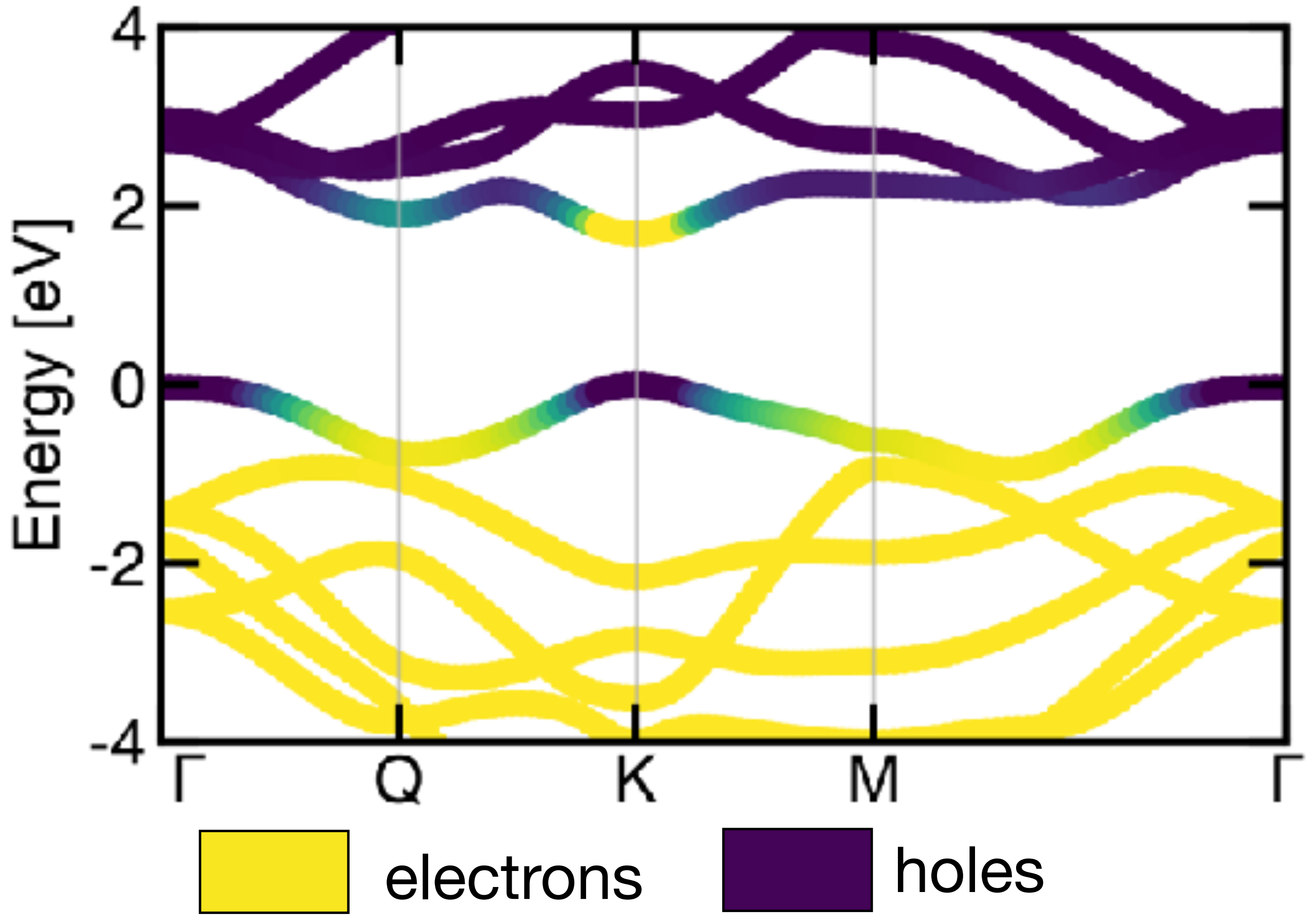
Rethfeld, Kaiser et al., Phys. Rev. B 65, 214303 (2002)
Bernardi, Louie et al., PNAS (2015)
FC, J. Phys. Chem. Lett. (2021)
Tong, Bernardi, Phys. Rev. Res. (2021)

Review: FC, Novko, Adv. Phys. X (2022)

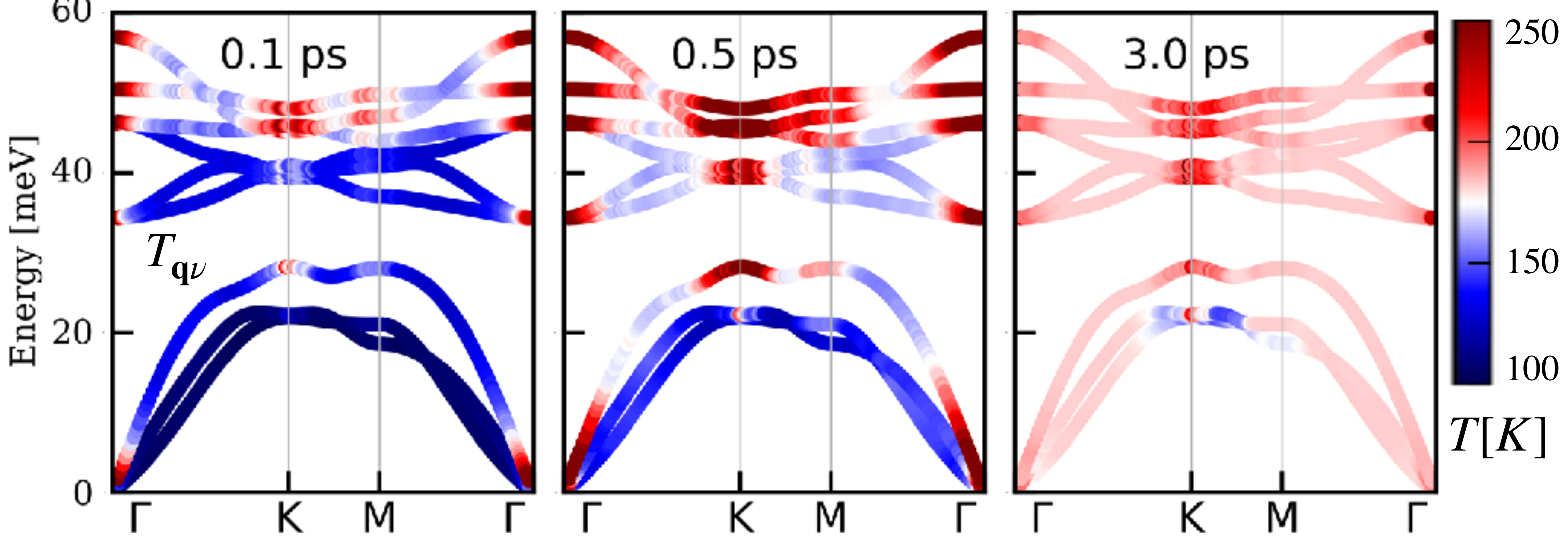
Non-equilibrium phonon dynamics from the time-dependent Boltzmann equation (TDBE)

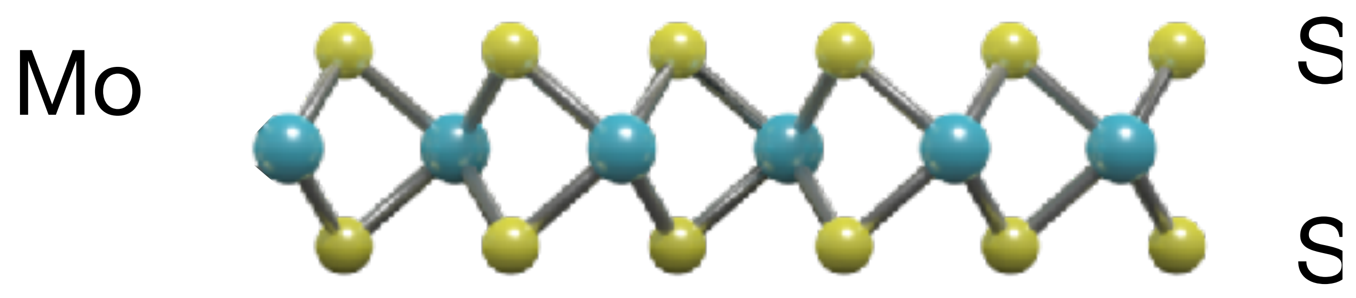
Coupled electron-phonon dynamics in monolayer MoS_2

Electronic excitation

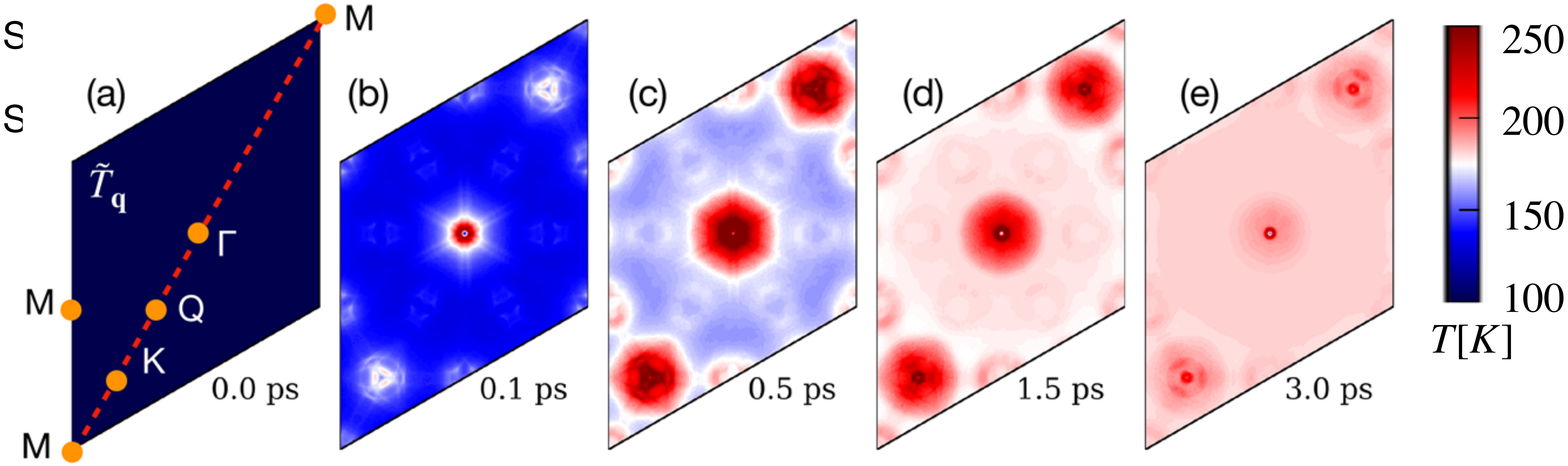


Phonon excited upon electron thermalization





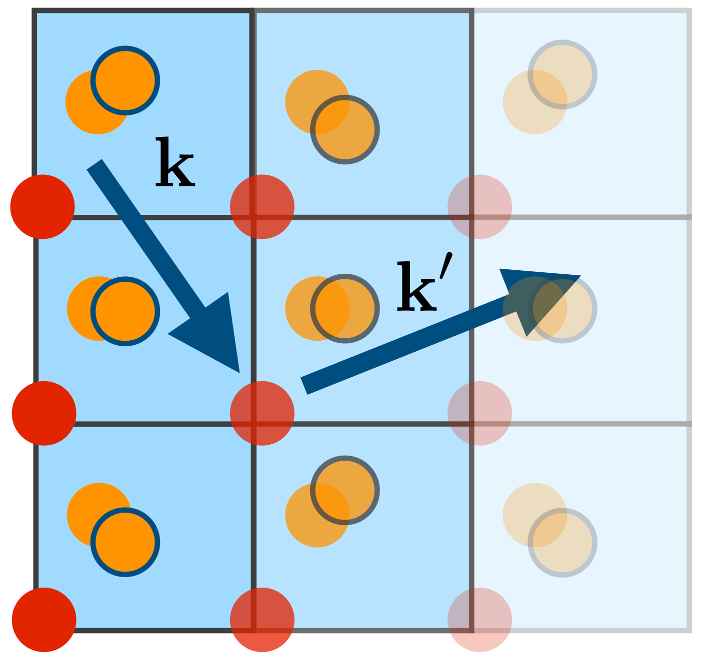
Non-equilibrium phonon population in 2D MoS_2



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

Modelling ultrafast diffuse scattering from first principles

Vibrating lattice (>1 atom per cell)



Scattering intensity of a vibrating lattice:

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

Zero-phonon → I_0; One-phonon → I_1

Zero-phonon term:

$$\langle I_0(\mathbf{S})\rangle_T = N_p^2 |f_0|^2 \exp(-2W_T)\delta_{\mathbf{S},\mathbf{G}}$$

One-phonon term:

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

phonon occupation (available from the TDBE)

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

1-phonon structure factor

\+

The TDBE:

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

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

=

ultrafast diffuse scattering from first principles

G.L. Squires, Introduction to the Theory of Thermal Neutron Scattering (Cambridge University Press)

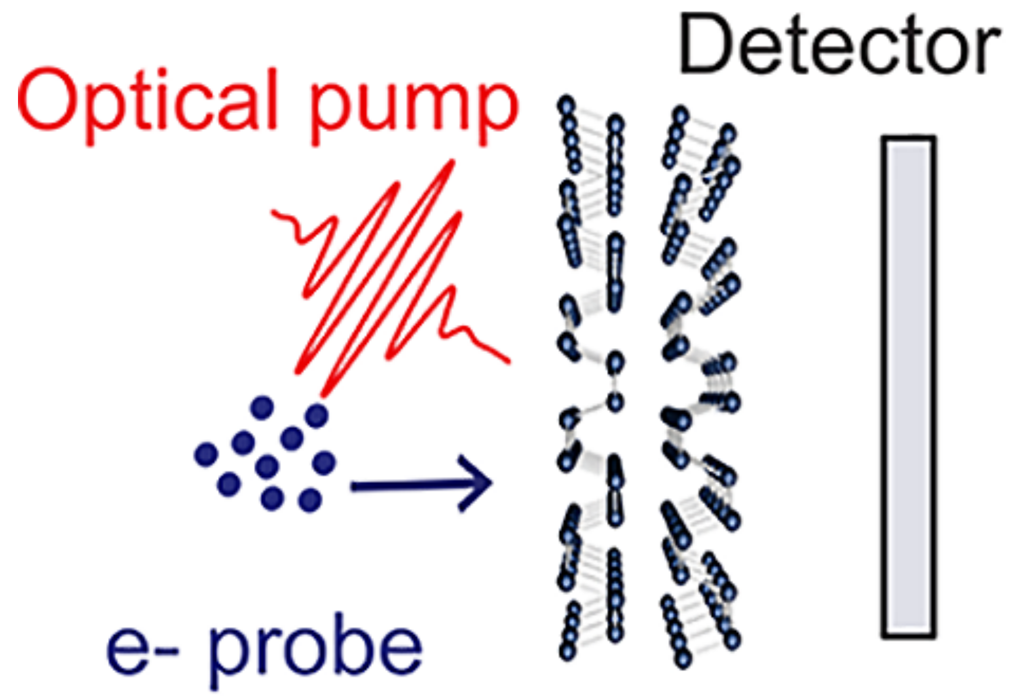
NANO LETTERS

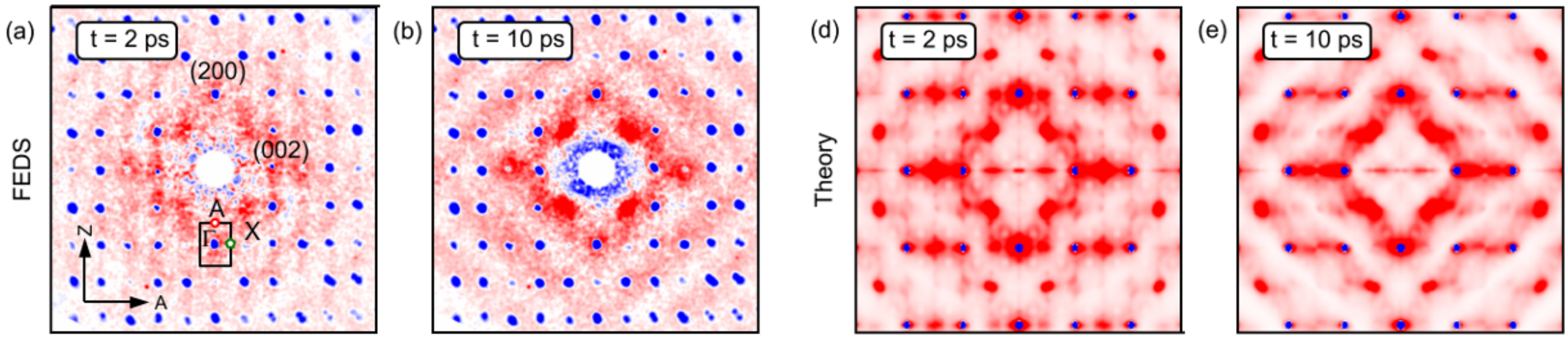
pubs.acs.org/NanoLett Letter

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*

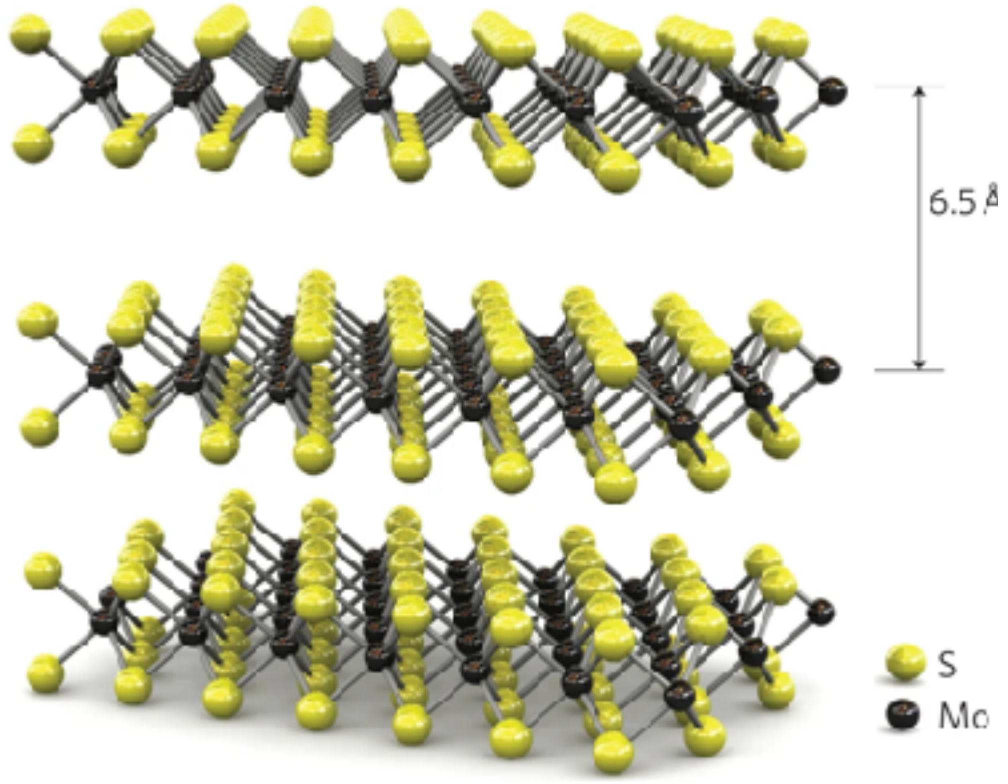
Cite This: *Nano Lett.* 2021, 21, 6171–6178 Read Online

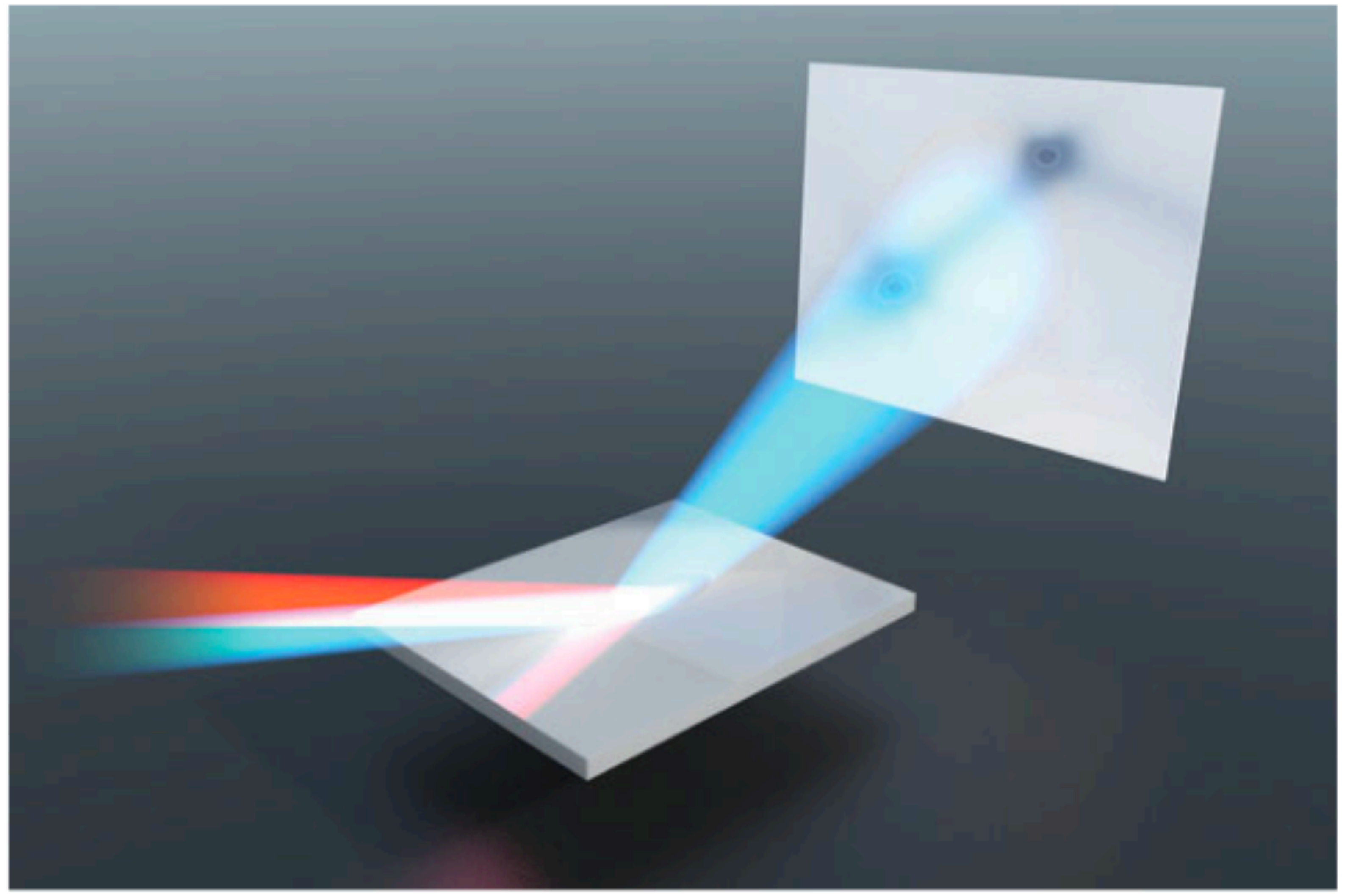


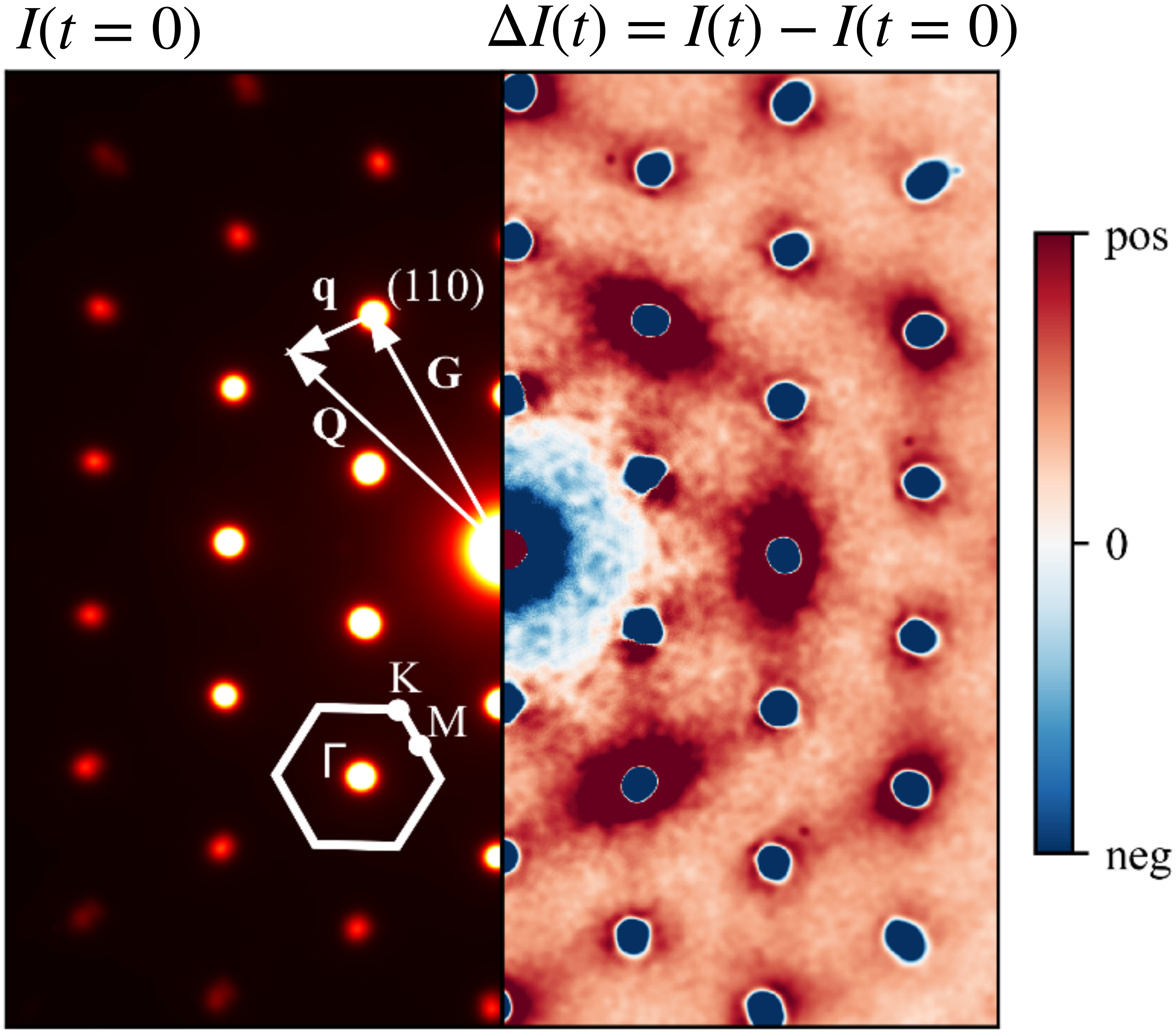


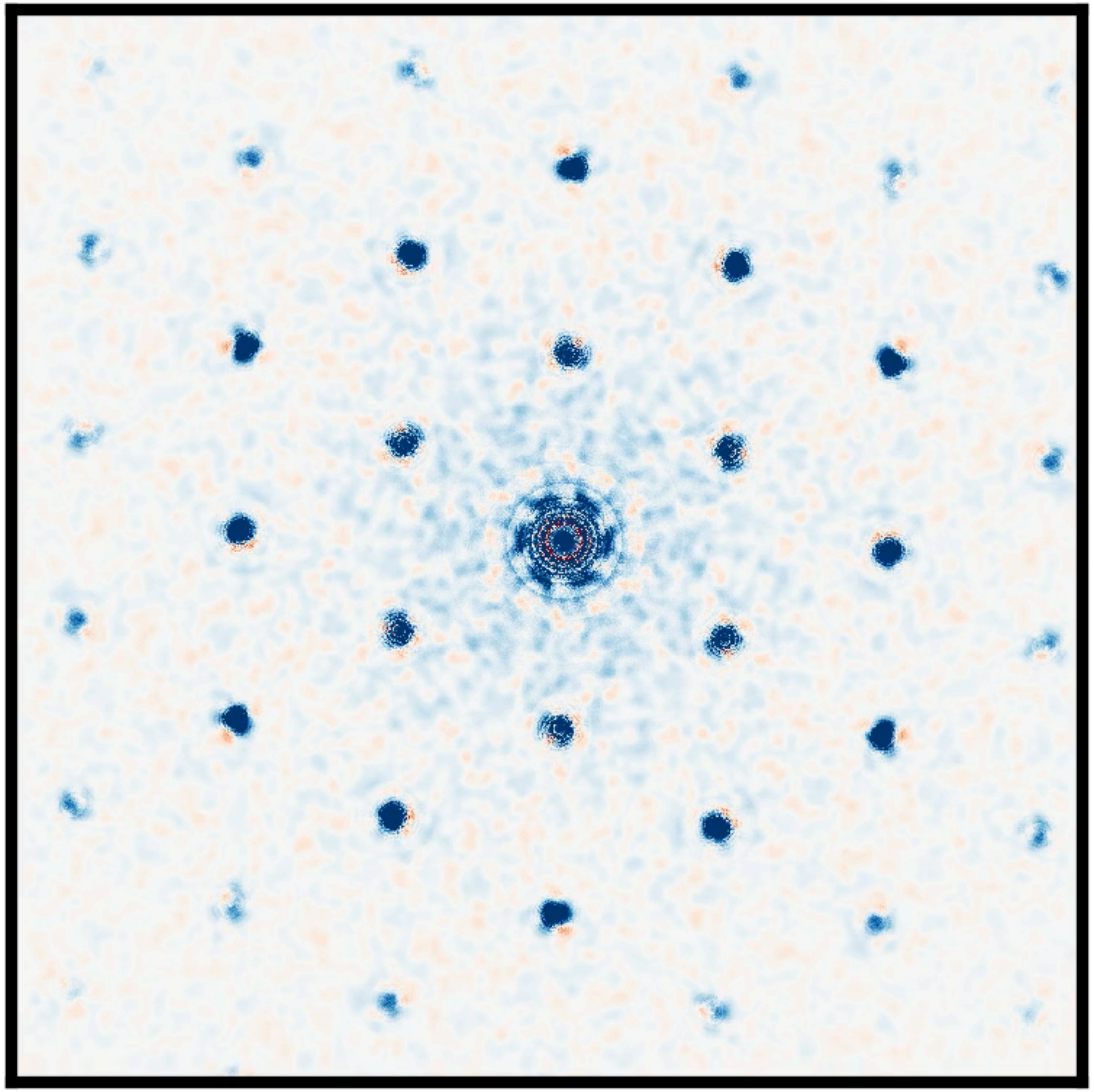
Ultrafast electron diffuse scattering: the case of bulk MoS_2

H. Seiler (FU Berlin)



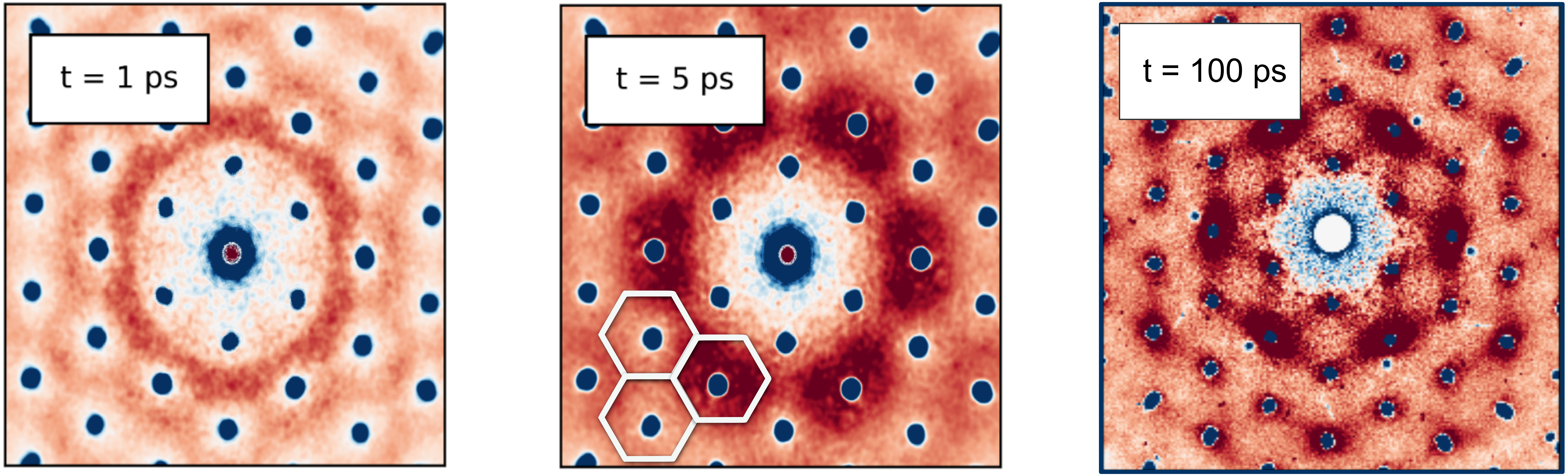






0.0 ps

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 from non-equilibrium phonon populations

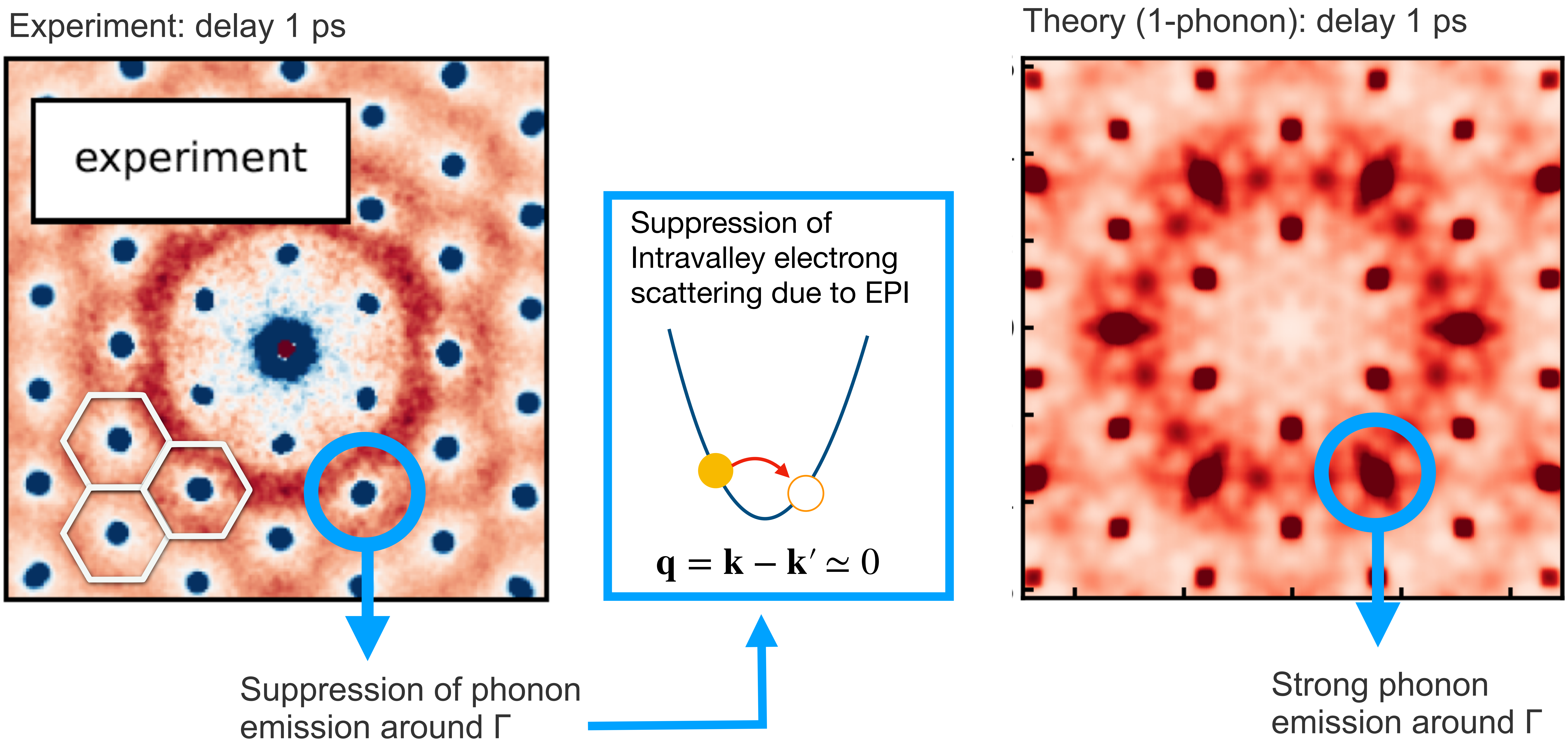
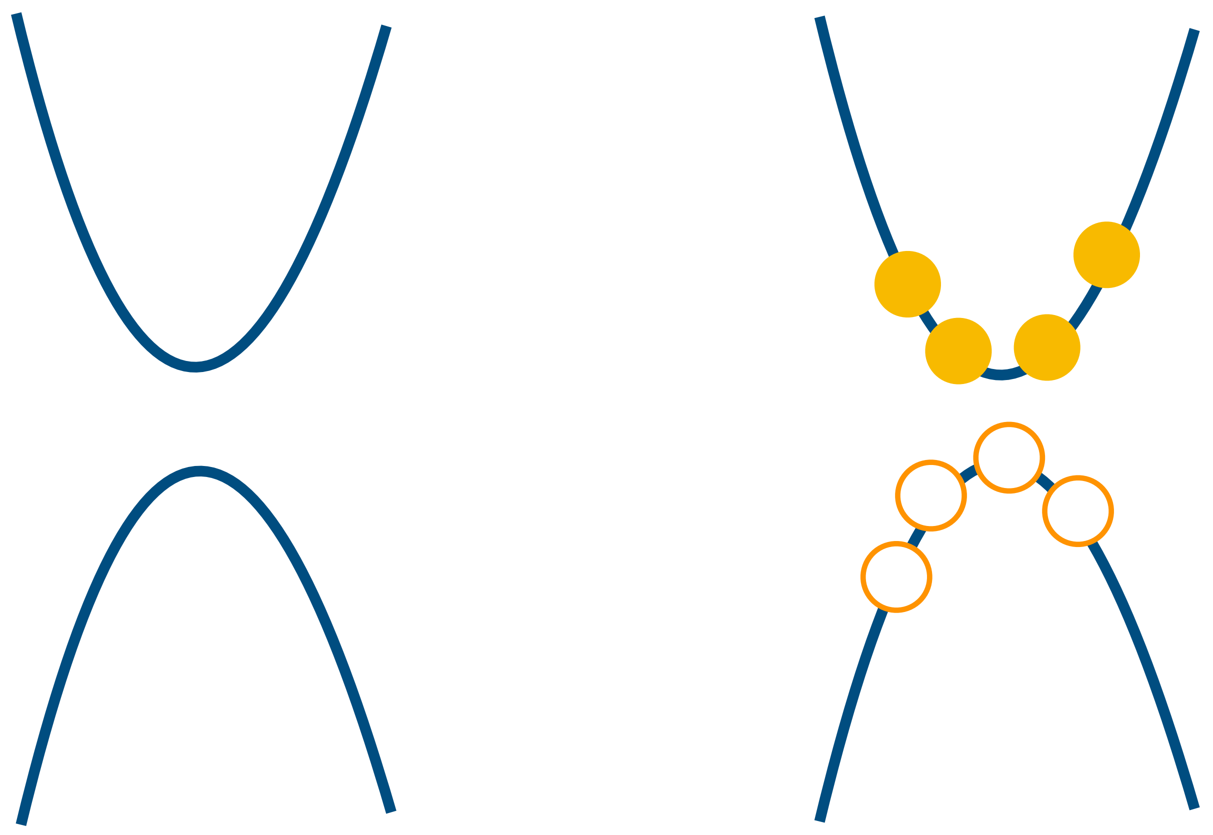


Photo-screening of the electron-phonon interaction

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



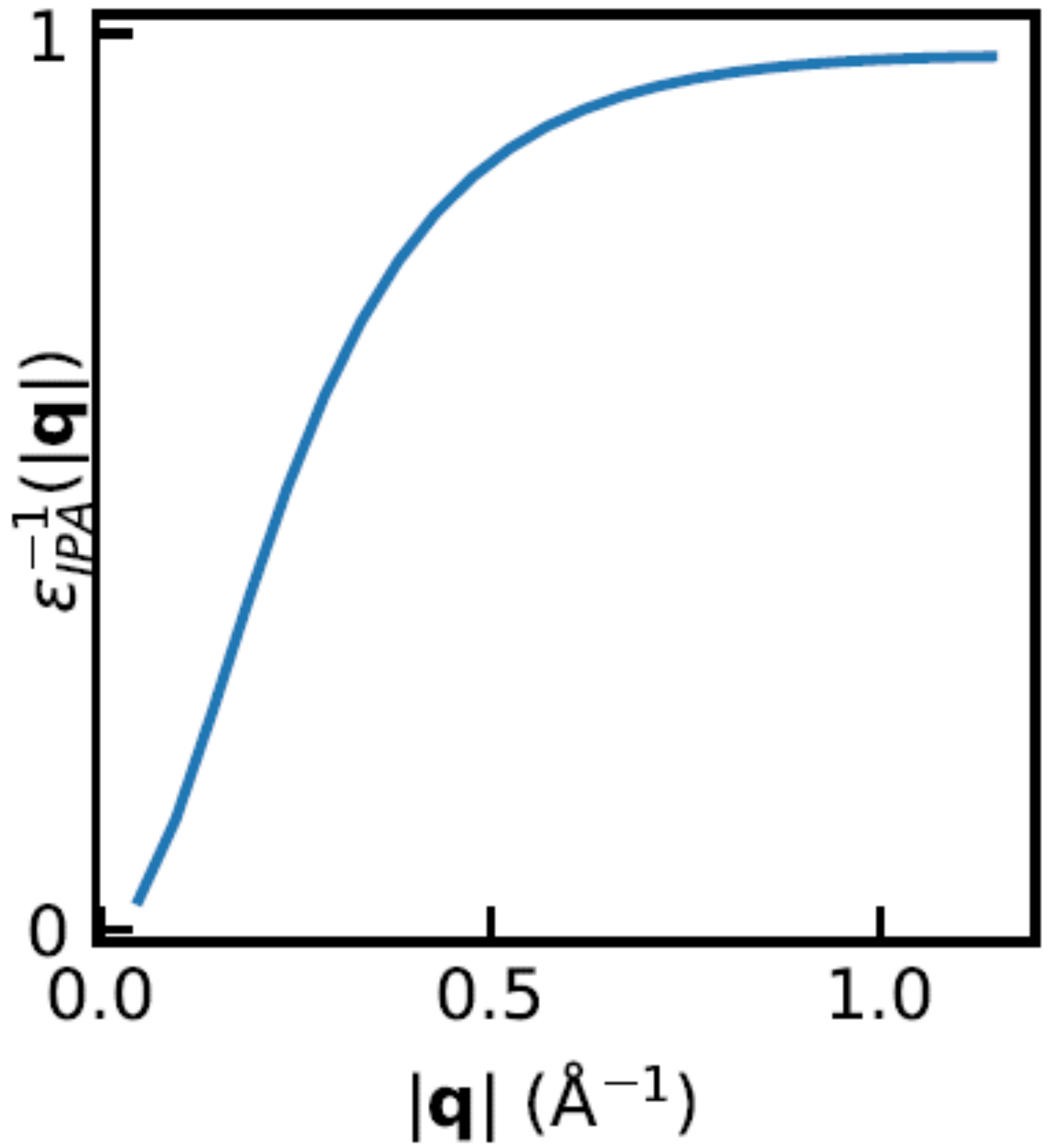
$$g^{\nu}_{mn}(\mathbf{k},\mathbf{q}) \neq \tilde{g}^{\nu}_{mn}(\mathbf{k},\mathbf{q})$$

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

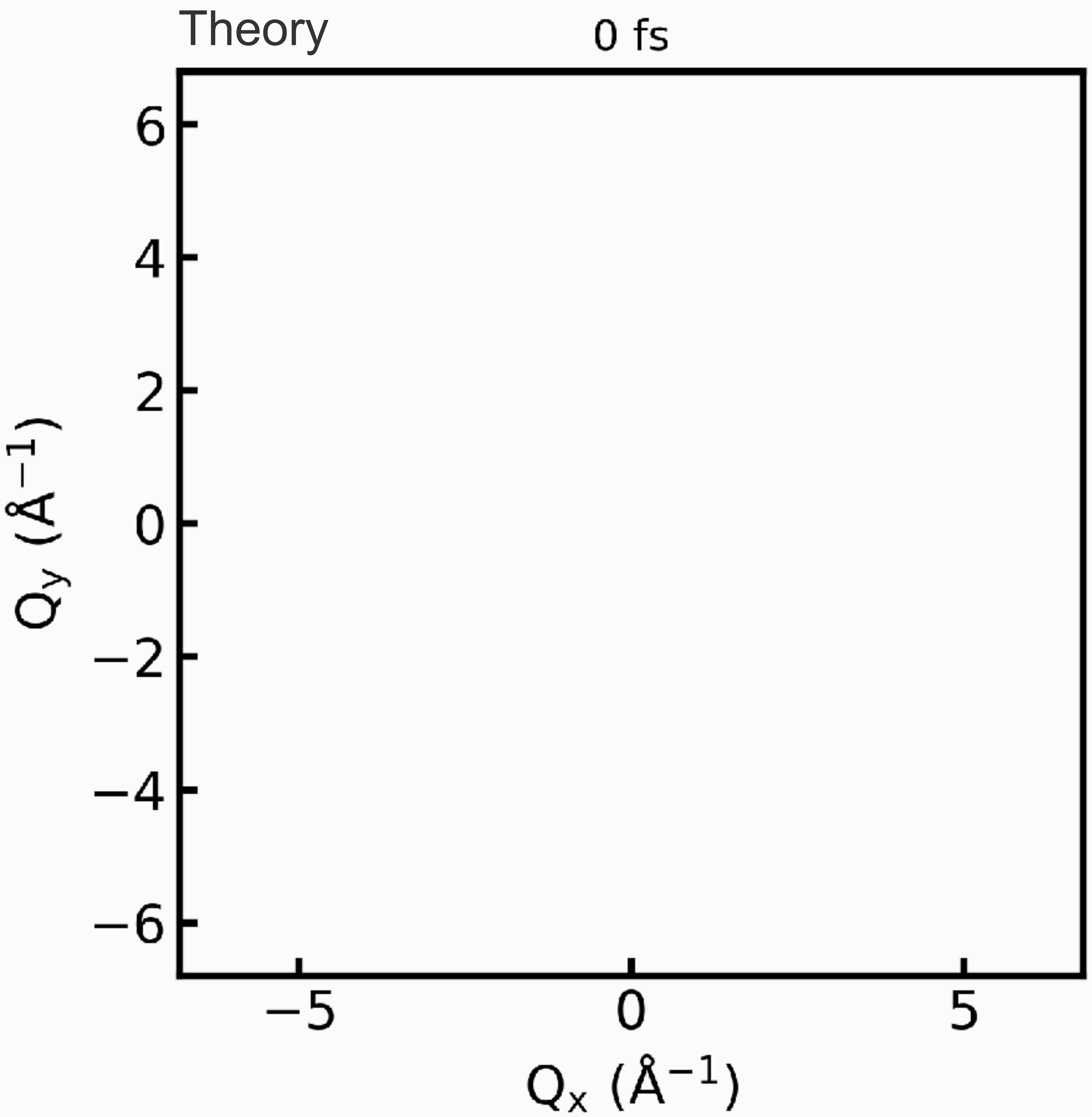
$$\delta\chi_0(\mathbf{q}) = \sum_{mm'\mathbf{k}} \frac{\delta f_{m\mathbf{k}}(t) - \delta f_{m'\mathbf{k}+\mathbf{q}}(t)}{\varepsilon_{m\mathbf{k}} - \varepsilon_{m'\mathbf{k}+\mathbf{q}}} \,|\langle u_{m'\mathbf{k}+\mathbf{q}} \,|\, u_{m\mathbf{k}} \rangle|^2$$

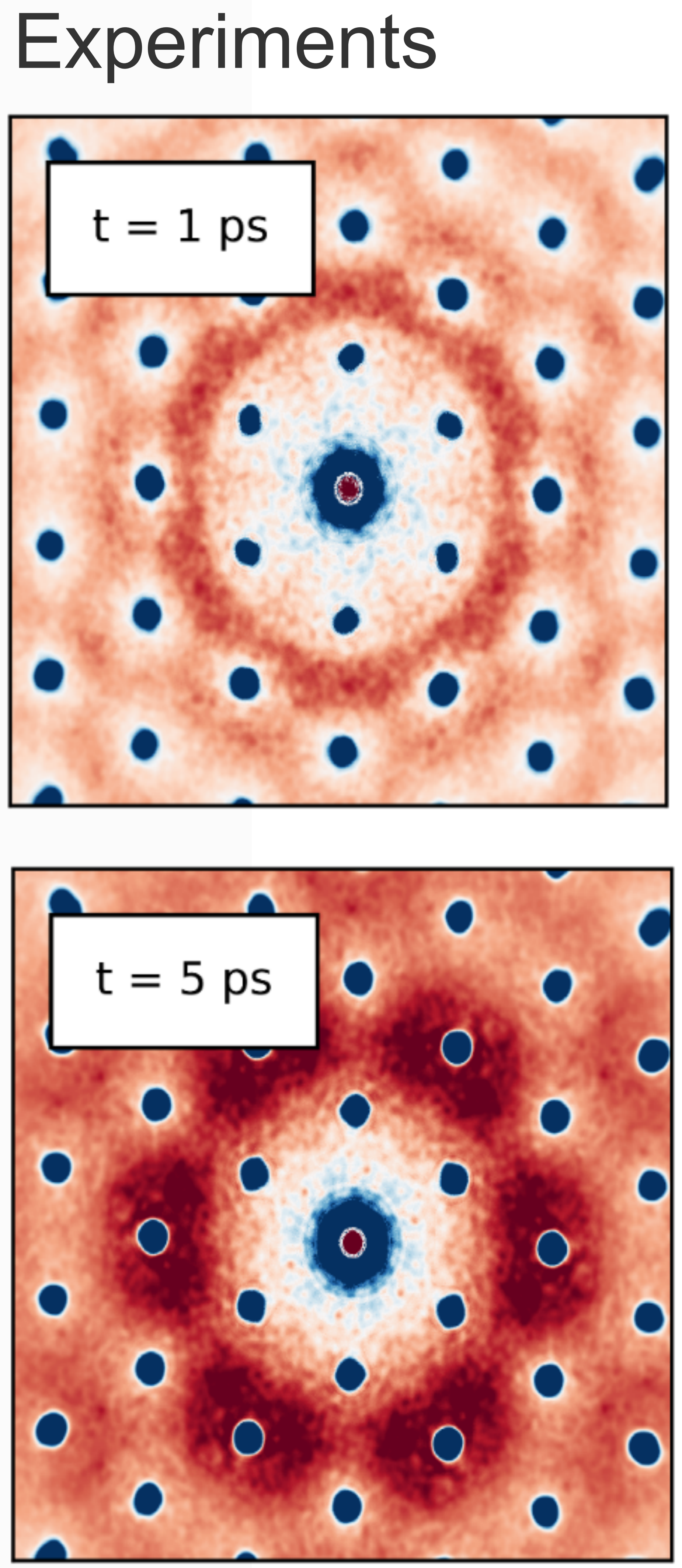
Electron-phonon matrix elements for photo-doping:

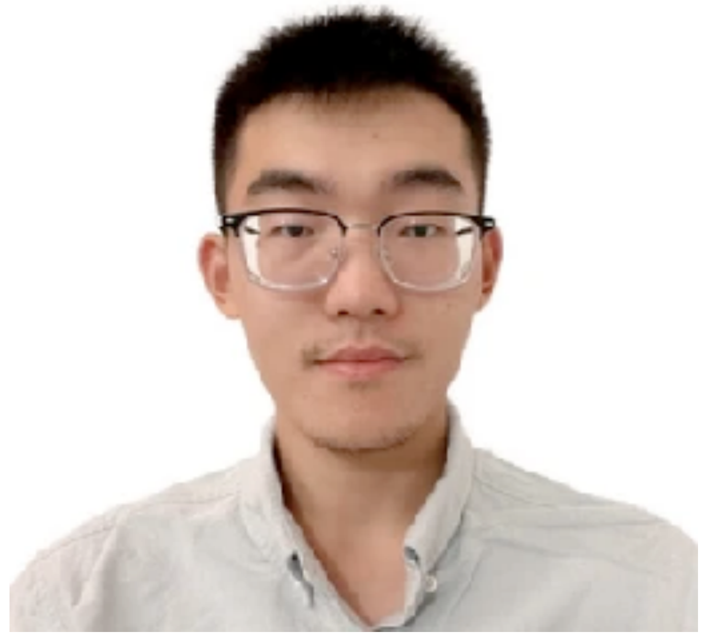
$$\tilde{g}^{\nu}_{mn}(\mathbf{k},\mathbf{q}) = \frac{g^{\nu}_{mn}(\mathbf{k},\mathbf{q})}{1 - \dfrac{4\pi e^2}{|\mathbf{q}|^2 \epsilon_{\mathrm{undoped}}}\delta\chi_0(\mathbf{q})}$$



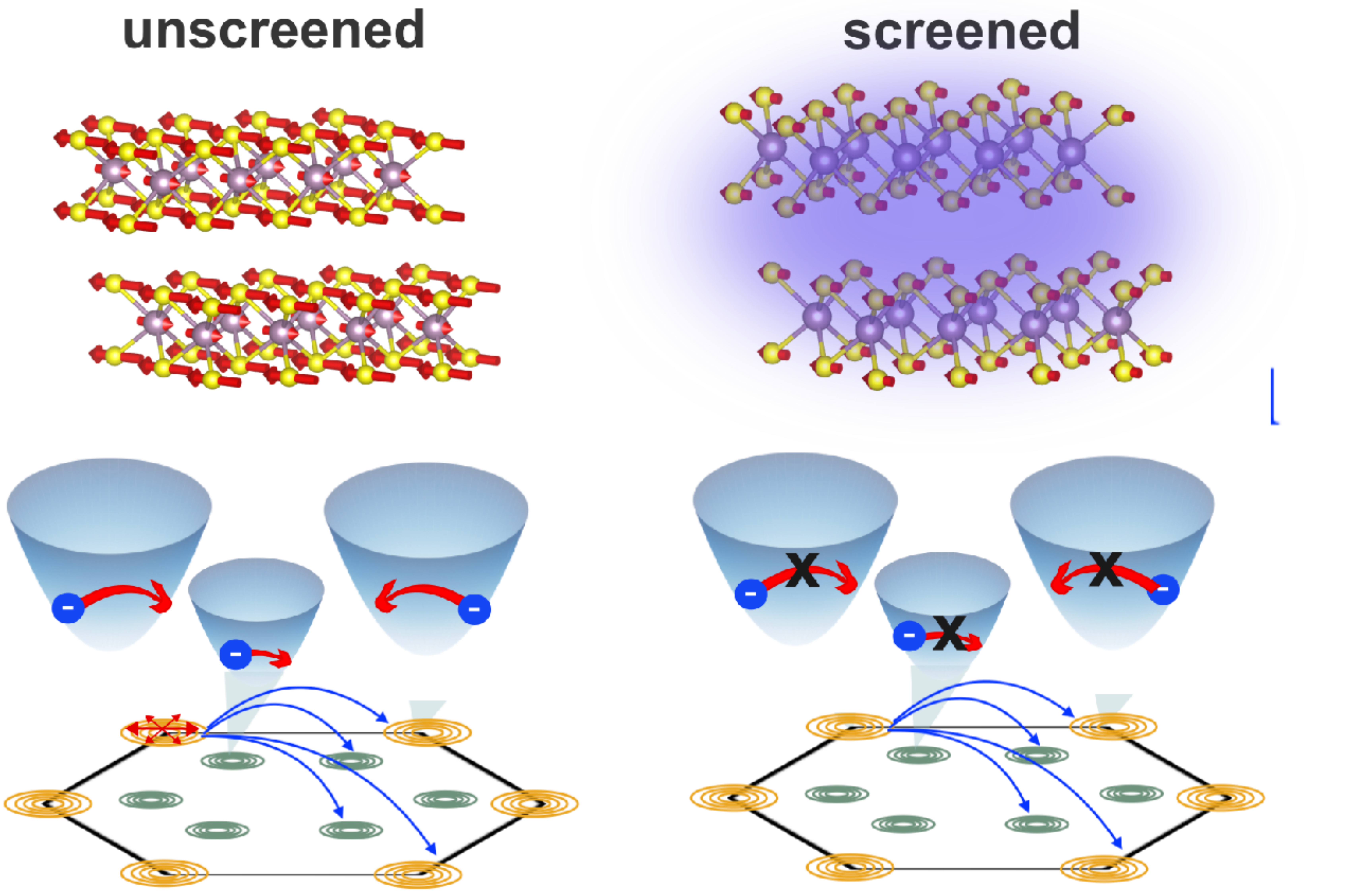
Ultrafast diffuse scattering in MoS_2







Poster by Y. Pan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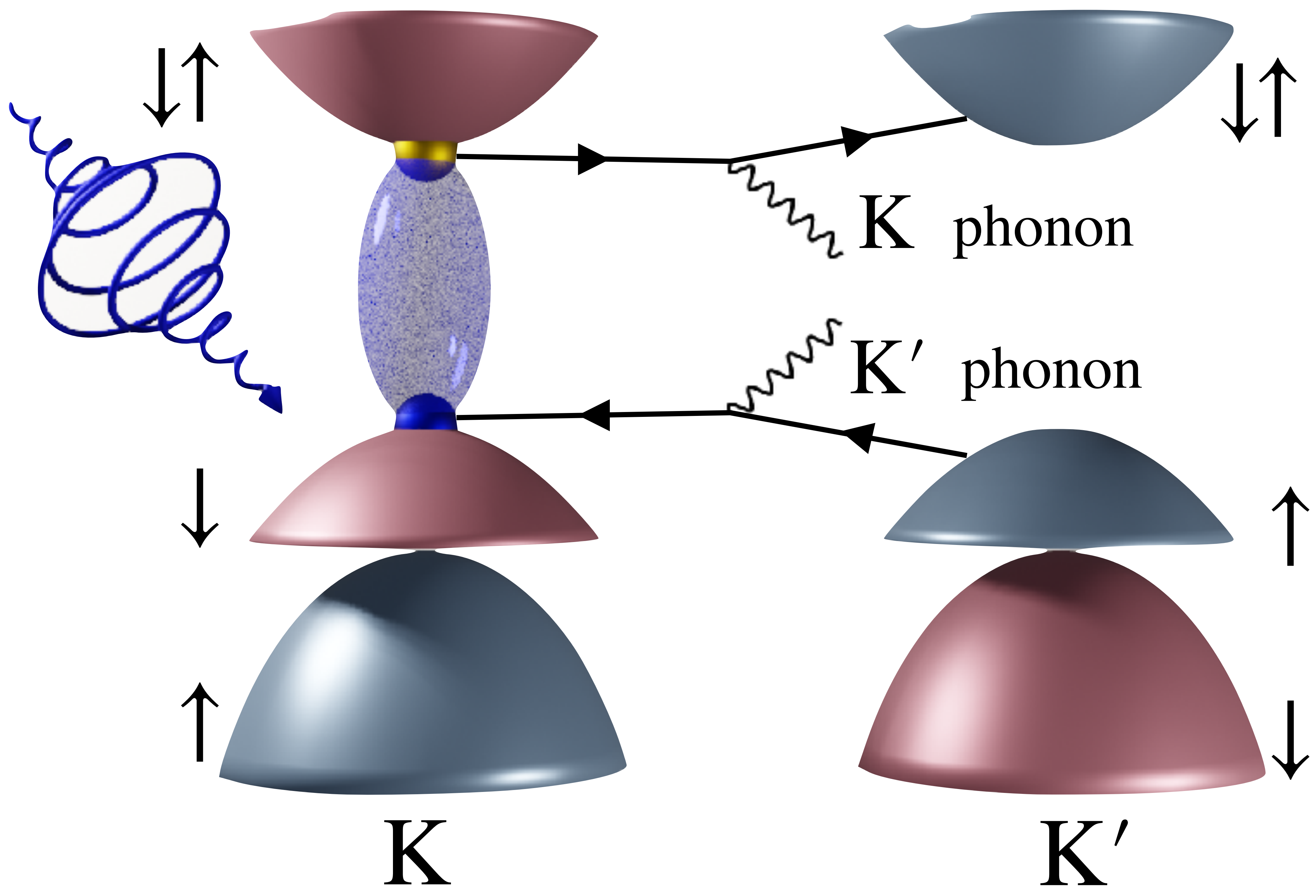
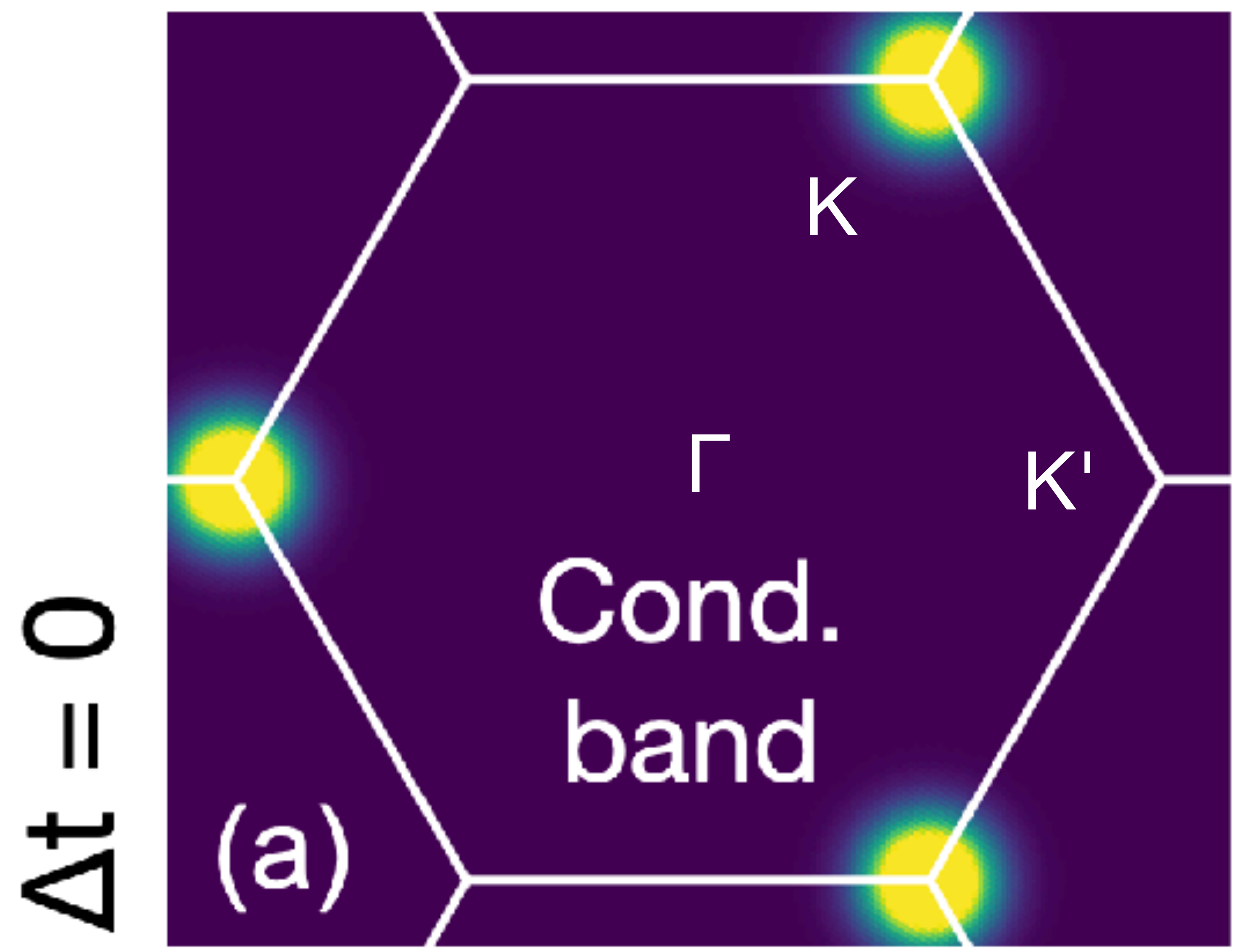
screened

Part 2

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

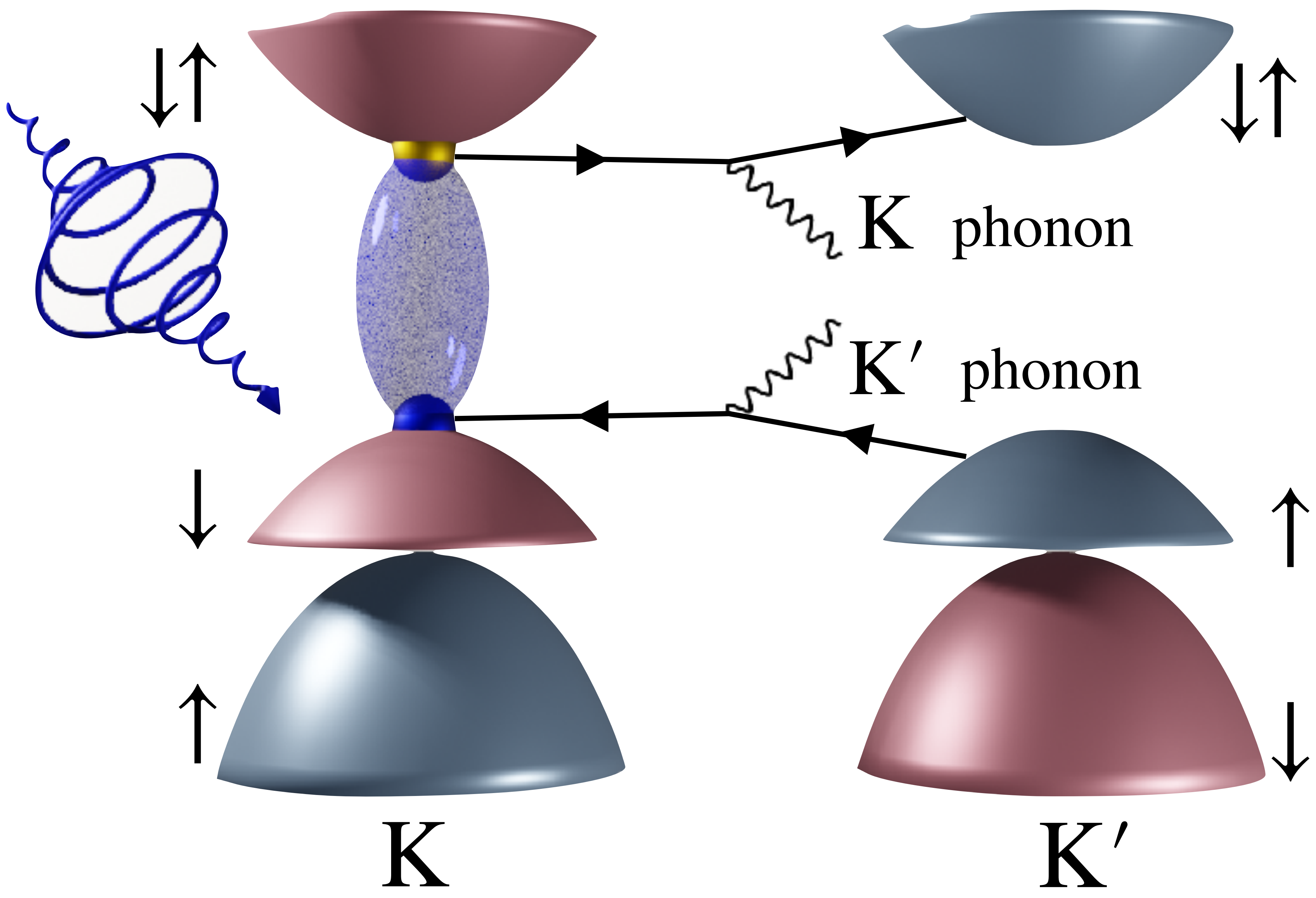
Valley selective optical excitation in MoS_2

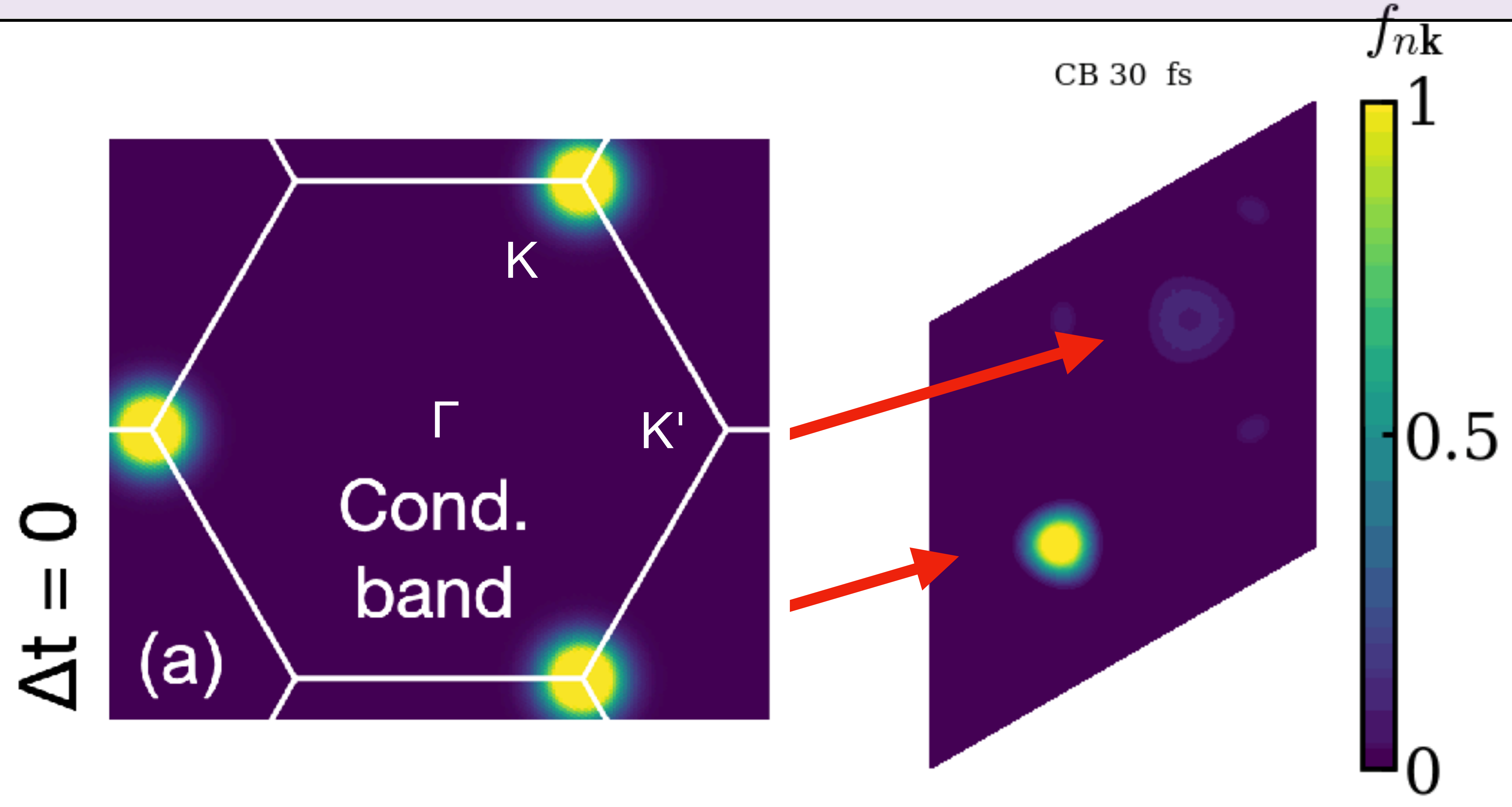




- three-fold rotational invariance
- non-centrosymmetryc crystal structure

Valley selective optical excitation in MoS_2



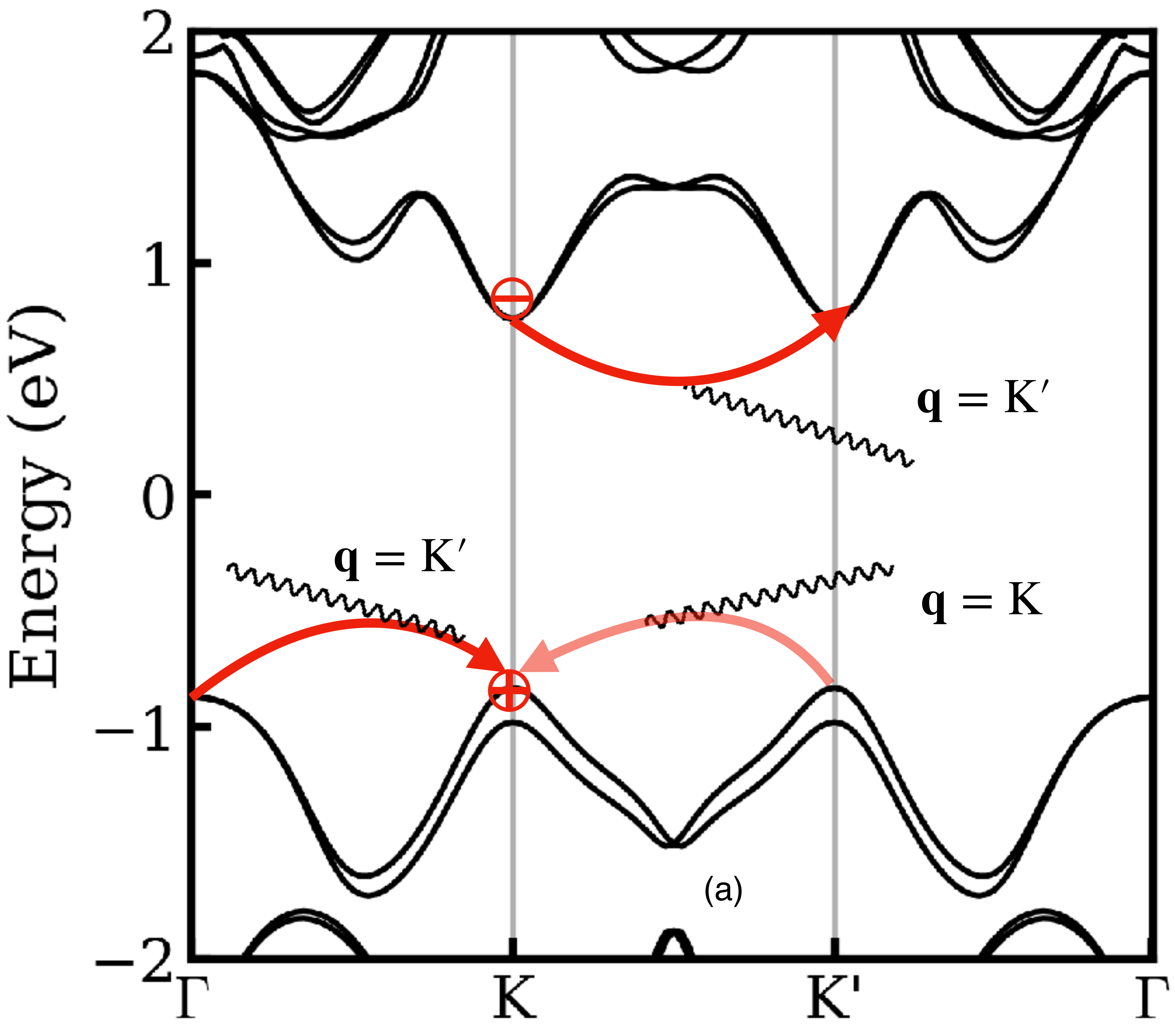


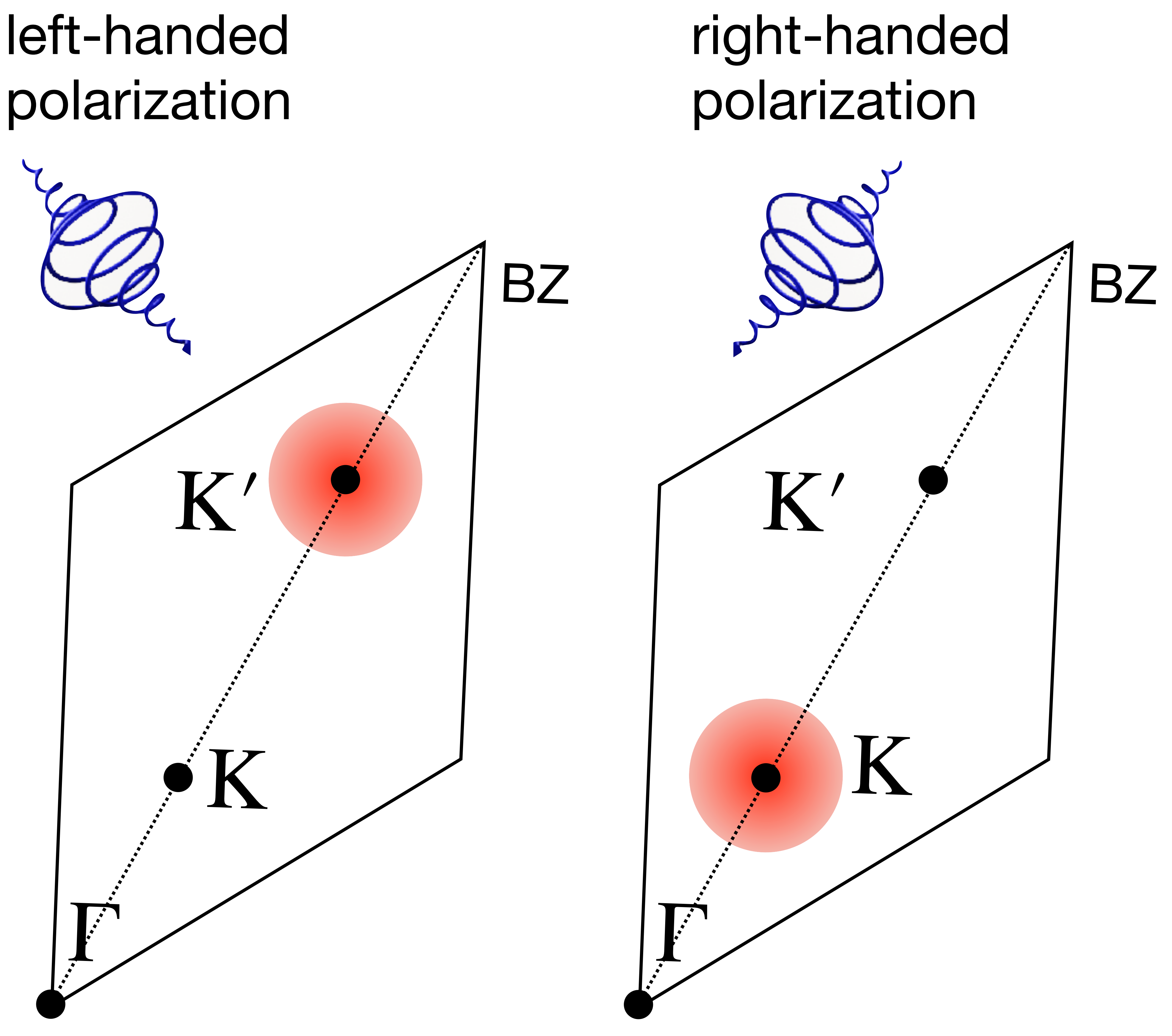
- three-fold rotational invariance
- non-centrosymmetryc crystal structure

- **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

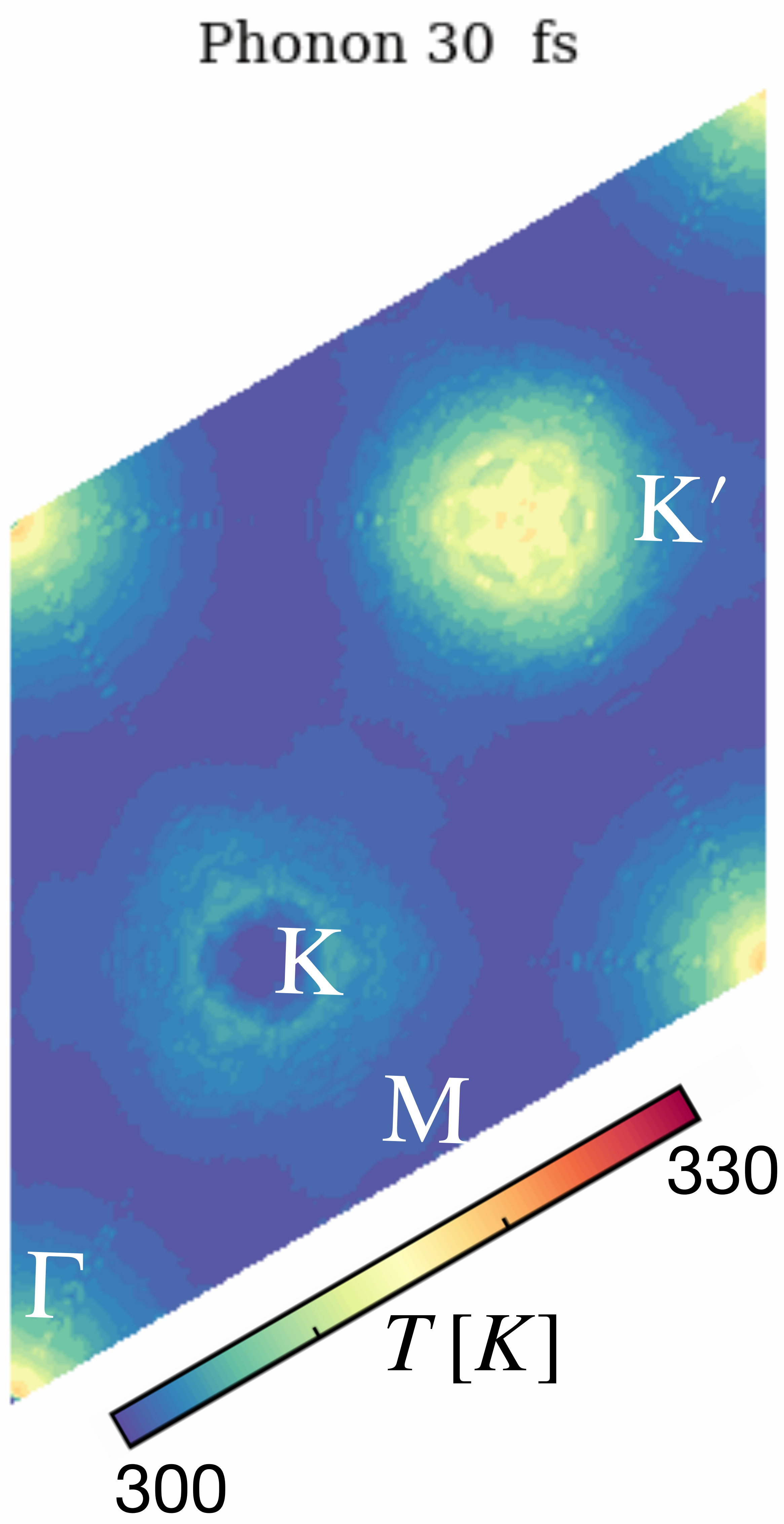




valley-polarized phonons at the K and -K high-symmetry points

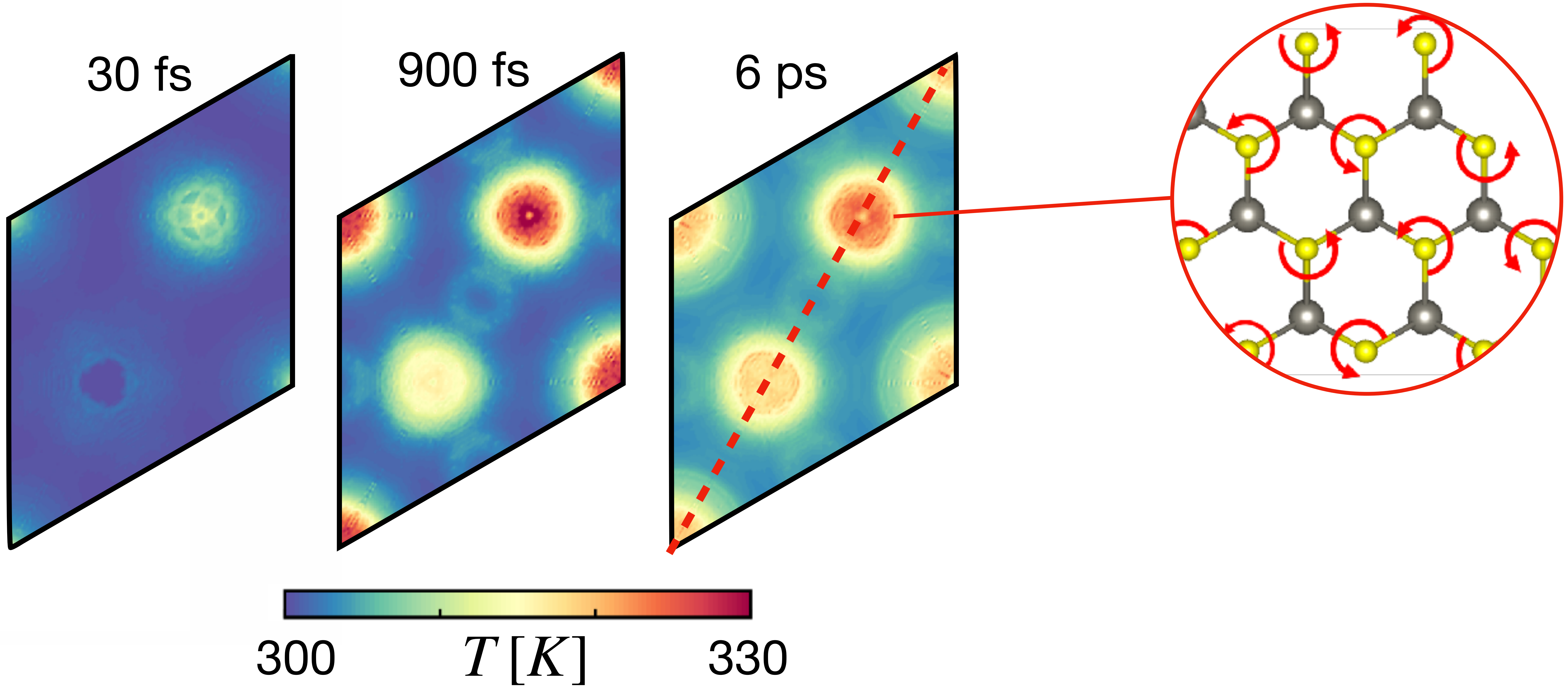
Pan, FC, Nano Lett. **23**, 7463 (2023)

Valley-polarized non-equilibrium phonon populations in MoS_2



Pan, FC, Nano Lett. **23**, 7463 (2023)

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

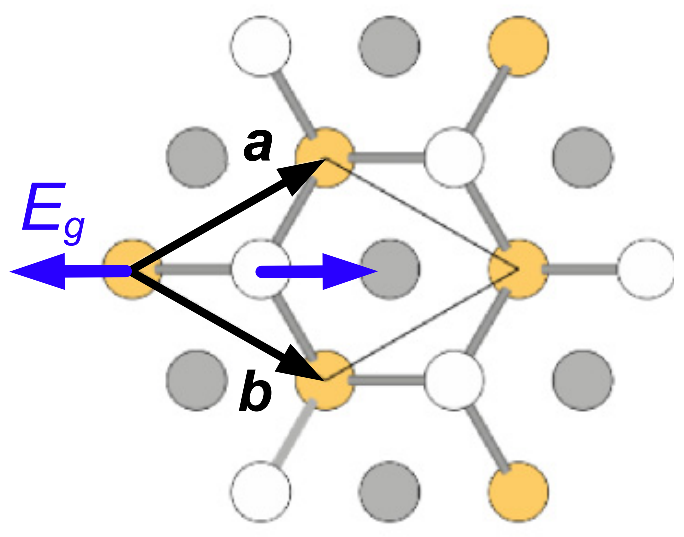


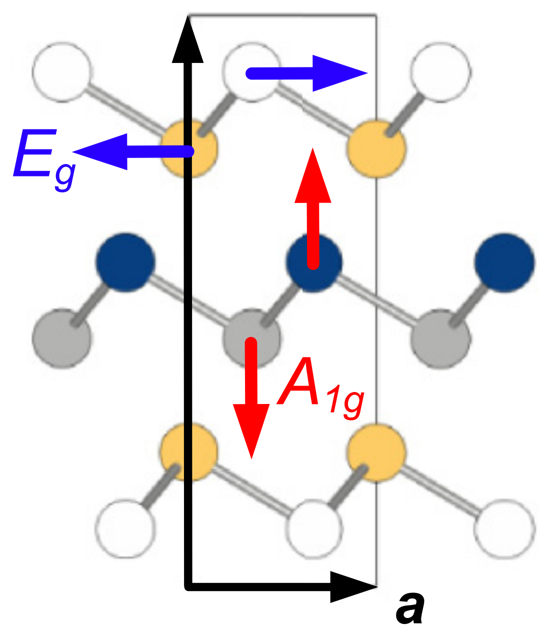
Part 2

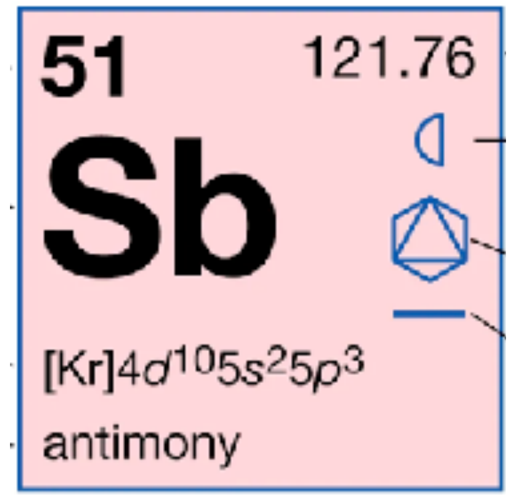
Ab-initio theory of coherent phonons in semimetals

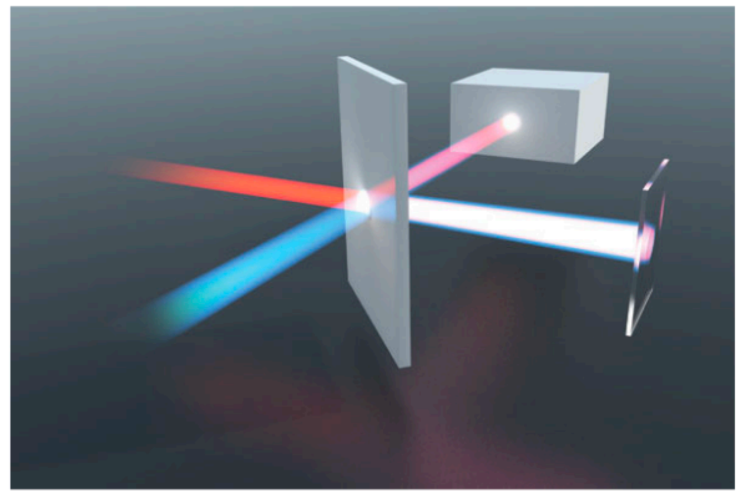
Coherent phonons in experiments: the example of antimony

Antimony (Sb)

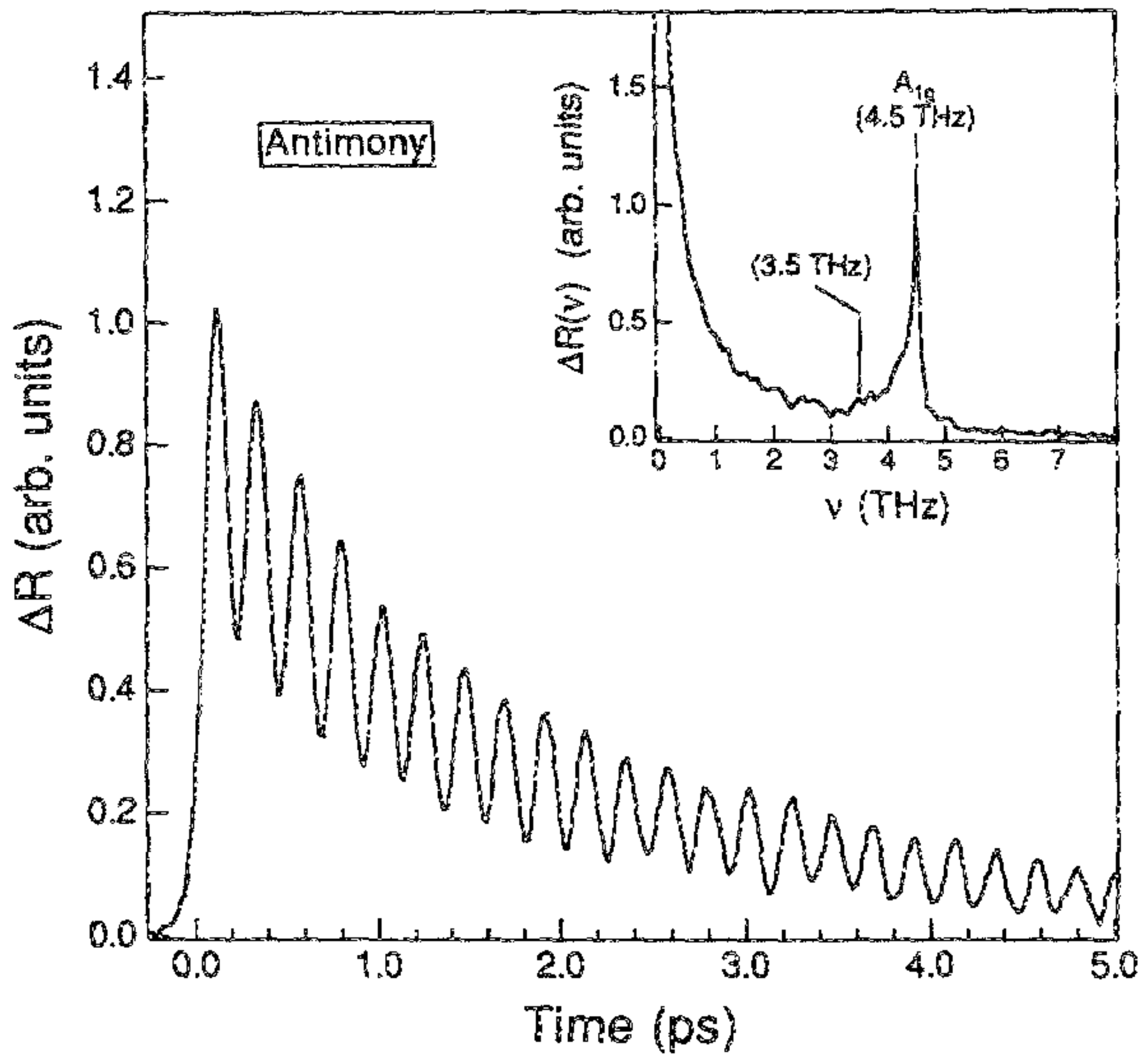




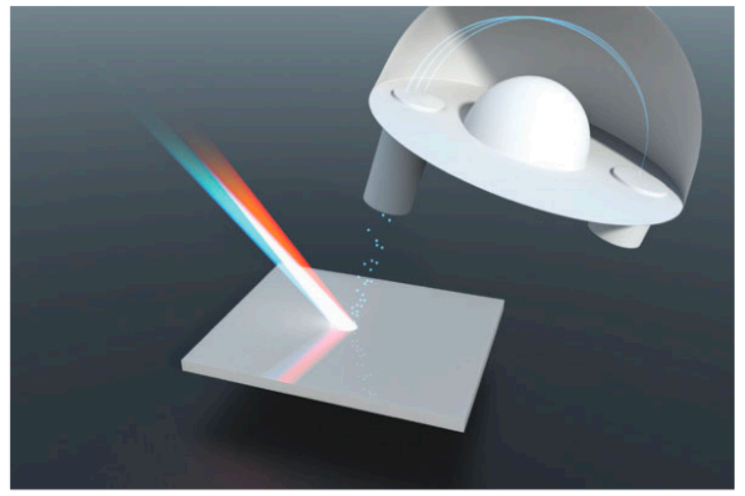




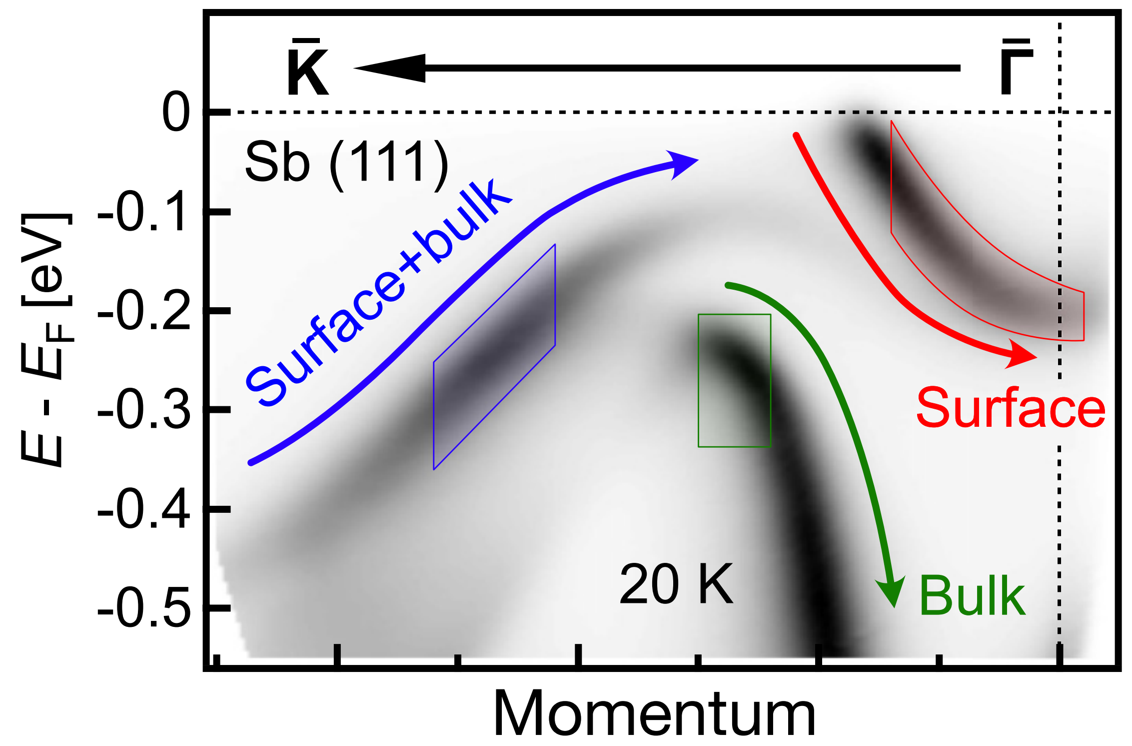
Transient Reflectivity

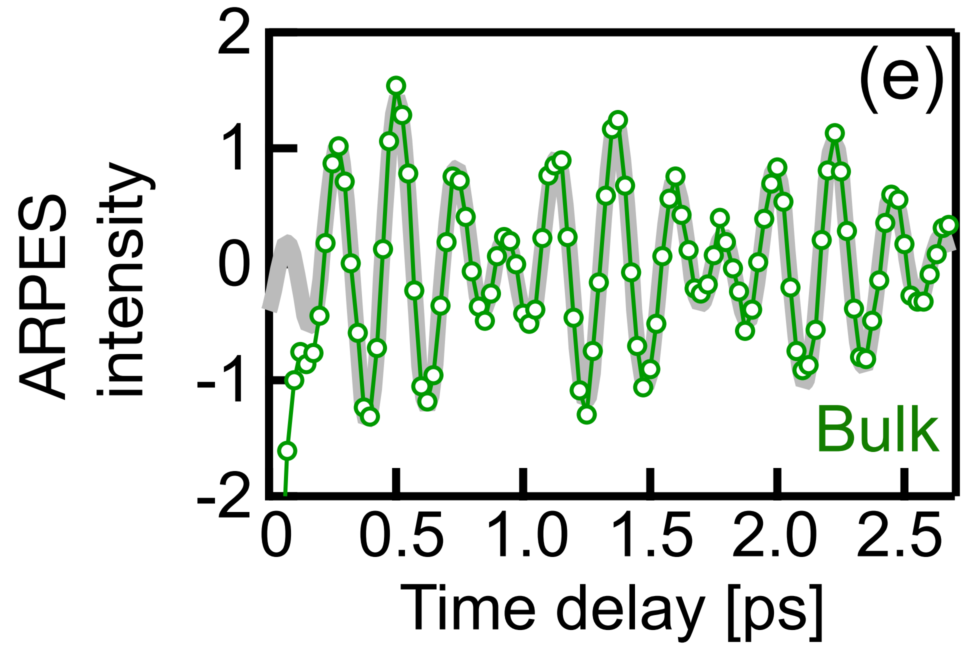


Cheng et al,
Appl. Phys. Lett. **57**, 1004 (1990)

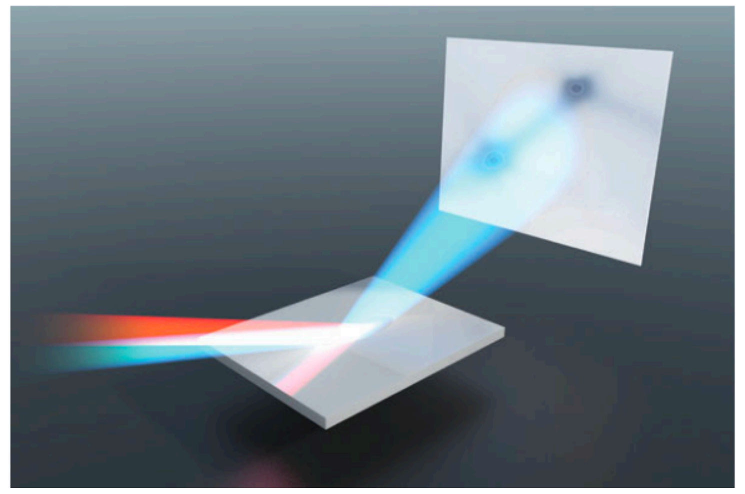


Pump-probe ARPES

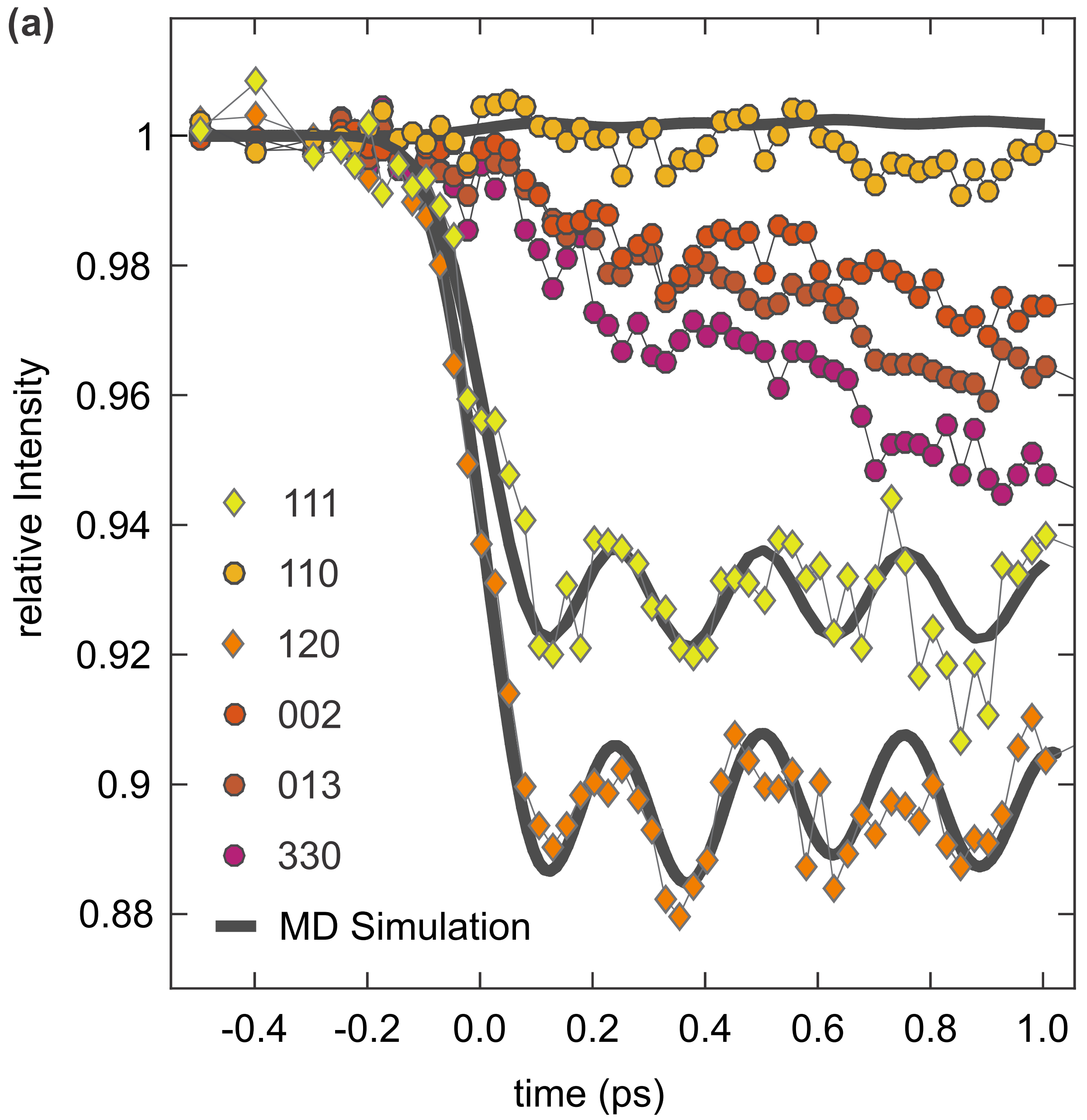




Sakamoto et al,
Phys. Rev. B **105**, L161107 (2022)

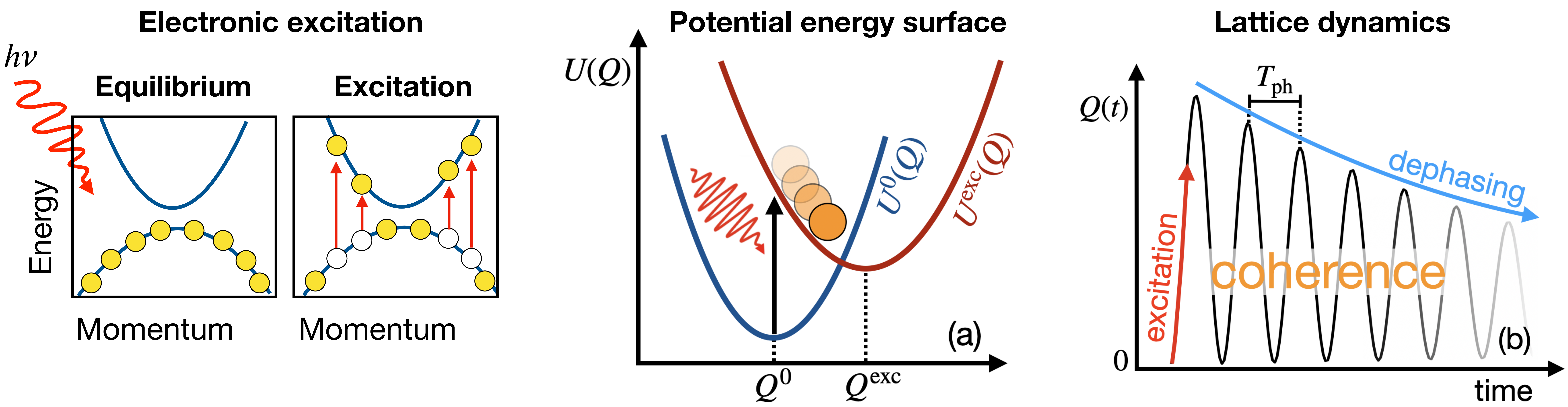


Ultrafast Diffraction



Waldecker, Ernstorfer, et al,
Phys. Rev. B **95**, 054302 (2017)

The displacive excitation of coherent phonons (DECP)



Comprehensive understanding of coherent-phonon excitation mechanisms:

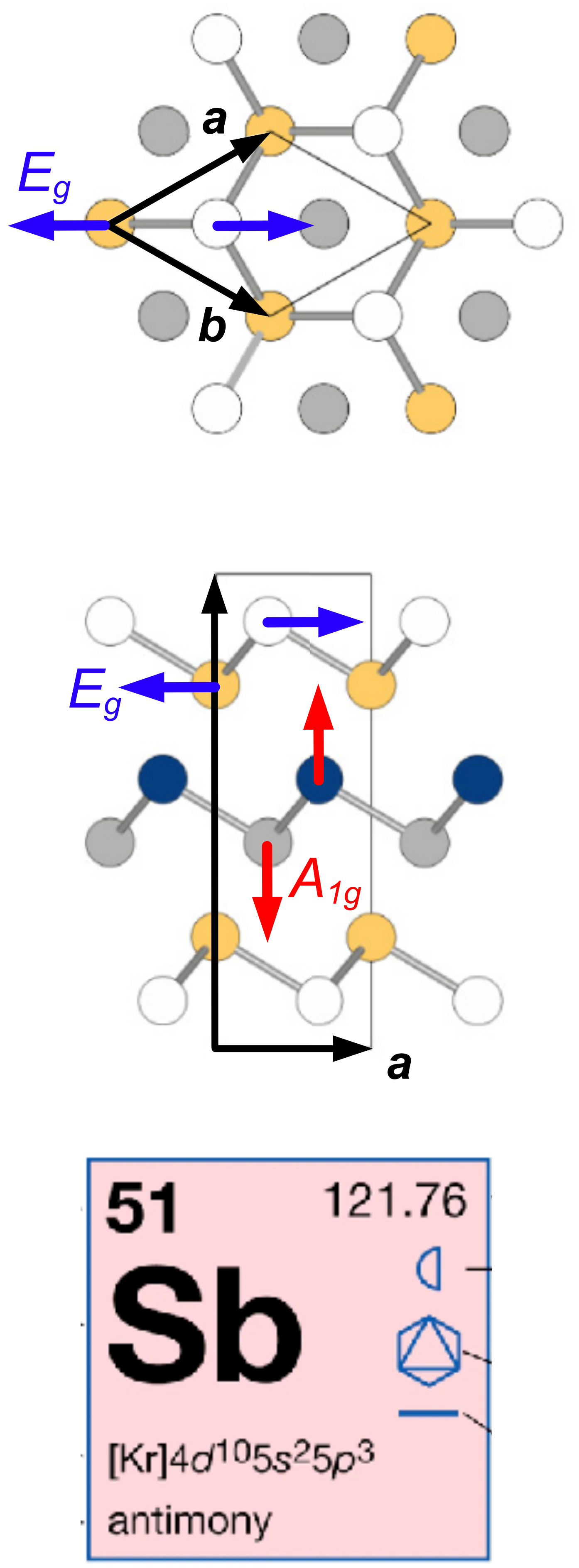
Zeiger, Dresselhaus et al., Phys. Rev. B **45**, 768 (1992)
Dhar, Nelson et al., Chem. Rev. **94**, 157 (1994)
Kutsenov, Stanton, Phys. Rev. Lett. **73**, 3243 (1994)
Garrett, Merlin et al., Phys. Rev. Lett. **77**, 3661 (1996)
Rossi, Kuhn, Rev. Mod. Phys. **74**, 895 (2002)
Hase, Petek et al., Nature (2003)
Wu, Meng, Nature Comm. **15**, 2804 (2024)
... and many more!!!

Challenges for ab-initio many-body methods:

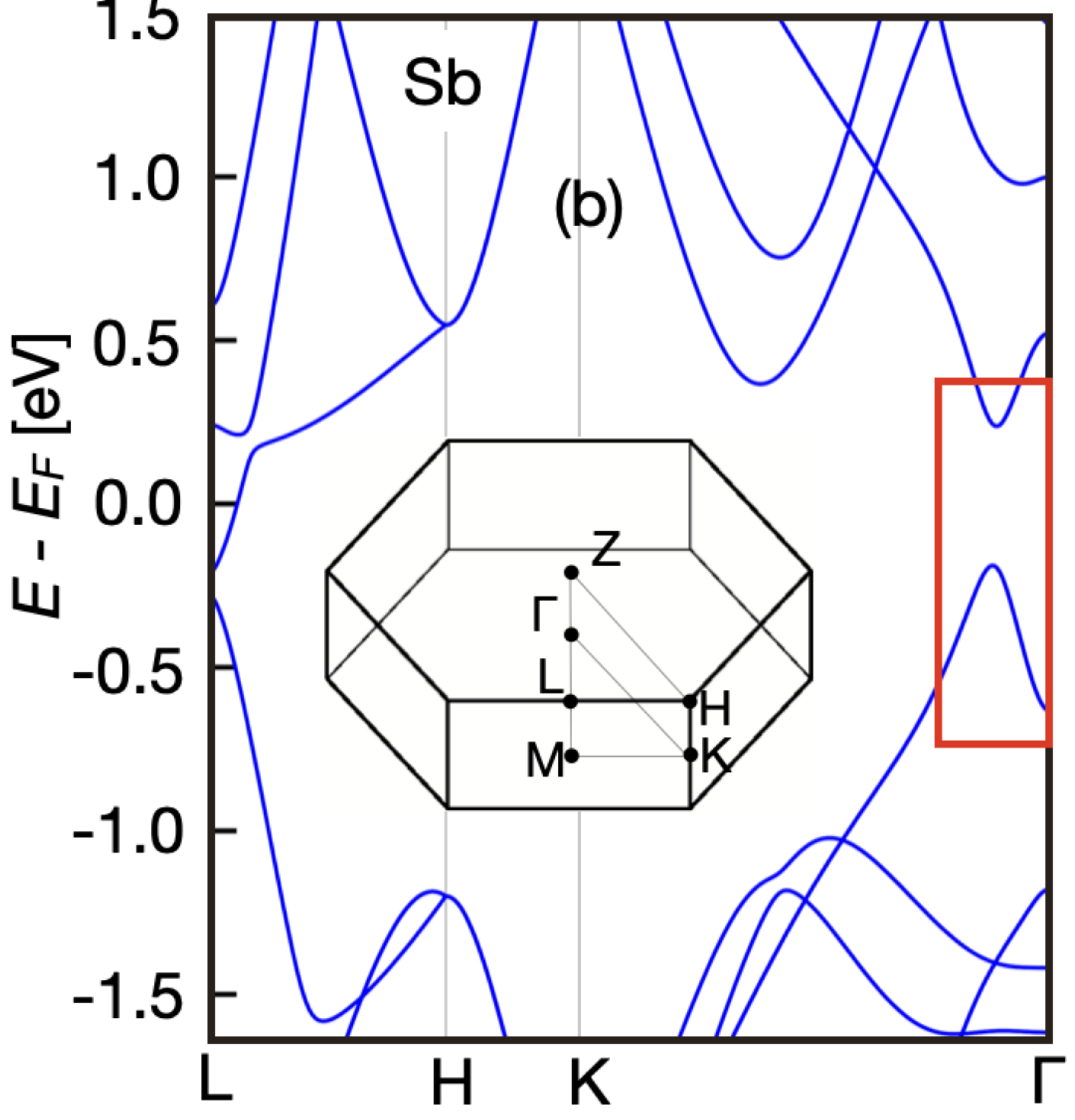
1. Capture emergence of coherent phonons in materials
2. Account quasiparticle renormalization due to coherent phonons
3. Develop a theory of phonon decoherence

The electronic structure of antimony

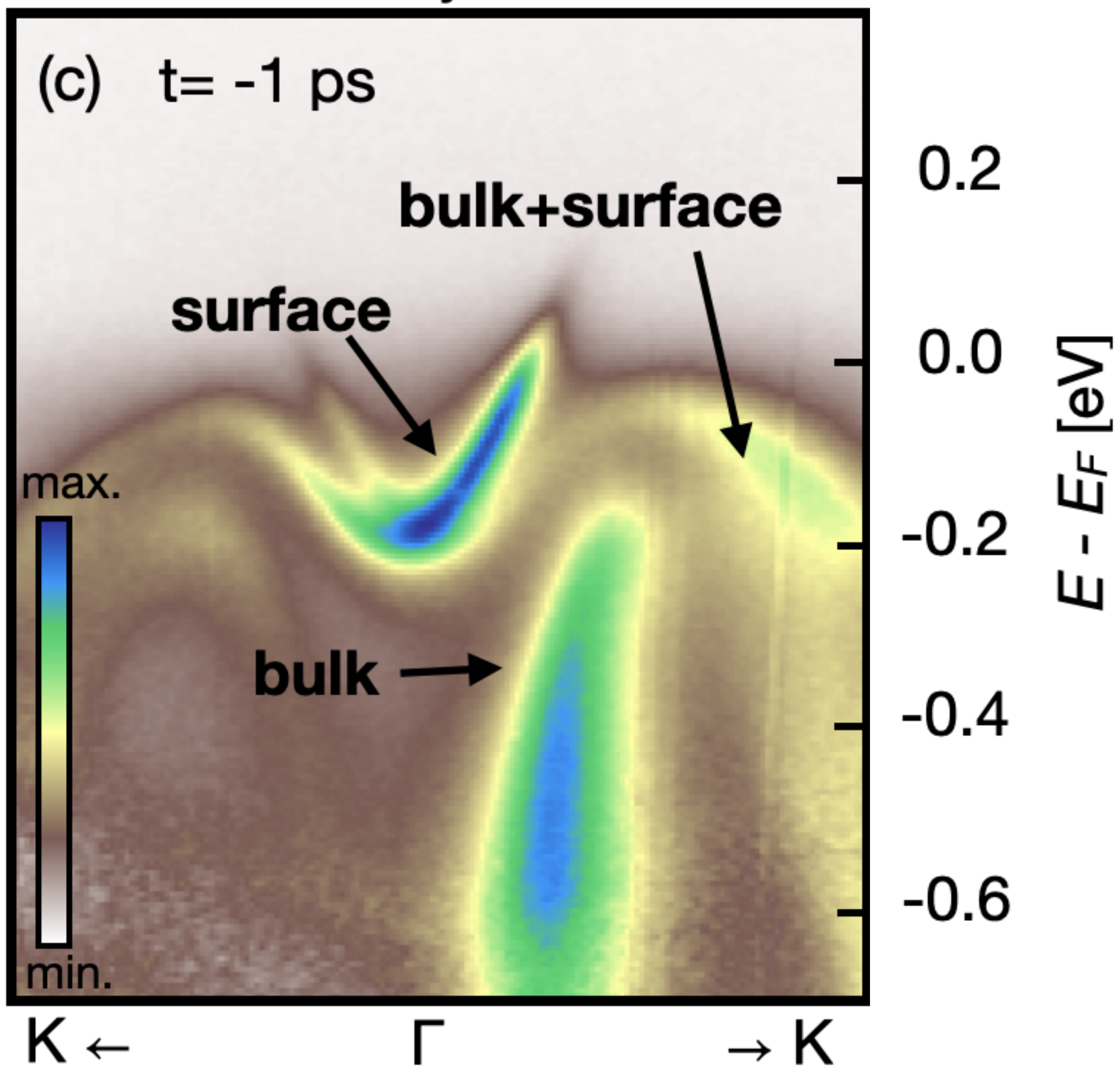
Antimony (Sb)



KS-DFT bands

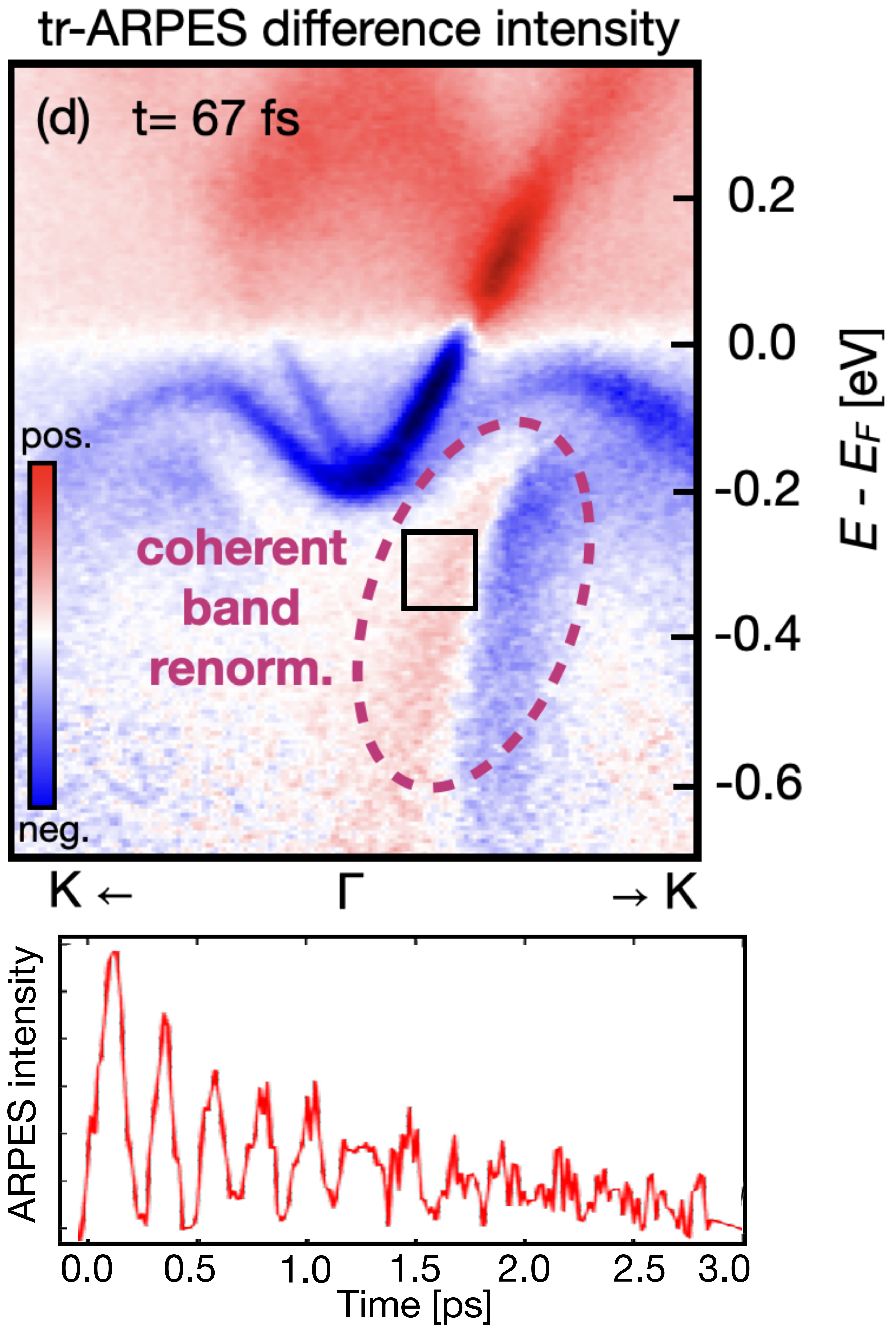


ARPES intensity



Measurements: S. Jauernik, Sunil, C. E. Jensen, P. Hein, M. Bauer

Probing coherent phonons via tr-ARPES



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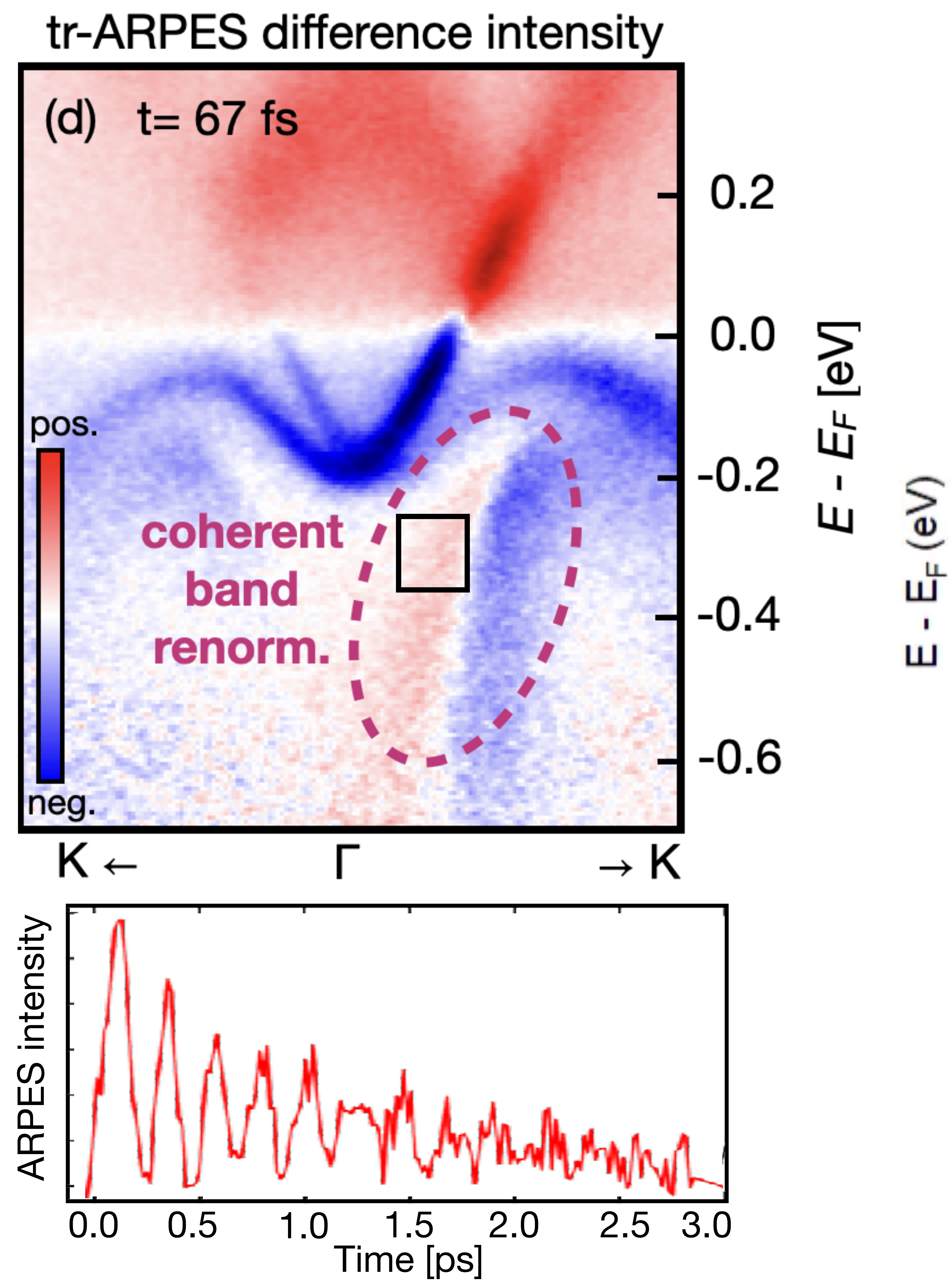
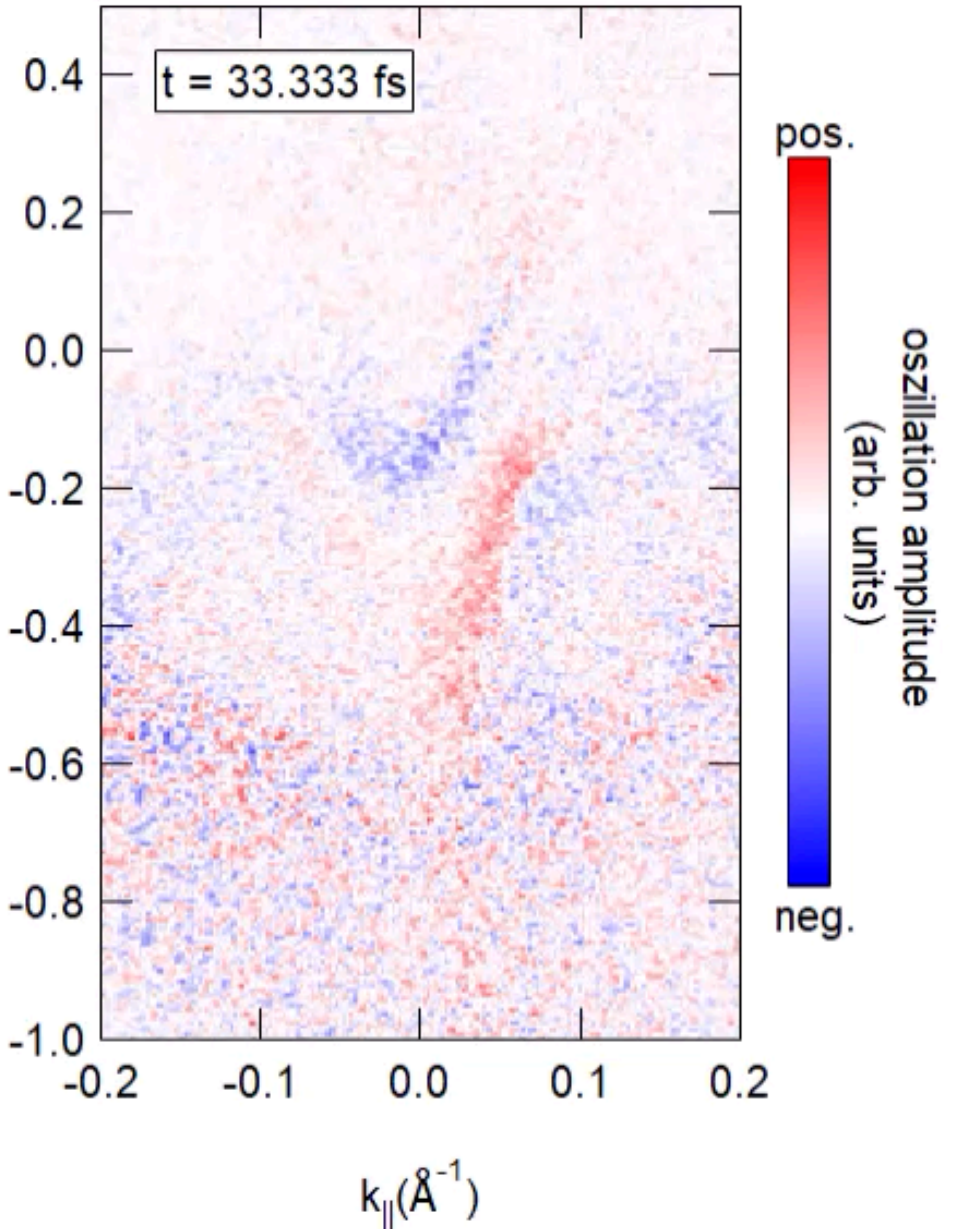
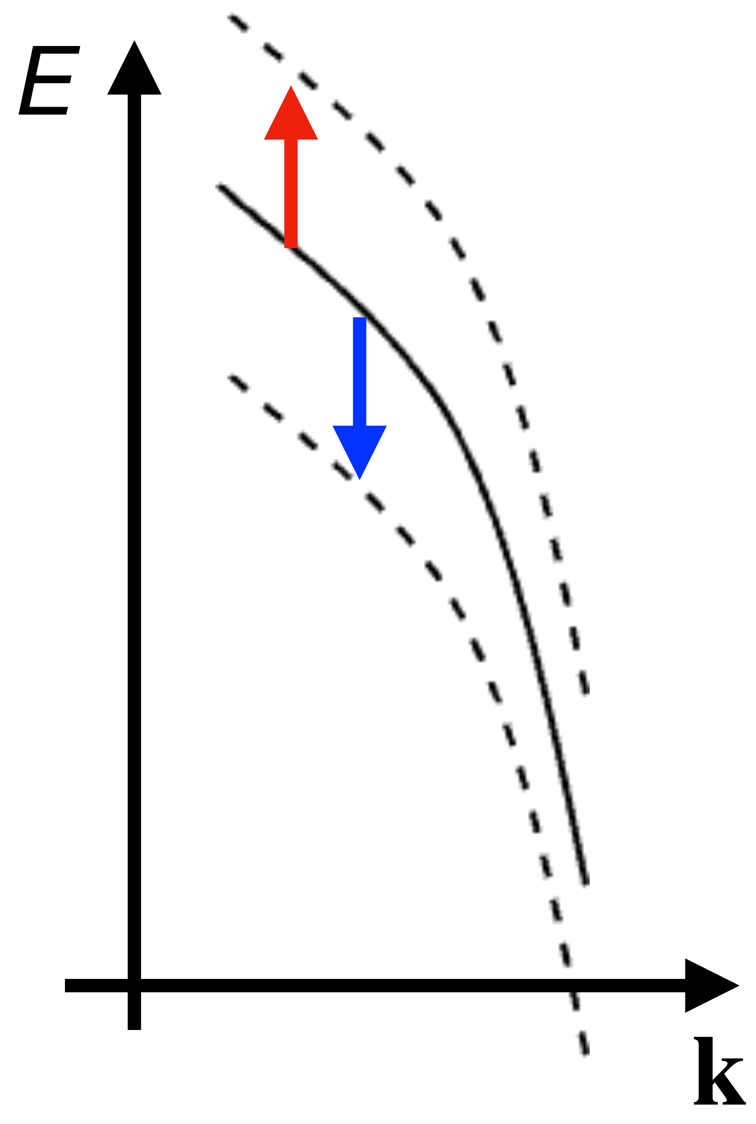
Probing coherent phonons via tr-ARPES

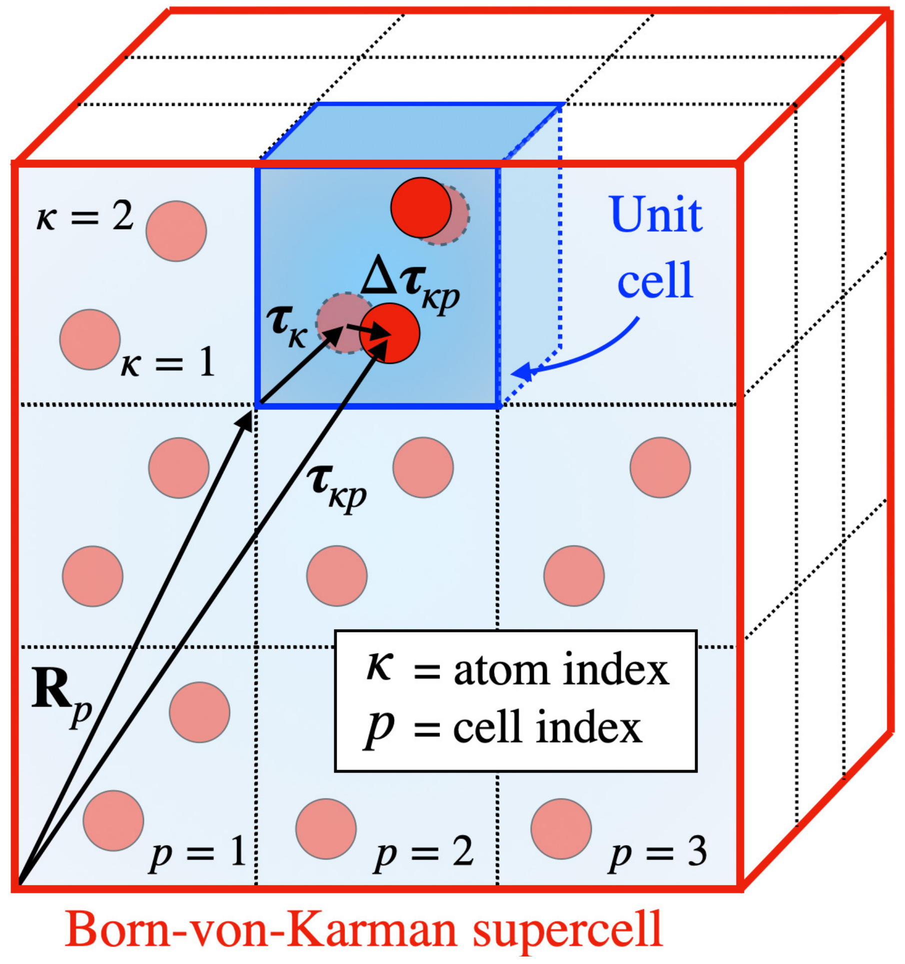






Measurements: S. Jauernik, Sunil, C. E. Jensen, P. Hein, M. Bauer

Ab-initio approach to coherent phonons in materials (DECP)



Displacement along phonon $\mathbf{q}\nu$: $\hat{Q}_{\mathbf{q}\nu}$

Lattice Hamiltonian: $\hat{H} = \hat{H}_{\mathrm{ph}} + \hat{H}_{\mathrm{e-ph}} + \hat{H}_{\mathrm{ph-ph}}$

Heisenberg eq. of motion: $i\hbar \frac{\partial \hat{Q}_{\mathbf{q}\nu}(t)}{\partial t} = [\hat{Q}_{\mathbf{q}\nu}, \hat{H}]$ →

Driven-damped harmonic oscillator:

$$\ddot{Q}_{\mathbf{q}\nu} + \gamma_{\mathbf{q}\nu}\dot{Q}_{\mathbf{q}\nu} + \omega^2_{\mathbf{q}\nu}Q_{\mathbf{q}\nu} = D_{\mathbf{q}\nu}(t)$$

damping — driving force

Displacive excitation of coherent phonons (DECP):

$$D^{\mathrm{eph}}_{\mathbf{q}\nu}(t) = -\frac{\omega_{\mathbf{q}\nu}}{\hbar N_p^{\frac{1}{2}}} \sum_{n\mathbf{k}} g^{\nu}_{nn}(\mathbf{k},\mathbf{q})[f_{n\mathbf{k}}(t) - f_{n\mathbf{k}}(0)]\delta_{\mathbf{q},0}$$

matrix element (EPI) — change of electron distribution (TDBE)

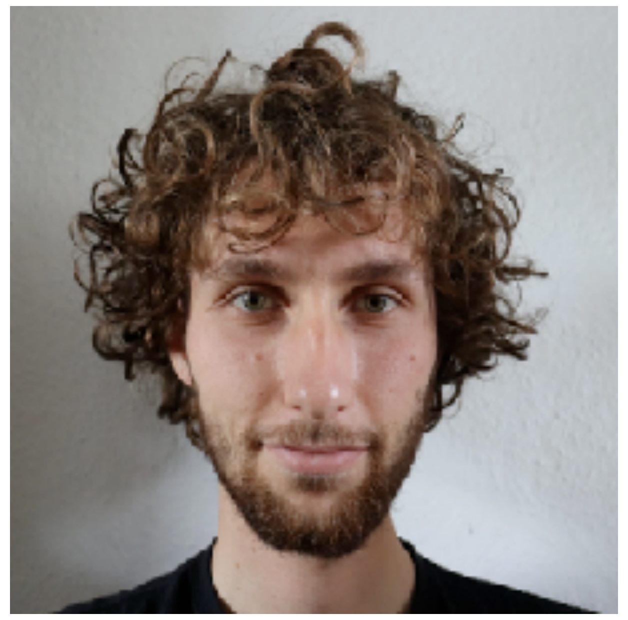
Quasiparticle energy renormalization due to coherent phonons:

$$\varepsilon^{\mathrm{QP}}_{n\mathbf{k}} = \varepsilon_{n\mathbf{k}} + \Sigma^{\mathrm{p}}_{n\mathbf{k}} \qquad \Sigma^{p}_{n\mathbf{k}}(t) = \sum_{\mathbf{q}\nu} g^{\nu}_{nn}(\mathbf{k},\mathbf{q})Q_{\mathbf{q}\nu}(t)$$

FC, M. Zacharias, PRB **107**, 054102 (2023)

Workflow for DECP simulations

DFT, DFPT:	electron and phonon energies → $\varepsilon_{n\mathbf{k}}, \hbar\omega_{\mathbf{q}\nu}$
excited-state Ansatz:	$f^{\mathrm{exc}}_{n\mathbf{k}} = [\exp((\varepsilon_{\mathrm{n}\mathbf{k}} - \mu)/\mathrm{k_B T_{exc}}) + 1]^{-1}$
time-dependent Boltzmann eq. (TDBE):	e-ph occupations $f_{n\mathbf{k}}(t), n_{\mathbf{q}\nu}(t)$
coherent-phonon EOM:	displacement amplitude $Q_{\mathbf{q}\nu}(t)$
polaron self-energy and quasiparticle renormalization:	$\Sigma^{\mathrm{p}}_{n\mathbf{k}}(t), A(\omega; t)$



C. Emeis

EPW®

Emeis, Jauernik, Sunil, Pan, Jensen, Hein, Bauer, FC, ArXiv 2407.17118 (2024)

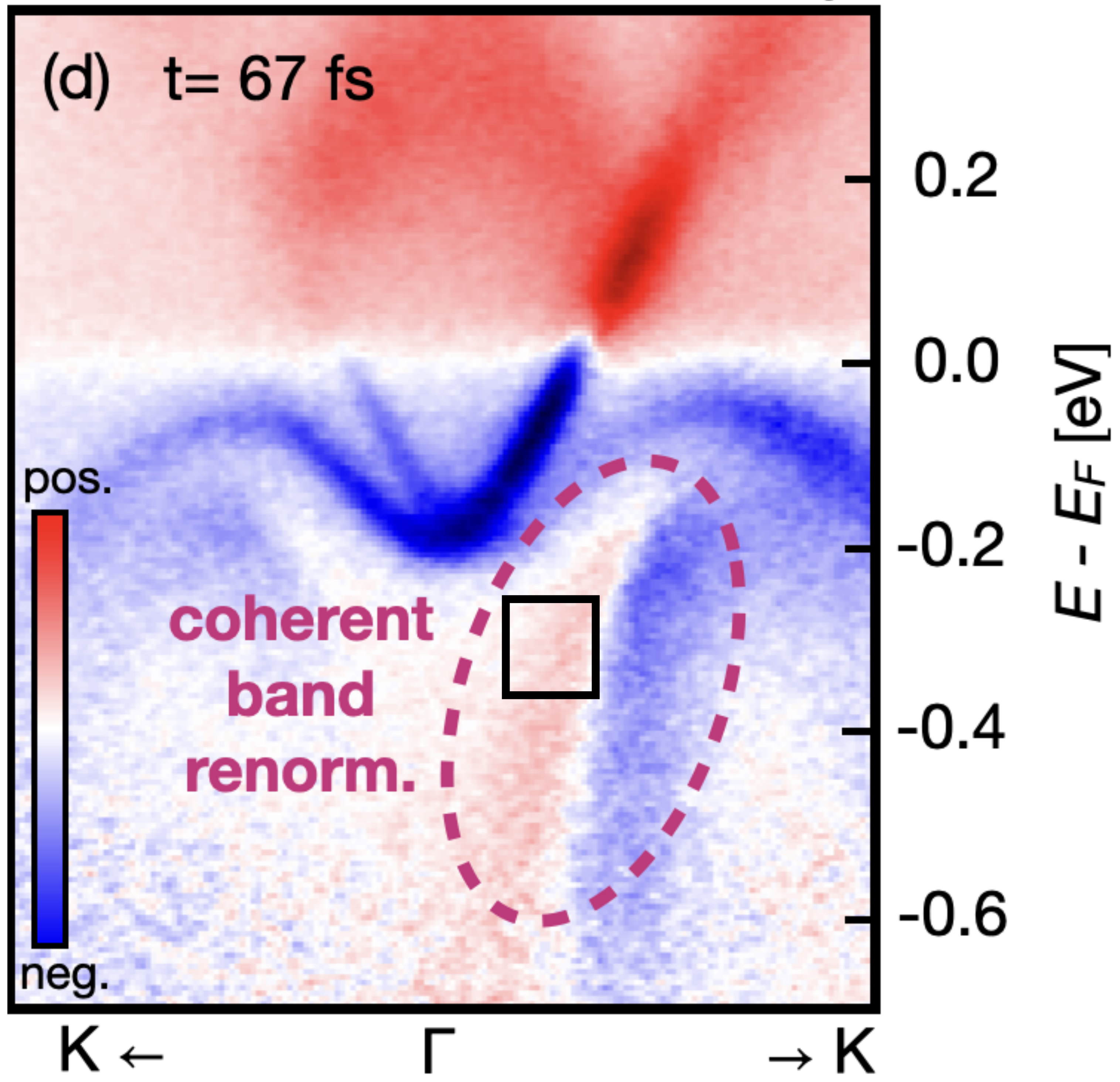
Coherent phonons in antimony from first principles

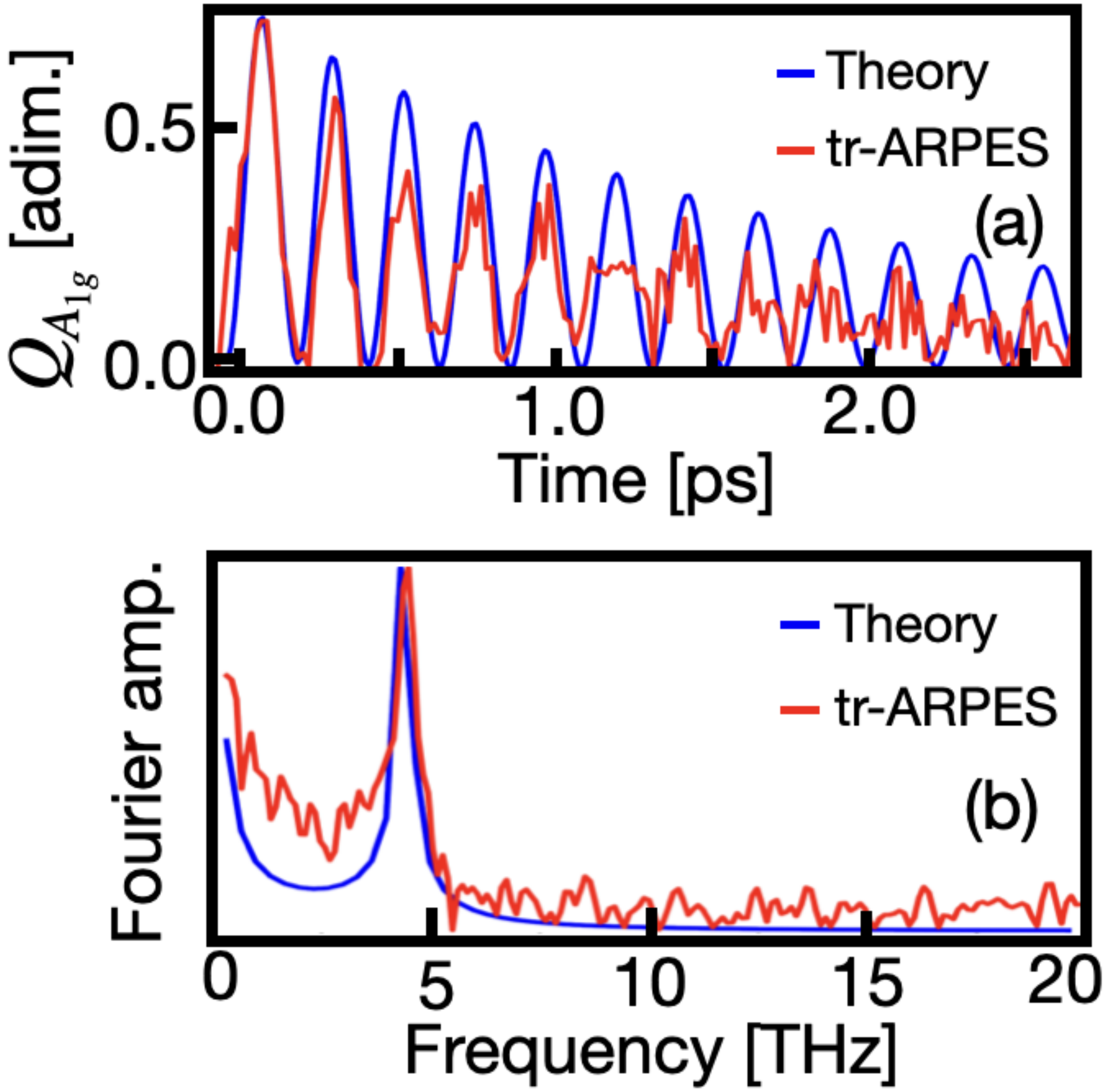
Coherent phonons EOM (DECP):

$$\ddot{Q}_{\mathbf{q}\nu} + \gamma_{\mathbf{q}\nu}\dot{Q}_{\mathbf{q}\nu} + \omega^2_{\mathbf{q}\nu} Q_{\mathbf{q}\nu} = D_{\mathbf{q}\nu}(t)$$

from first principles

tr-ARPES difference intensity





Band structure renormalization due to coherent phonons

(Static) spectral function due to the electron-phonon interaction

$$A_{n\mathbf{k}}(\omega) = -\frac{1}{\pi}\frac{\mathrm{Im}\Sigma^{(2)}_{n\mathbf{k}}(\omega)}{[\hbar\omega - \varepsilon_{n\mathbf{k}} - \mathrm{Re}\Sigma^{(2)}_{n\mathbf{k}}(\omega)]^2 + [\mathrm{Im}\Sigma^{(2)}_{n\mathbf{k}}(\omega)]^2}$$

Fan-Migdal self-energy (second-order)

Changes due to coherent phonons:

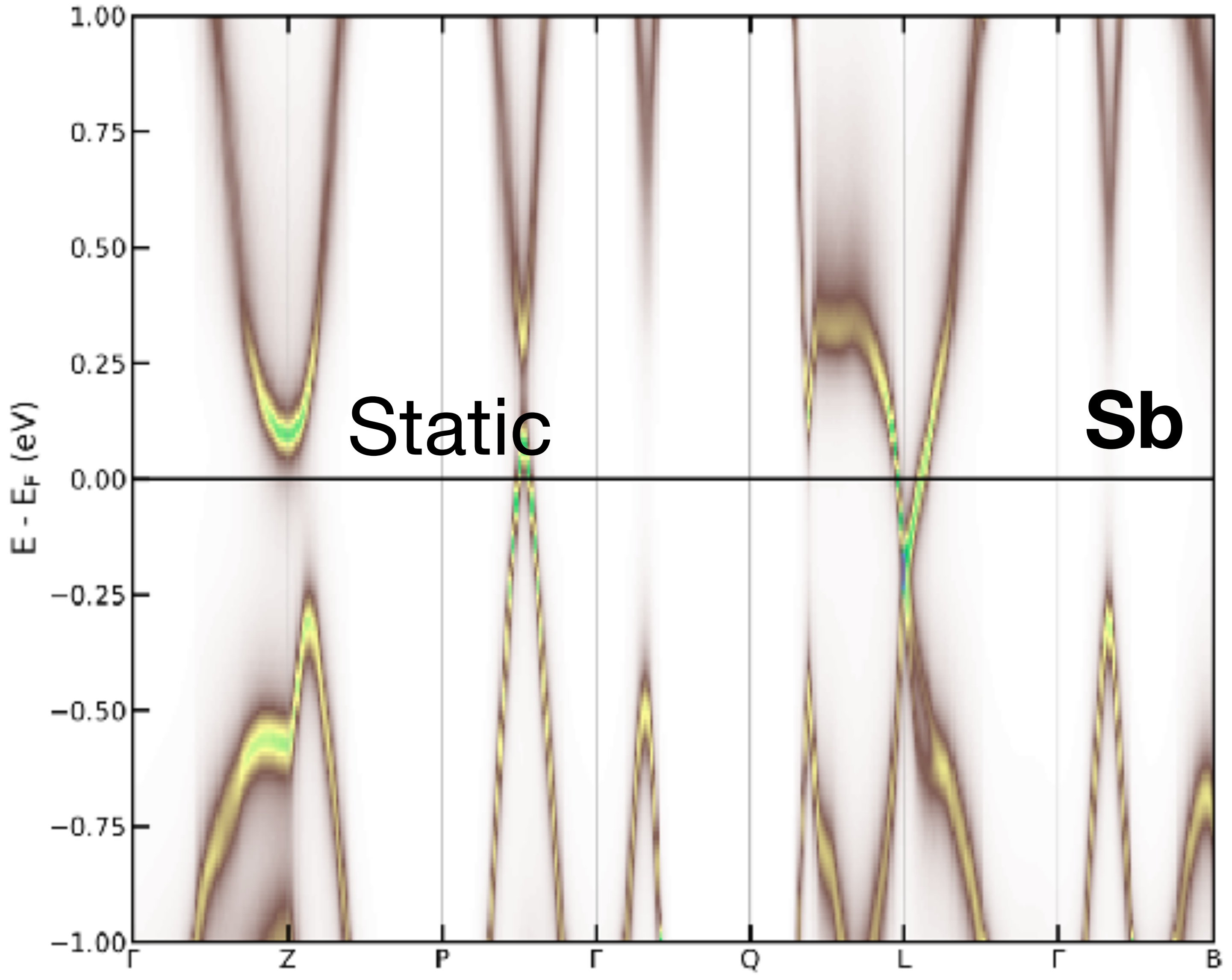
$$\varepsilon_{n\mathbf{k}}(t) = \sum_{\nu} g^{\nu}_{mn}(\mathbf{k},\mathbf{q})Q_{\mathbf{q}\nu}(t) = \Sigma^{\mathrm{P}}_{n\mathbf{k}}(t)$$

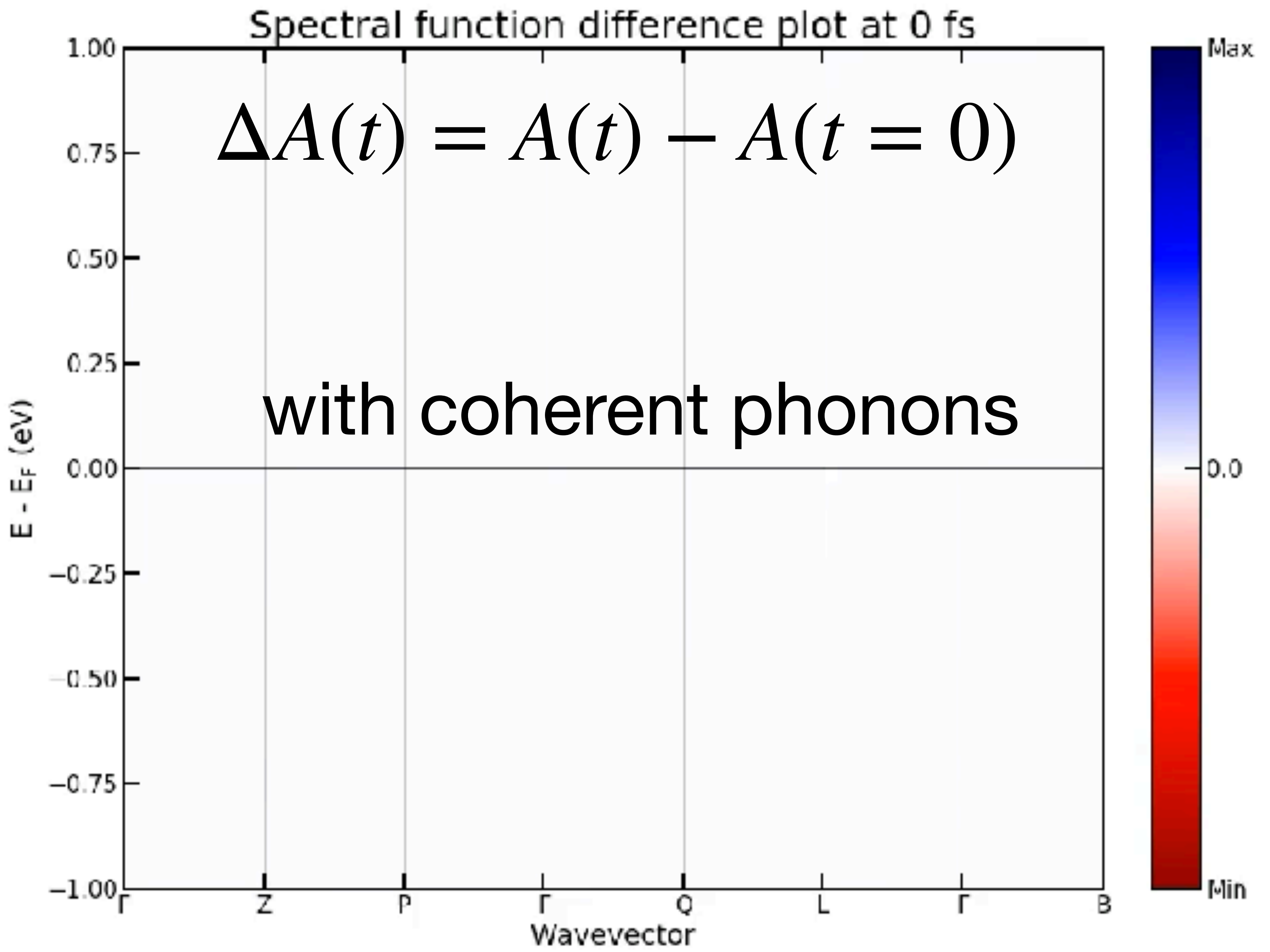
Polaron self-energy (first-order)

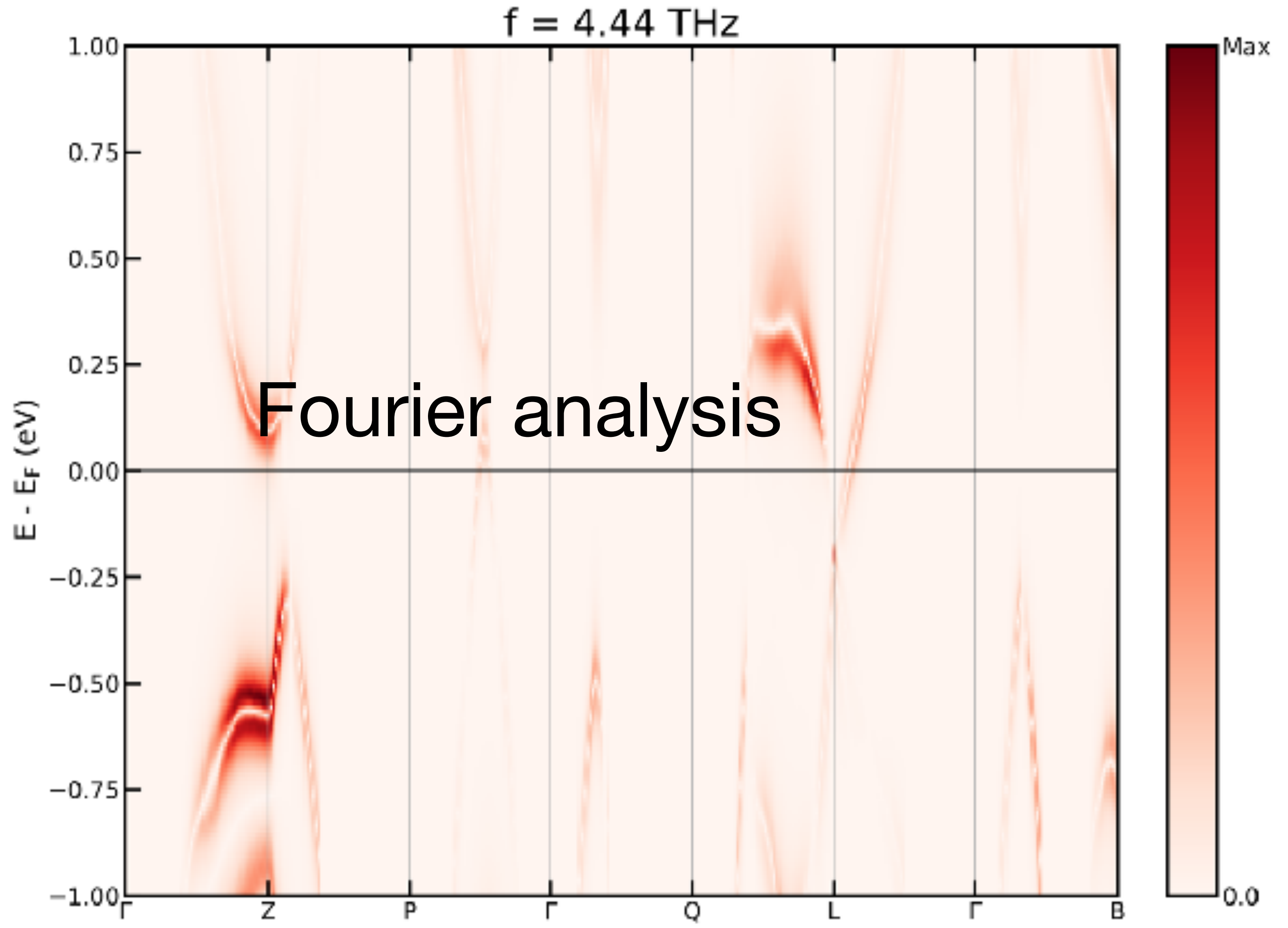
Lafuente-Bartolome, Giustino et al., Phys. Rev. Lett. **129**, 076402 (2022)

Time-dependent spectral function

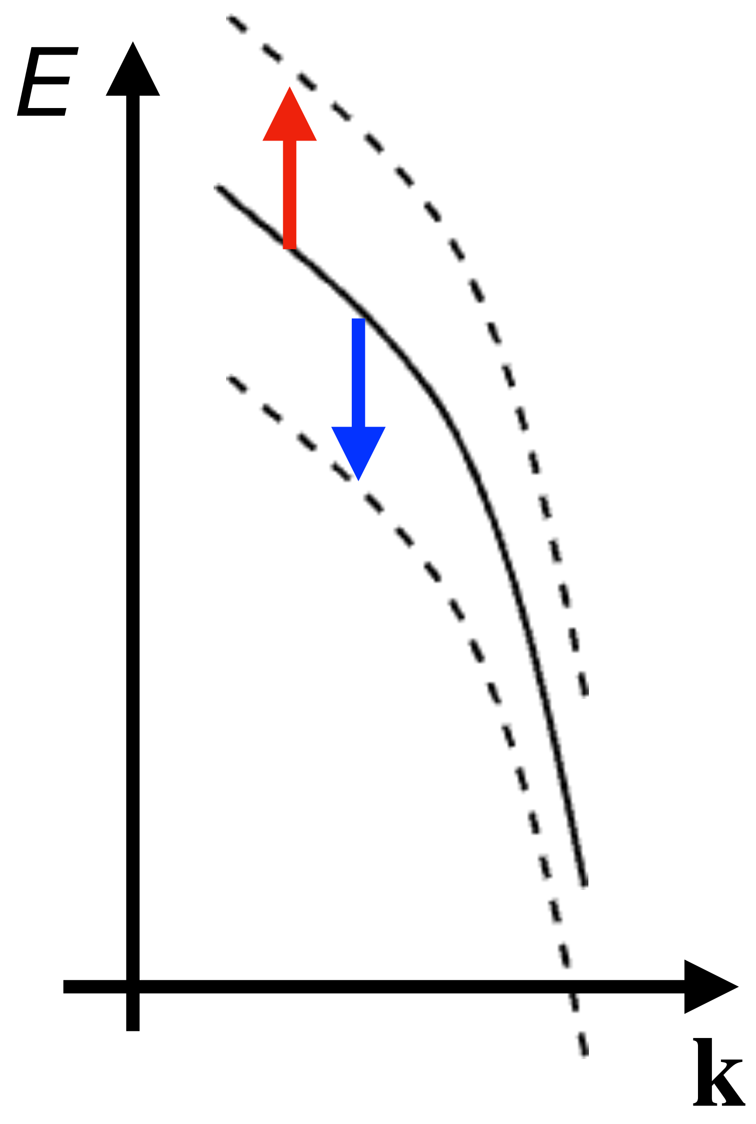
$$A_{n\mathbf{k}}(\omega; t) = -\frac{1}{\pi}\frac{\mathrm{Im}\Sigma^{(2)}_{n\mathbf{k}}(\omega)}{[\hbar\omega - \varepsilon_{n\mathbf{k}} - \Sigma^{\mathrm{P}}_{n\mathbf{k}}(t) - \mathrm{Re}\Sigma^{(2)}_{n\mathbf{k}}(\omega)]^2 + [\mathrm{Im}\Sigma^{(2)}_{n\mathbf{k}}(\omega)]^2}$$

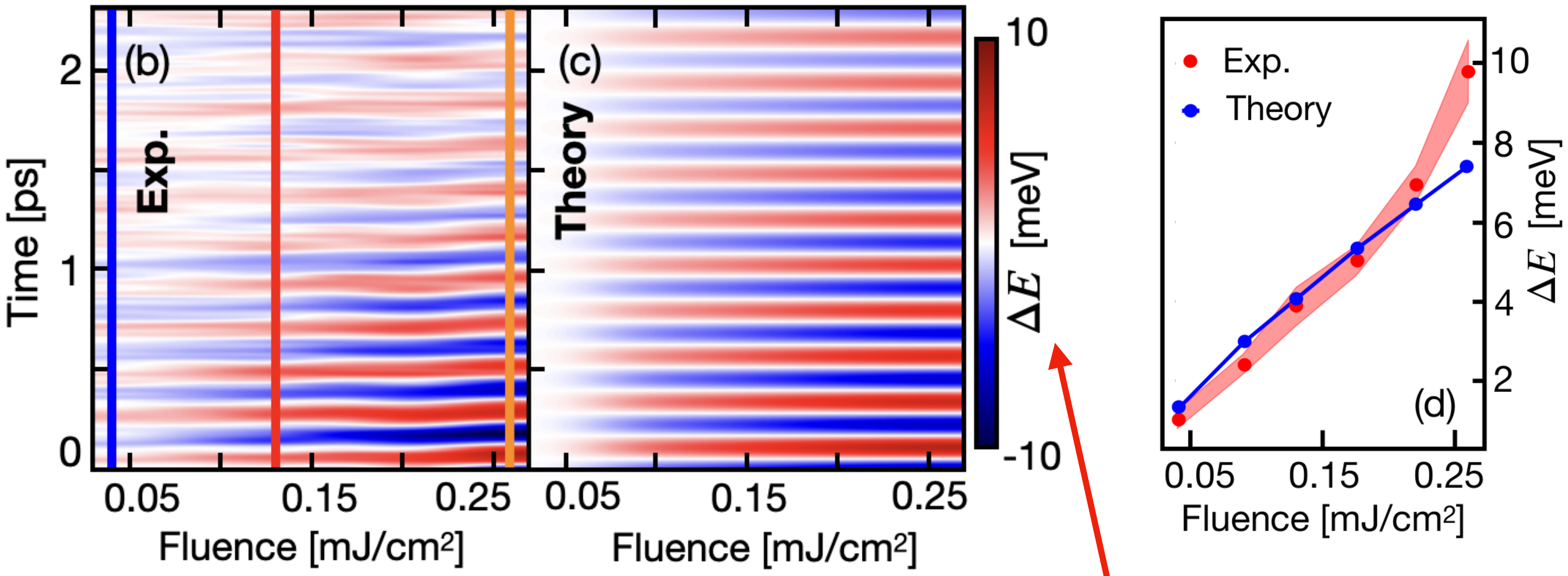






Fluence-dependent measurements of coherent phonons: theory and td-ARPES





band shift due to coherent phonon excitation

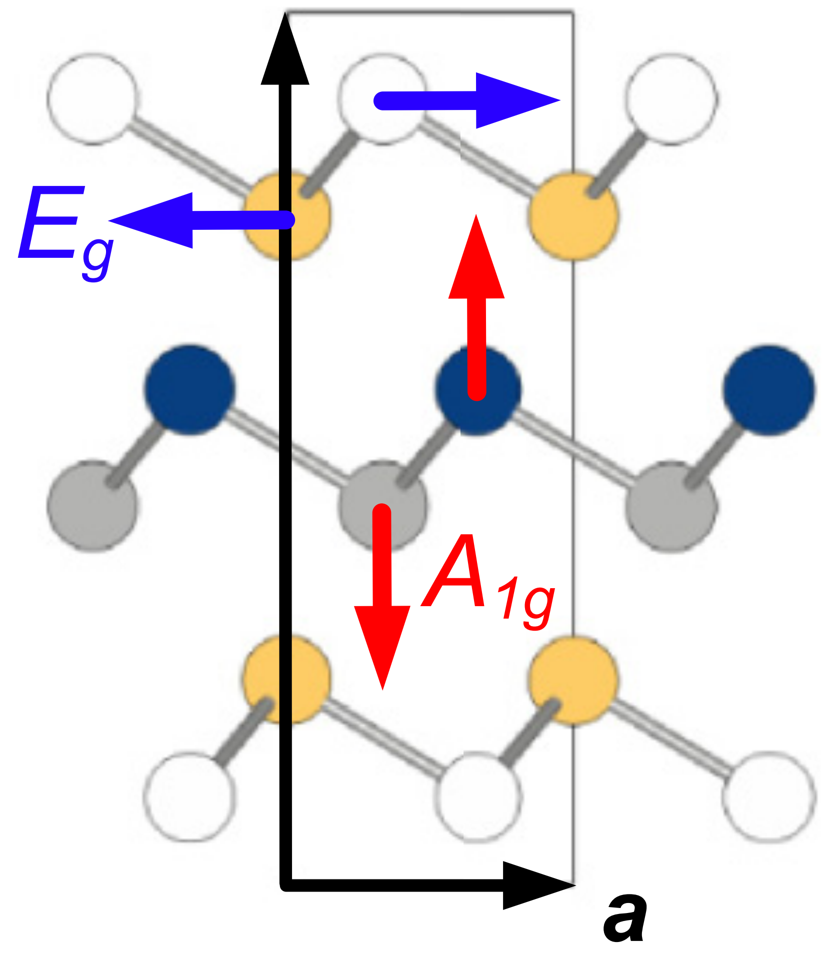
Retrieving nuclear trajectories from tr-ARPES

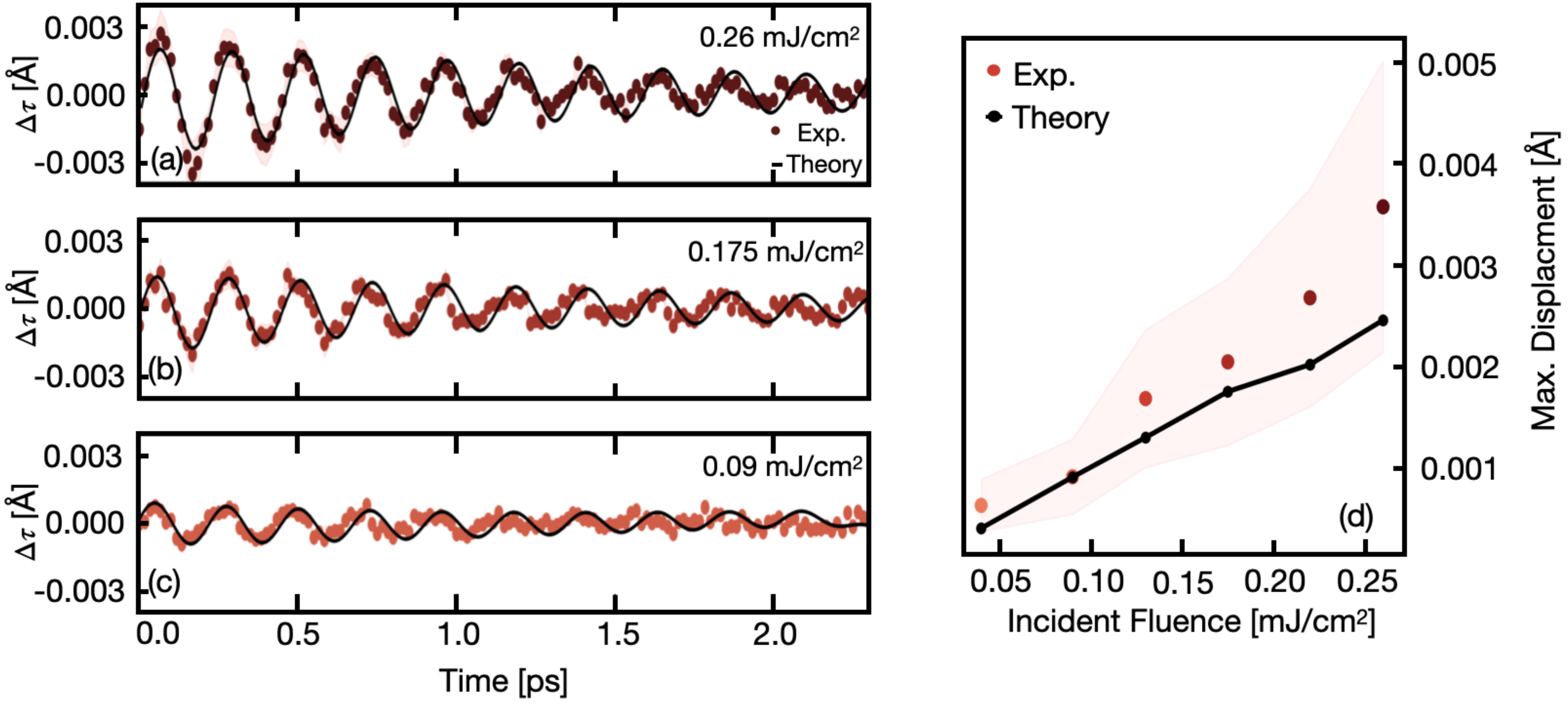
Quasiparticle renormalization due to coherent phonons:

$$\Delta\varepsilon_{n\mathbf{k}}(t) = g^{\nu}_{nn}(\mathbf{k},\mathbf{q})Q_{\nu}(t)$$

$$Q_{\nu}(t) = \Delta\varepsilon_{n\mathbf{k}}(t)[g^{\nu}_{nn}(\mathbf{k},\mathbf{q})]^{-1}$$

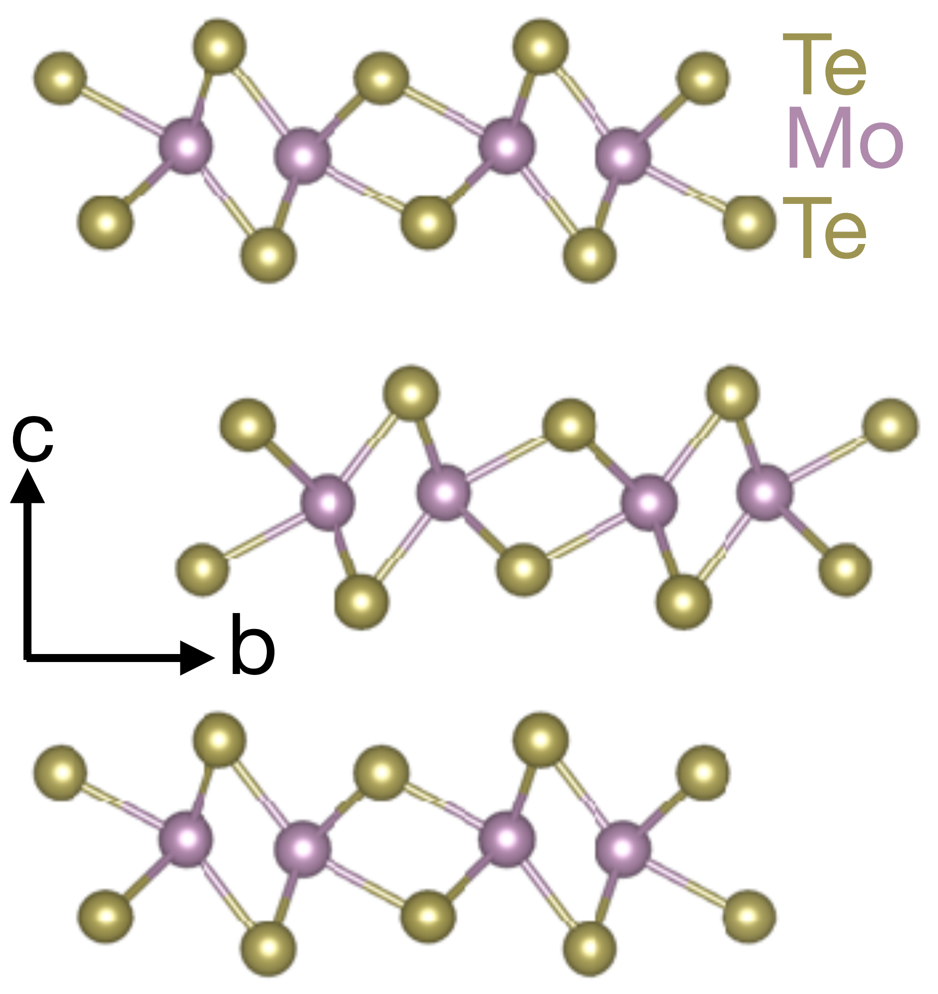
exp. theory

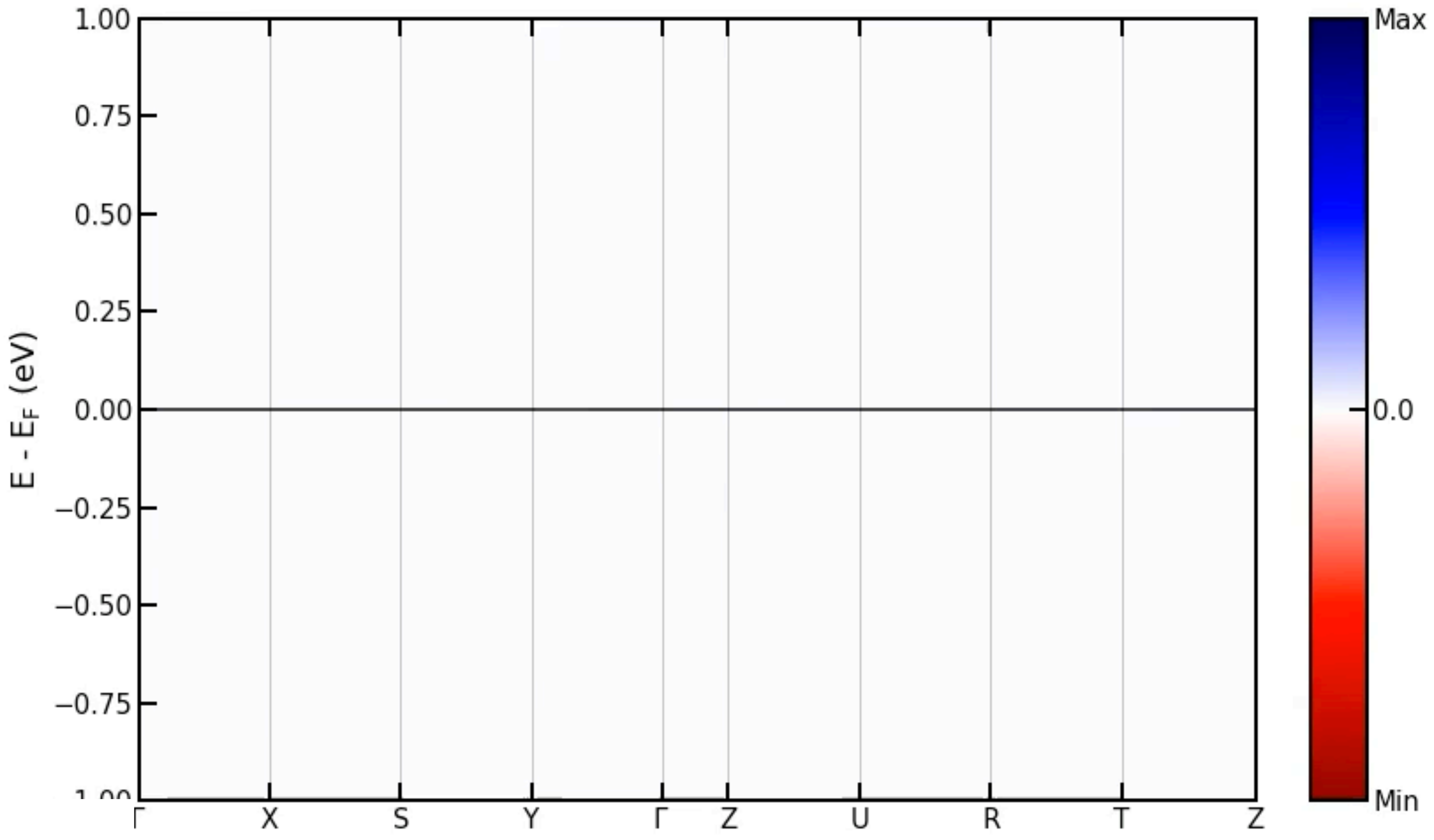


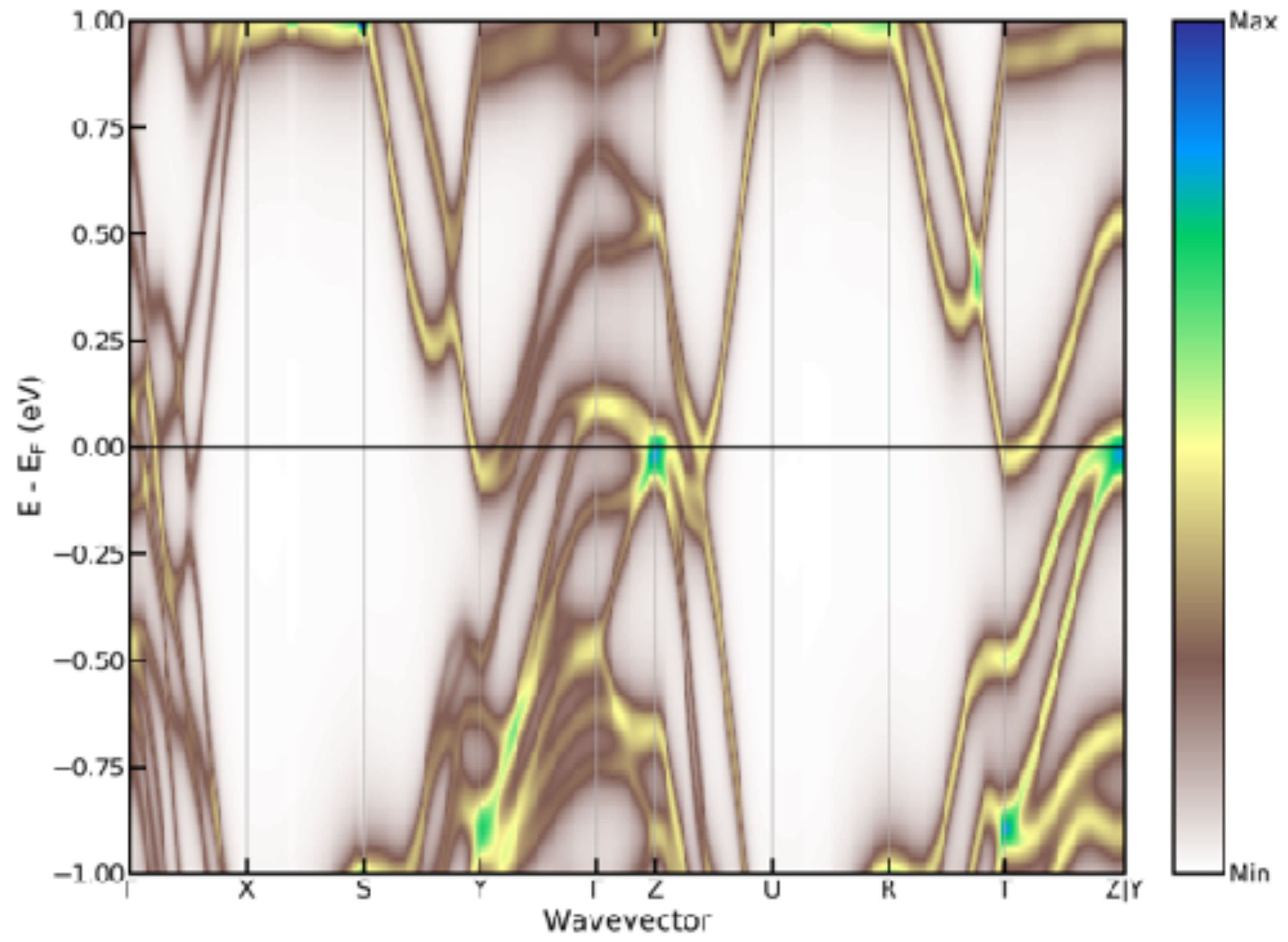


Coherent lattice dynamics in T_d-MoTe$_2$

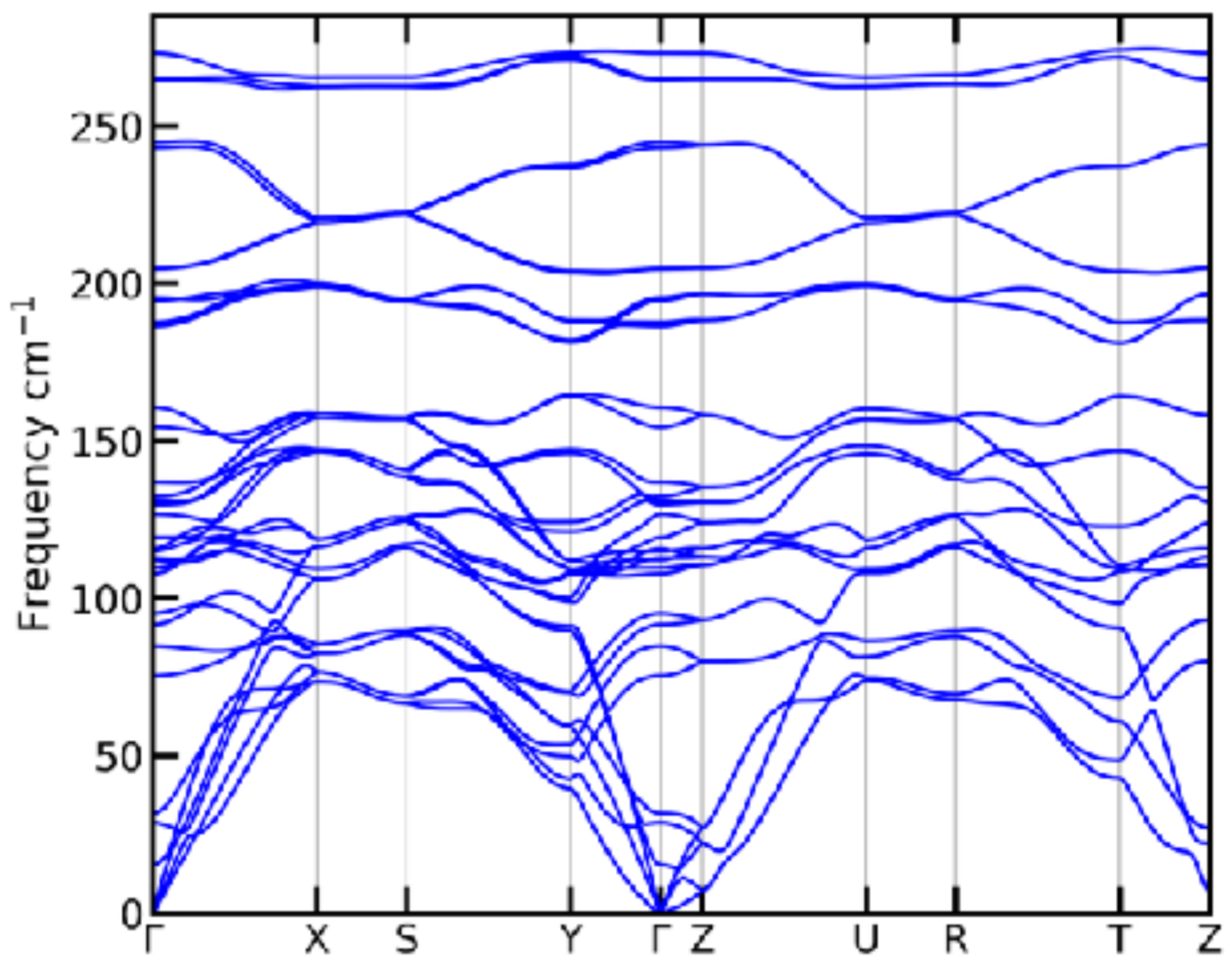
T_d-MoTe$_2$

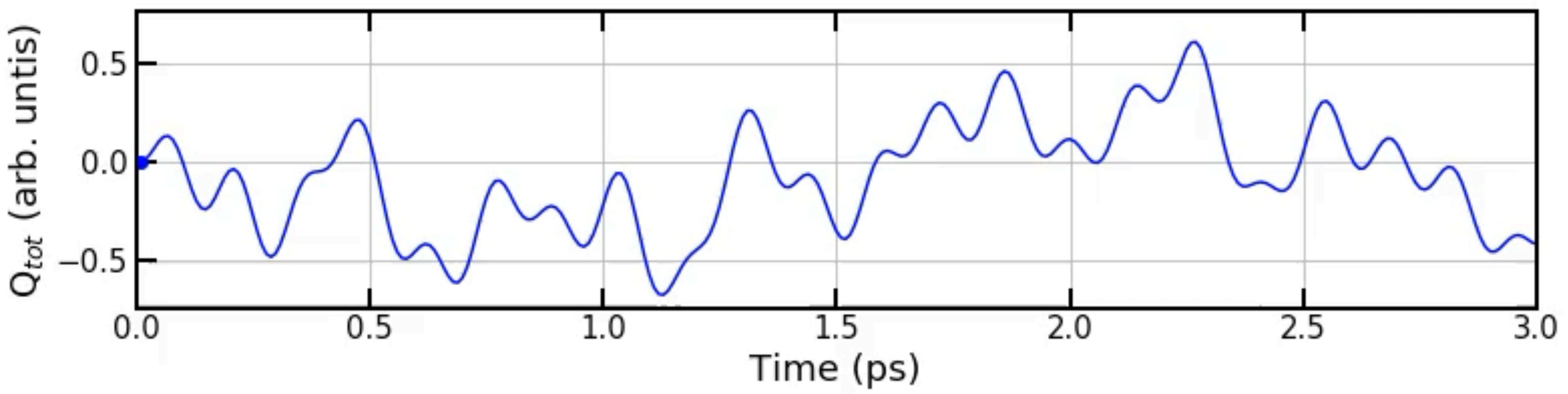






Phonon dispersion

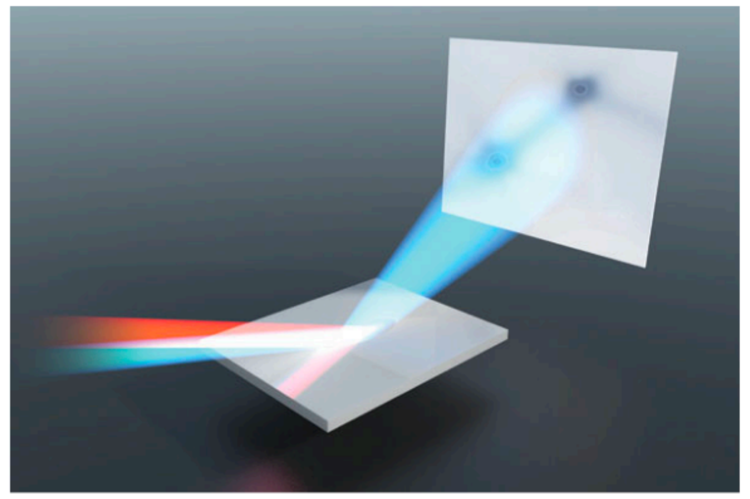


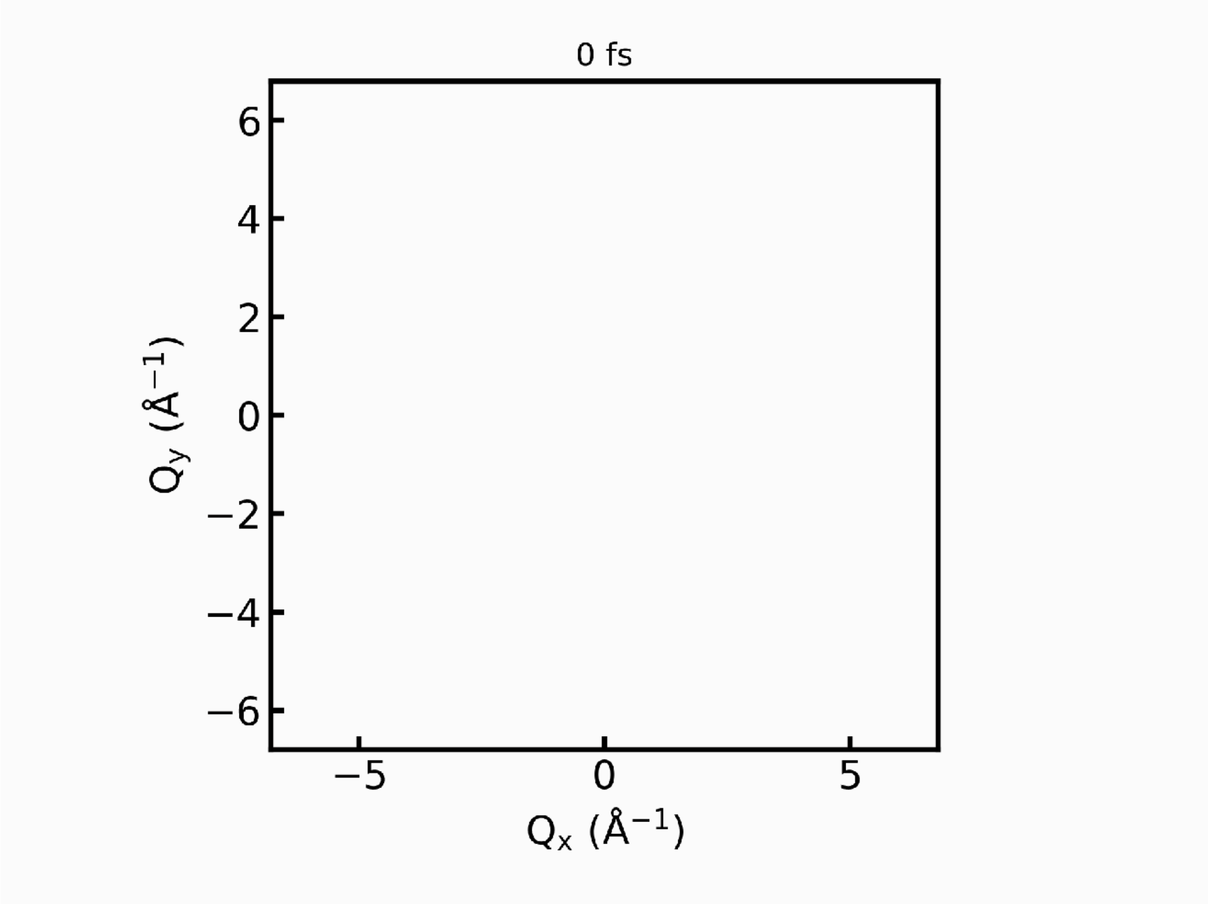


Final remarks

Part 1

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





Part 2

Coherent phonon and quasiparticle renormalization in semimetals from first principles

Emeis, Jauernik, Sunil, Pan, Jesper, Hein, Bauer, FC, ArXiv 2407.17118 (2024)

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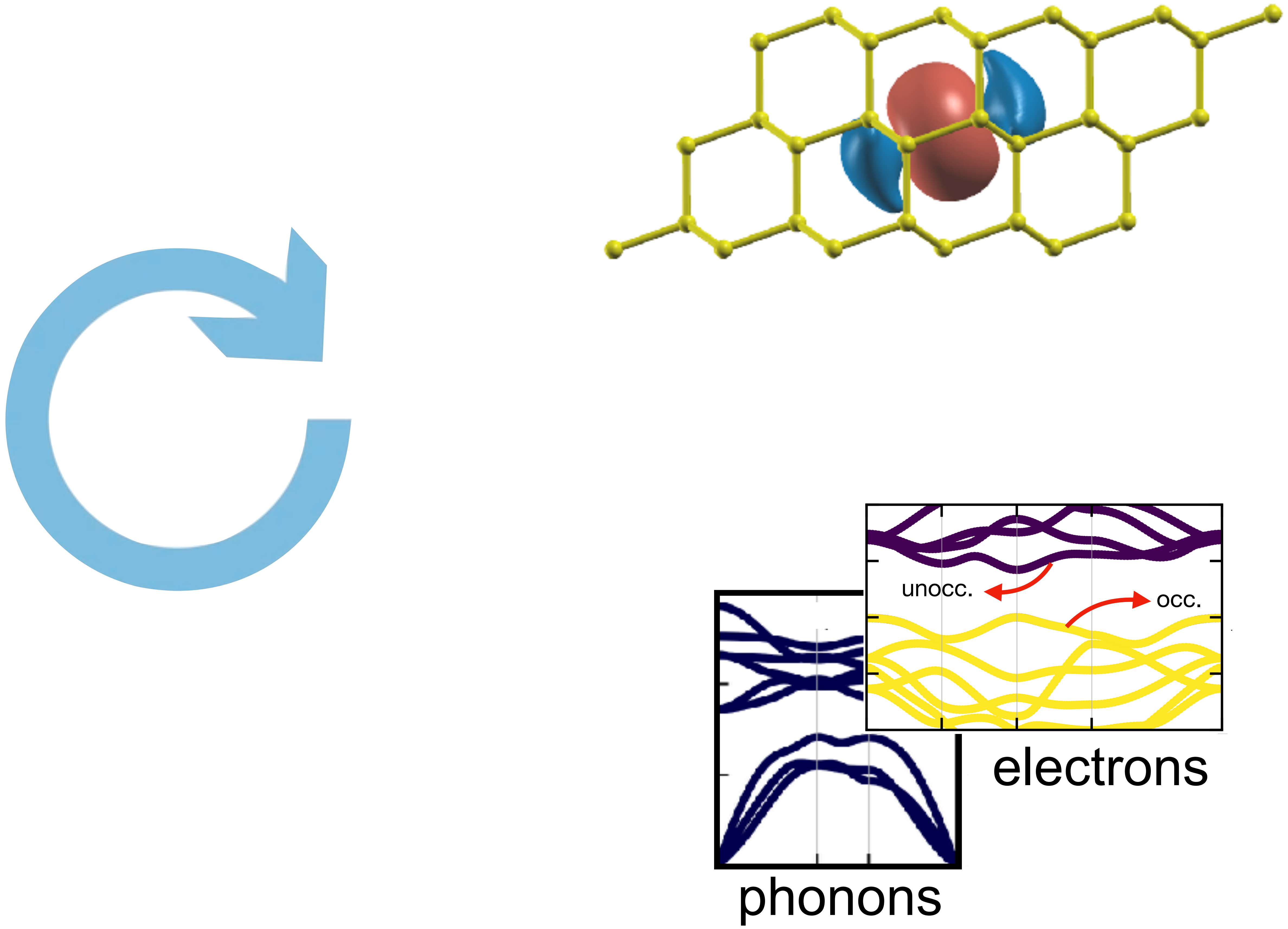
slides: cs2t.de/publications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