Phonons out of equilibrium: from ab-initio theory to pump-probe experiments

Fabio Caruso

September 25th, 2022

2D Materials and Hybrids: Hybrid Quasiparticles in Quantum Materials

Physikzentrum Bad Honnef
WE-Heraus Seminar

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Christian-Albrechts-Universität zu Kiel

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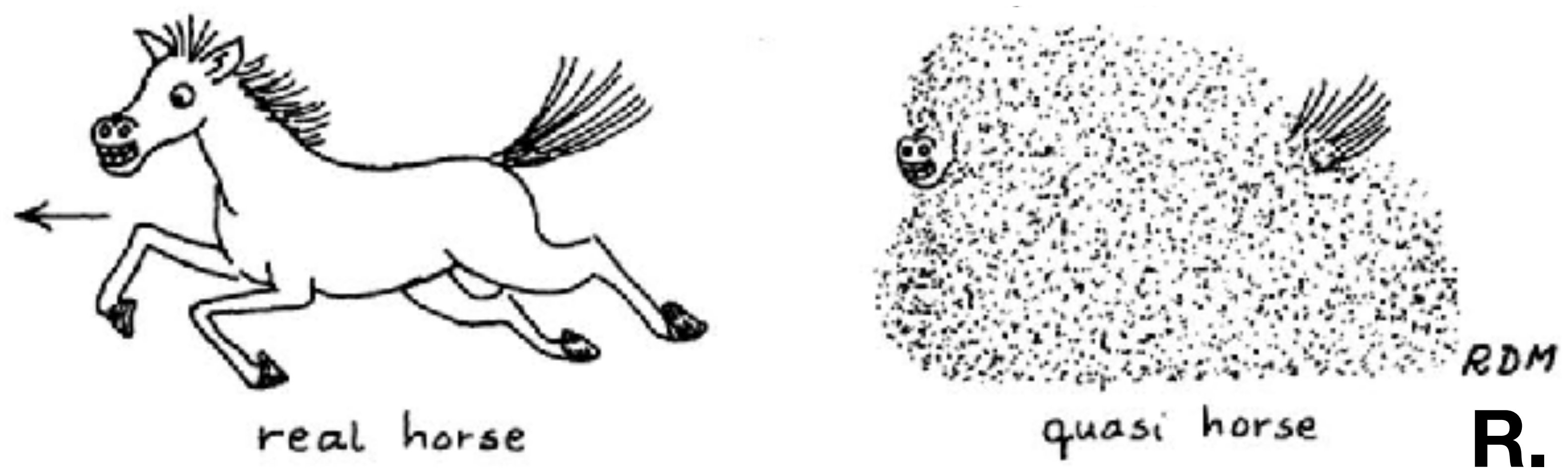
German Research Foundation

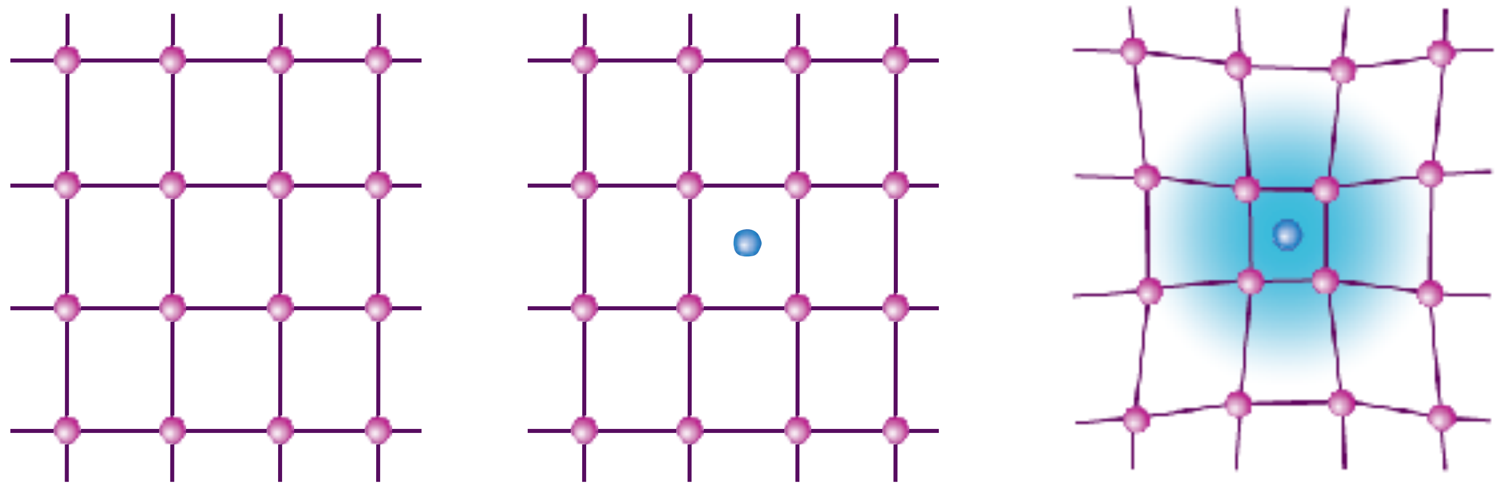
Quasiparticles and phonons



R. Mattuck

L. Landau, Electron motion in crystal lattices,
Phys. Z. Sowjetunion **3**, 664 (1933)

Phonon assisted quasiparticles



lattice external particle quasi-particle

example:

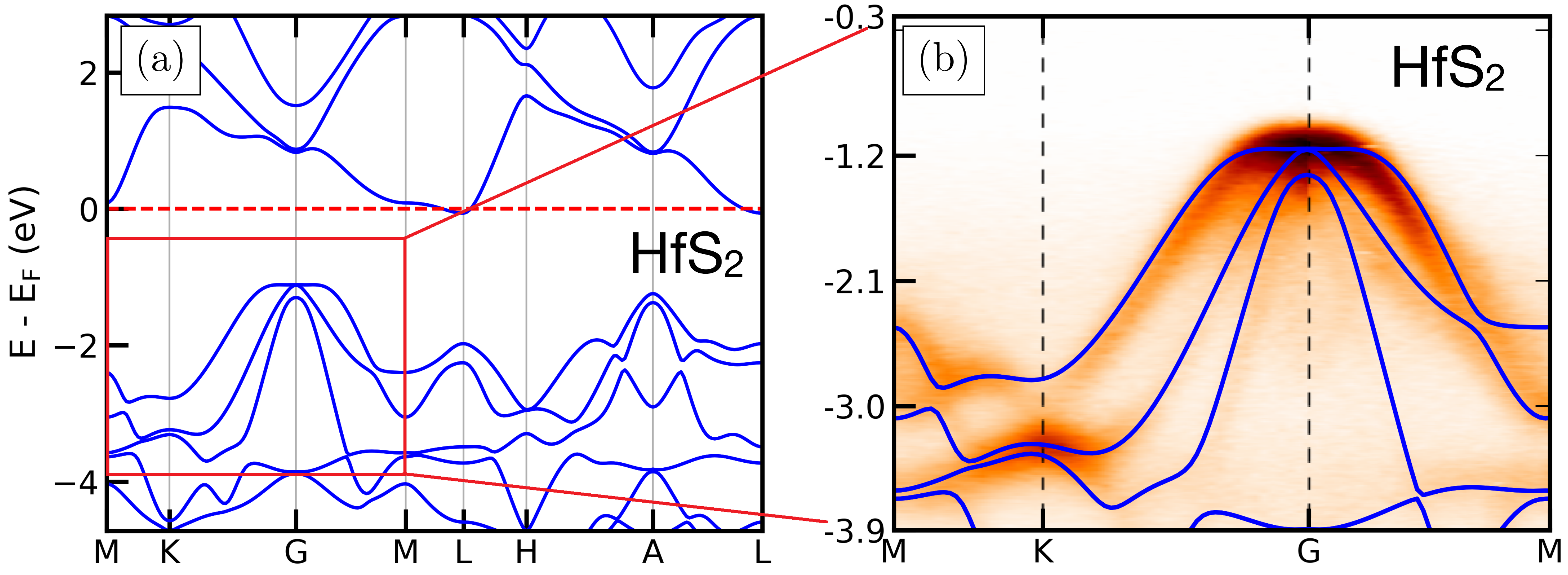
electron (hole) +
lattice distortion
= **polarons**

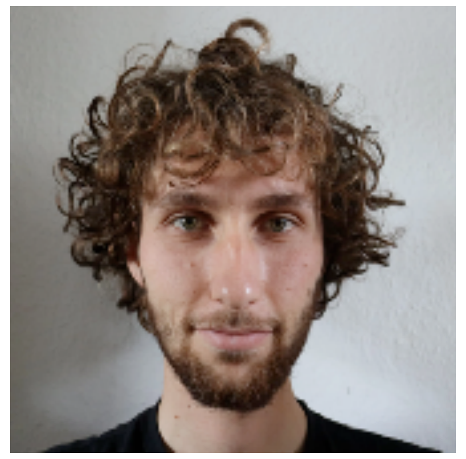
Introduction: Phonon assisted quasiparticles and ab-initio methods

Density functional theory: $n(r) \Leftrightarrow \Psi$ Hohenberg and Kohn, Phys. Rev. (1964) Kohn and Sham, Phys. Rev. (1965)

ELECTRONS

DFT band vs ARPES in HfS_2 (Measurements: S. Mahatha, K. Rossnagel)

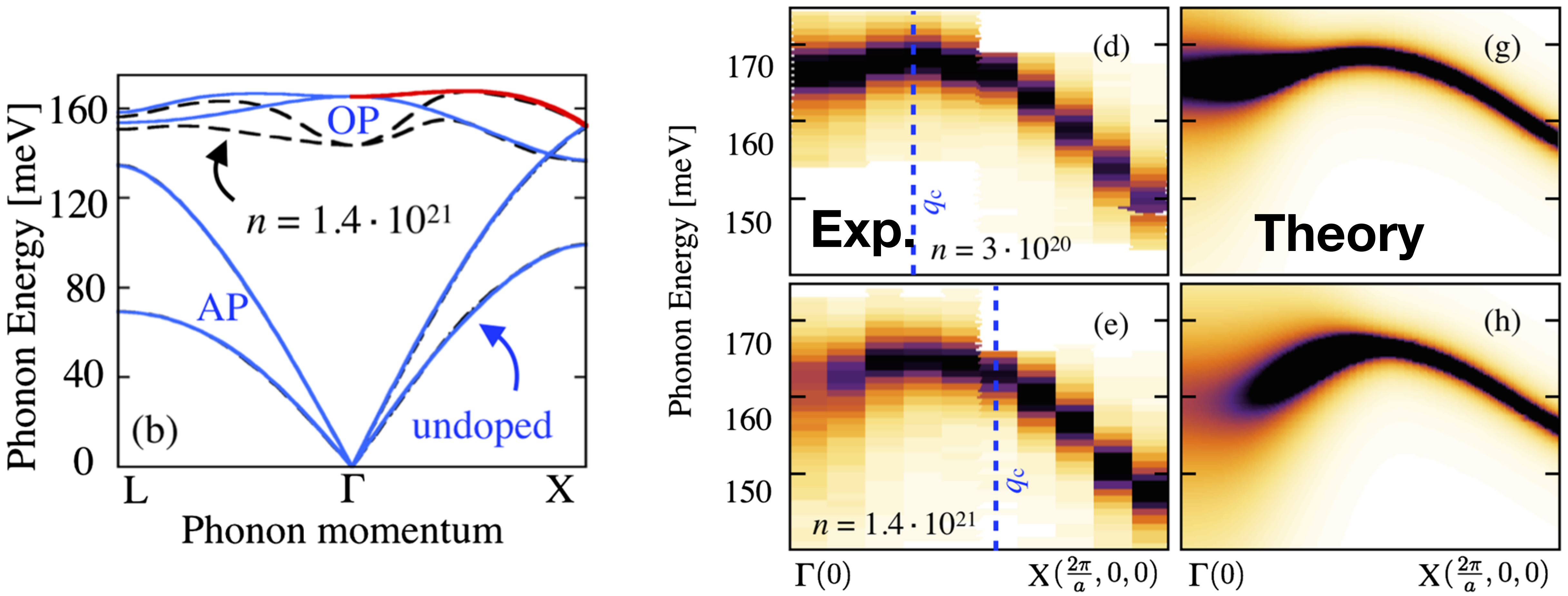




C. Emeis

PHONONS

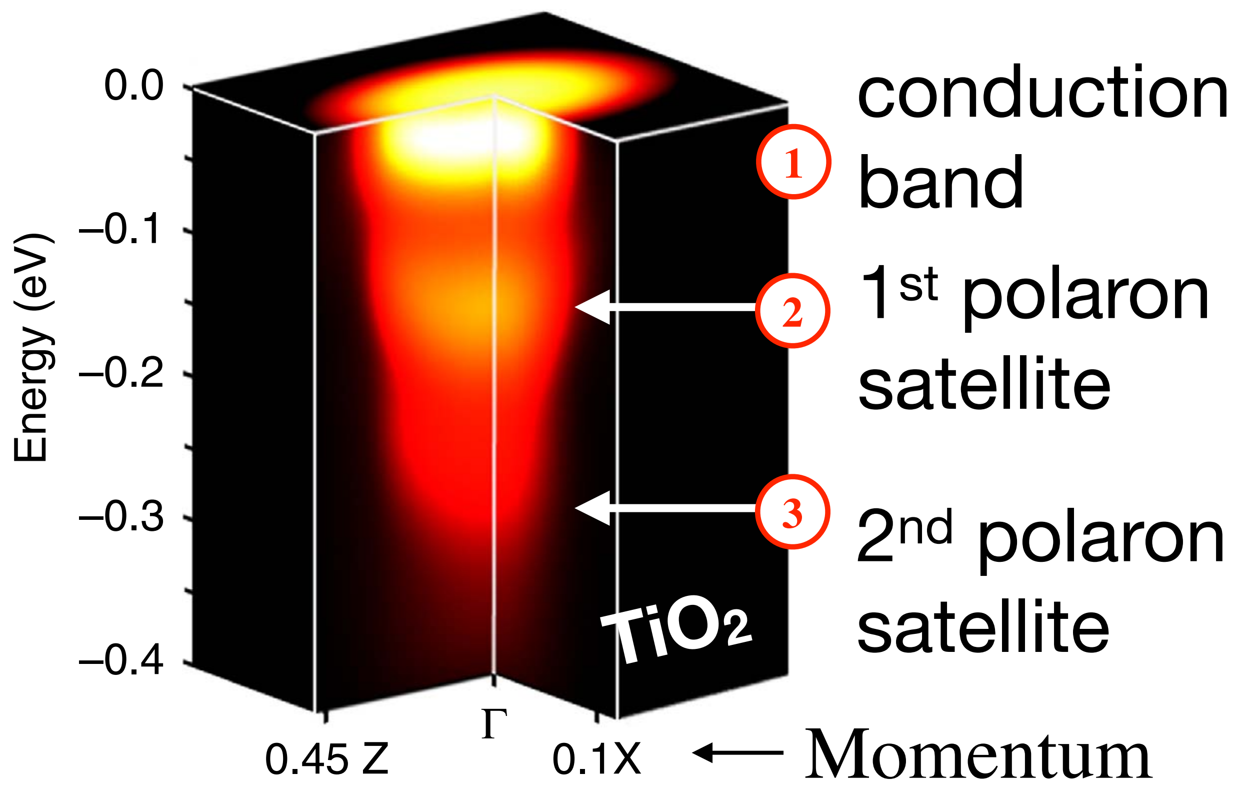
Doping-induced phonon softening in diamond (Measurements: M. Hoesch)



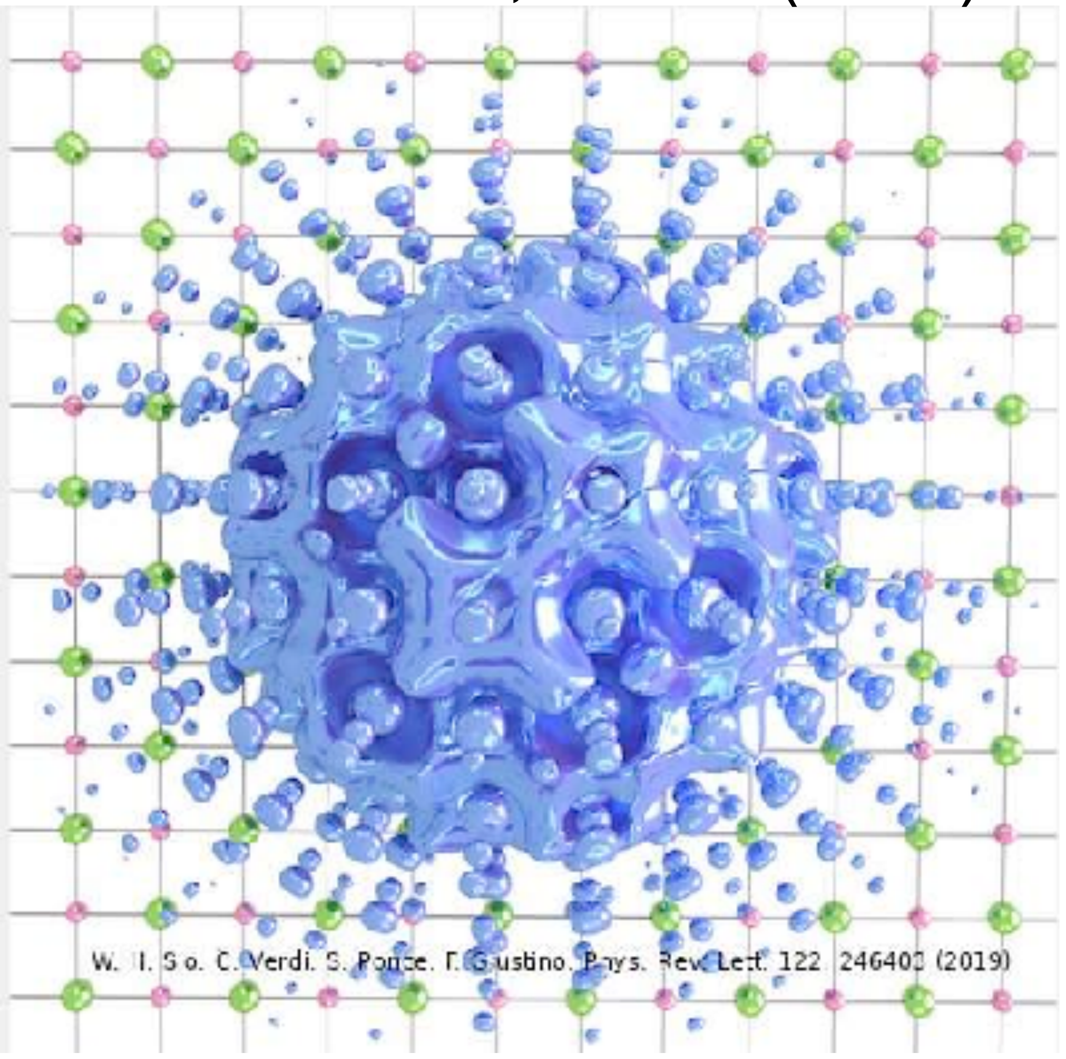
F. Caruso, M. Hoesch, et al. Phys. Rev. Lett. **119**, 017001 (2017)

ELECTRON-PHONON INTERACTIONS

Polarons in photoemission



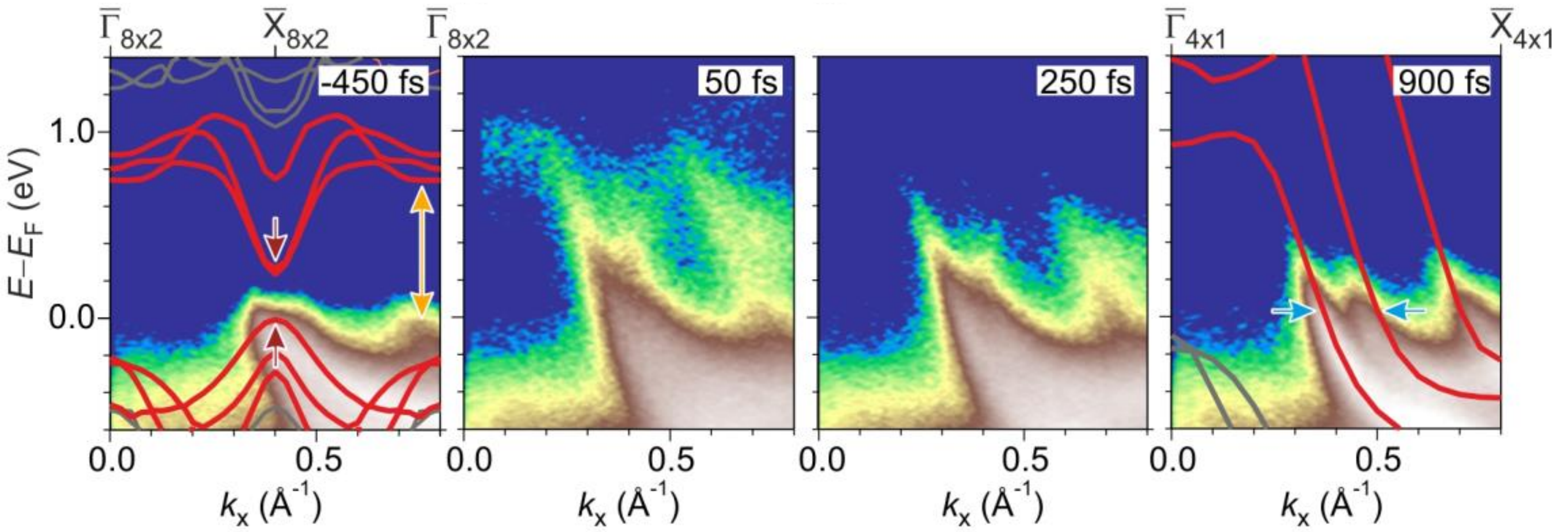
Verdi, Caruso, Giustino, Nature Comm. **8**, 15769 (2017)



Sio, Verdi, Ponce, Giustino, Phys. Rev. Lett. **122**, 246403 (2019)

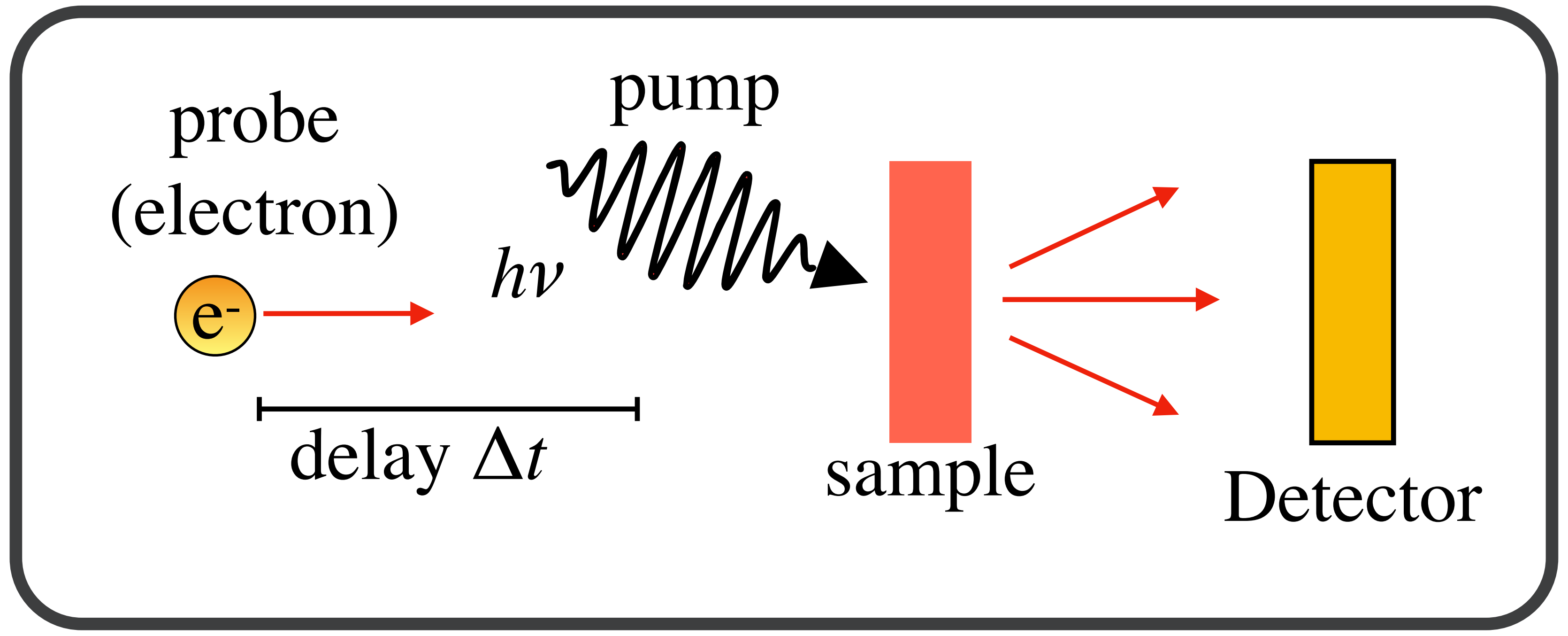
Electron-phonon interactions out of equilibrium: fingerprints in spectroscopy

1. Light-induced structural phase transitions (In nanowires)

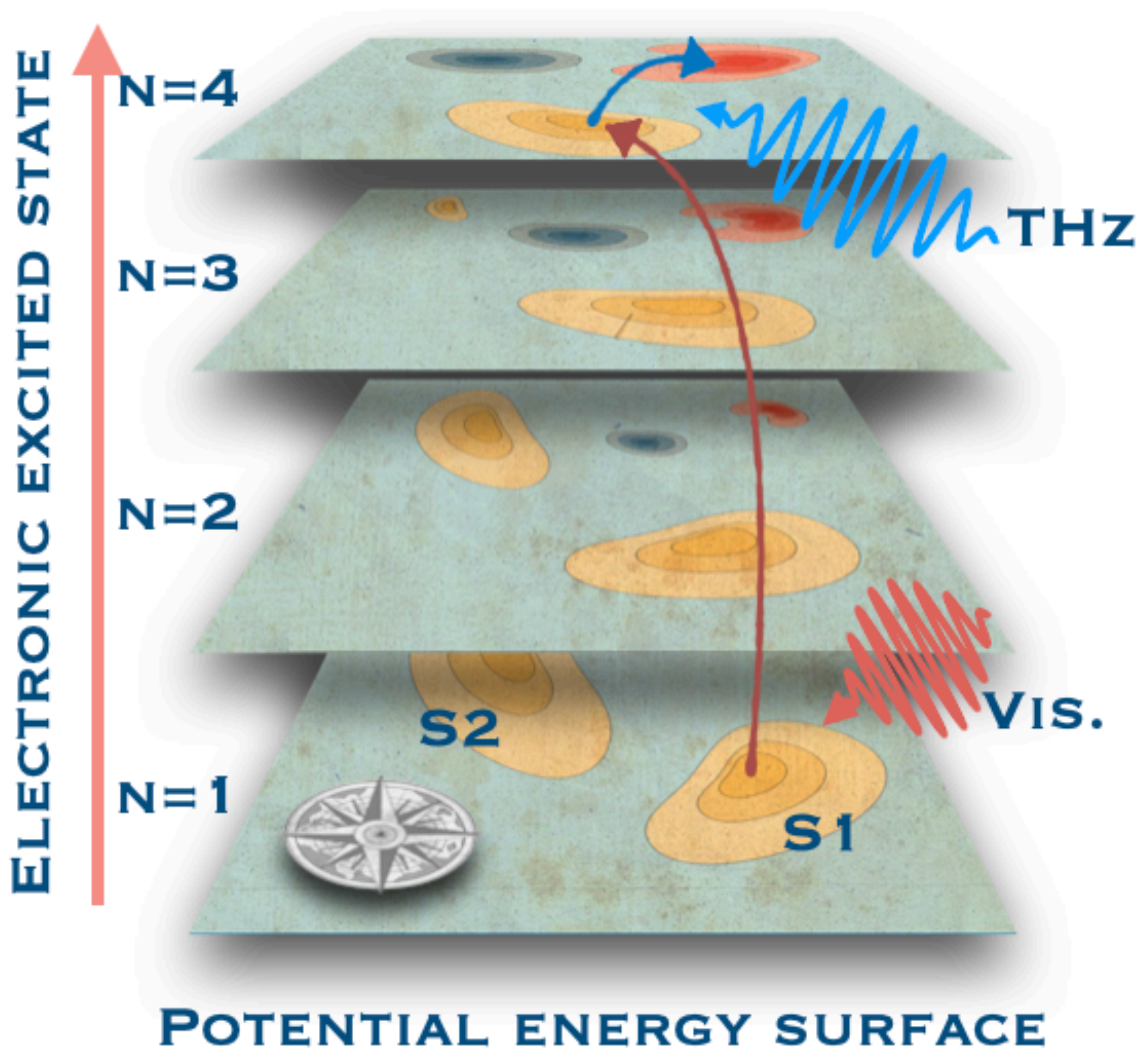


Nicholson, Lücke, Gero Schmidt, Puppin, Rettig, Ernstorfer, Wolf, Science **362**, 821 (2018)

2. Direct imaging of phonons out of equilibrium



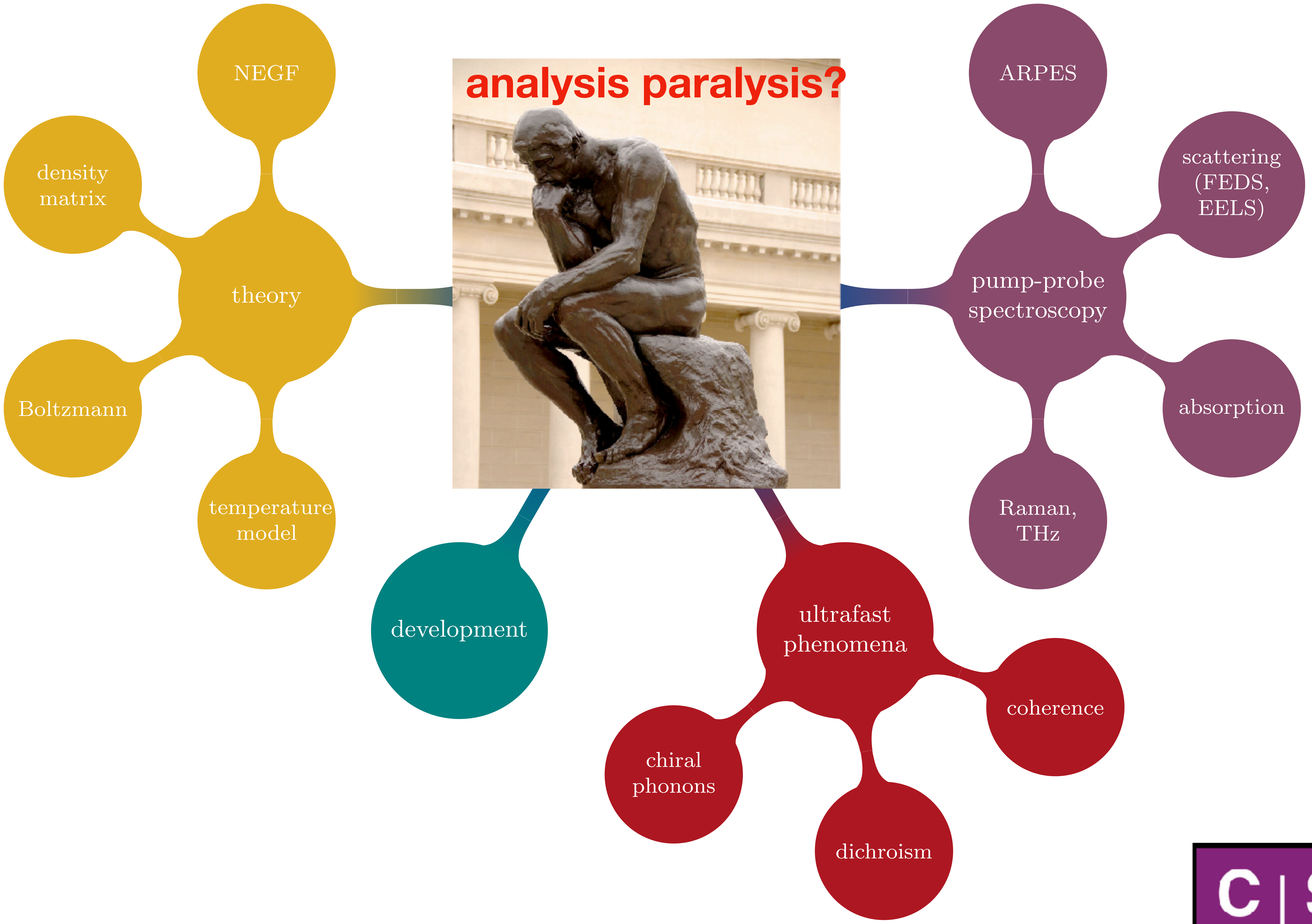
Ultrafast electron diffuse scattering (UEDS)



A challenge for ab-initio approaches:

- Highly-anharmonic lattices
- Phase transitions
- Highly-nonlinear effects (strong fields)
- Several competing mechanisms (electron, spin, phonon)

Challenges in ab-initio theory of ultrafast dynamics



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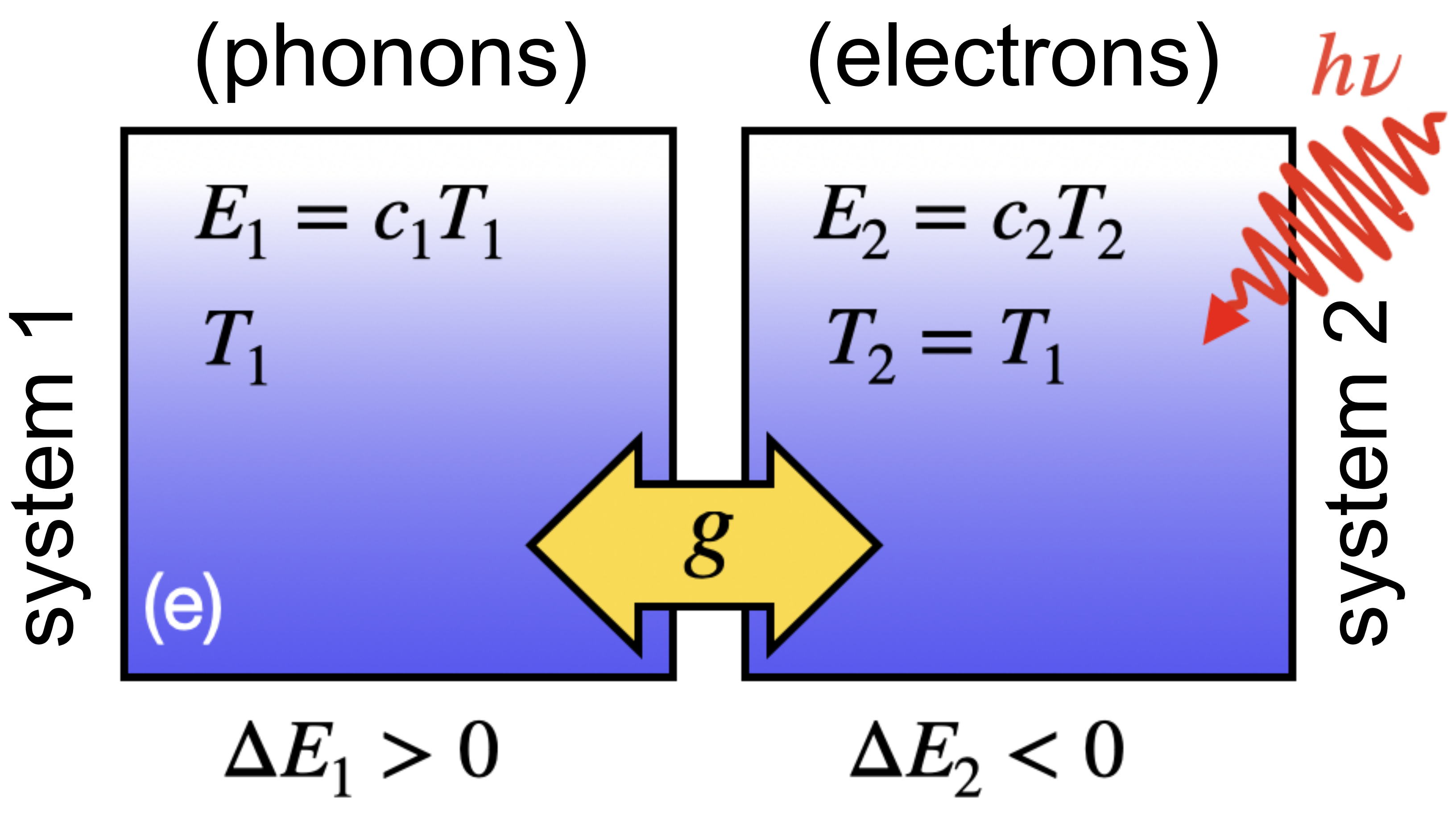
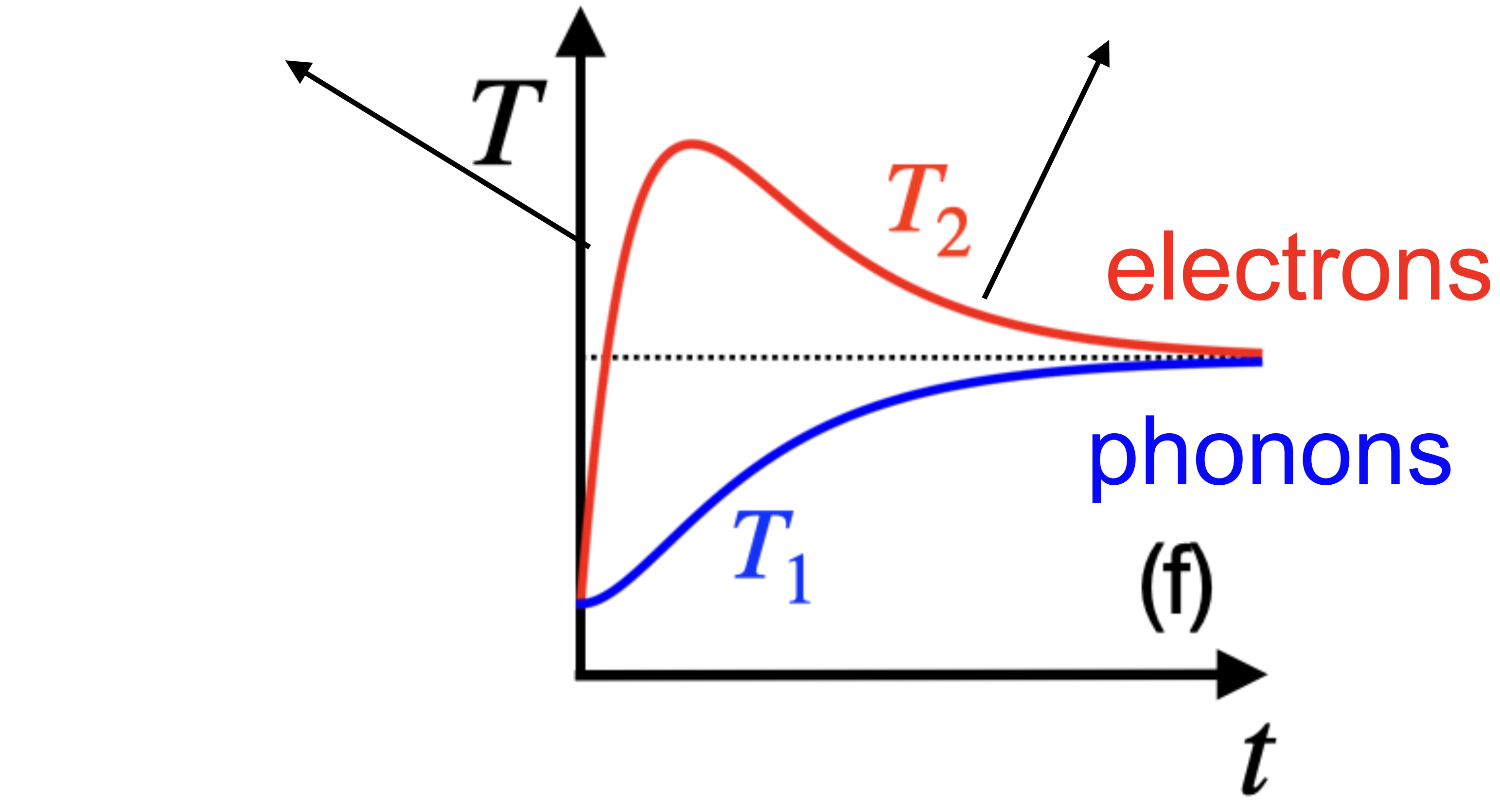
Part 1

Non-equilibrium dynamics of electrons and phonons from first (or second?) principles

Thermalization of electrons and lattice from the two-temperature model





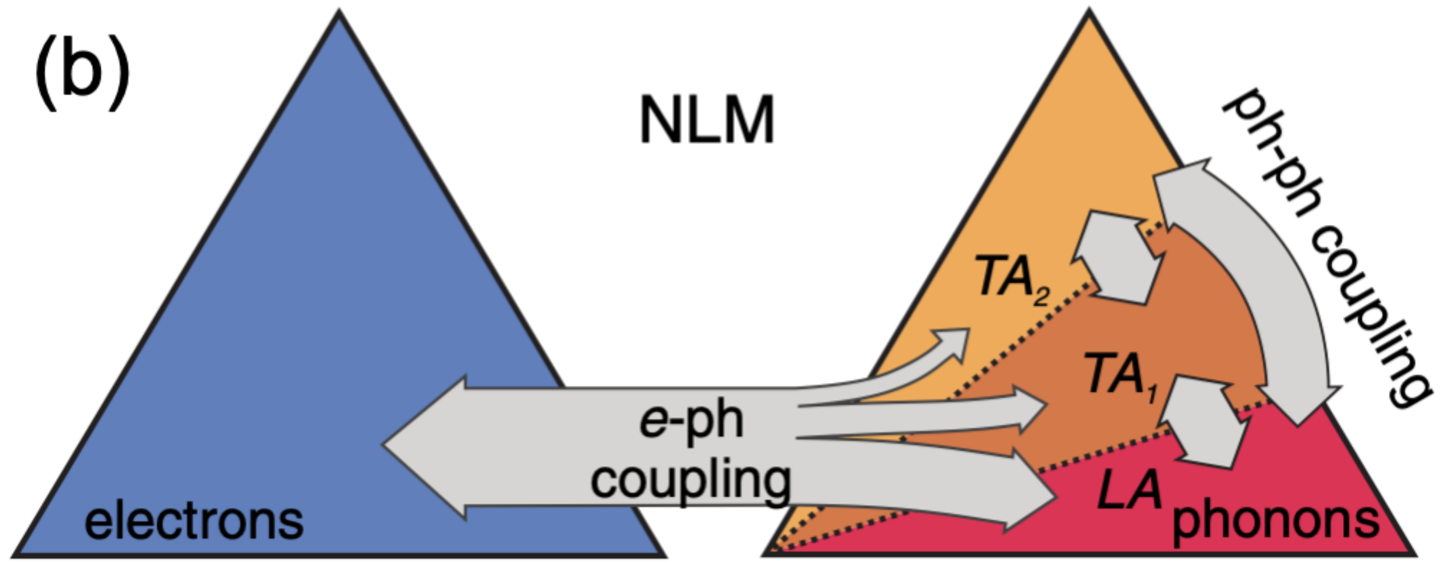
F. Caruso, D. Novko, Adv. Phys. X **7**, 2095925 (2022)

1. Two-temperature model (TTM)

$$\frac{\partial T_{\rm ph}}{\partial t} = \frac{g}{C_{\rm ph}}(T_{\rm el} - T_{\rm ph}) \quad ,$$
$$\frac{\partial T_{\rm el}}{\partial t} = \frac{g}{C_{\rm el}}(T_{\rm ph} - T_{\rm el}) + S(t)$$

Free parameters: g, $C_{\rm ph}$, $C_{\rm el}$
(can be obtained *ab initio*)

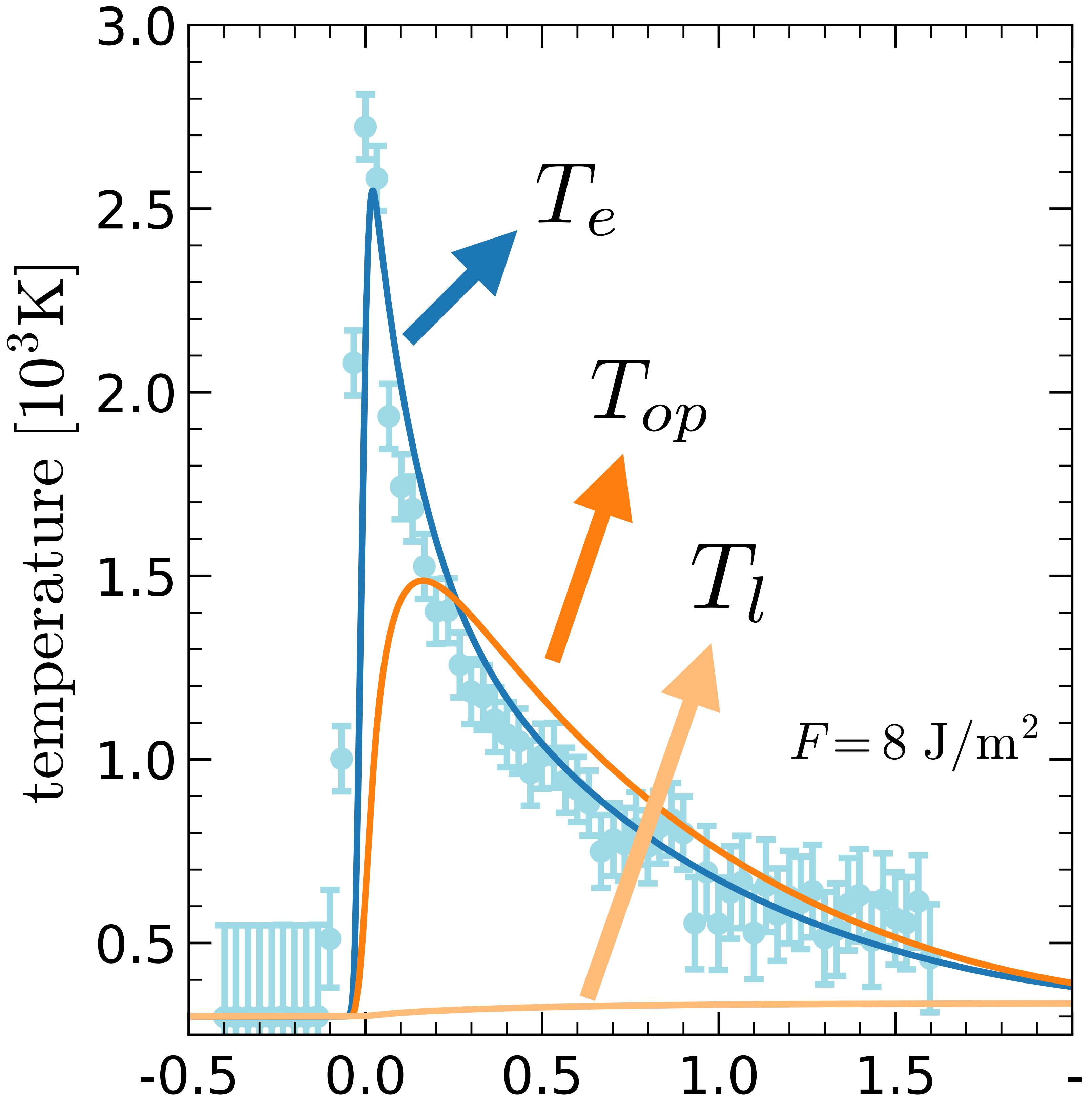
2. Non-thermal lattice model (NLM)



L. Waldecker, R. Bertoni, R. Ernstorfer, J. Vorberger, Phys. Rev. X **6**, 021003 (2016)

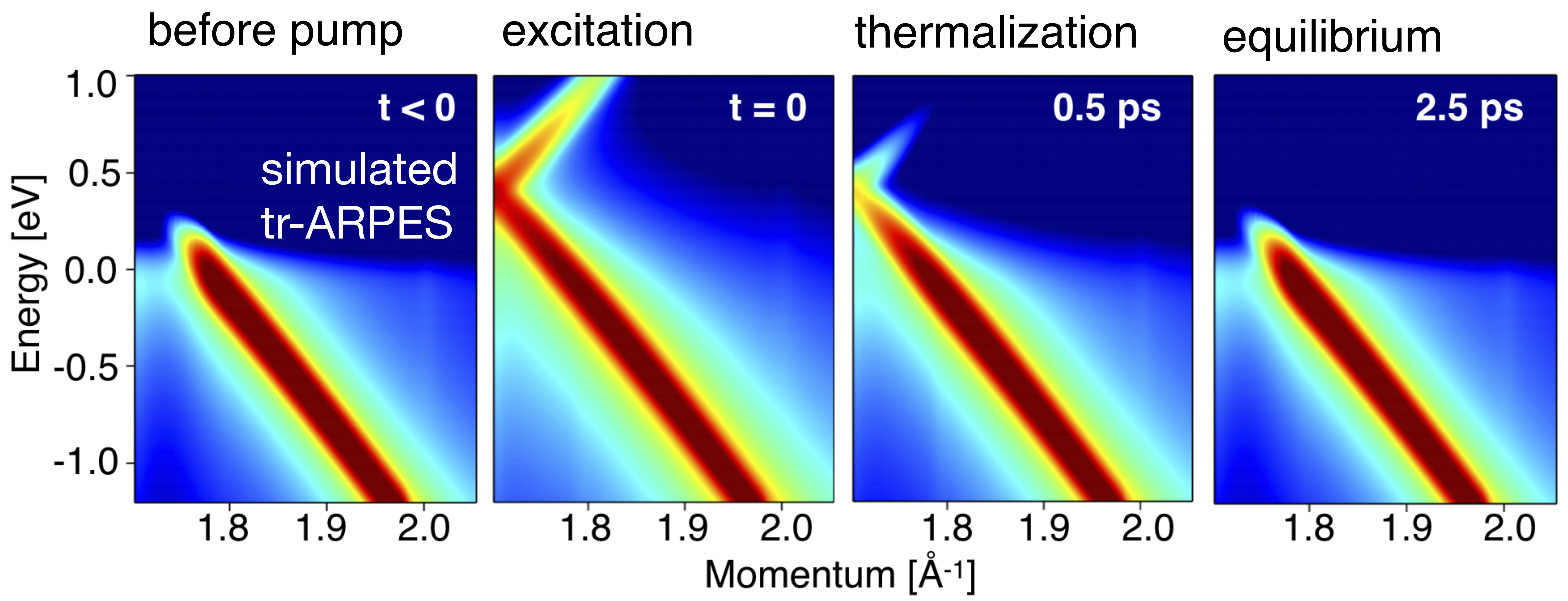
Non-thermal lattice models and ultrafast dynamics in graphene

Calculated vs measured electronic temperature



Delay [ps]

Exp: Gierz, et al. Nature Materials, 12, 1119 (2013)



1. Electron phonon coupling self-energy

Bose / Fermi occupations

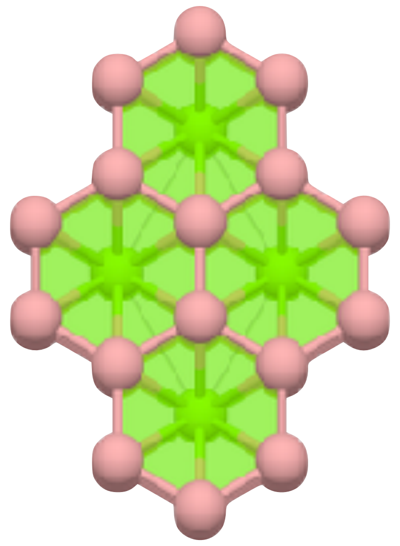
$$\Sigma^{\rm e-ph}_{n\mathbf{k}}(\omega) = \int \frac{d\mathbf{q}}{\Omega_{\rm BZ}} \sum_{m\nu} |g^{\rm e-ph}_{mn\nu}(\mathbf{k},\mathbf{q})|^2 \left[\frac{n_{\mathbf{q}\nu} + f_{m\mathbf{k}+\mathbf{q}}}{\omega - \varepsilon_{m\mathbf{k}+\mathbf{q}} + \omega_{\mathbf{q}\nu} - i\eta} + \frac{n_{\mathbf{q}\nu} + 1 - f_{m\mathbf{k}+\mathbf{q}}}{\omega - \varepsilon_{m\mathbf{k}+\mathbf{q}} - \omega_{\mathbf{q}\nu} - i\eta} \right]$$

2. Spectral function:

$$A_{n\mathbf{k}}(\omega, T) = \frac{1}{\pi} \frac{|{\rm Im}\,\Sigma^{\rm e-ph}_{n\mathbf{k}}(\omega)|}{[\omega - \varepsilon_{n\mathbf{k}} - {\rm Re}\,\Sigma^{\rm e-ph}_{n\mathbf{k}}(\omega)]^2 + [{\rm Im}\,\Sigma^{\rm e-ph}_{n\mathbf{k}}(\omega)]^2}$$

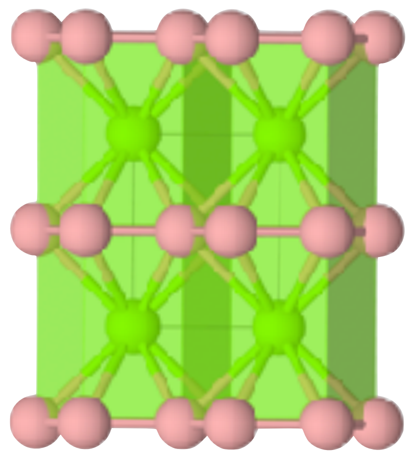
F. Caruso, D. Novko, C. Draxl, Phys. Rev. B **101,** 035128 (2020)

Transient phonon softening (Kohn anomaly) in MgB_2



Facts about MgB_2

- Metal with hexagonal crystal structure
- BCS superconductor (T_c = 39 K)
- Highest known T_c for BCS superconductivity
- Strongly coupled E_{2g} modes

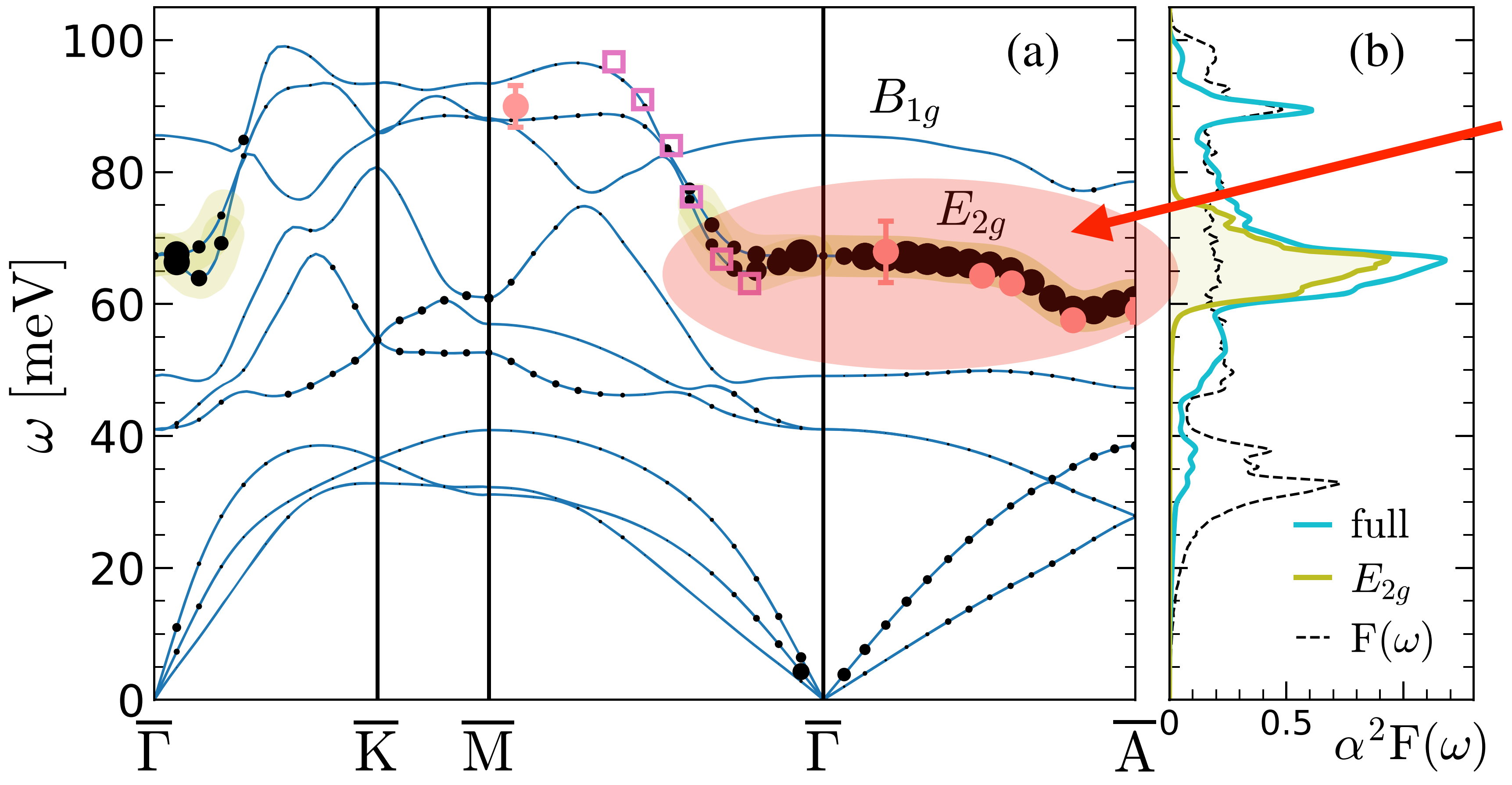


$$\Omega^2_{E_{2g}} = \omega^2_{E_{2g}} + 2\omega_{E_{2g}}\bar{\Pi}(\Omega_{E_{2g}};\{T\}),$$

"dressed" phonon frequency

bare phonon frequency

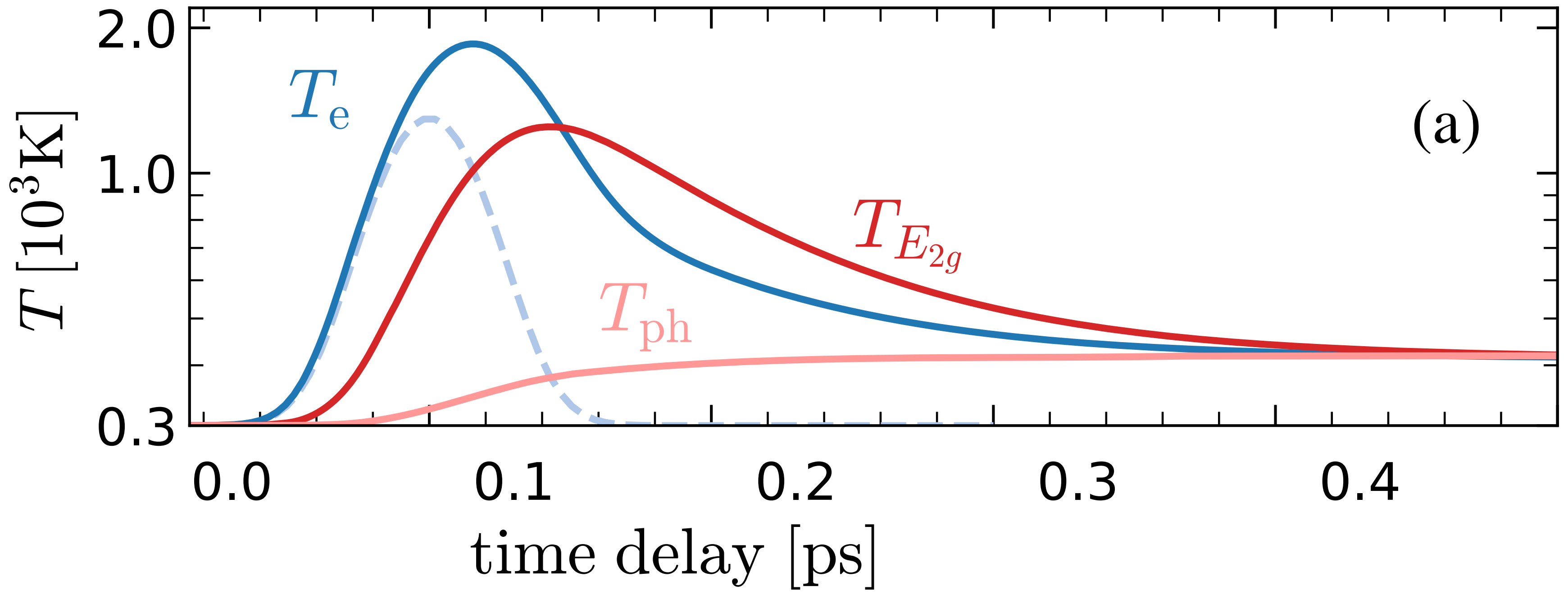
phonon self-energy



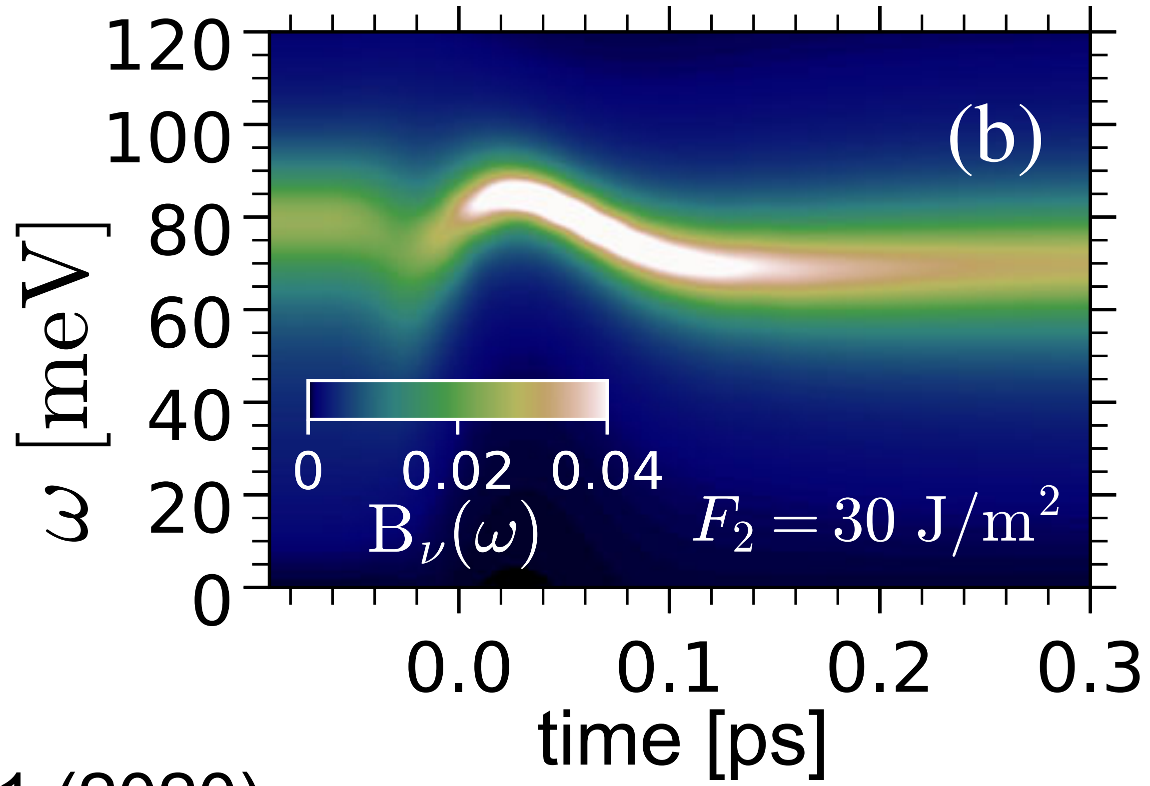
Kohn anomaly

Phonon spectral function

$$A_{\mathbf{q}\nu}(\omega) = \pi^{-1}\mathrm{Im}\left[\frac{2\omega_{\mathbf{q}\nu}}{\omega^2 - \omega^2_{\mathbf{q}\nu} - 2\omega_{\mathbf{q}\nu}\Pi^{\mathrm{NA}}_{\mathbf{q}\nu}(\omega)}\right]$$



Phonon spectral function @ Γ



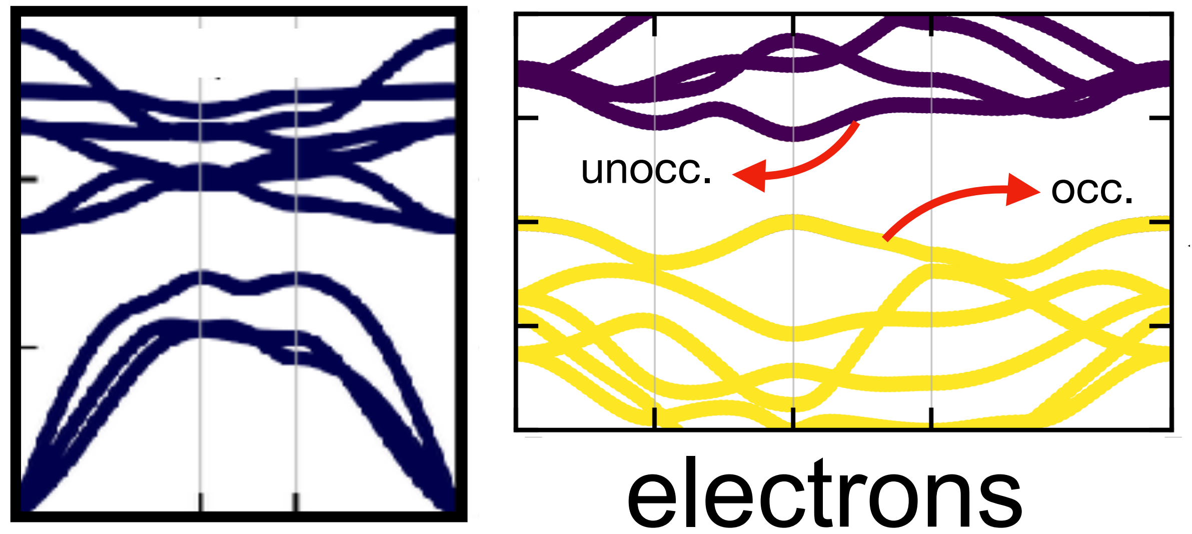
Transient change of the E_{2g} phonon frequency due to the EPI: **"transient Kohn anomaly"**

D. Novko, F. Caruso, C. Draxl, E. Cappelluti, Phys. Rev. Lett. **124**, 077001 (2020)

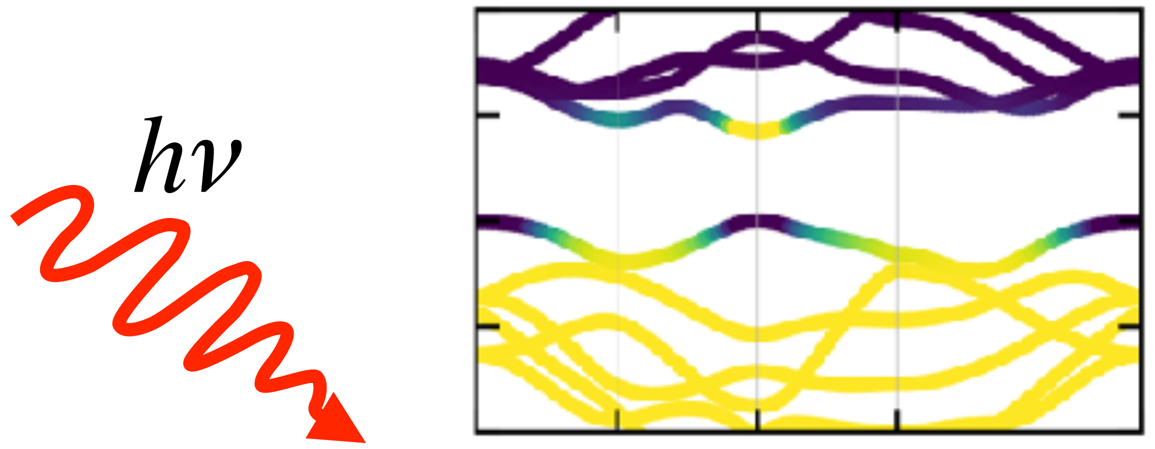
Workflow for ultrafast dynamics simulations

1. Band structure (DFT)
   Phonon dispersion (DFPT)
2. Model coupling to light pulse
   (or excited state Ansatz)
3. Time propagation
4. Transient many-body effects
   and emergent phenomena
5. Theoretical spectroscopy:
   tr-ARPES, Raman, scattering

1.



2.



3.

two-temperature model

time-dependent Boltzmann equation

non-equilibrium Green's functions

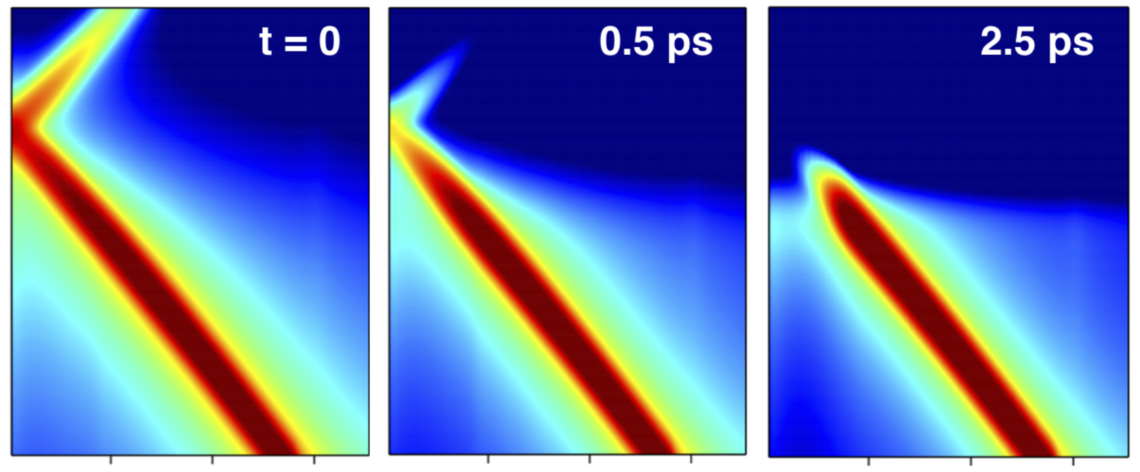
4.

Many-body perturbation theory and Green's functions

$$G(1,2) = -i\langle\Psi_0|\hat{T}\hat{\psi}(1)\hat{\psi}^\dagger(2)|\Psi_0\rangle$$

5.

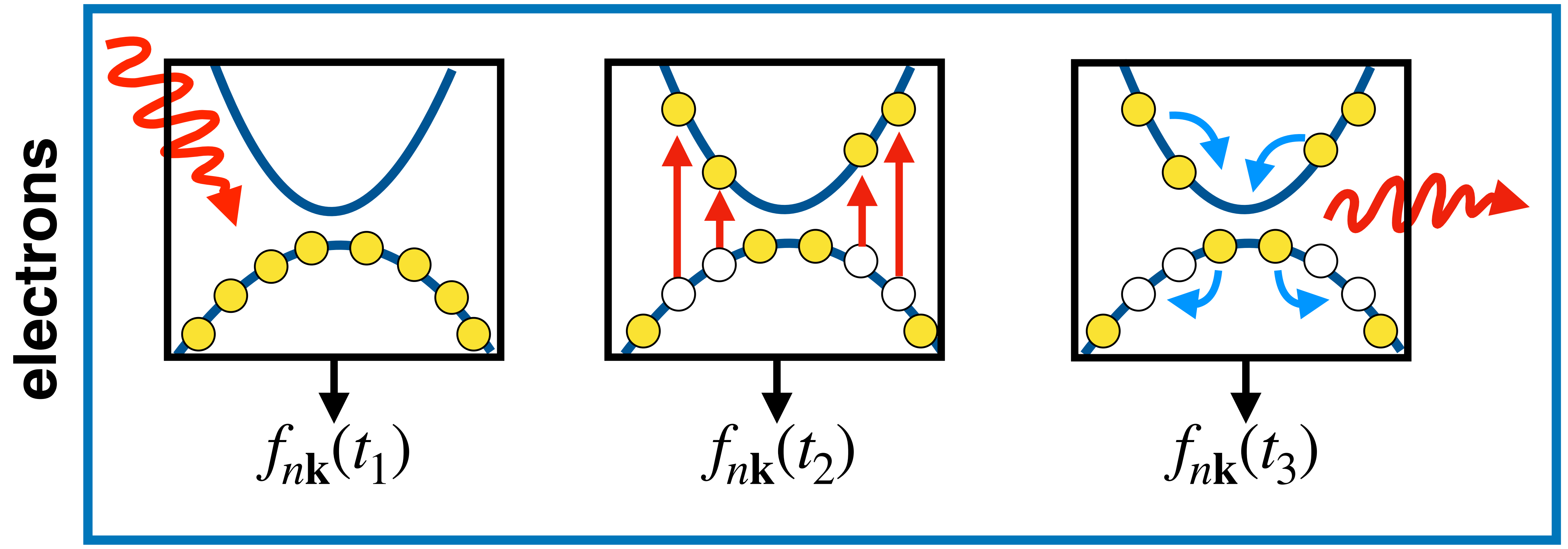
tr-ARPES

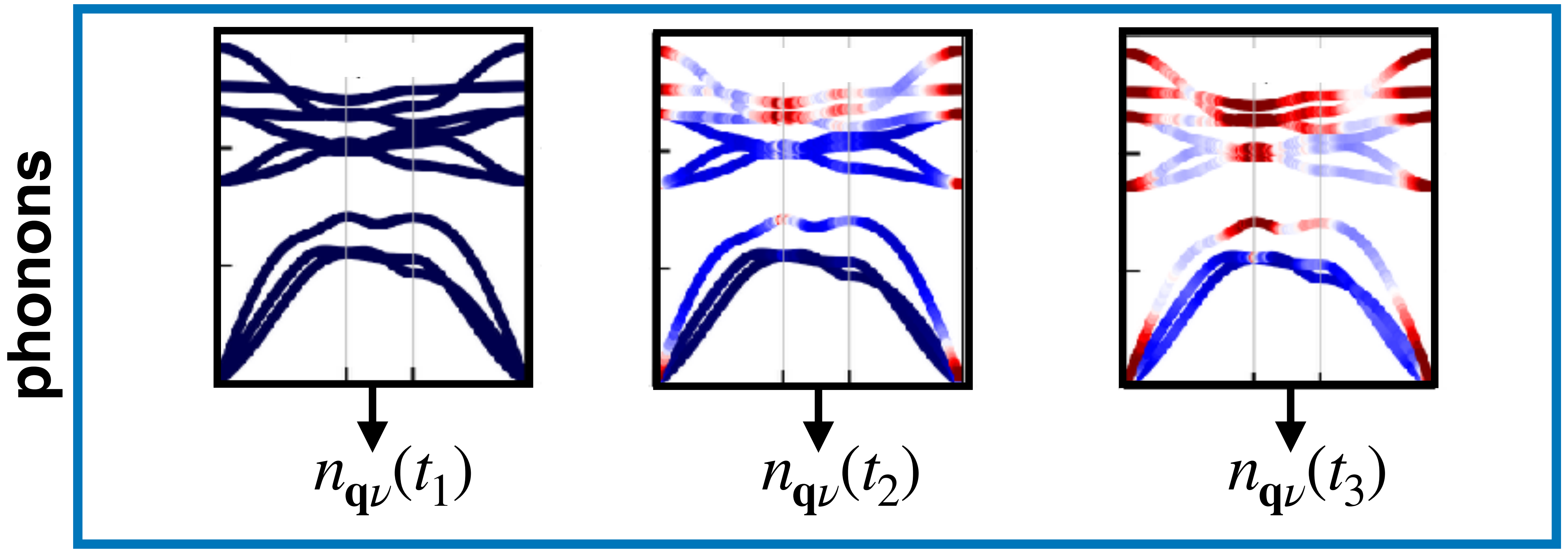


Review: F. Caruso, D. Novko, Adv. Phys. X **7**, 2095925 (2022)

Time-dependent Boltzmann equation

Key assumption: Electronic and vibrational excitations are described via the corresponding distribution functions





Time-dep. Boltzmann equation (TDBE)

$$\frac{\partial f_{n\mathbf{k}}}{\partial t} = I^{\mathrm{e-ph}}_{n\mathbf{k}}[f,n] + I^{\mathrm{light}}_{n\mathbf{k}}[f] + I^{\mathrm{e-e}}_{n\mathbf{k}}[f]$$

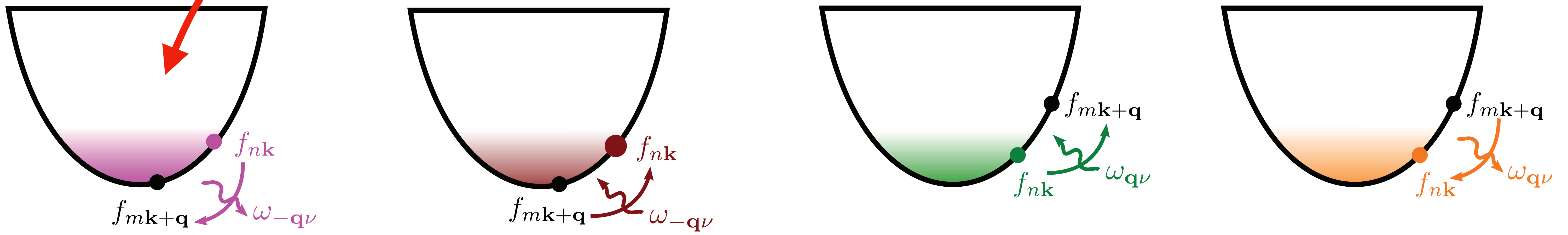
$$\frac{\partial n_{\mathbf{q}\nu}}{\partial t} = I^{\mathrm{e-ph}}_{\mathbf{q}\nu}[f,n] + I^{\mathrm{ph-ph}}_{\mathbf{q}\nu}[n]$$

- solved via Euler or Runge-Kutta algorithms.
- scattering rates are obtained ab initio

Example: electron-phonon scattering rate

$$I^{\mathrm{e-ph}}_{n\mathbf{k}}[f,n] = \frac{2\pi}{\hbar}\sum_{m\nu}\int \frac{d\mathbf{q}}{\Omega_{\mathrm{BZ}}} |g_{mn\nu}(\mathbf{k},\mathbf{q})|^2$$
$$\times\{(1-f_{n\mathbf{k}})f_{m\mathbf{k}+\mathbf{q}}\delta(\varepsilon_{n\mathbf{k}}-\varepsilon_{m\mathbf{k}+\mathbf{q}}+\hbar\omega_{\mathbf{q}\nu})(1+n_{\mathbf{q}\nu})$$
$$+(1-f_{n\mathbf{k}})f_{m\mathbf{k}+\mathbf{q}}\delta(\varepsilon_{n\mathbf{k}}-\varepsilon_{m\mathbf{k}+\mathbf{q}}-\hbar\omega_{\mathbf{q}\nu})n_{\mathbf{q}\nu}$$
$$-f_{n\mathbf{k}}(1-f_{m\mathbf{k}+\mathbf{q}})\delta(\varepsilon_{n\mathbf{k}}-\varepsilon_{m\mathbf{k}+\mathbf{q}}-\hbar\omega_{\mathbf{q}\nu})(1+n_{\mathbf{q}\nu})$$
$$-f_{n\mathbf{k}}(1-f_{m\mathbf{k}+\mathbf{q}})\delta(\varepsilon_{n\mathbf{k}}-\varepsilon_{m\mathbf{k}+\mathbf{q}}+\hbar\omega_{\mathbf{q}\nu})n_{\mathbf{q}\nu}\}$$

electron-phonon matrix element



Review: F. Caruso, D. Novko, Adv. Phys. X **7**, 2095925 (2022)

Ultrafast dynamics from first principles

WANNIER90

Wannier-function interpolation

G. Pizzi et al.,
J. Phys. Cond. Matt. 32, 165902 (2020)

EPW

Electron-phonon coupling

S. Poncé et al.,
Comp. Phys. Comm. (2017)

Coupled-dynamics of electrons and phonons

$$\frac{\partial f_{n\mathbf{k}}}{\partial t} = I_{n\mathbf{k}}^{\mathrm{e-ph}}[f, n]$$

$$\frac{\partial n_{\mathbf{q}\nu}}{\partial t} = I_{\mathbf{q}\nu}^{\mathrm{e-ph}}[f, n] + I_{\mathbf{q}\nu}^{\mathrm{ph-ph}}[n]$$

QUANTUMESPRESSO

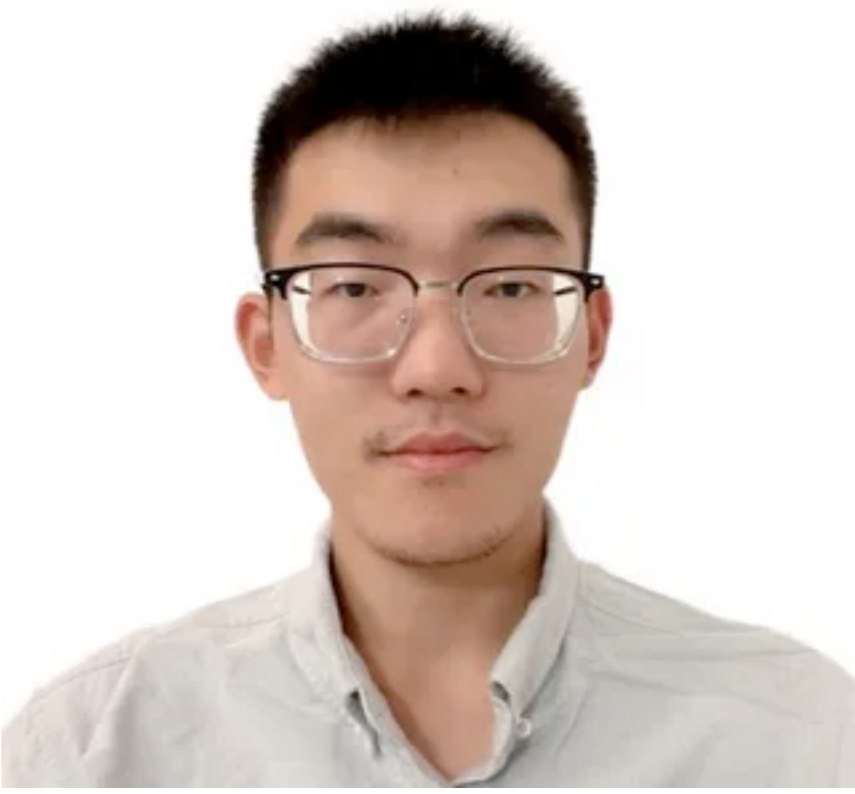
Density functional theory

P. Giannozzi et al.,
J. Phys.: Condens. Matter 29, 465901 (2017)

声 BTE

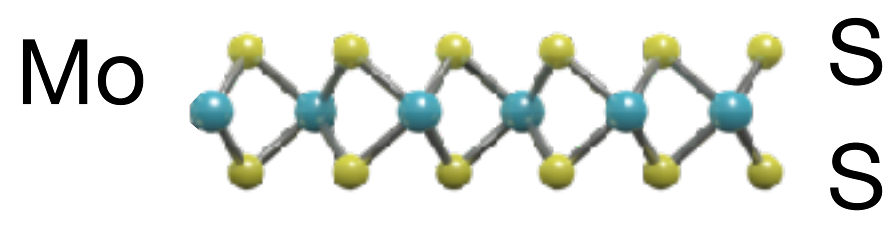
Third-order force constant

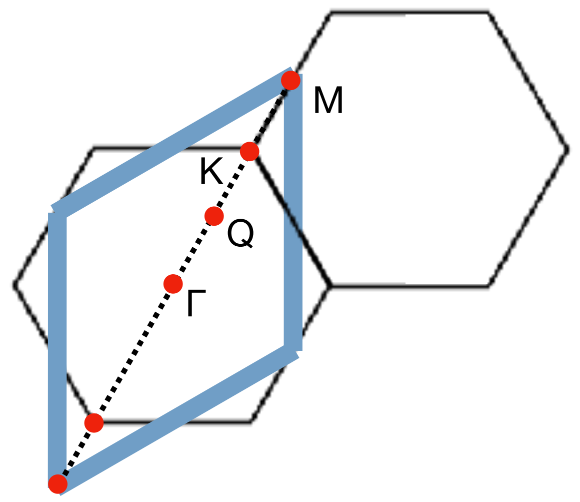
W. Li et al., Comp. Phys. Comm. 185, 1747 (2014)

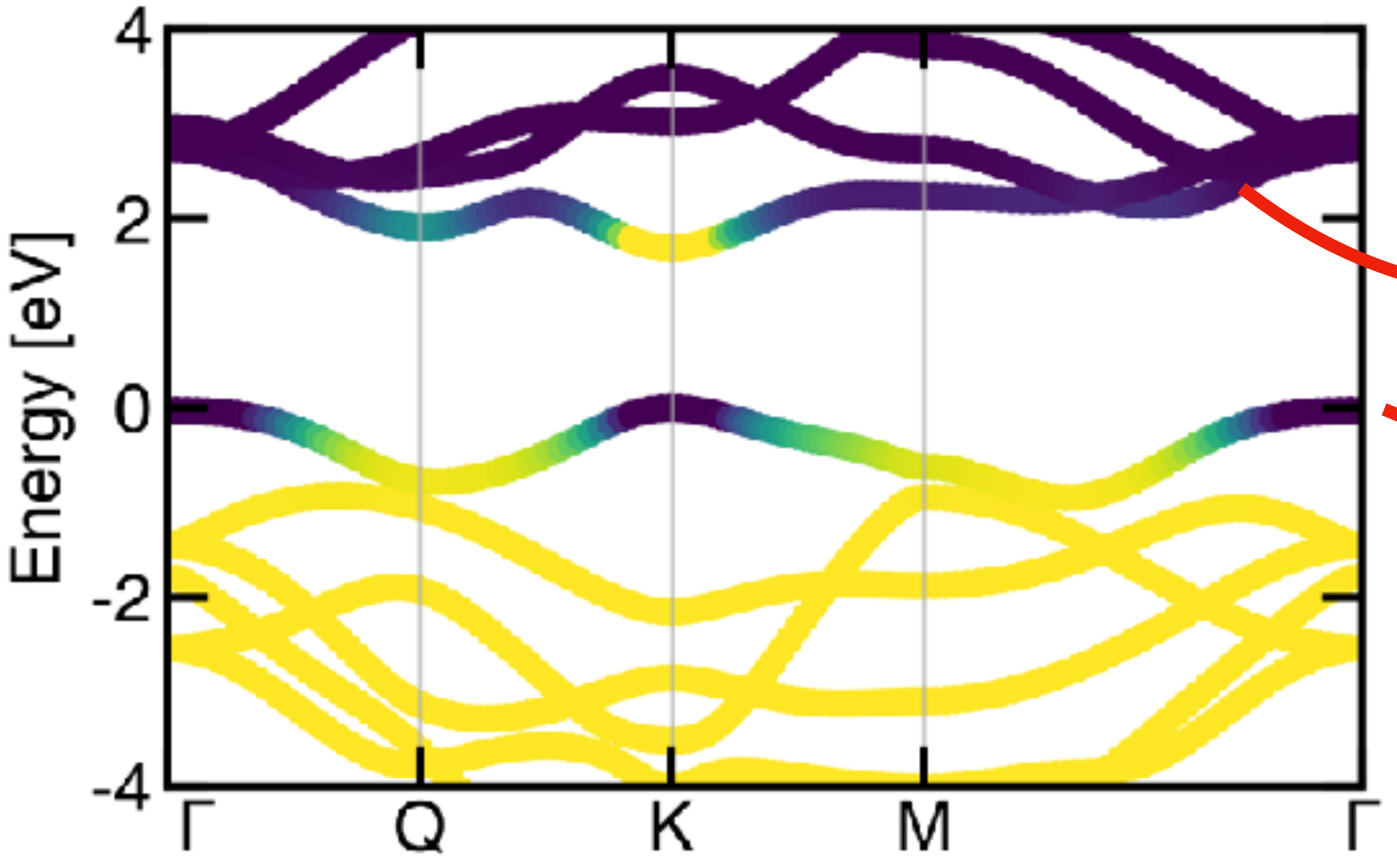


Yiming Pan

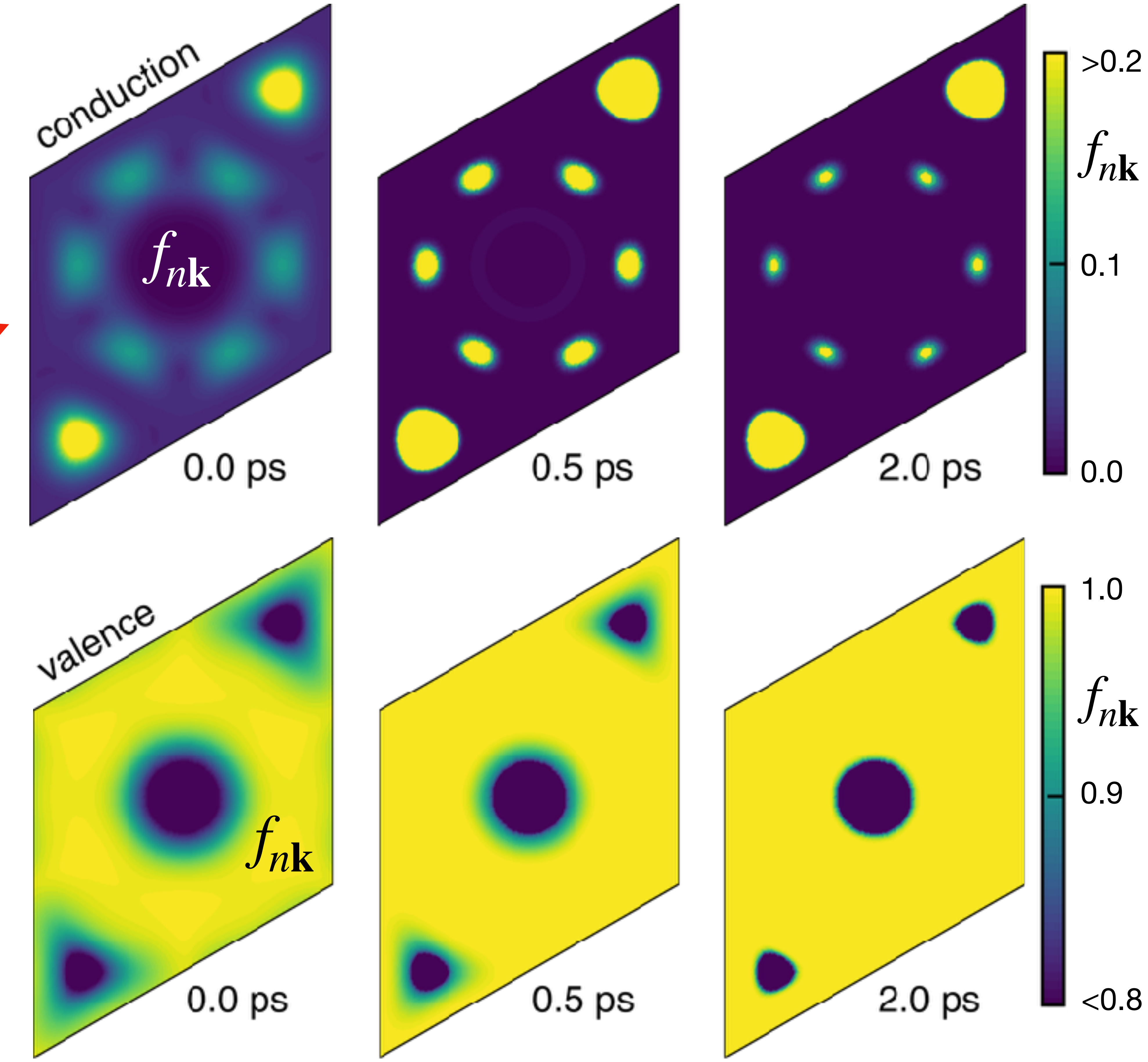
Nonequilibrium ELECTRON dynamics in monolayer MoS_2

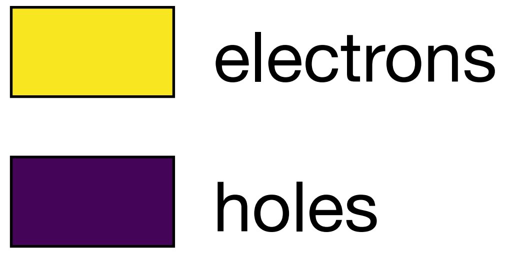






Electron and hole occupation in the Brillouin zone





Estimated timescales for electron relaxation:	$< 2\,\mathrm{ps}$
Experiments from (*) (fs-electron diffraction):	$1.7 \pm 0.3\,\mathrm{ps}$

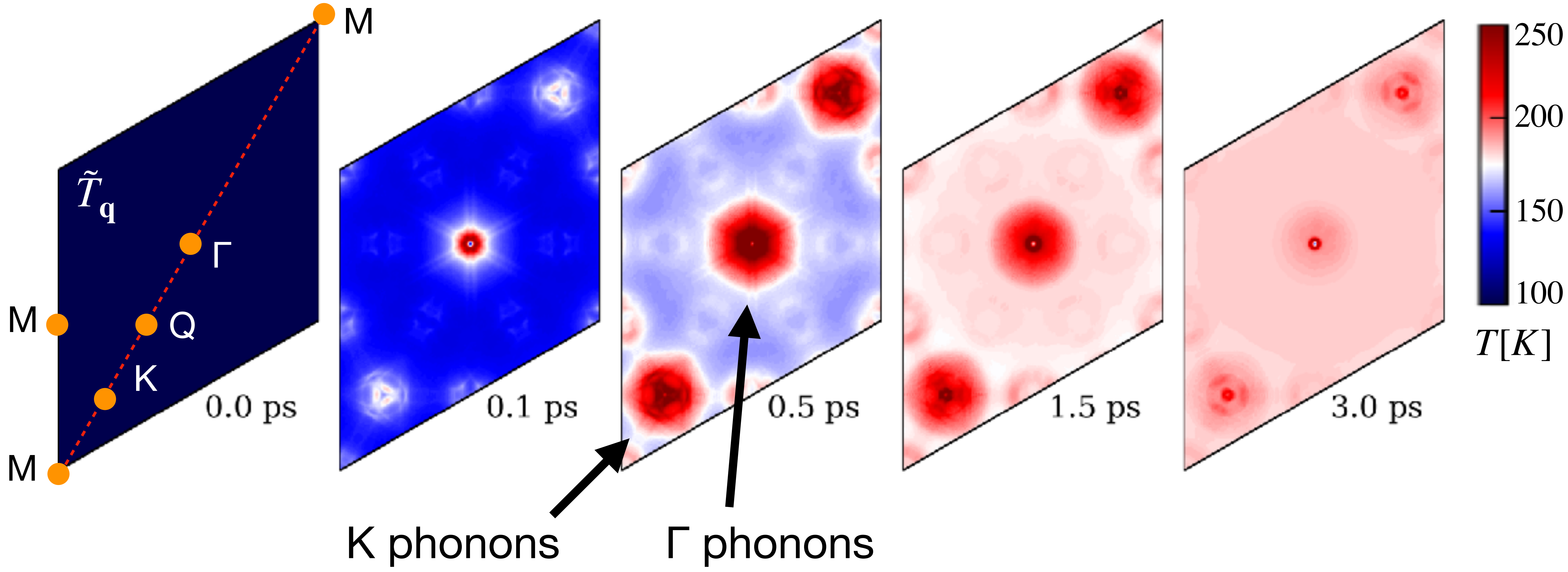
(*) Mannebach et al., Nano Lett. 15, 6889 (2015)

$$\frac{\partial f_{n\mathbf{k}}}{\partial t} = I^{\mathrm{e-ph}}_{n\mathbf{k}}[f, n]$$

F. Caruso, J. Phys. Chem. Lett. **12**, 1274 (2021)

Nonequilibrium PHONON dynamics in monolayer MoS_2

Phonon TEMPERATURE in the Brillouin zone

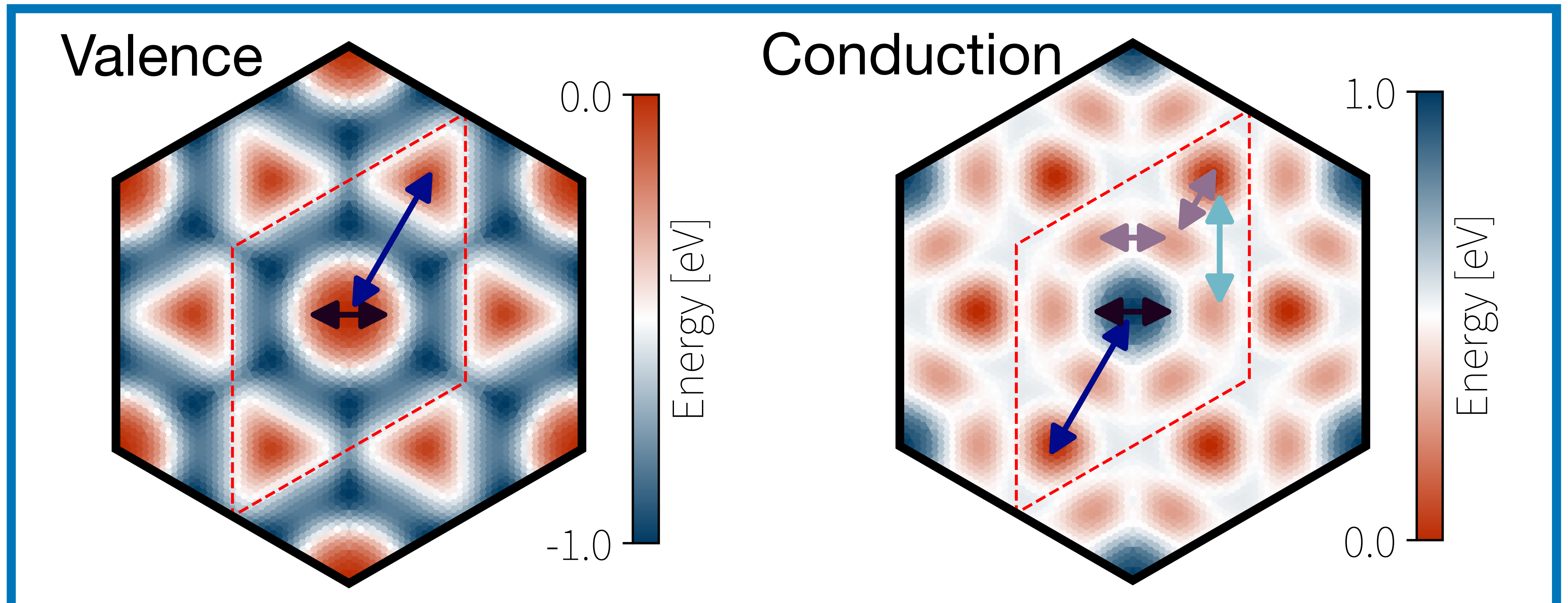


Phonon temperature

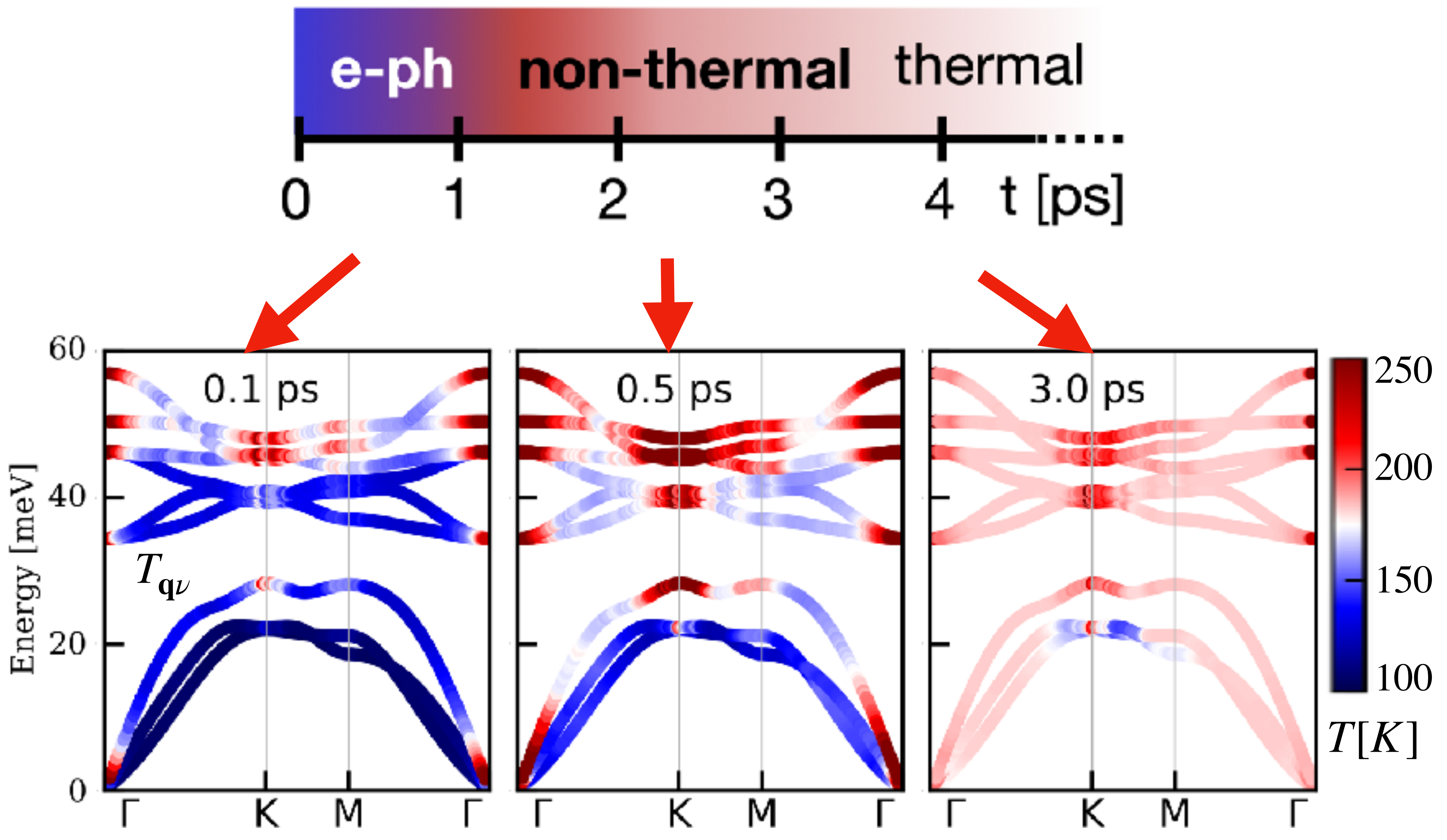
$$T_{\mathbf{q}\nu} = \hbar\omega_{\mathbf{q}\nu}[k_{\mathrm{B}}\ln(1+n_{\mathbf{q}\nu})]^{-1}$$

(averaged for all phonon polarizations)

Momentum selectivity in the phonon emission:



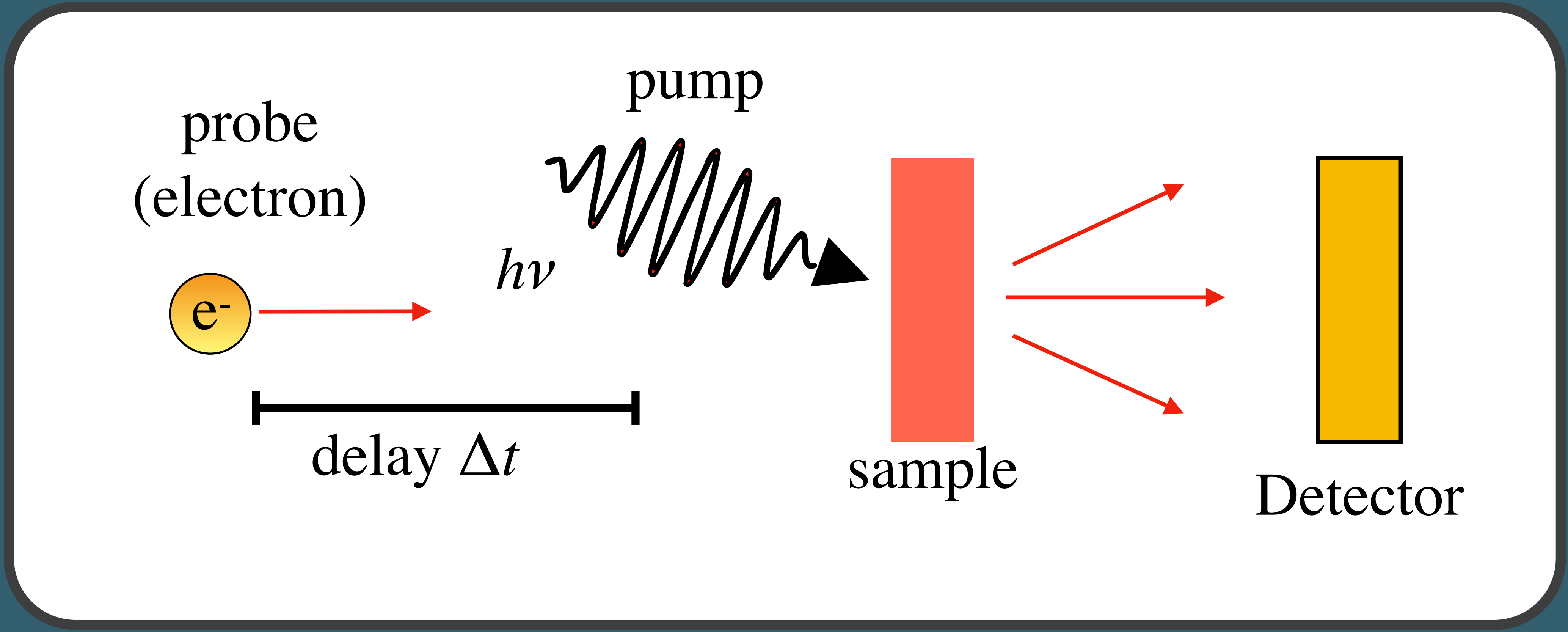
Emission K and Γ phonons are allowed



F. Caruso, J. Phys. Chem. Lett. **12**, 1274 (2021)

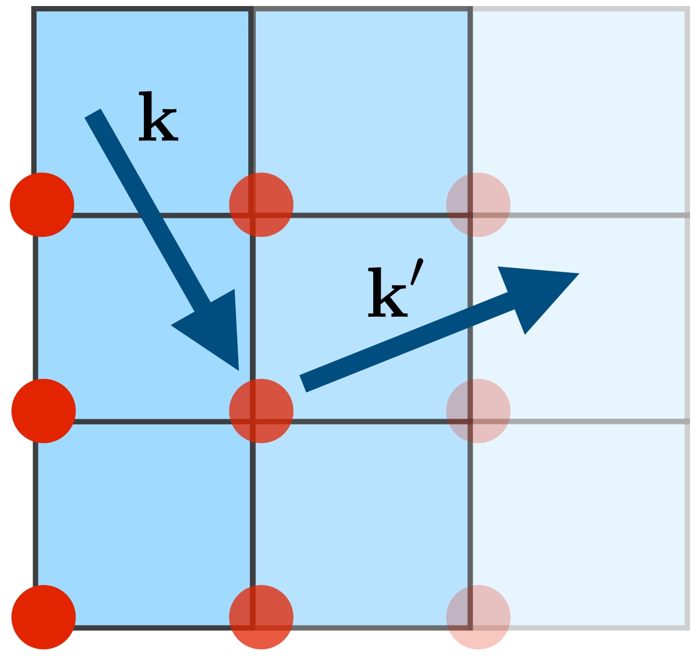
Part 2

Direct imaging non-equilibrium phonon populations via ultrafast electron diffuse scattering (UEDS)



A crash course in the theory diffraction

Static lattice (1 atom per cell)



Bragg's law:

$$A(\mathbf{q}) = \sum_{p}^{N_p} f_0 \, \exp\left[i\mathbf{q}\cdot\mathbf{R}_p\right]$$

amplitude

Scattering cross section

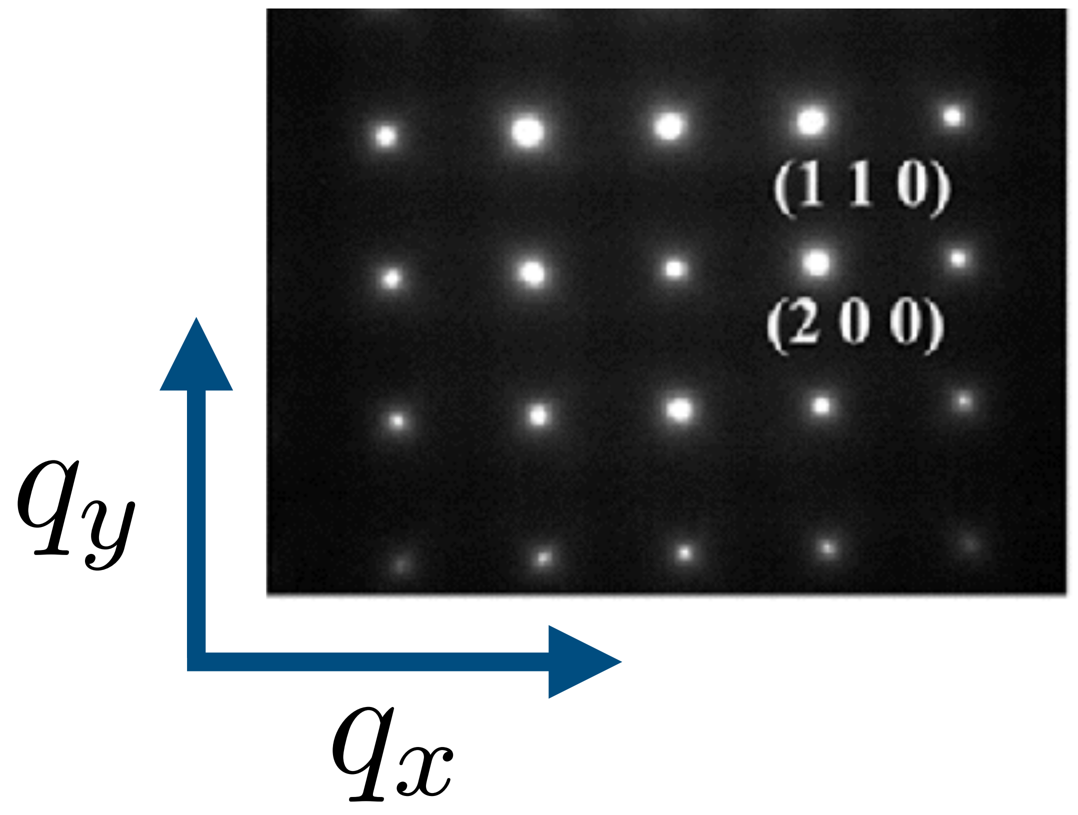
Lattice vector

Scattering intensity for a perfect lattice:

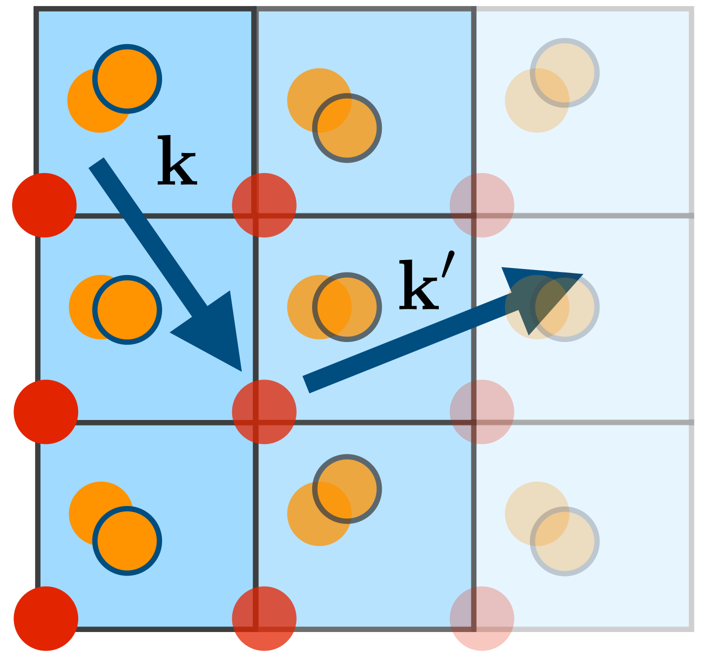
$$I(\mathbf{q}) = |A(\mathbf{q})|^2 \simeq \delta_{\mathbf{q},\mathbf{G}}$$

Reciprocal lattice vector

example: Cubic lattice



Vibrating lattice (>1 atom per cell)



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

Zero-phonon

One-phonon

Zero-phonon term:

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

One-phonon term:

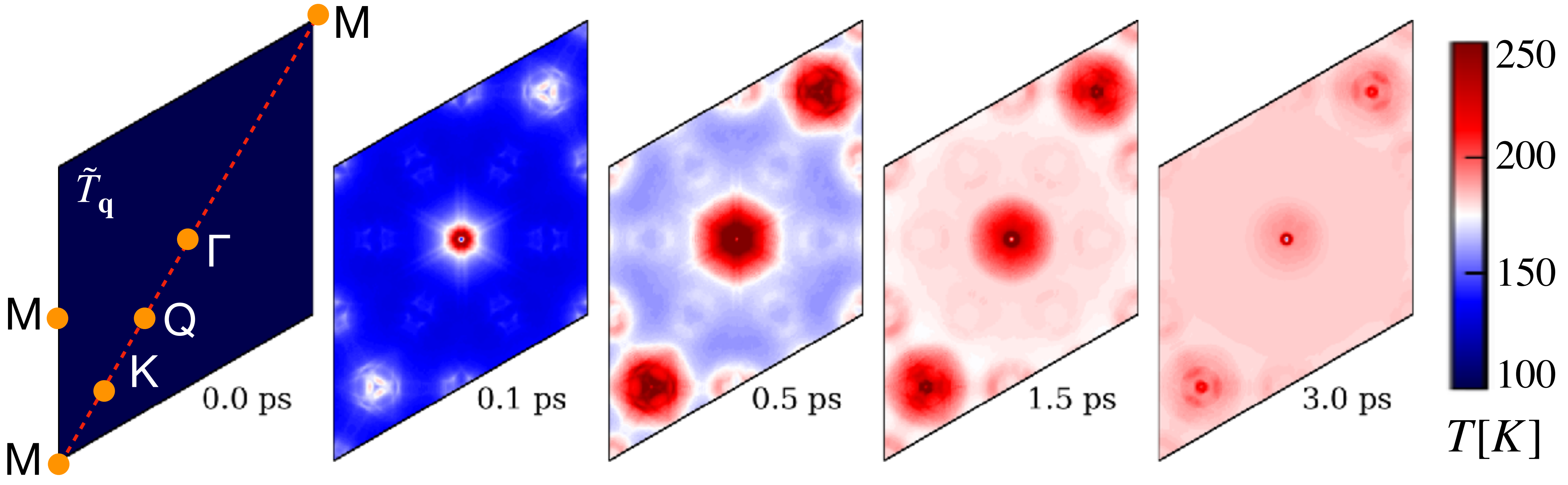
$$\begin{aligned}\langle I_1(\mathbf{S},E)\rangle_T \quad &= \quad |f_0|^2 \exp(-2W_T)\frac{\hbar^2 N_p}{2M_\kappa}\sum_{\mathbf{q}\nu}\left[\sum_{\alpha\alpha'} S_\alpha S_{\alpha'} e^{\nu}_{\kappa\alpha}(\mathbf{q}) e^{\nu *}_{\kappa\alpha'}(\mathbf{q})\right]\frac{1}{\hbar\omega_{\mathbf{q}\nu}} \qquad (16)\\ &\times \quad \left[\delta(\mathbf{S}+\mathbf{q})n_{\mathbf{q}\nu,T}\,\delta(E+\hbar\omega_{\mathbf{q}\nu}) + \delta(\mathbf{S}-\mathbf{q})(n_{\mathbf{q}\nu,T}+1)\delta(E-\hbar\omega_{\mathbf{q}\nu})\right].\end{aligned}$$

phonon occupation (available from the TDBE)

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

Recipe for ab-initio simulation of UEDS intensities

Step 1: Obtain the non-equilibrium phonon population from the TDBE



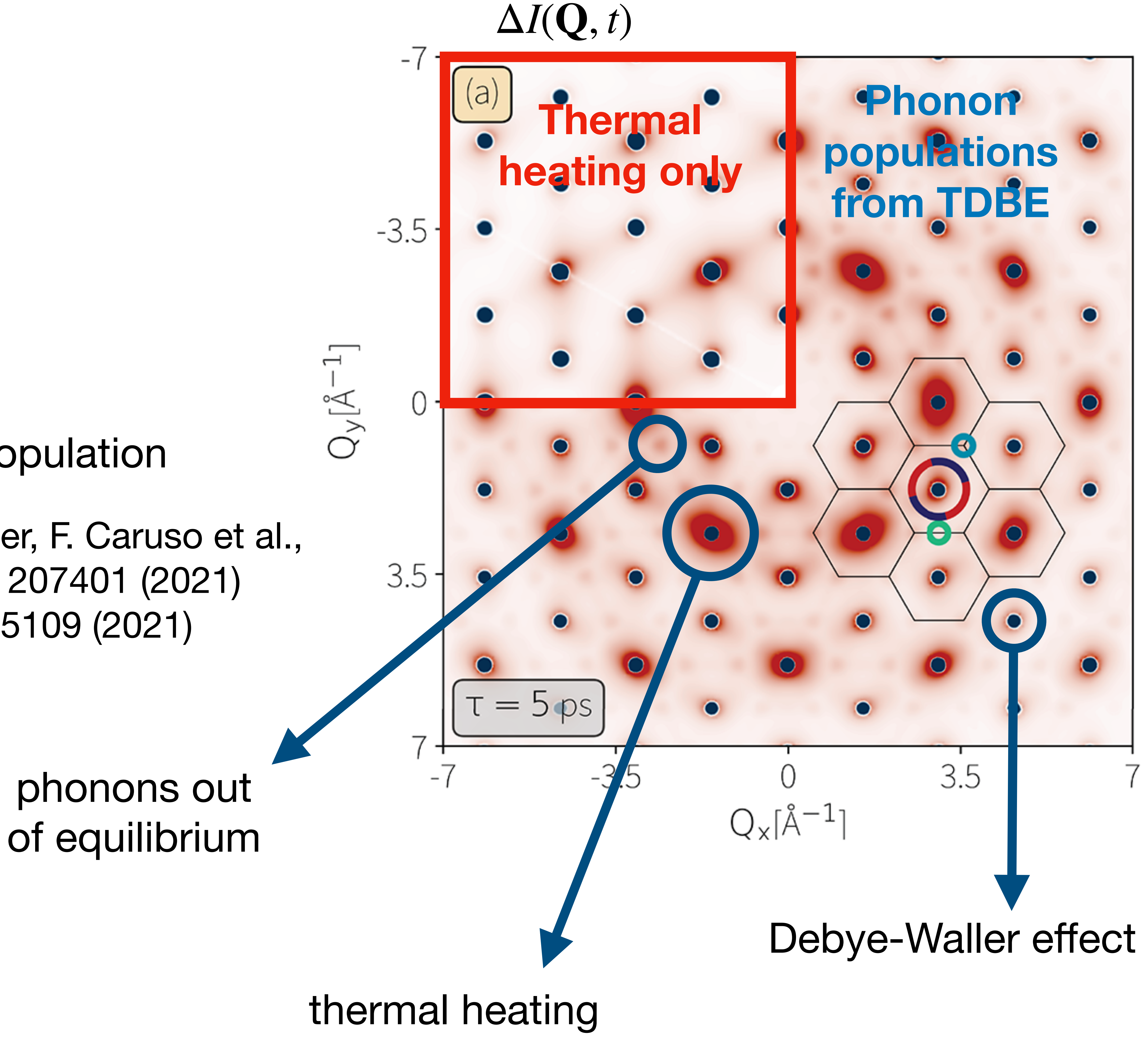
Step 2: Evaluate the structure factor for the instantaneous phonon population

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

Zero-phonon One-phonon

M. Zacharias, H. Seiler, F. Caruso et al.,
Phys. Rev. Lett. **127**, 207401 (2021)
Phys. Rev. B **104**, 205109 (2021)

Simulated UEDS for monolayer MoS_2



Step 3: Subtract the initial structure factor

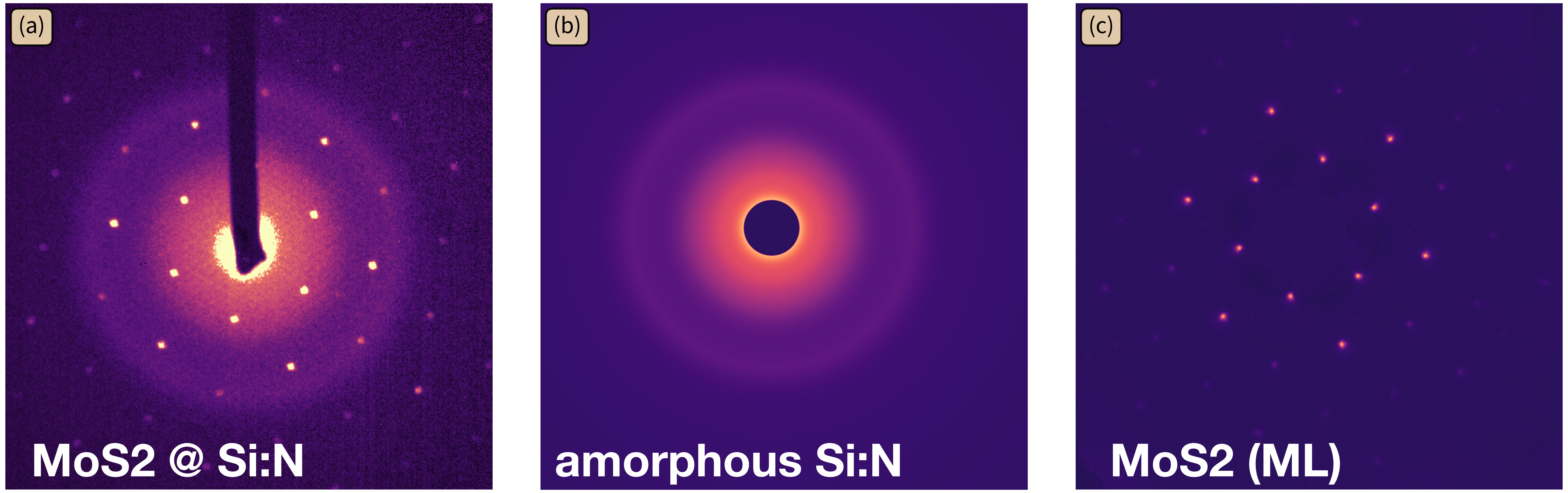
Intensity difference: $\Delta I(\mathbf{Q},t) = I(\mathbf{Q},t) - I(\mathbf{Q},t=0)$

T. Britt, (...) F. Caruso, XYZ, B. Siwick, Nano Lett. **22**, 4718 (2022)

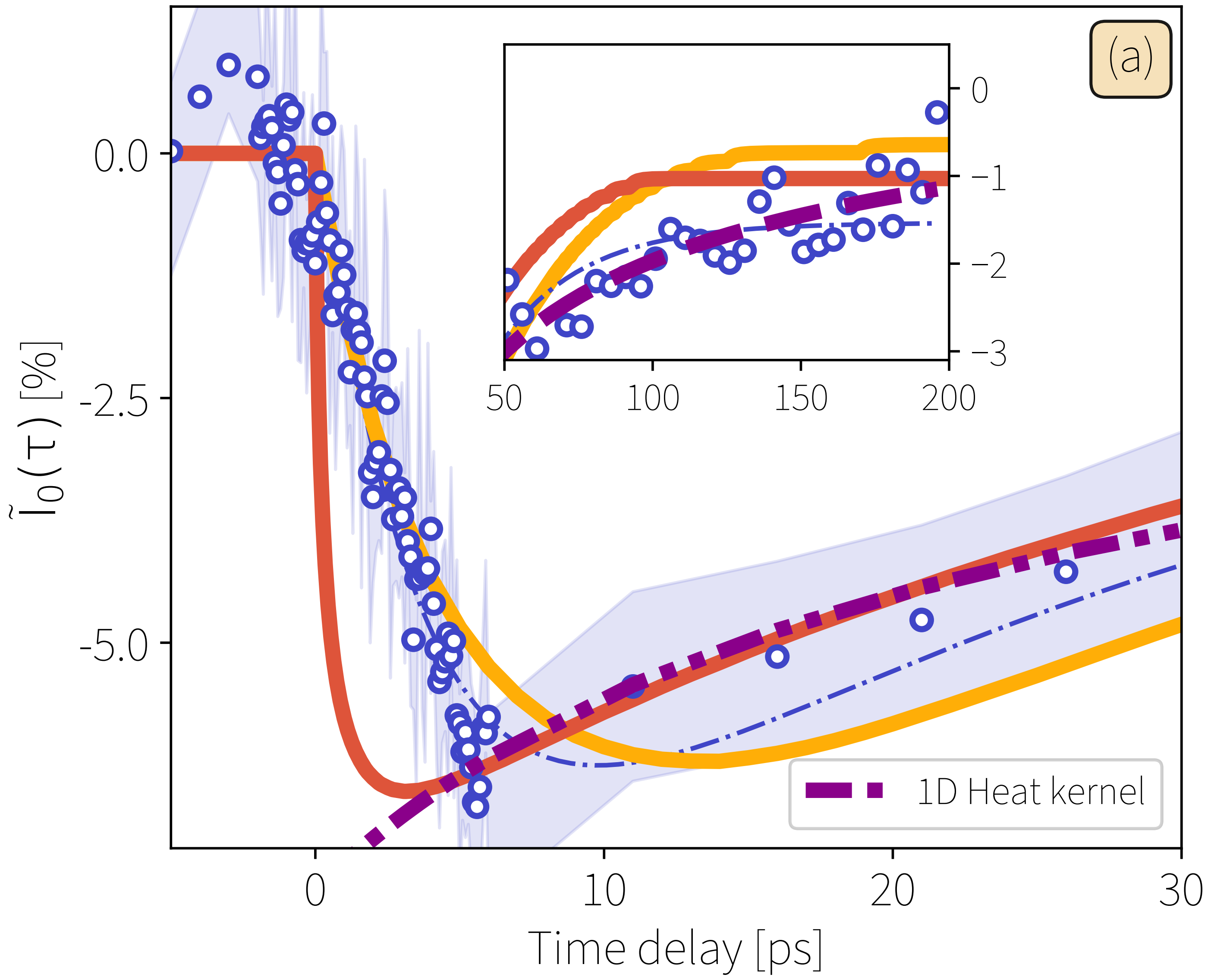
Direct view of phonon dynamics in MoS_2 monolayer

Experiments

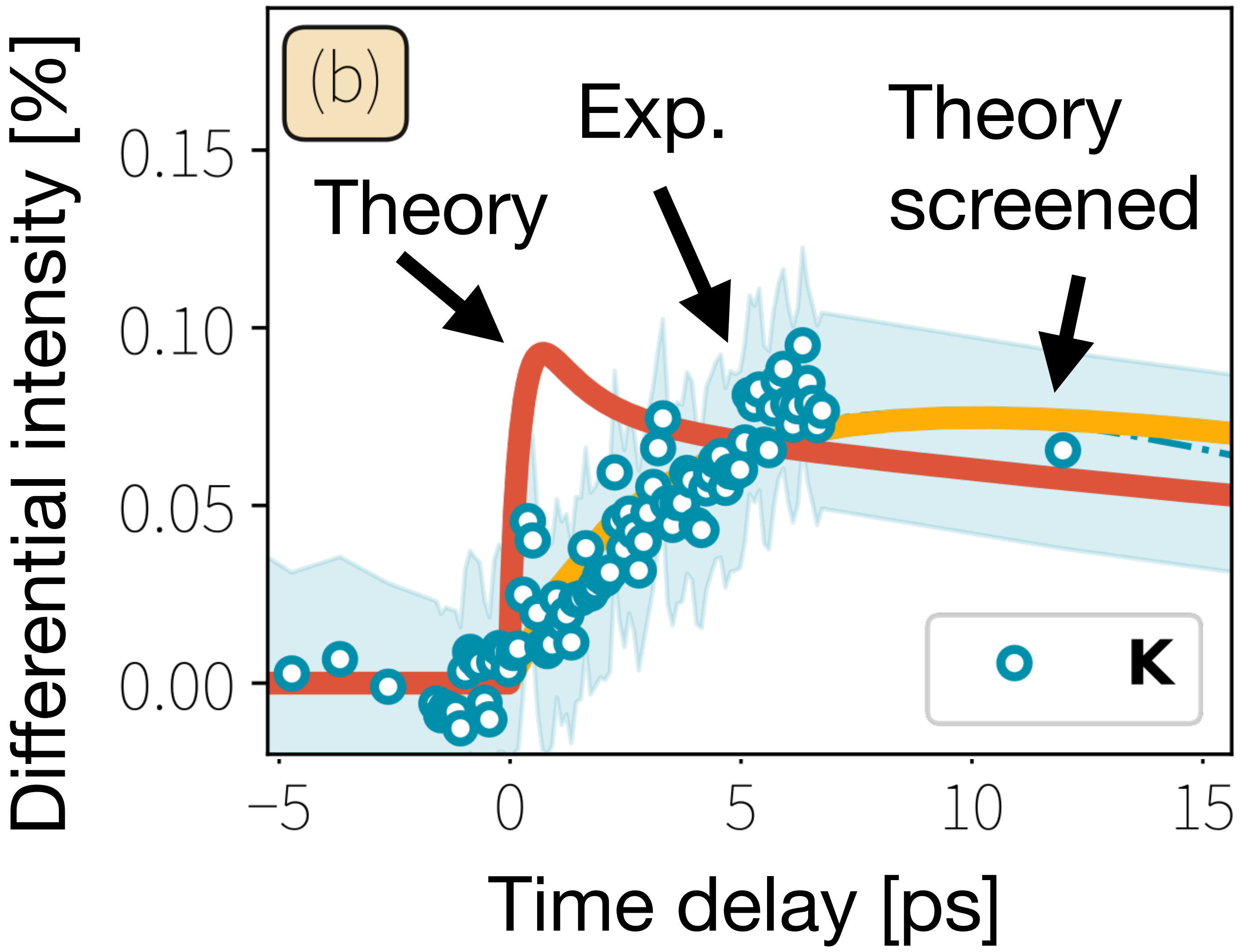
Tristan Britt
Bradley Siwick
(McGill University)



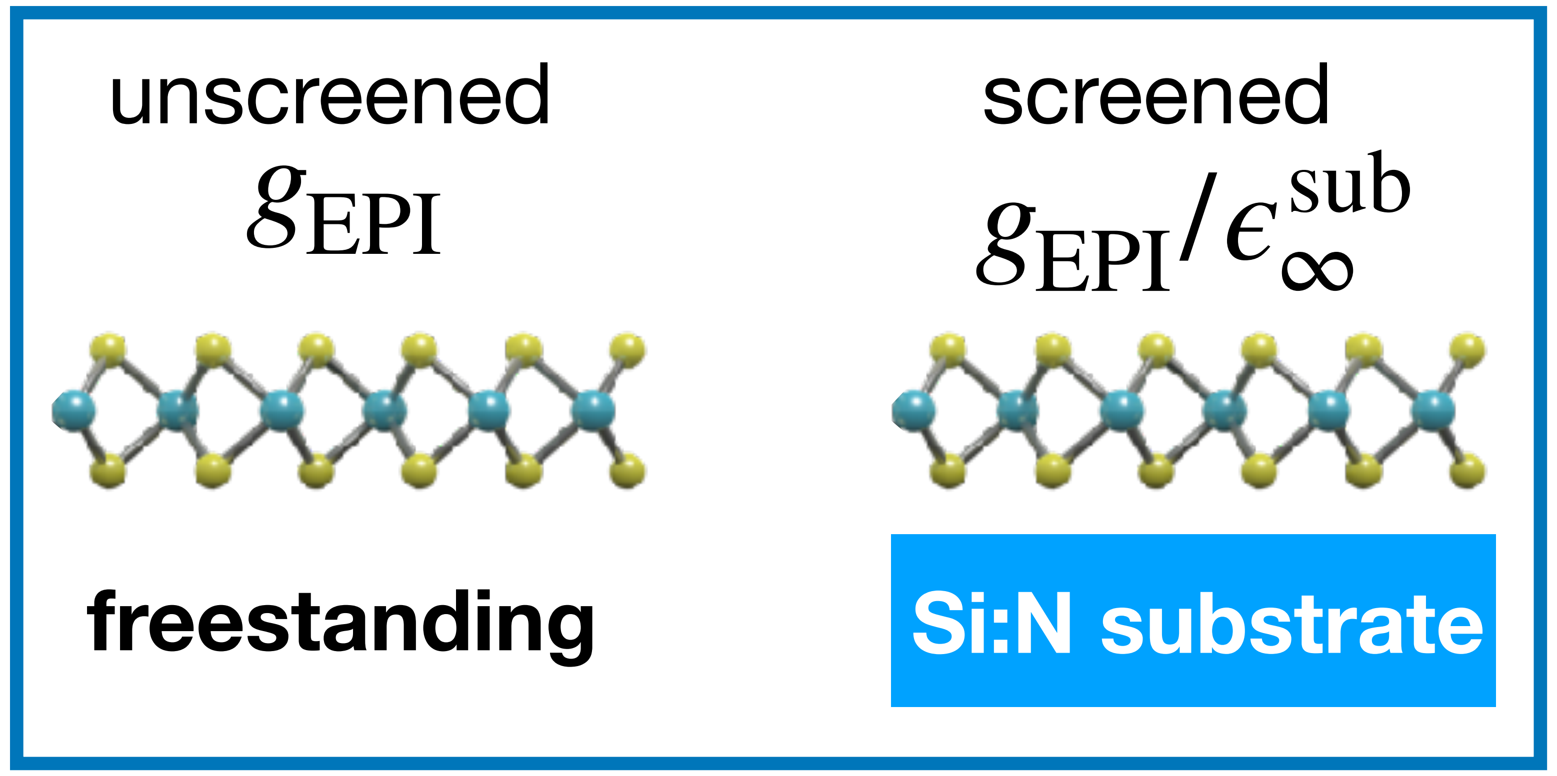
Bragg peak dynamics (Debye Waller effect)



Diffuse scattering at K

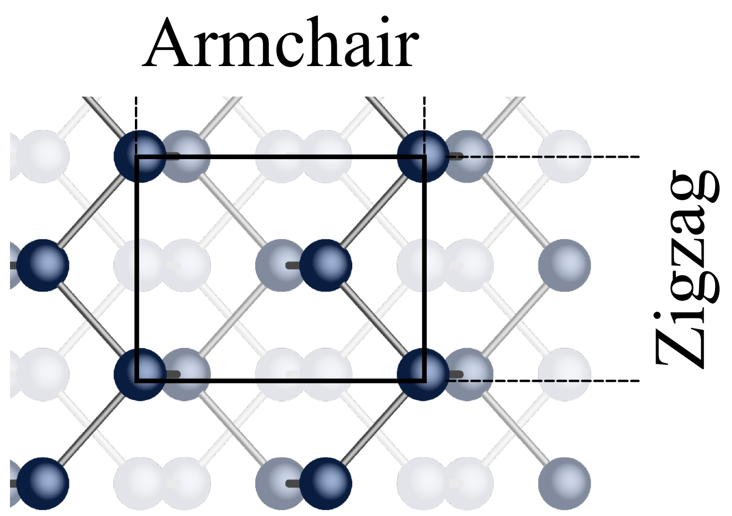


Measured energy transfer to the lattice: ~7 times slower than theory.

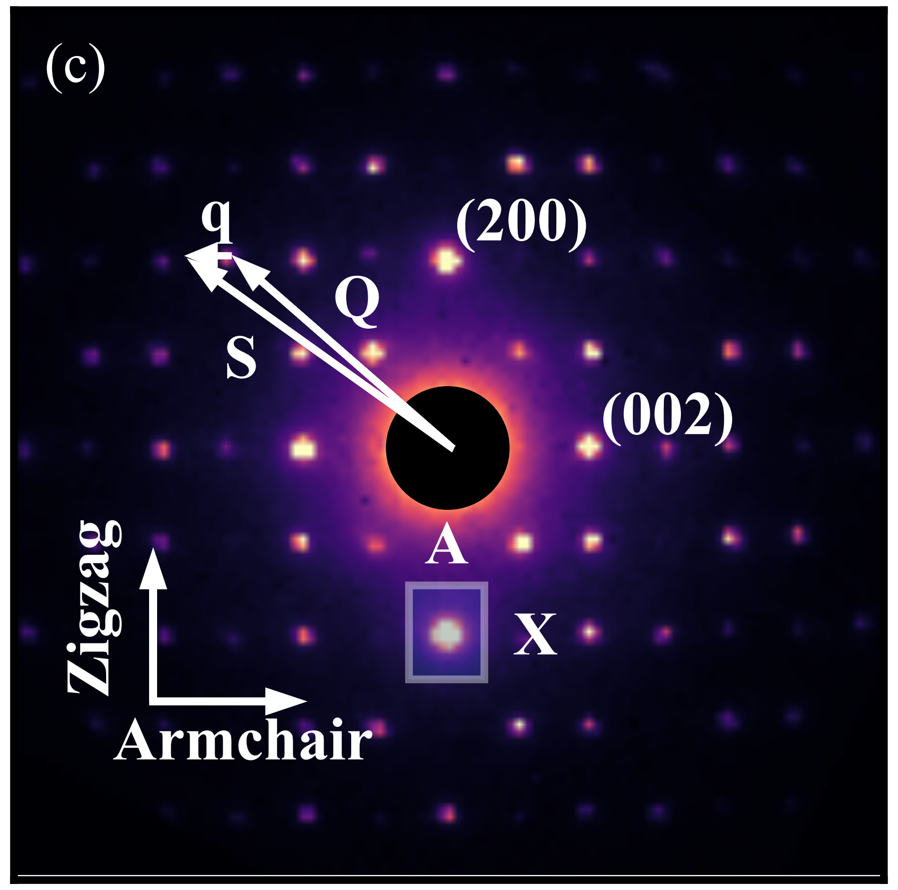


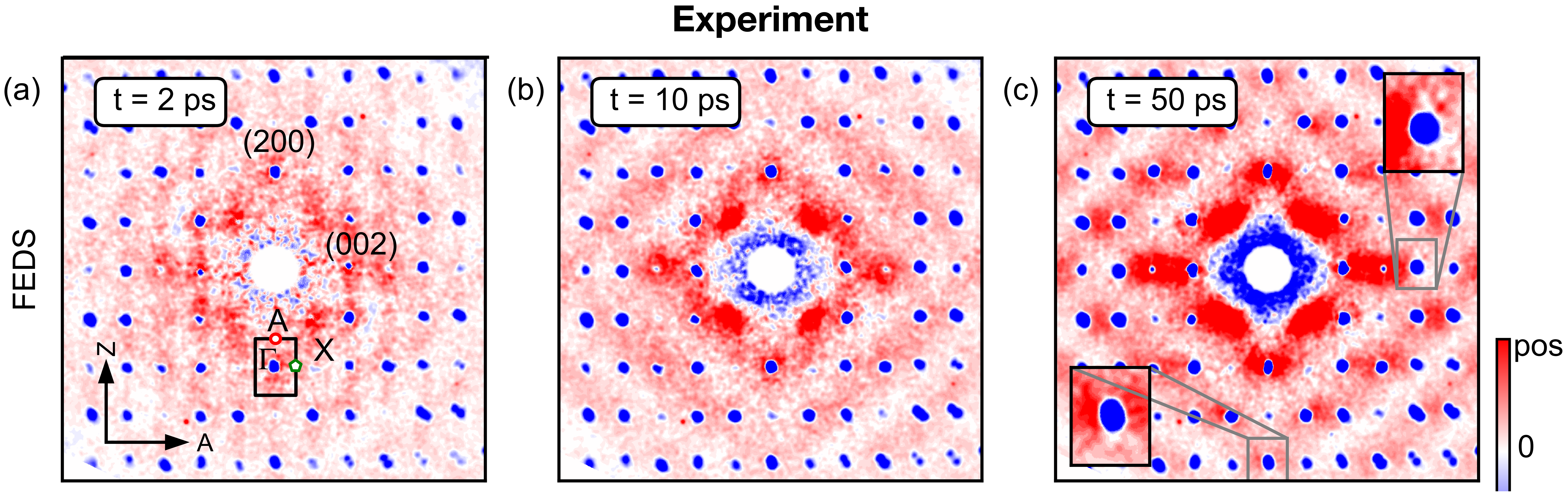
T. Britt, (...) F. Caruso, XYZ, B. Siwick, Nano Lett. **22**, 4718 (2022)

Ultrafast electron diffuse scattering: black Phosphorus



Black phosphorus





Experiments: FHI Berlin

Helene Seiler

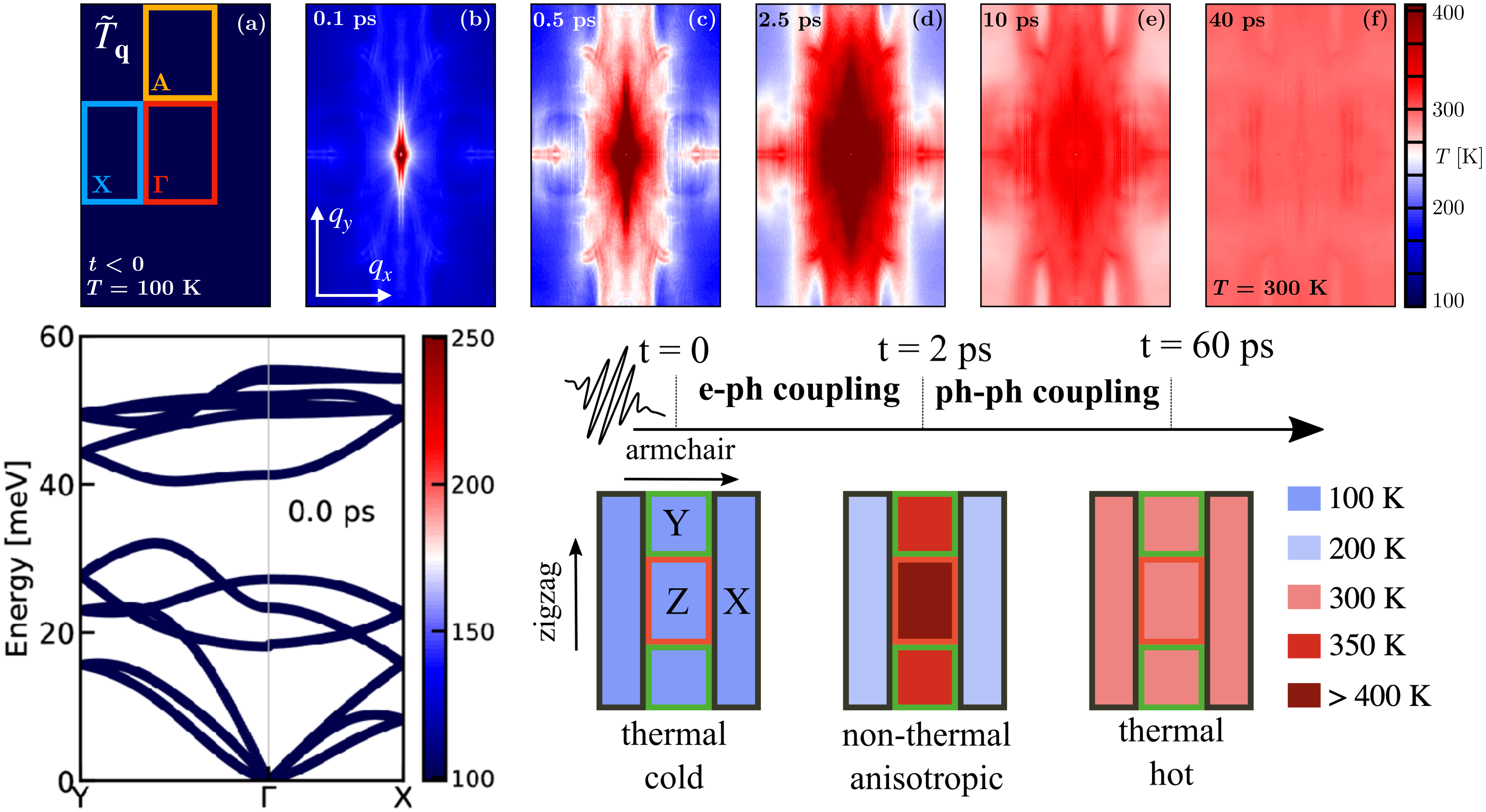
Ralph Ernstorfer

H. Seiler, D. Zahn, M. Zacharias, (...) R. Ernstorfer, F. Caruso, Nano Lett. **21**, 6171 (2021)

Non-equilibrium lattice dynamics in bP from first-principles

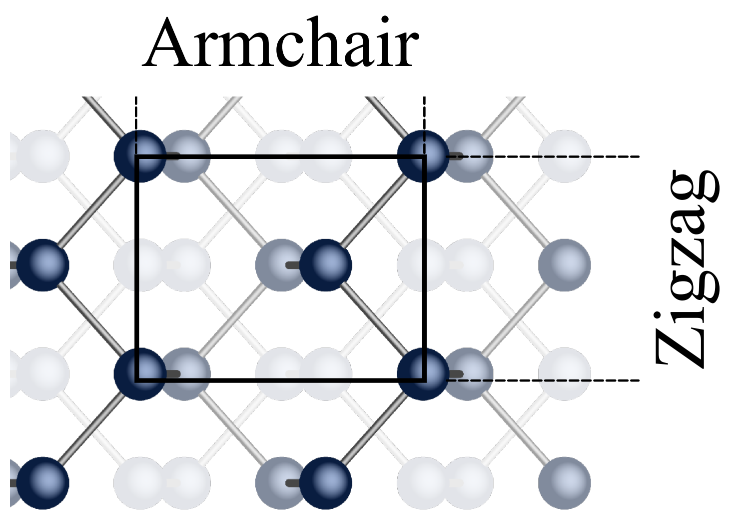
$$\frac{\partial n_{\mathbf{q}\nu}}{\partial t} = I^{\mathrm{e-ph}}_{\mathbf{q}\nu}[f,n] + I^{\mathrm{ph-ph}}_{\mathbf{q}\nu}[n]$$

Effective vibrational temperature

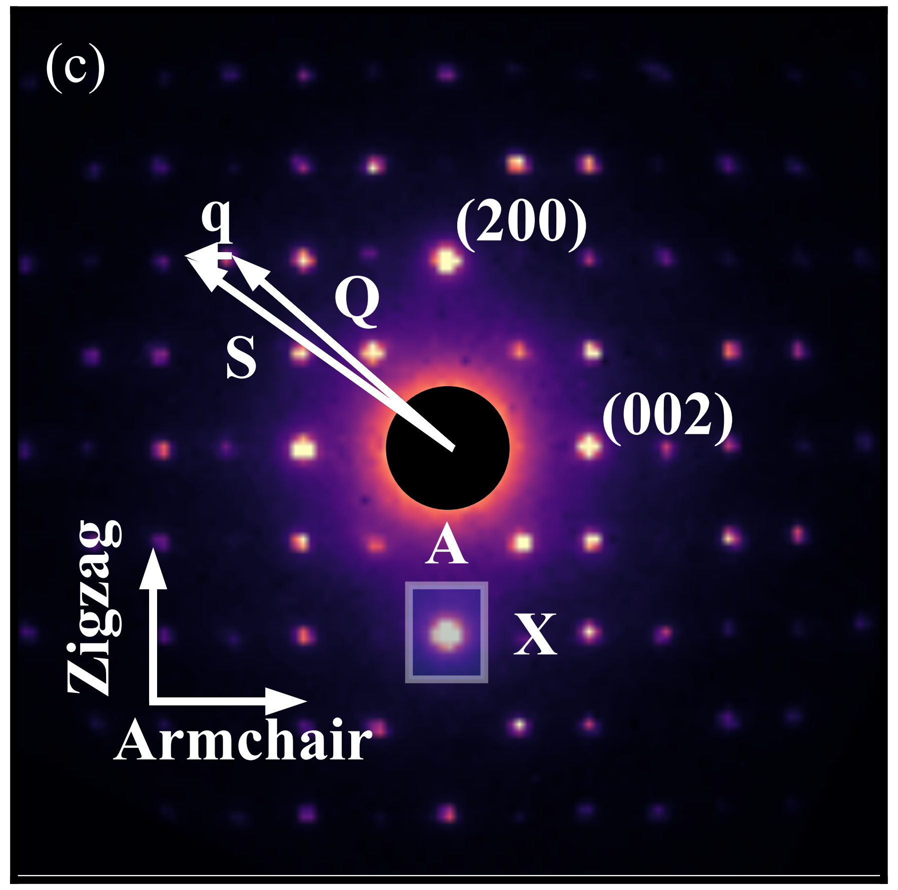


H. Seiler, D. Zahn, M. Zacharias, (...) R. Ernstorfer, F. Caruso, Nano Lett. **21**, 6171 (2021)

Ultrafast electron diffuse scattering: black Phosphorus



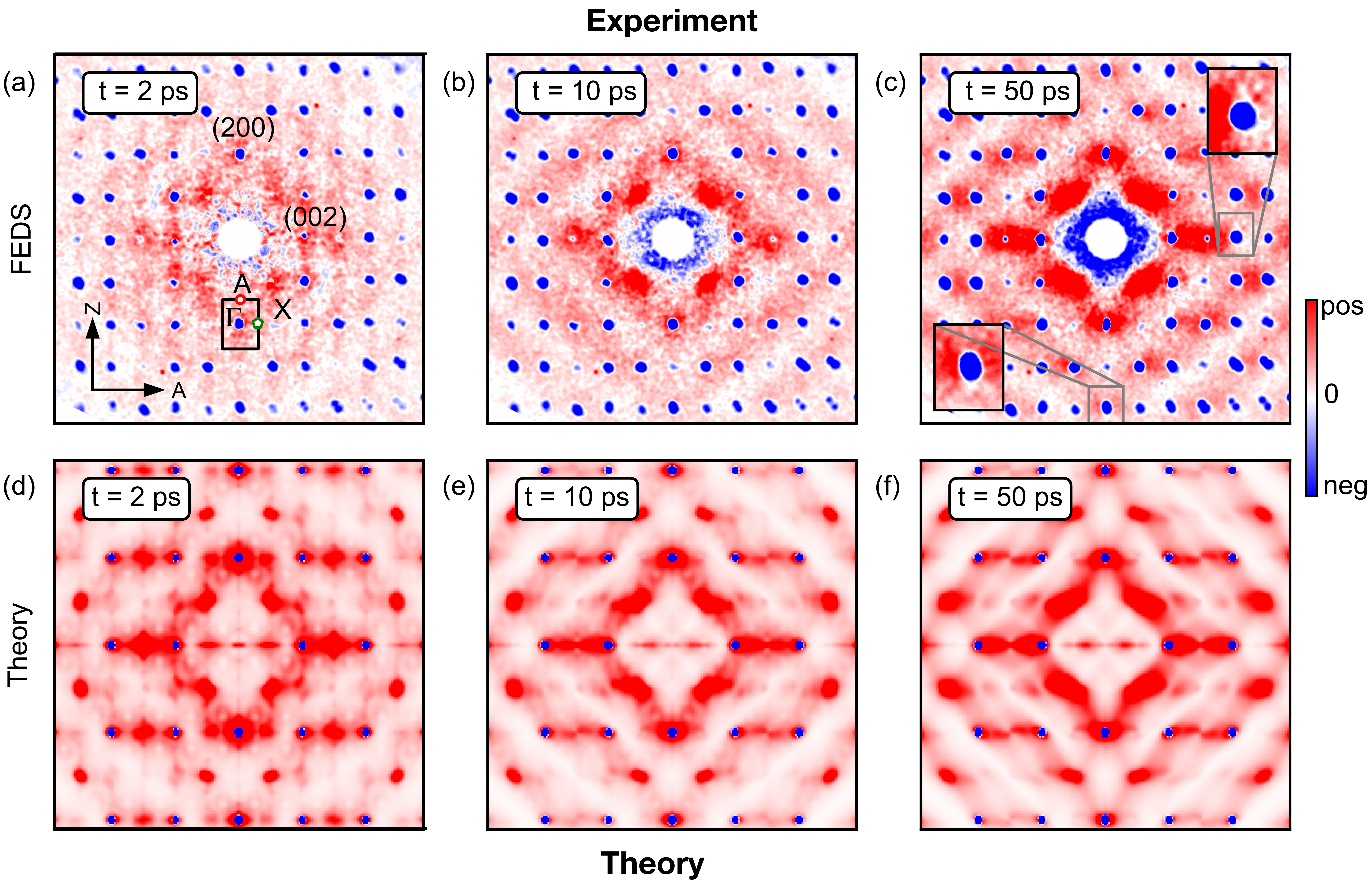
Black phosphorus



Experiments: FHI Berlin

Helene Seiler

Ralph Ernstorfer



H. Seiler, D. Zahn, M. Zacharias, (...) R. Ernstorfer, F. Caruso, Nano Lett. **21**, 6171 (2021)

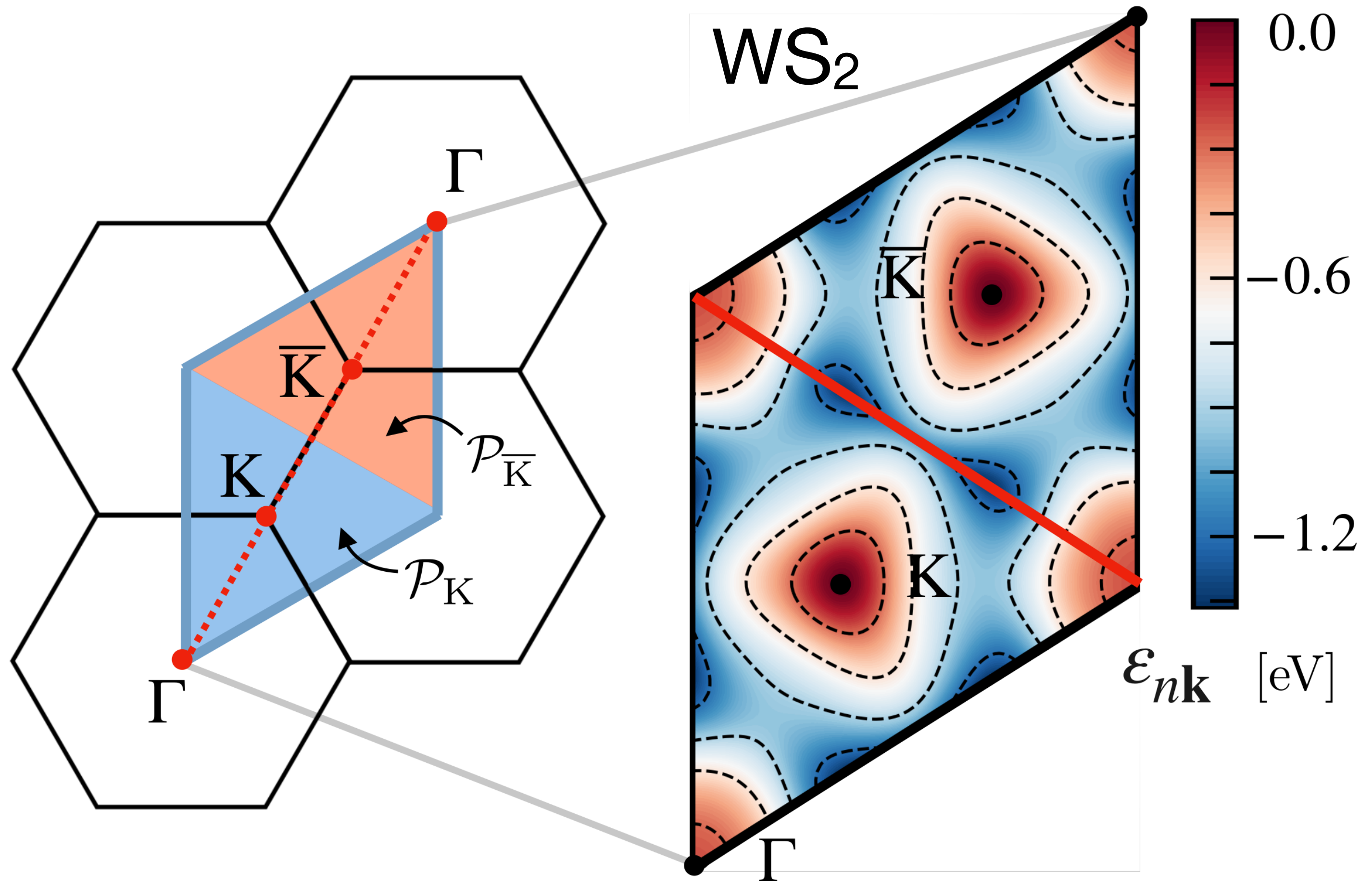
Part 3

Valley-selective circular dichroism in WS_2 and chiral phonon dynamics

(Outlook)

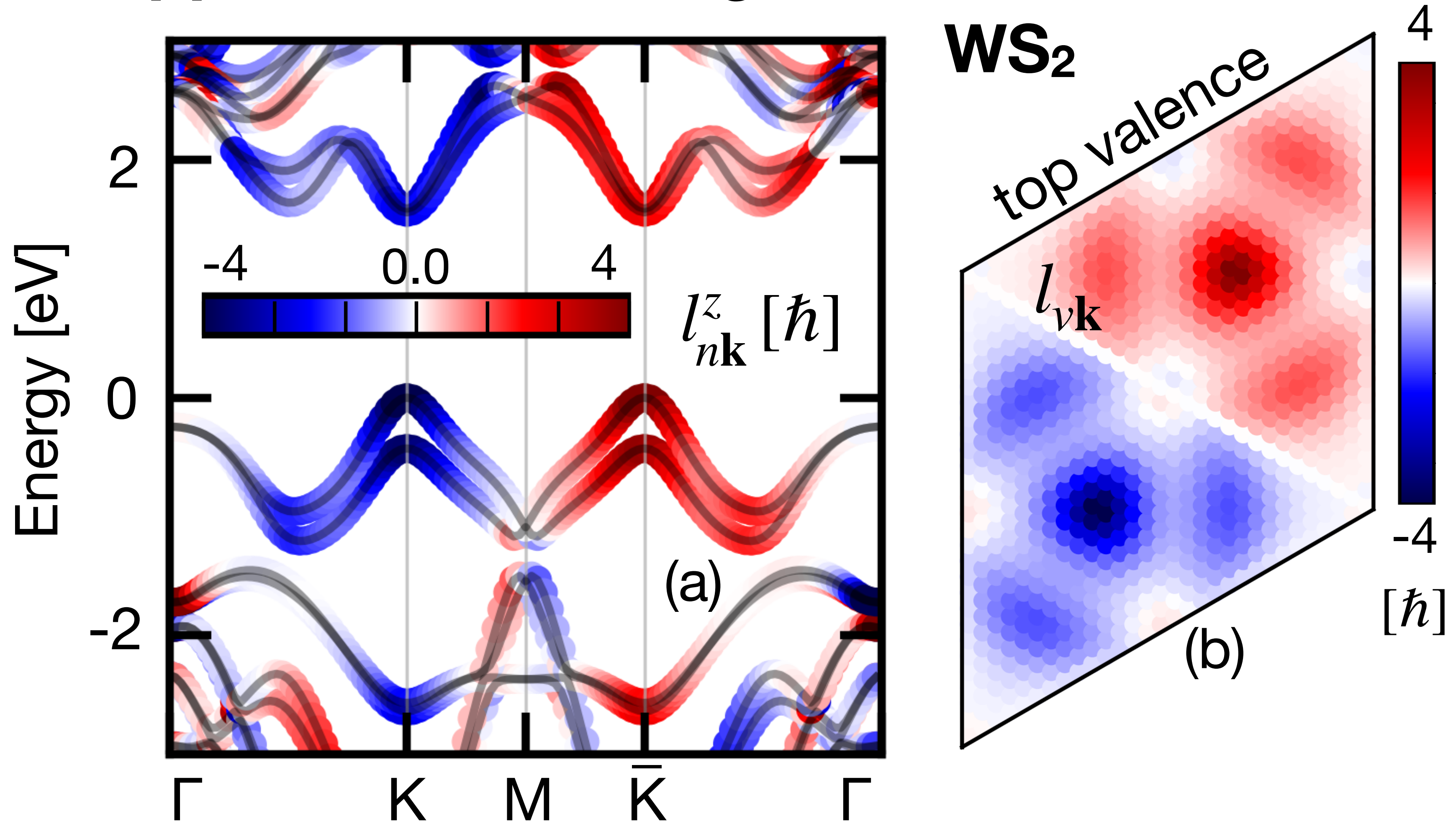
Valley dichroism in transition metal dichalchogenide monolayers

Non-centrosymmetric honeycomb lattices

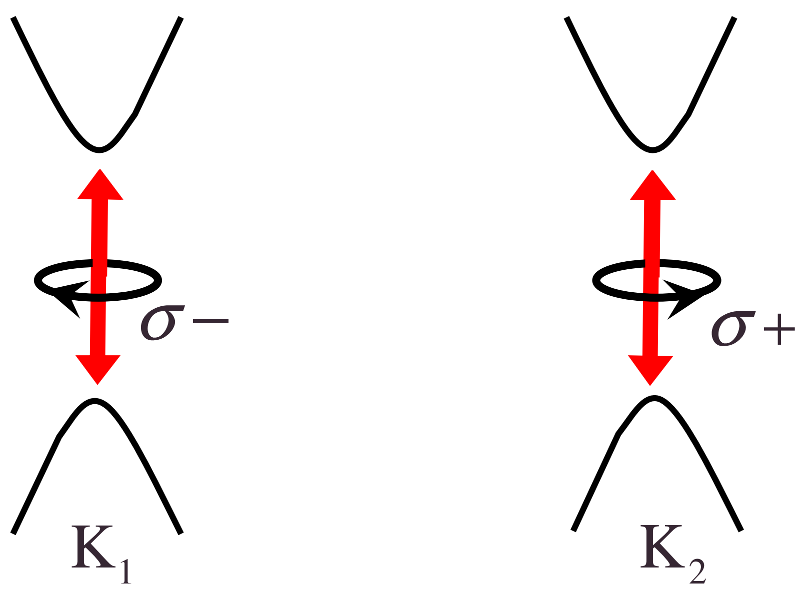


- Twofold degeneracy at the high-symmetry points K and -K
- K and -K are inequivalent in absence of an inversion center

1. Opposite Orbital angular momentum at K and -K



2. Valley-selective circular dichroism



Yao, Xiao, Niu, PRB **77**, 235406 (2008)
Zeng et al., Nature Nanotec. **7**, 490 (2012)
Souza, Vanderbilt, PRB **77**, 054438 (2008)

Ab-initio theory of valley-selective circular dichroism & valley excitons

BSE 2-particle Hamiltonian

eigenvectors

eigenvalues

Bethe-Salpeter equation:

$$H_{vc\mathbf{k},v'c'\mathbf{k}'} A^{\lambda}_{v'c'\mathbf{k}'} = E^{\lambda} A^{\lambda}_{vc\mathbf{k}}$$

$$A^{\lambda}_{vc\mathbf{k}} = \langle \psi^{h}_{v\mathbf{k}} \psi^{e}_{c\mathbf{k}} | \psi_{\lambda} \rangle$$

Imaginary part of the dielectric function (optical absorption):

$$\varepsilon_2(\omega) \propto \sum_{\lambda} \left| \hat{\boldsymbol{\epsilon}} \cdot \mathbf{t}^{\lambda} \right|^2 \delta(E^{\lambda} - \hbar\omega)$$

exciton eigenstates

$$|\lambda\rangle = \sum_{vc\mathbf{k}} A^{\lambda}_{vc\mathbf{k}} |v_{\mathbf{k}} c_{\mathbf{k}}\rangle$$

Recipe for ab-initio calculation of valley excitons:

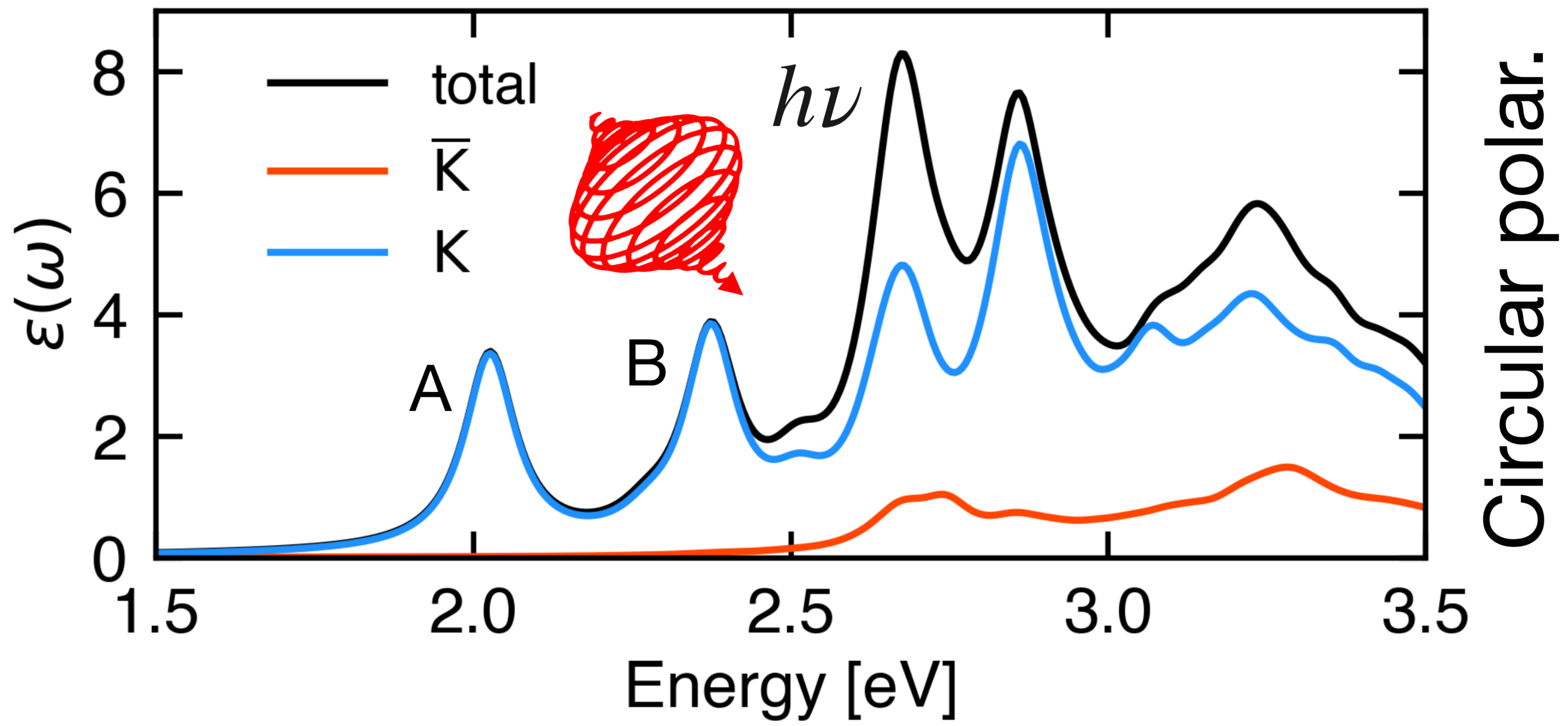
1. Explicit treatment of valley degrees of freedom
2. Account for the circular-polarization vector of light

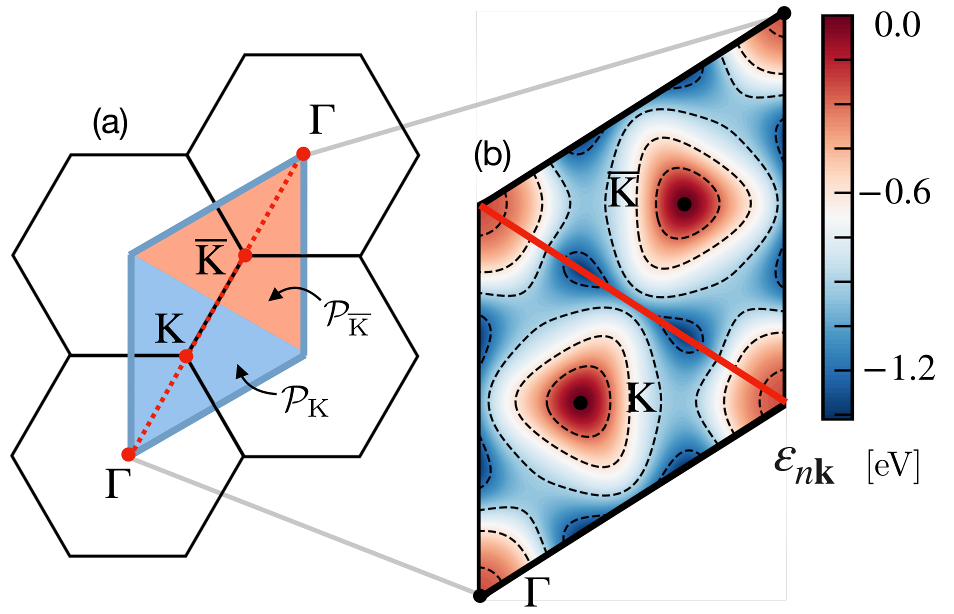
$$\hat{\boldsymbol{\epsilon}} = 1/\sqrt{2}(\hat{\mathbf{x}} \pm \hat{\mathbf{y}})$$

F. Caruso, M. Schebek, Y. Pan, C. Vona, C. Draxl, J. Phys. Chem Lett. **13**, 5894 (2022)

Ab-initio description of valley excitons in WS_2

Formation of valley excitons upon absorption of light with circular polarization





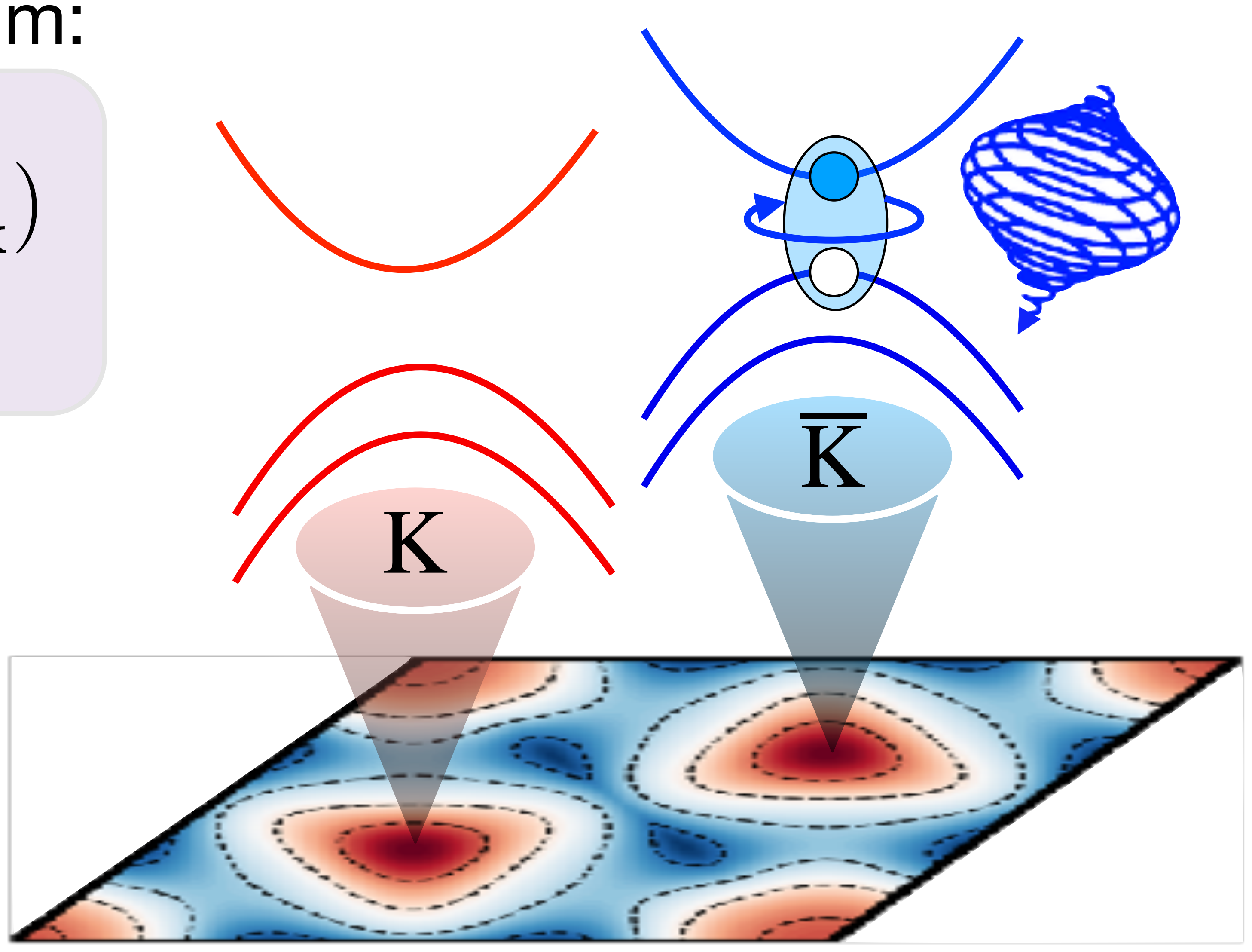
Exciton orbital angular momentum:

$$L^z_\lambda = \sum_{vc\mathbf{k}} \left|A^\lambda_{vc\mathbf{k}}\right|^2 (l^z_{c\mathbf{k}} + l^z_{v-\mathbf{k}})$$

Excitons inherit the OAM from the underlying band structure

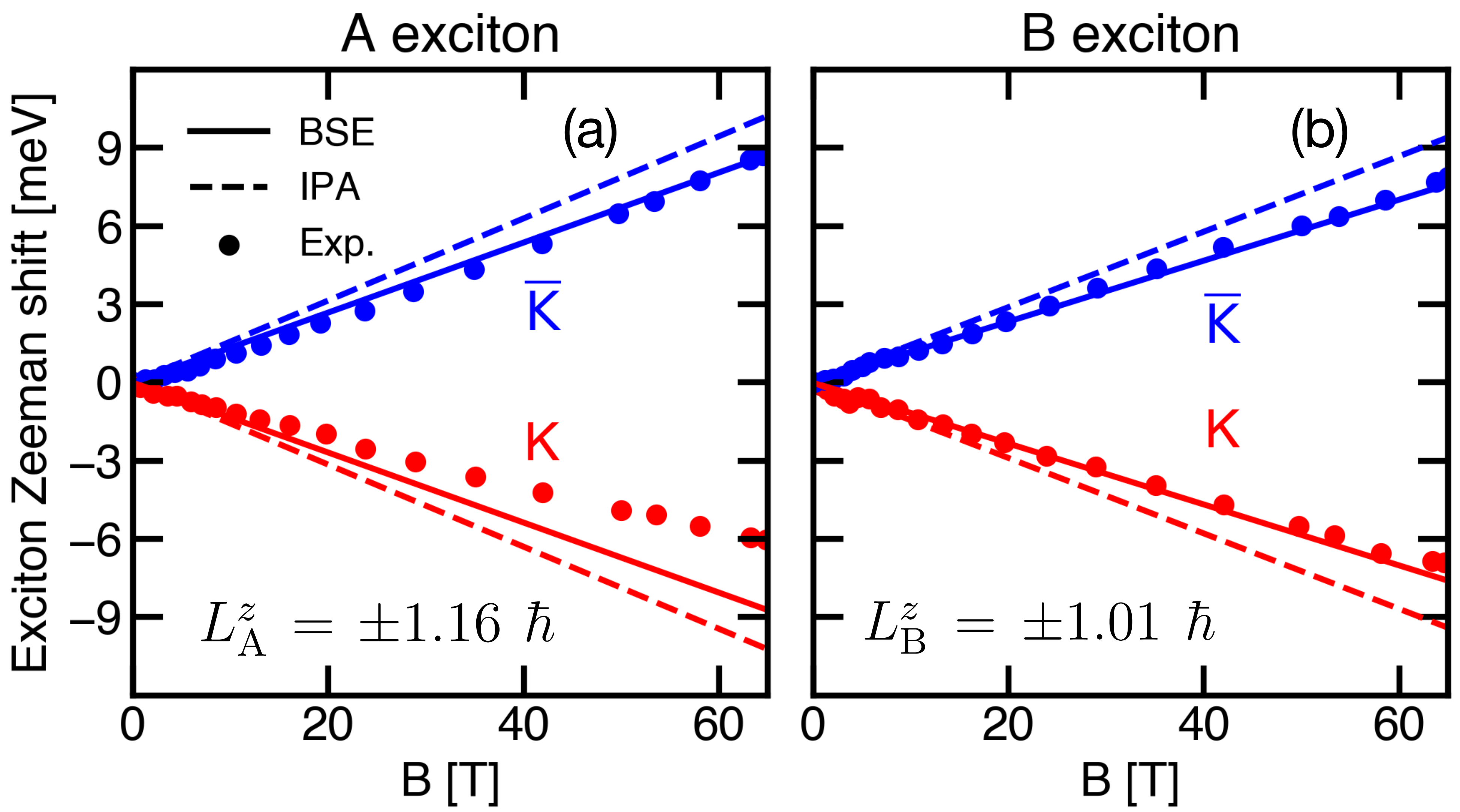
$$L^z_{\mathrm{A}} = \pm 1.16\ \hbar$$

$$L^z_{\mathrm{B}} = \pm 1.01\ \hbar$$



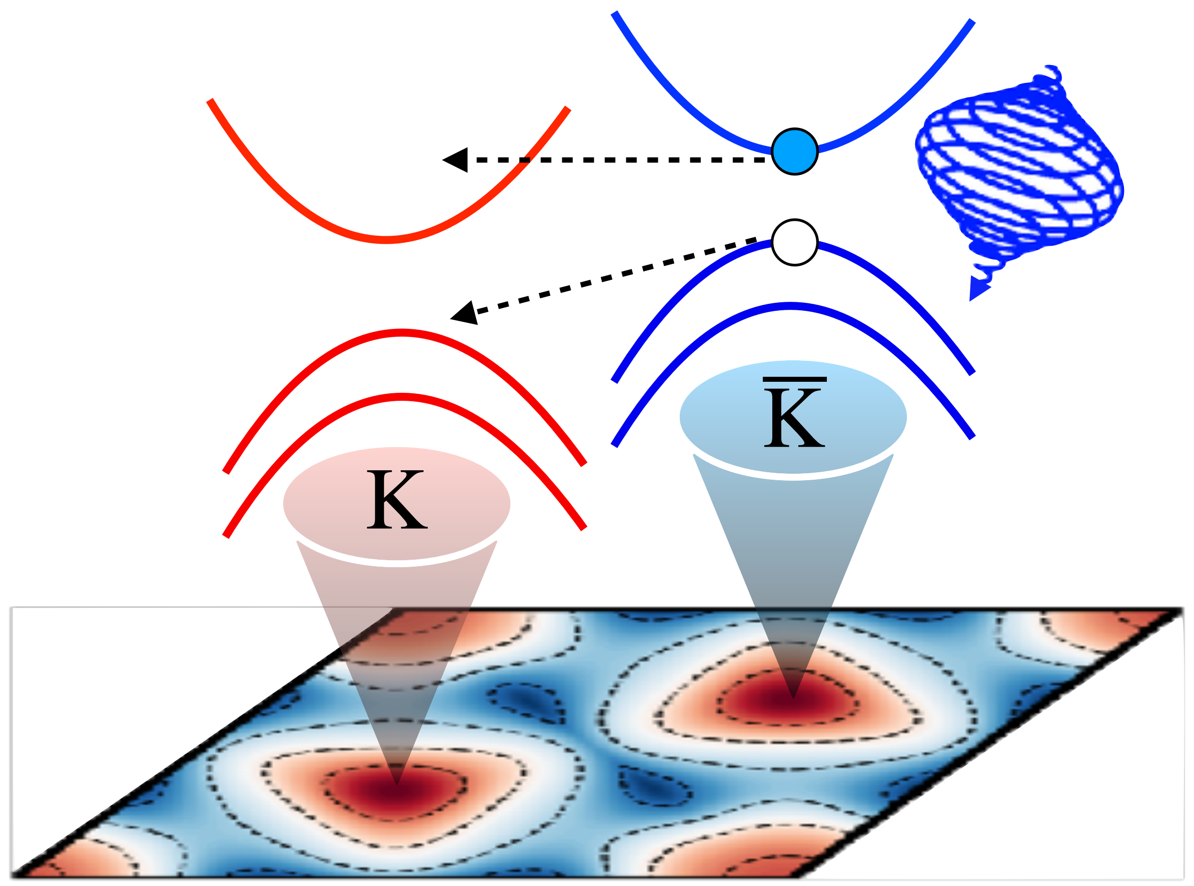
Zeeman splitting in a magnetic field

$$\Delta E_b = -\mathbf{M}_\lambda \cdot \mathbf{B}$$



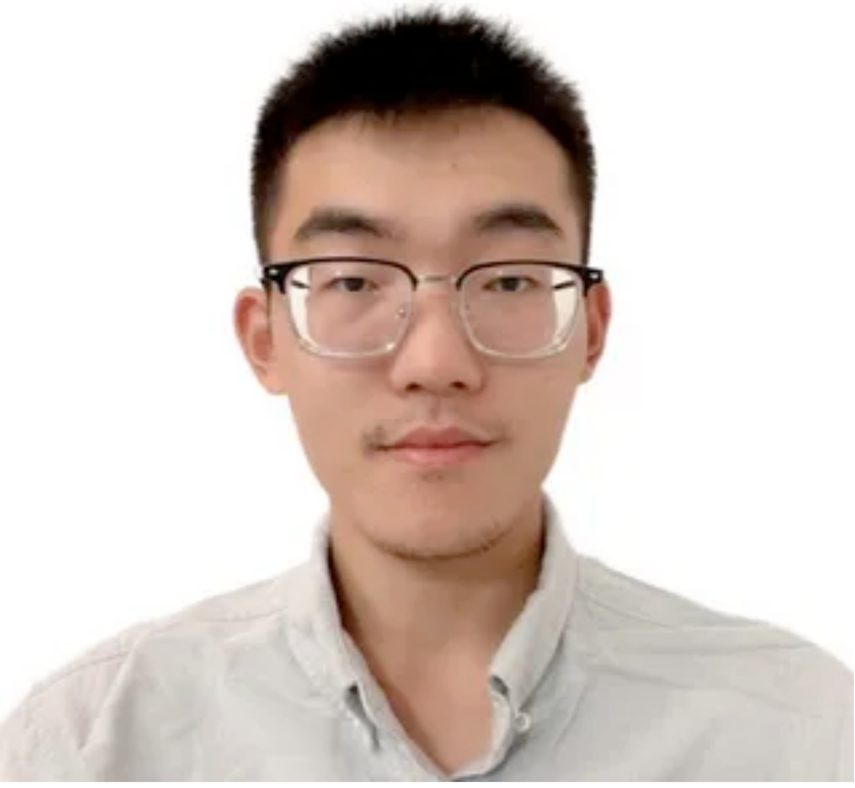
F. Caruso, M. Schebek, Y. Pan, C. Vona, C. Draxl, J. Phys. Chem Lett. **13**, 5894 (2022)

Chiral phonon excitation upon valley depolarization

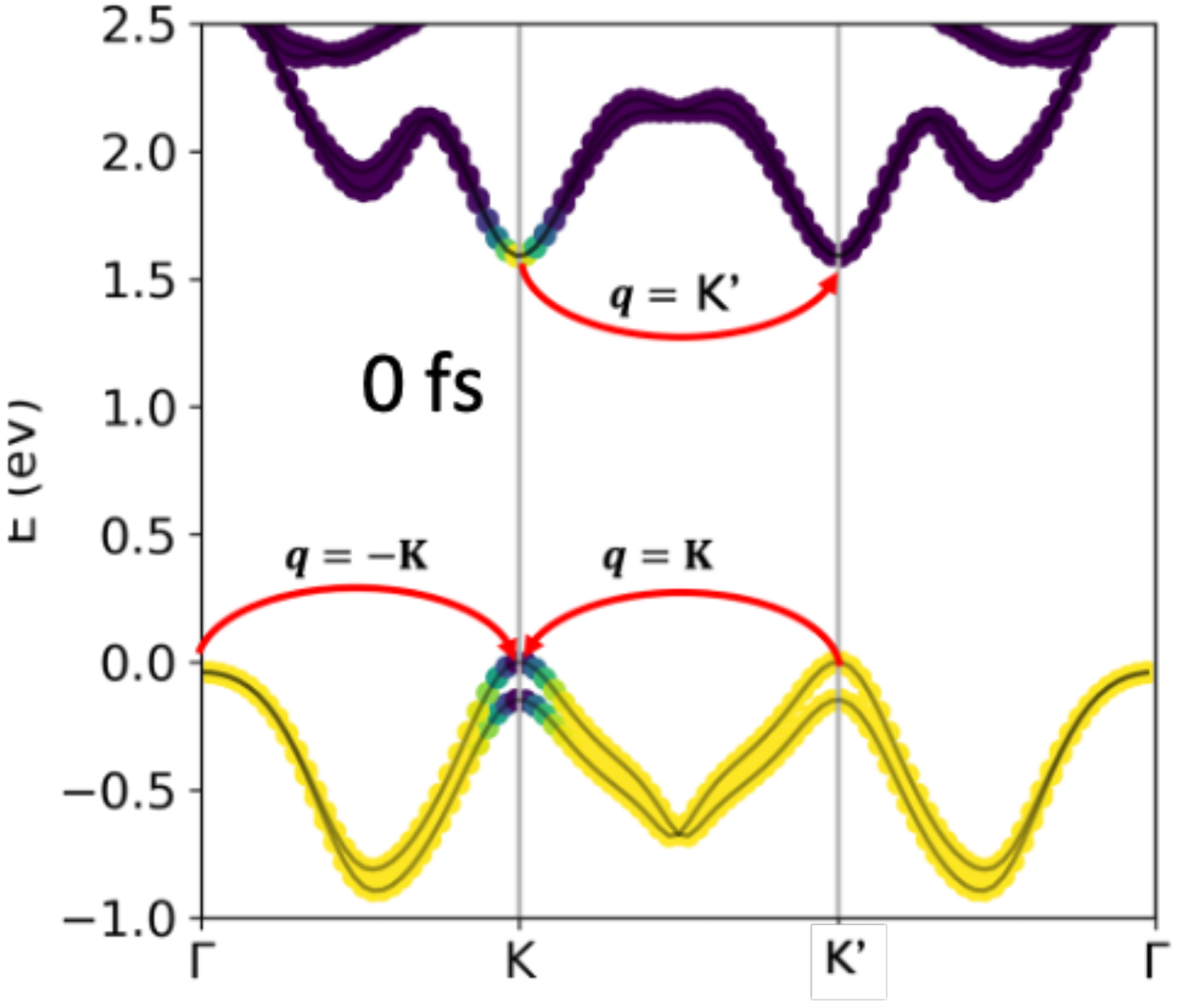


Open questions:

Q1: Phonon population established upon valley depolarization?
Q2: Selection rules?
Q3: Phonon angular momentum (PAM) and chiral phonons?
Q4: Time-scales of PAM transfer and decay?

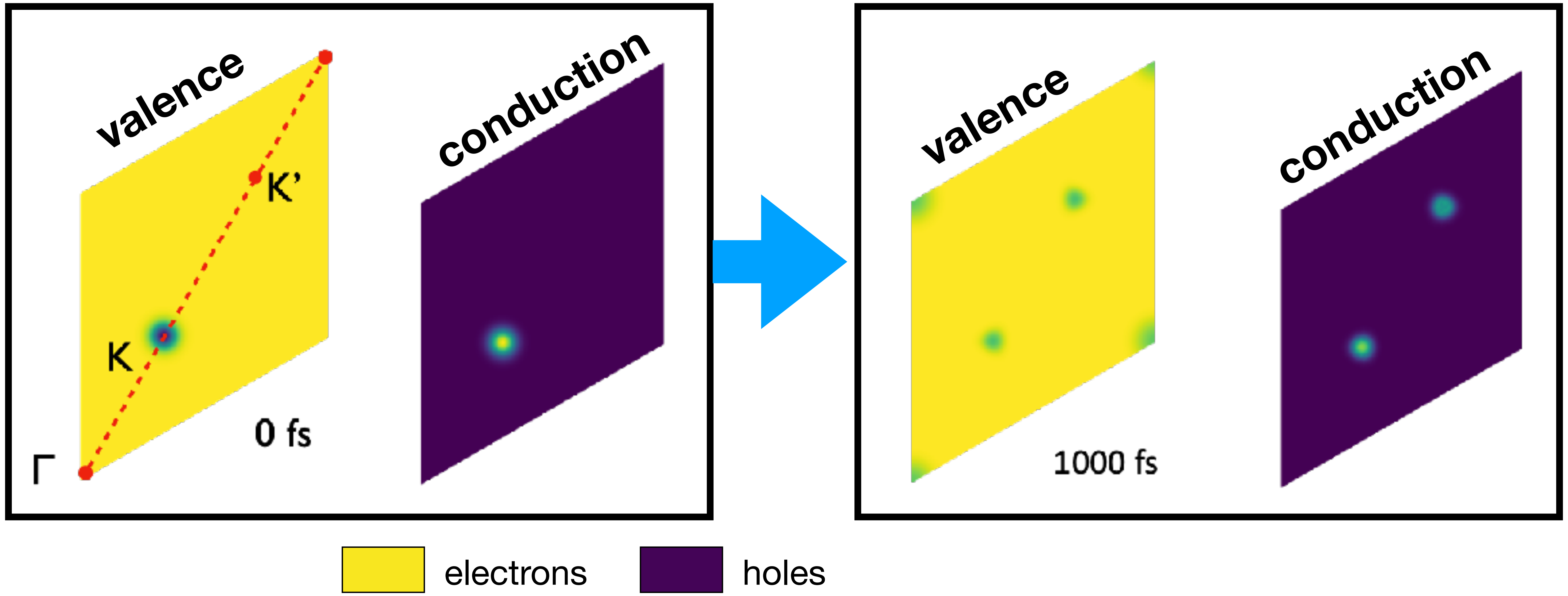


Yiming Pan



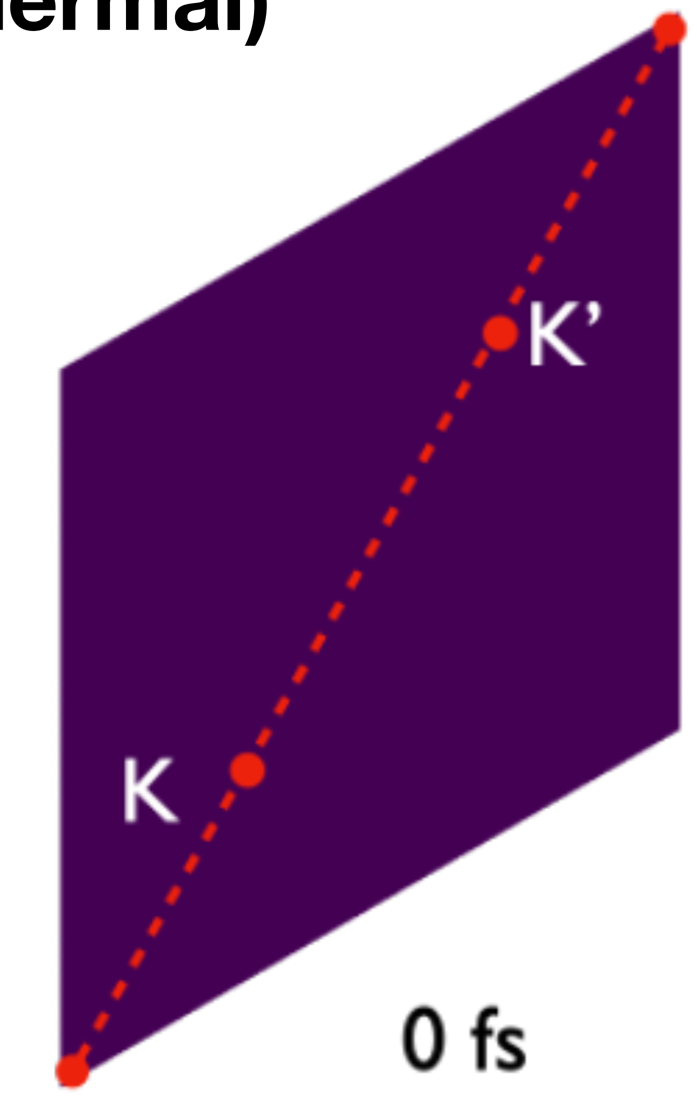
initial electronic state (valley polarized)

final electronic state (depolarized)

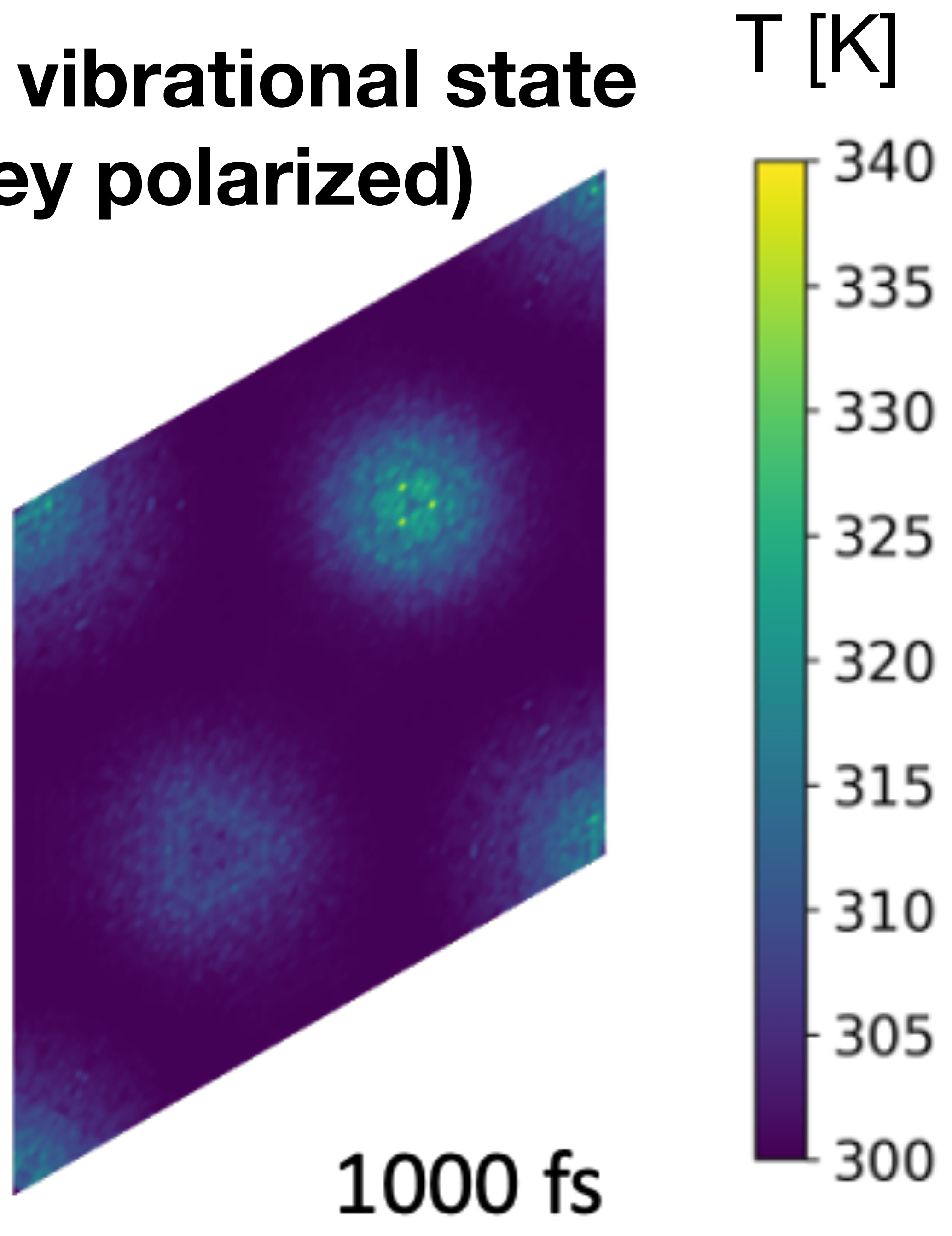


Chiral phonon excitation upon valley depolarization

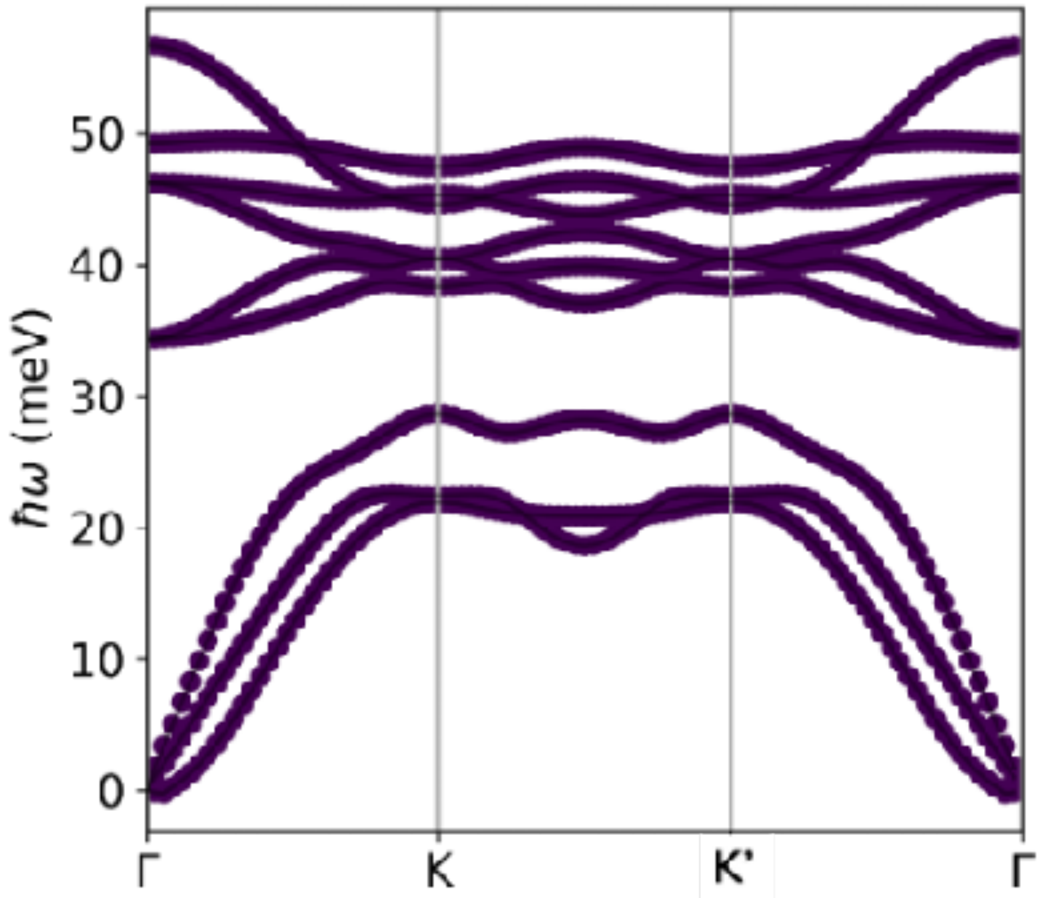
initial vibrational state (thermal)



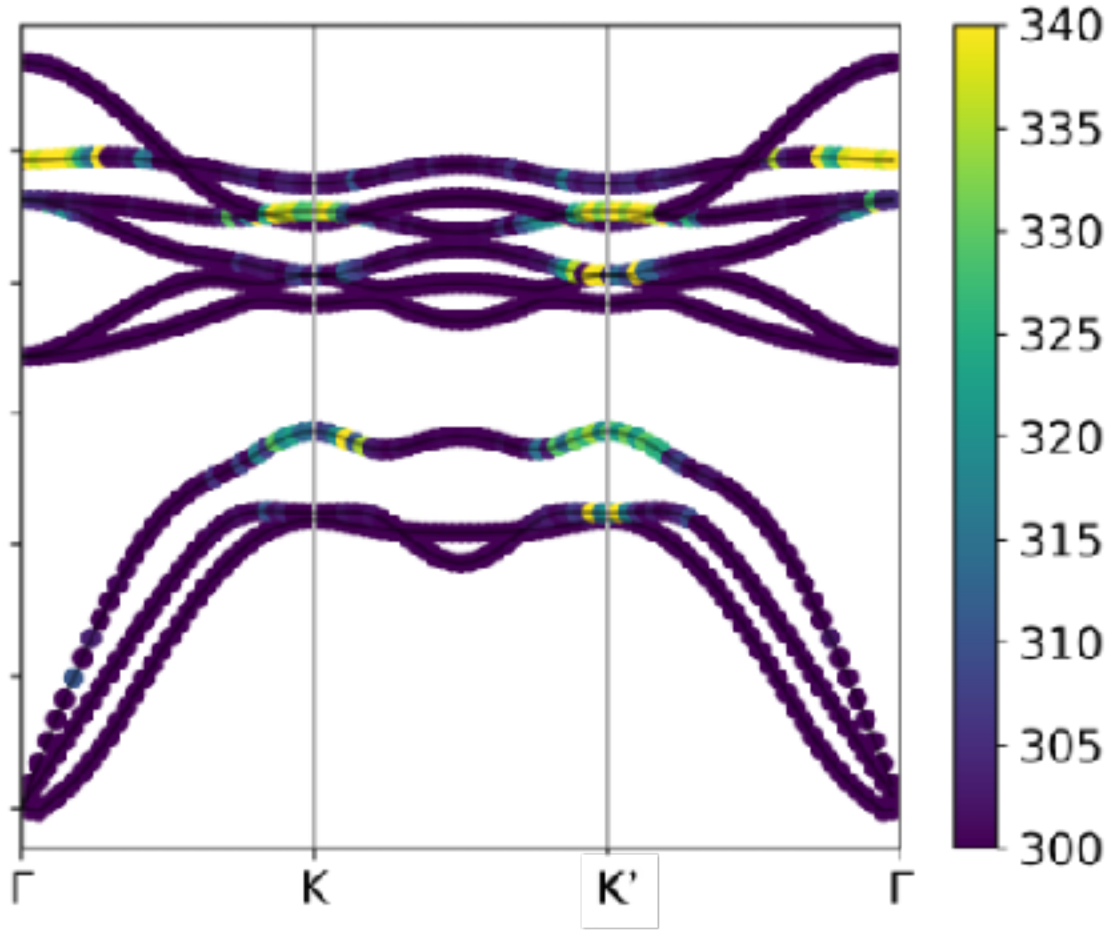
final vibrational state (valley polarized)



Thermal

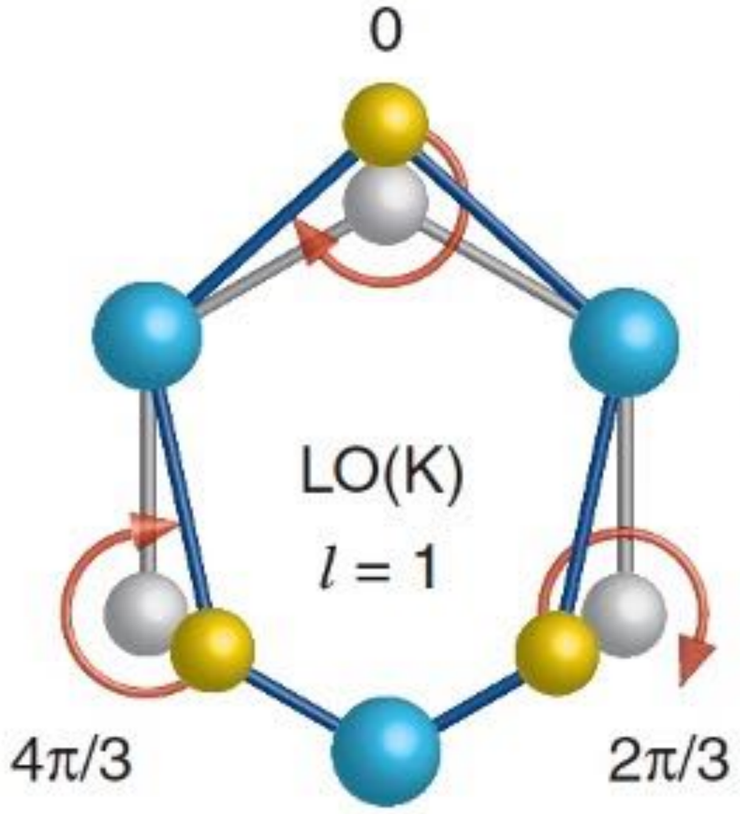


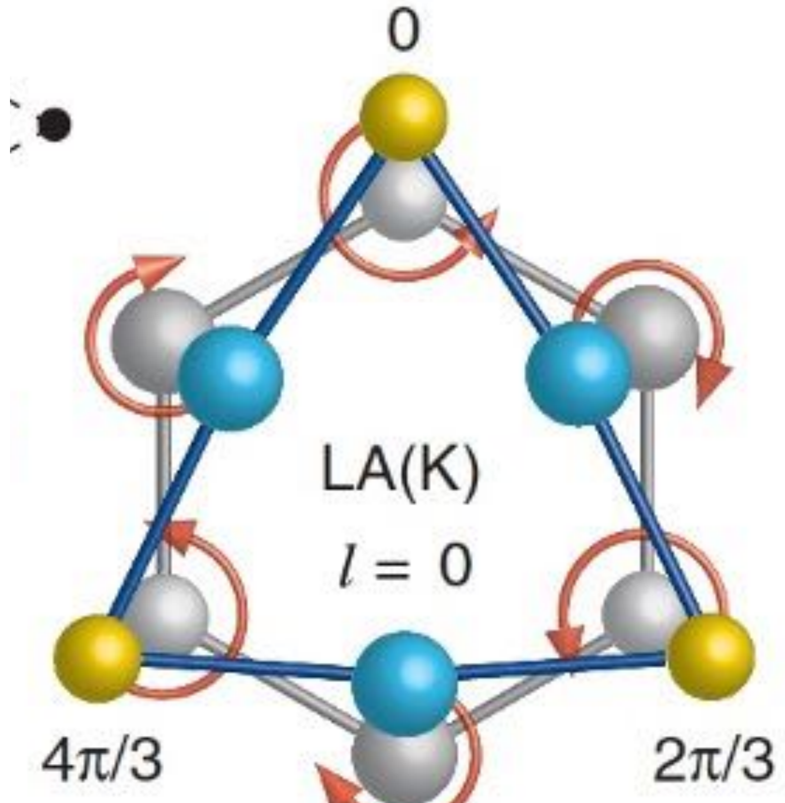
Valley polarized



Phonon angular momentum

$$\mathbf{L}_{\mathbf{q}\nu} = i\hbar \sum_\kappa \mathbf{e}^{\kappa}_{\mathbf{q}\nu} \times [\mathbf{e}^{\kappa}_{\mathbf{q}\nu}]^*$$



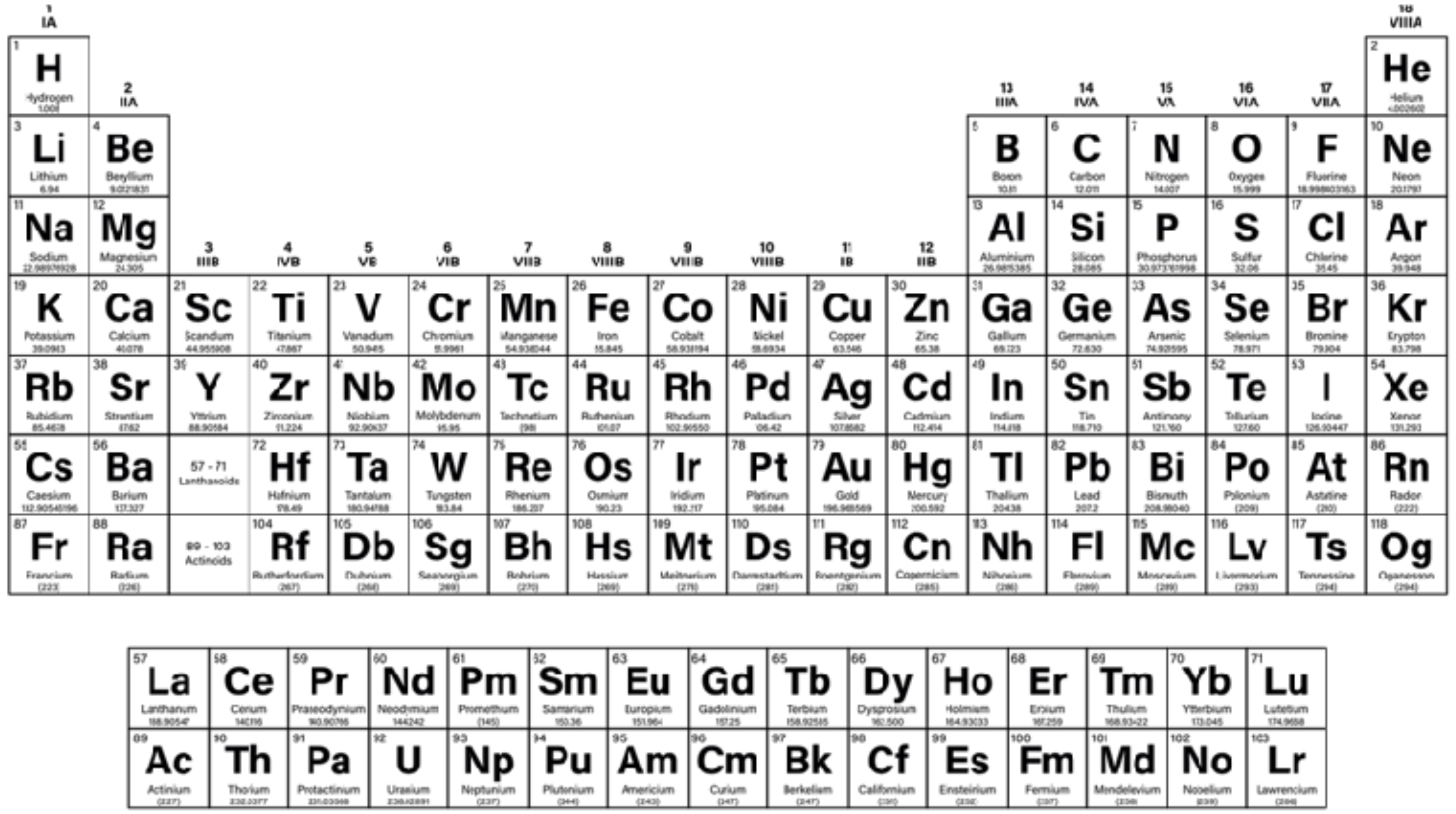


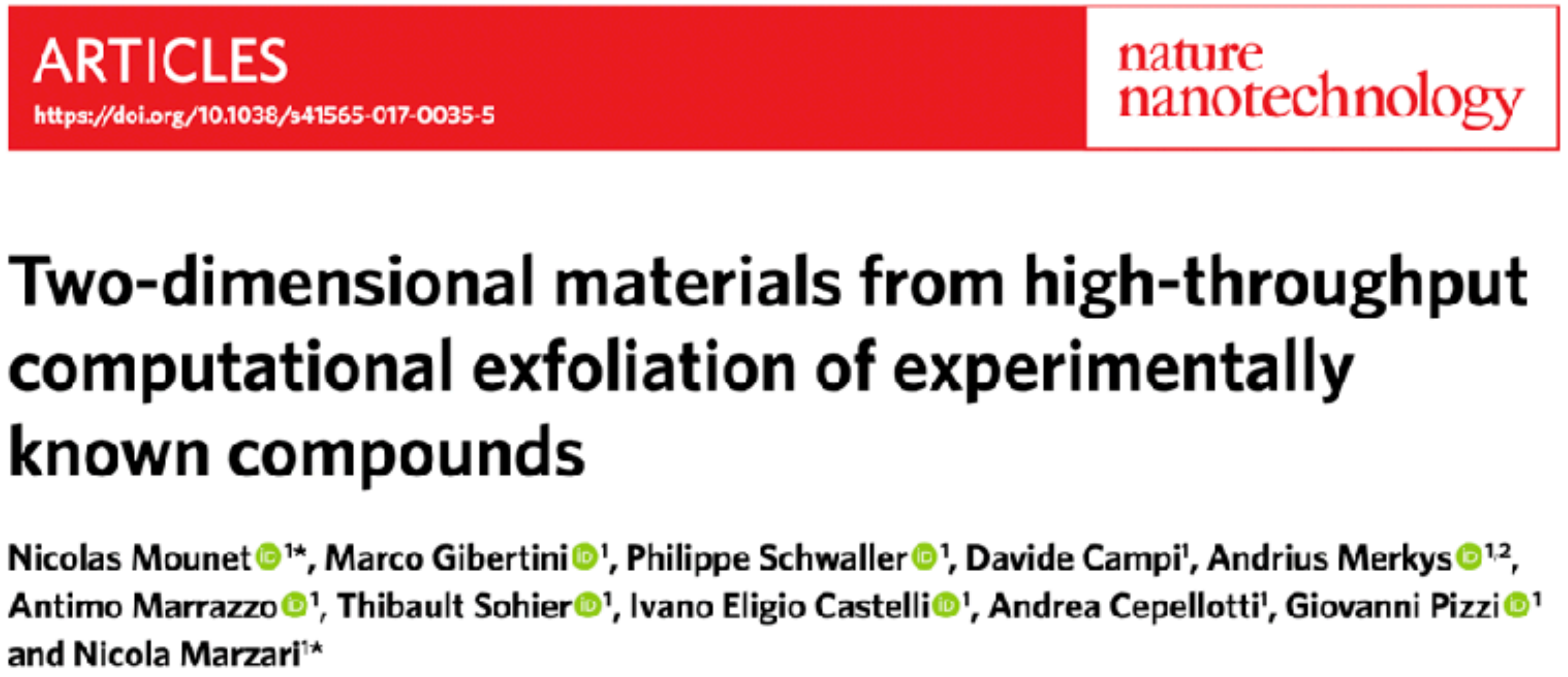
Part 4

Computational screening of
novel 2D materials valleytronics

(Outlook)

Computational screening of novel 2D materials for valleytronics




ARTICLES
https://doi.org/10.1038/s41565-017-0035-5
nature nanotechnology

Two-dimensional materials from high-throughput computational exfoliation of experimentally known compounds

Nicolas Mounet[1*], Marco Gibertini[1], Philippe Schwaller[1], Davide Campi[1], Andrius Merkys[1,2], Antimo Marrazzo[1], Thibault Sohier[1], Ivano Eligio Castelli[1], Andrea Cepellotti[1], Giovanni Pizzi[1] and Nicola Marzari[1*]

[1] Mounet et al., Nature Nanotec. **13**, 246 (2018)

Ingredients for (accessible) multi-valley semiconductors

- three-fold rotation symmetry
- lack of an inversion center
- non-metallic

How many non-centrosymmetric 2D honeycomb lattices are thermodynamically stable?

High-throughput search:

~200.000 — Entries in the ICSD database:

↓ [1]

1.825 — 2D materials which can be easily exfoliated

↓

258 — have less than 7 atoms per unit cell

↓

10 — obey the symmetry requirements

Computational screening of novel 2D materials for valleytronics

The list of 10:

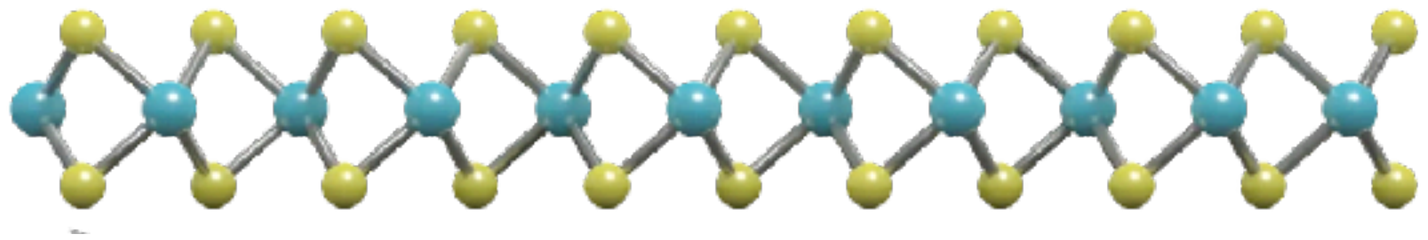
- hBN
- MoS_2
- $MoSe_2$
- WS_2
- WSe_2

Already known!

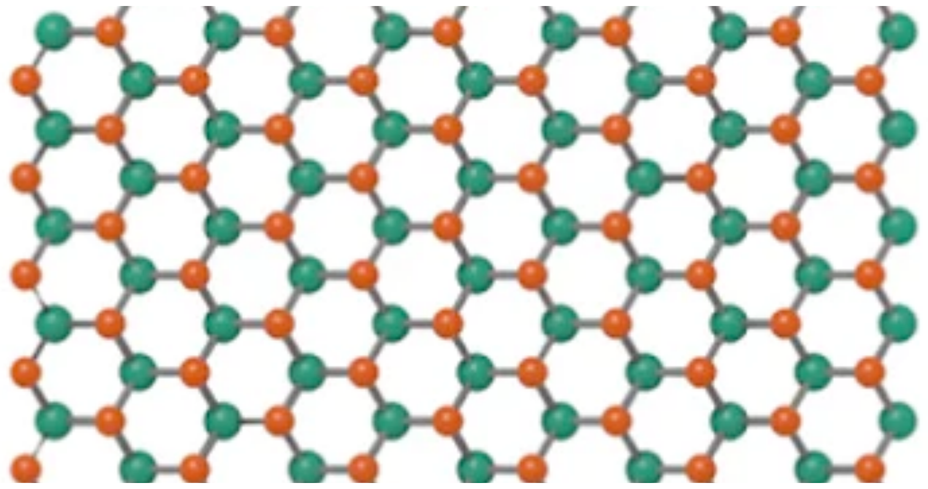
- $ZrCl_2$
- ZrClN
- ZrBrN
- GeI_2
- OTl_2

New!

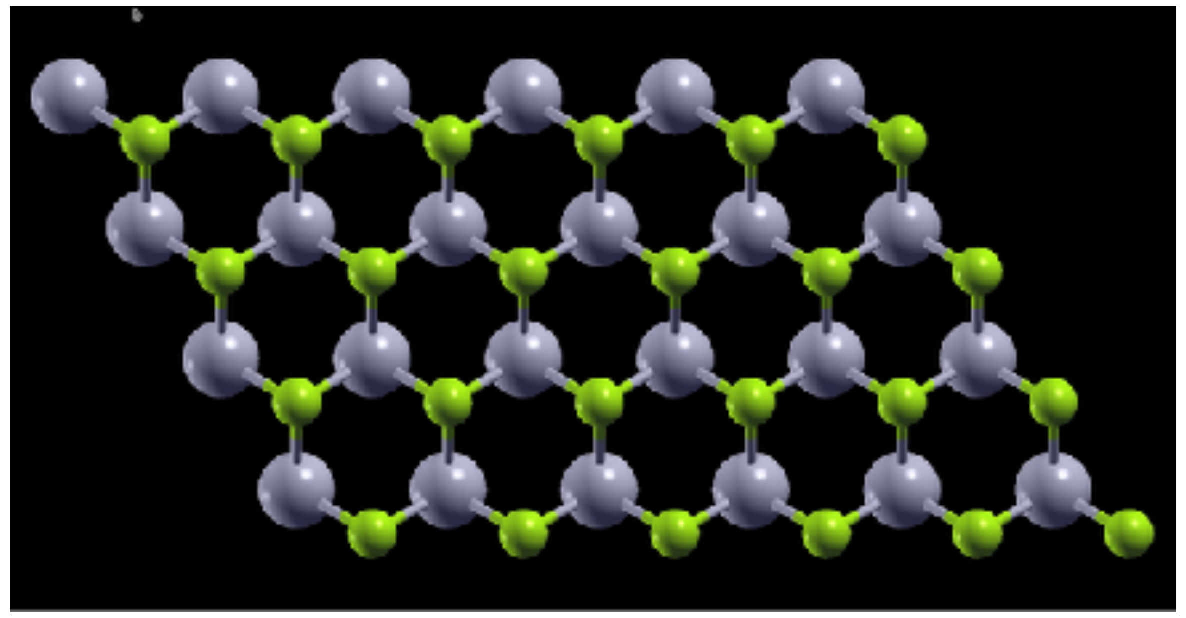
MX_2 (M=Mo,W and X=S,Se)



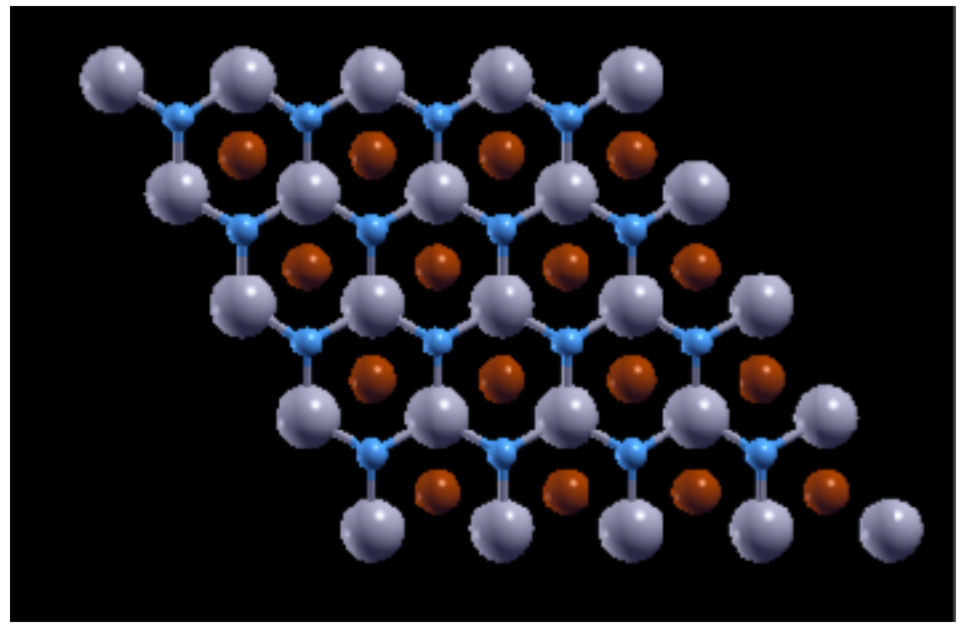
hBN monolayer



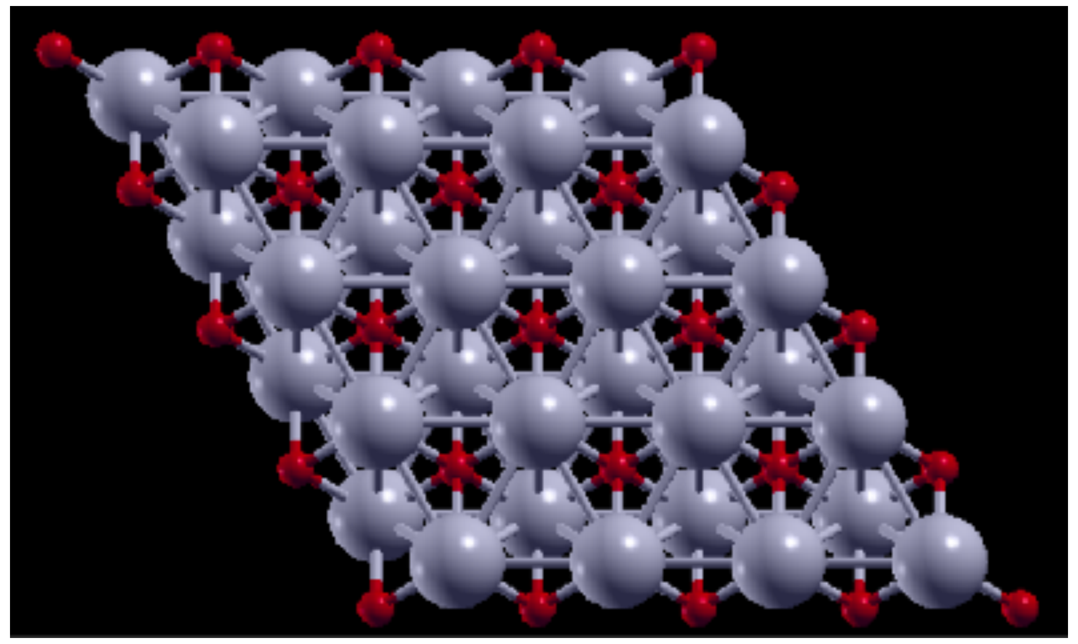
$ZrCl_2$



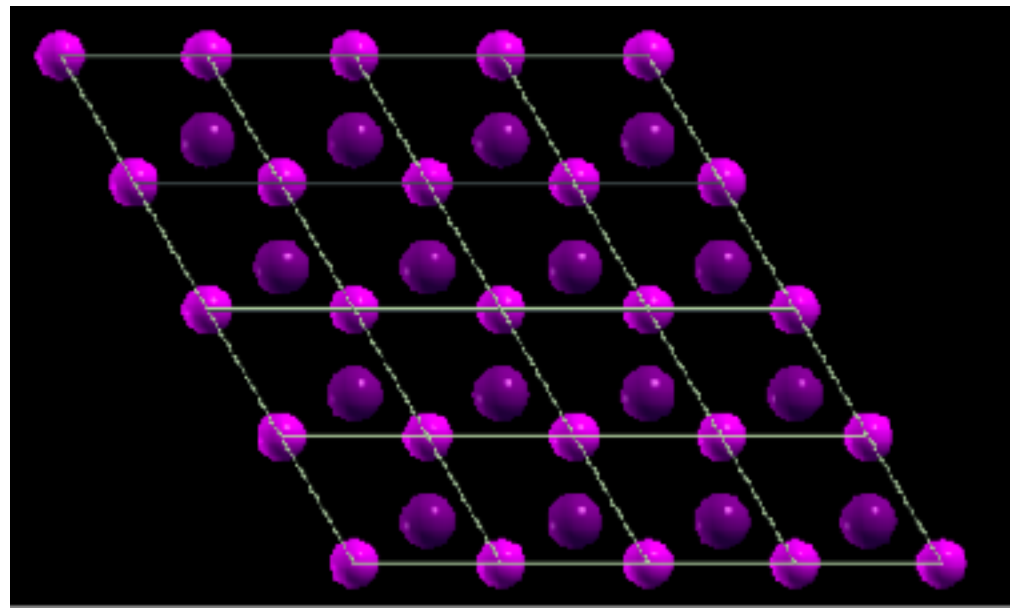
ZrNCl & ZrNBr



OTl_2



GeI_2



Optical properties of $ZrCl_2$

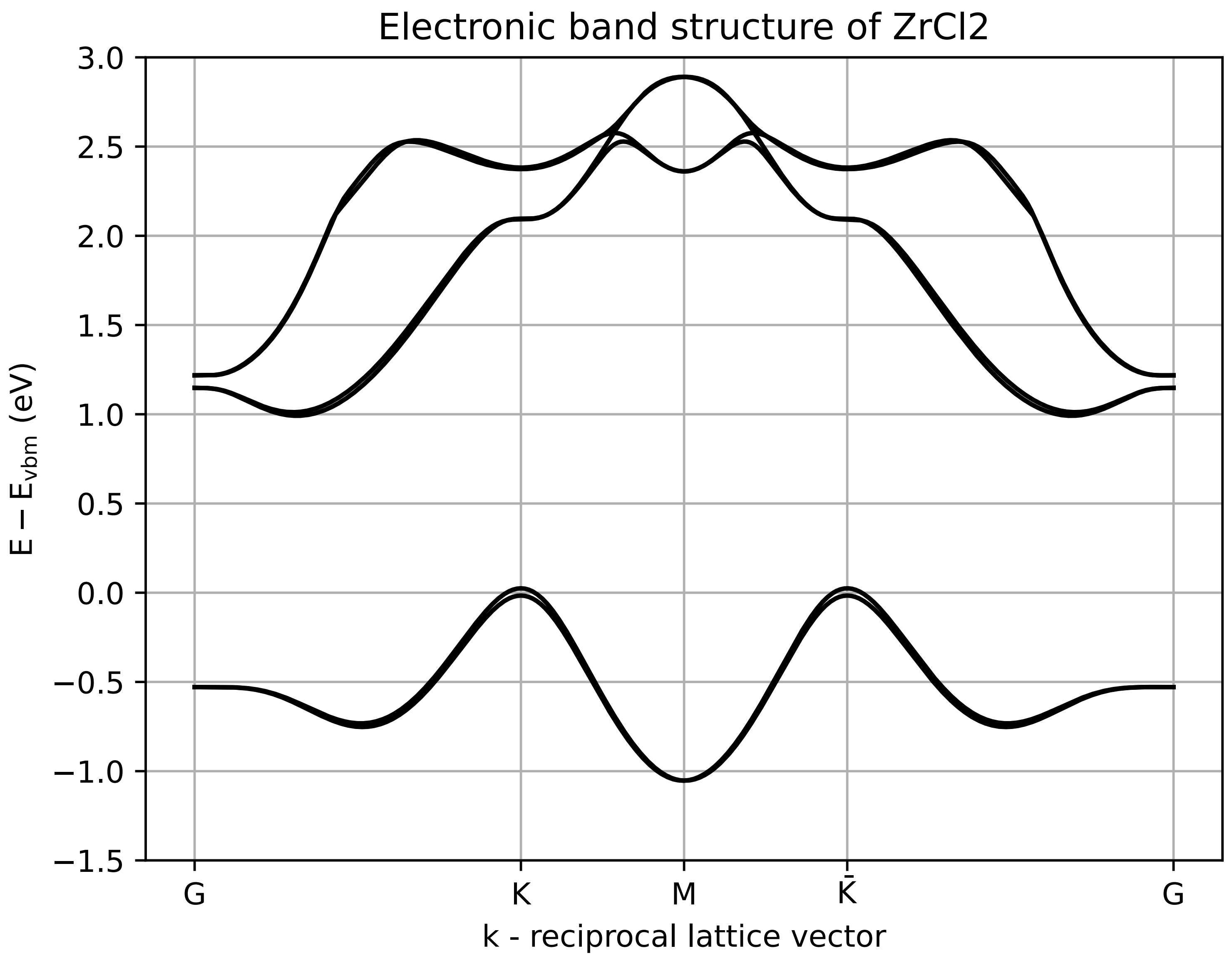
Bethe-Salpeter equation:

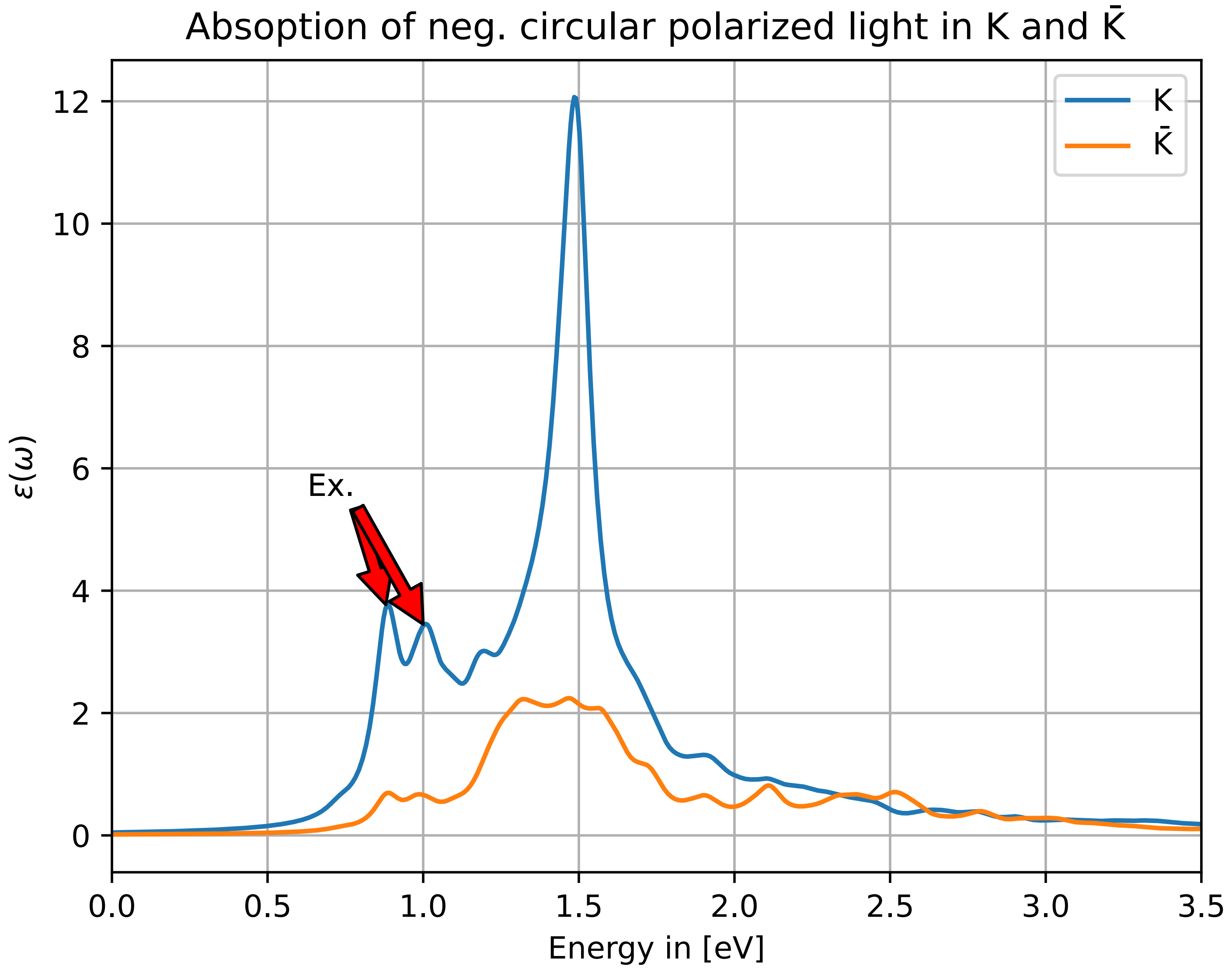
$$H_{vc\mathbf{k},v'c'\mathbf{k}'}A^{\lambda}_{v'c'\mathbf{k}'} = E^{\lambda}A^{\lambda}_{vc\mathbf{k}}$$

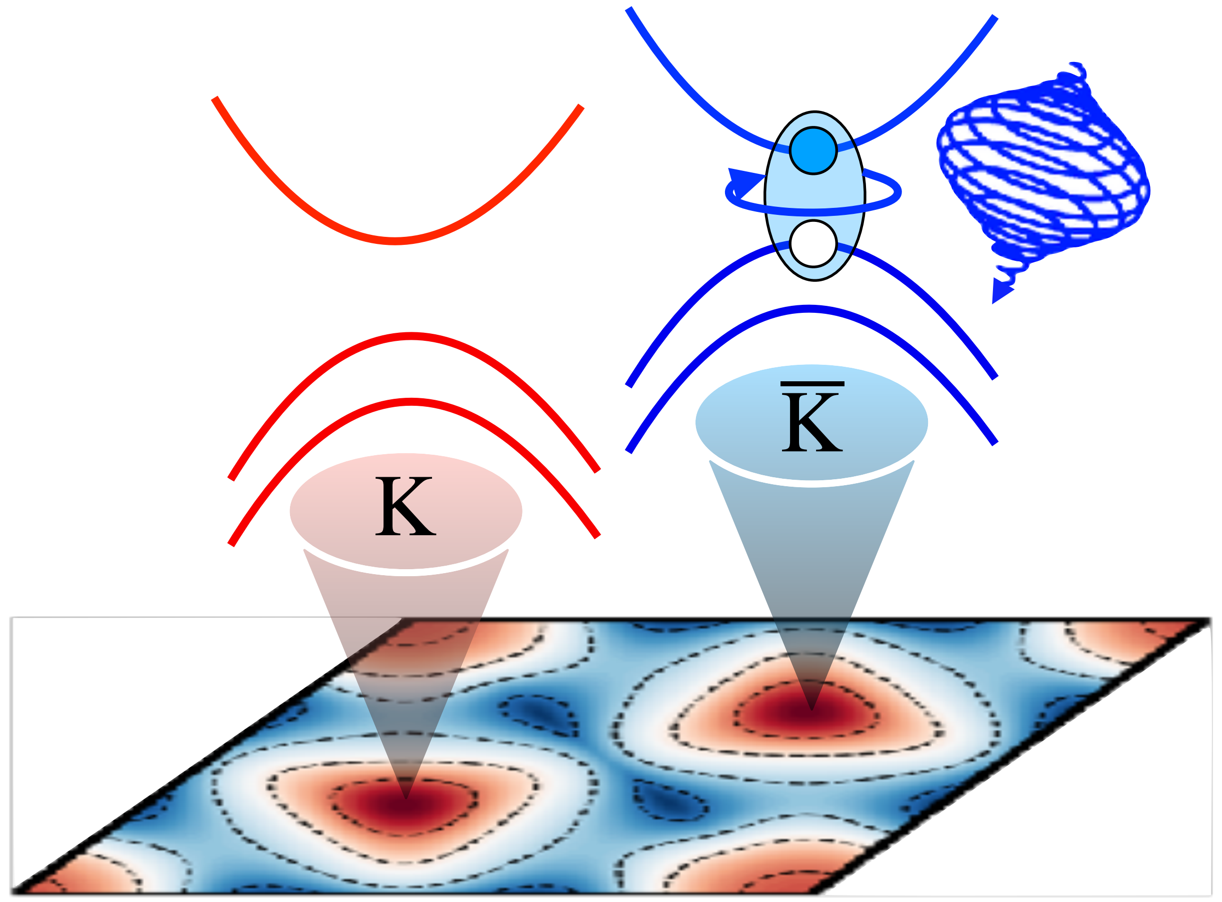
Imaginary part of the dielectric function (optical absorption):

$$\varepsilon_2(\omega) \propto \sum_{\lambda} \left| \hat{\boldsymbol{\epsilon}} \cdot \mathbf{t}^{\lambda} \right|^2 \delta(E^{\lambda} - \hbar\omega)$$

Philipp Lauwen

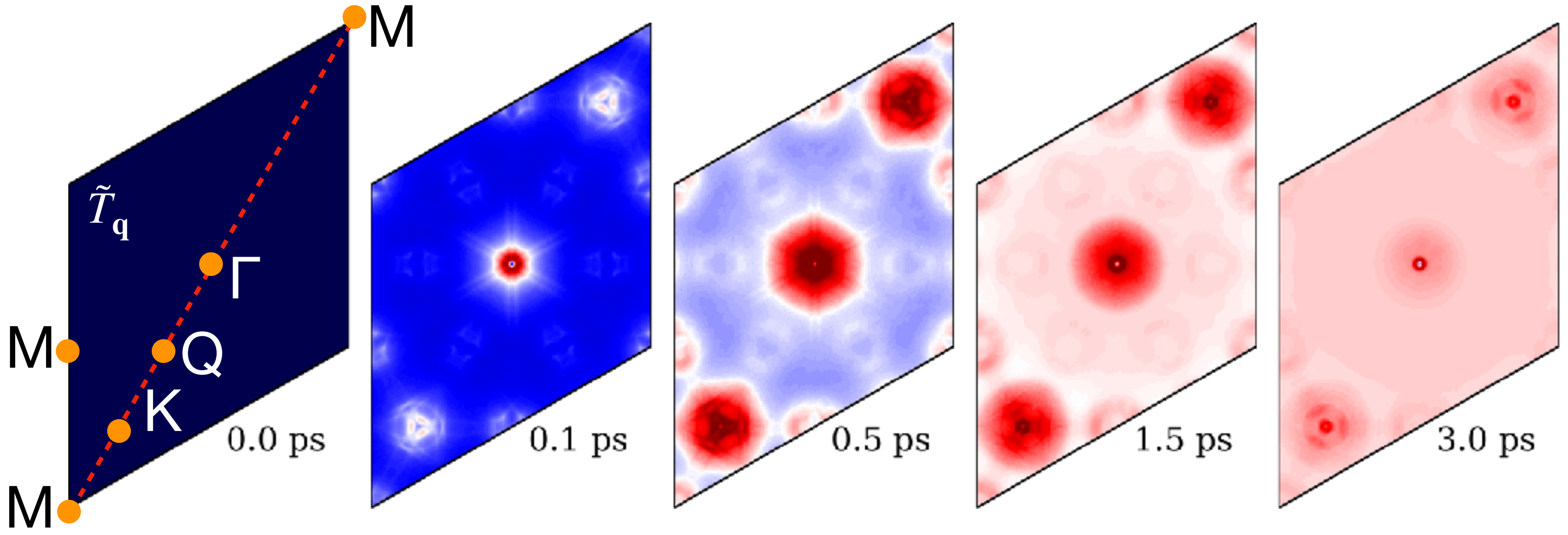






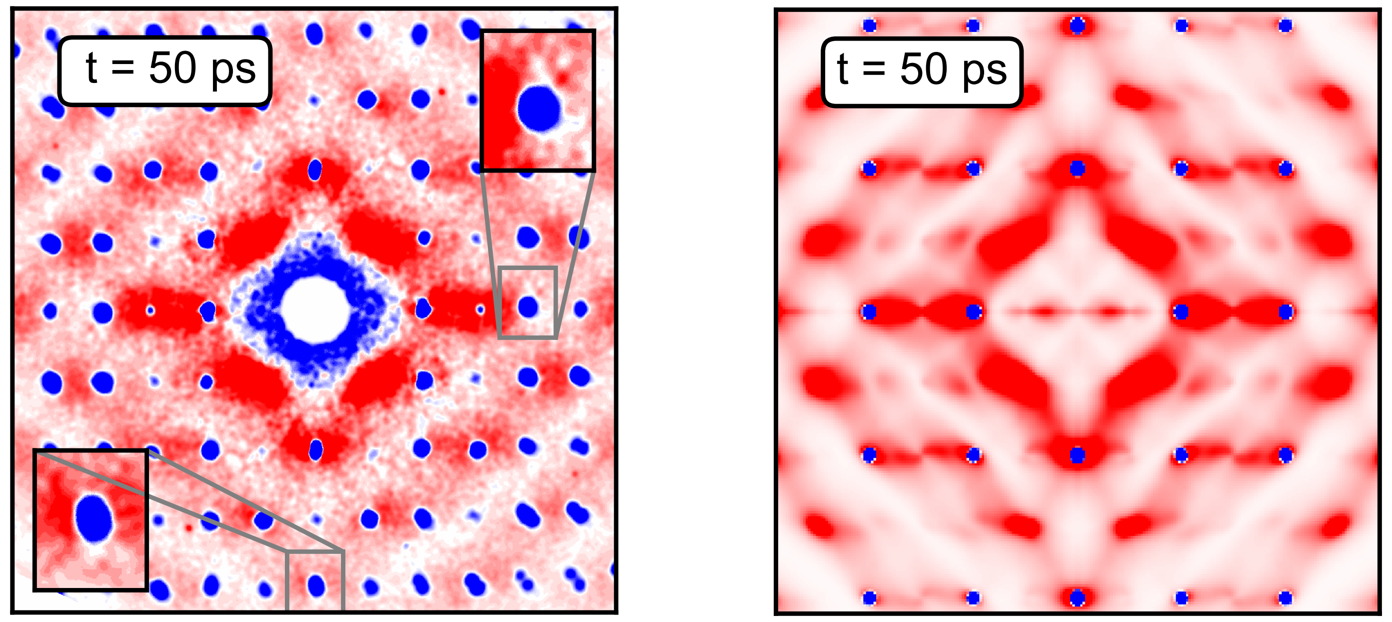
Summary

- **Coupled electron-phonon dynamics in (2D) systems out of equilibrium**



F. Caruso, D. Novko, C. Draxl, Phys. Rev. B **101,** 035128 (2020)
F. Caruso, J. Phys. Chem. Lett. **12**, 1274 (2021)
F. Caruso, D. Novko, Adv. Phys. X **7**, 2095925 (2022)

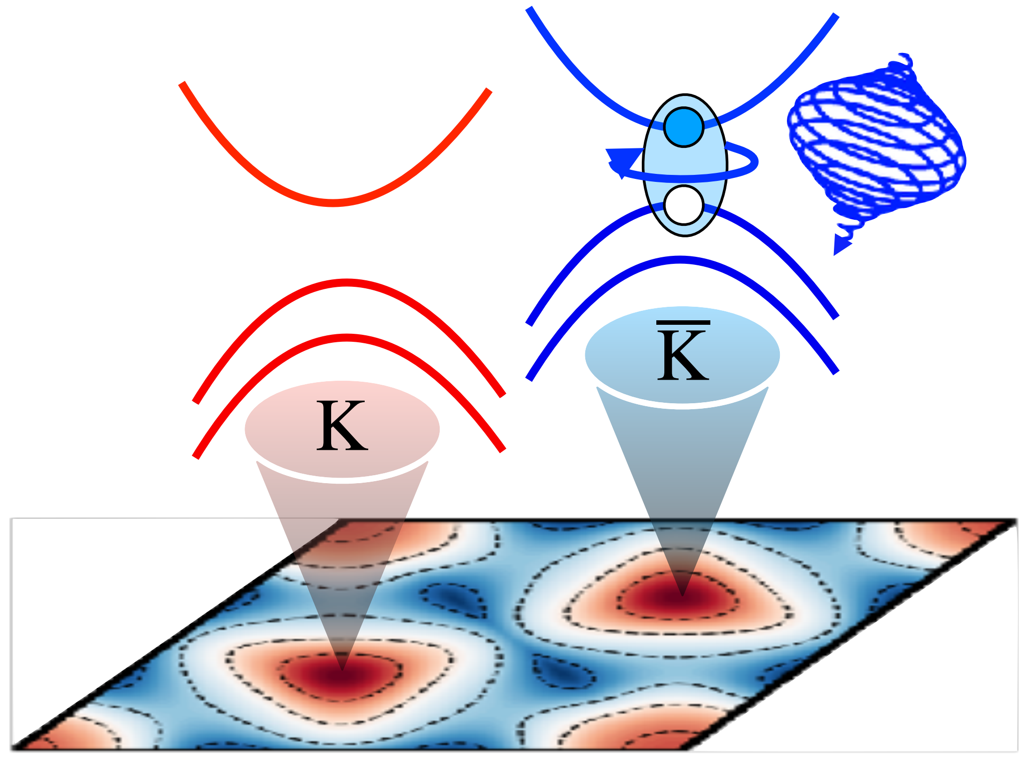
- **Fingerprints of non-equilibrium lattice dynamics in Ultrafast Diffuse Scattering**

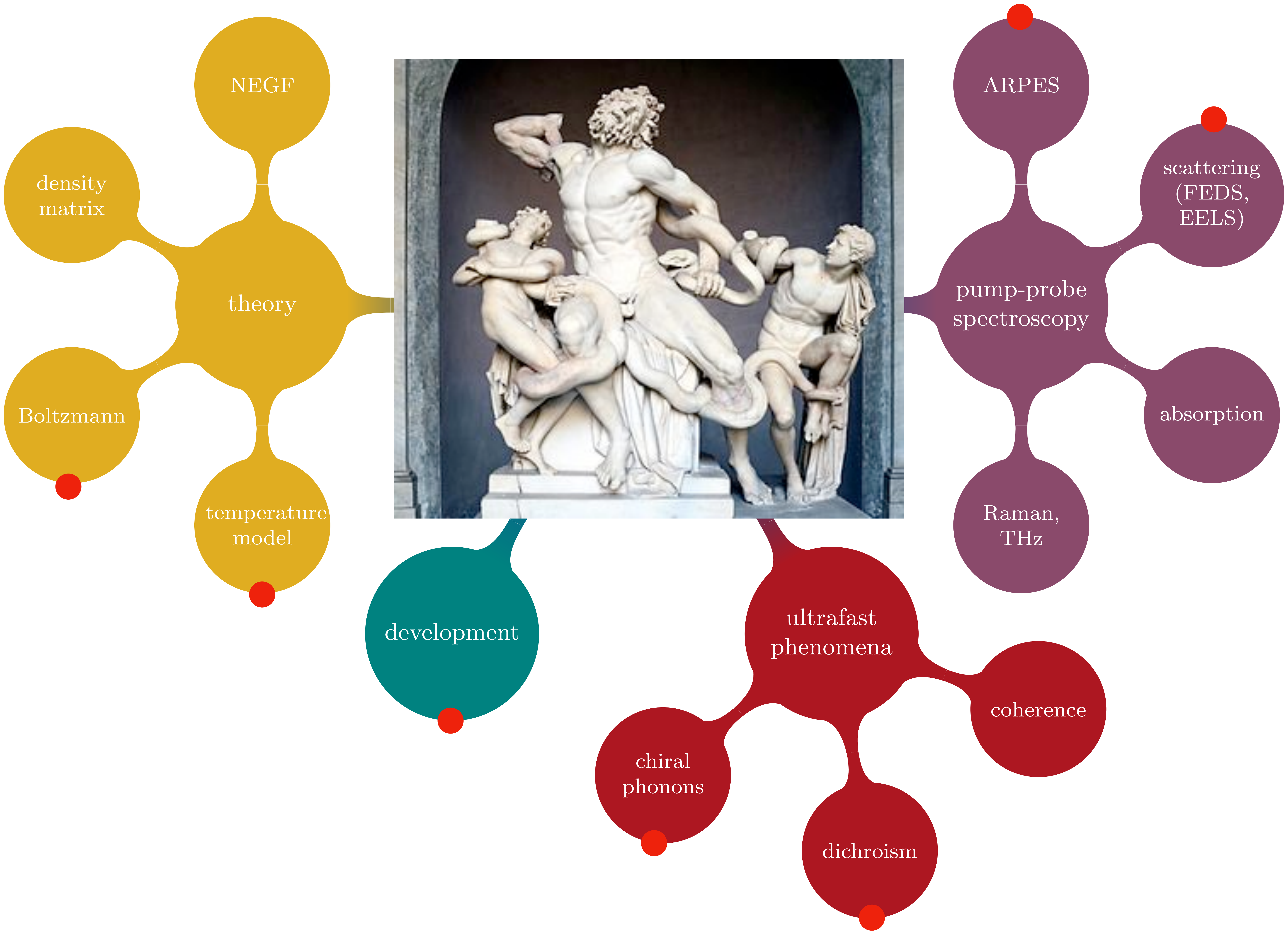


H. Seiler, (...) R. Ernstorfer, F. Caruso, Nano Lett. **21**, 6171 (2021)
T. Britt, (...), F. Caruso, XYZ, B. Siwick, Nano Lett. **22**, 4718 (2022)
M. Zacharias, H. Seiler, F. Caruso et al., Phys. Rev. Lett. **127,** 207401 (2021)
M. Zacharias, H. Seiler, F. Caruso et al., Phys. Rev. B **104,** 205109 (2021)

- **Chiral valley excitons and ultrafast dynamics of chiral phonon**

F. Caruso, M. Schebek, Y. Pan, C. Vona, C. Draxl, J. Phys. Chem Lett. **13**, 5894 (2022)

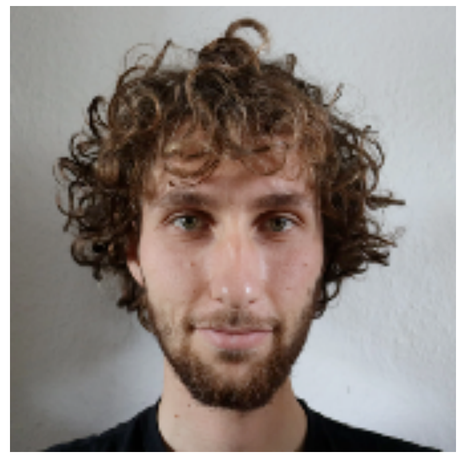



NEGF
density matrix
theory
Boltzmann
temperature model
development
ARPES
scattering (FEDS, EELS)
pump-probe spectroscopy
absorption
Raman, THz
ultrafast phenomena
coherence
chiral phonons
dichroism

Acknowledgements

The CS2T group @ Uni. Kiel:

Yiming Pan



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Mate Capin

Philipp Lauwen

Melina Seidel

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- Ralph Ernstorfer (FHI Berlin)
- Tristan Britt (McGill, Montreal)
- Bradley Siwick (McGill, Montreal)
- Sanjoy Mahatha (Desy)
- Kai Rossnagel (Uni Kiel)

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W. Li et al., Comp. Phys. Comm. 185, 1747 (2014)

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- Ignacio Gonzales-Oliva (HU Berlin)
- Maximilian Schebek (HU Berlin)
- Claudia Draxl (HU Berlin)

PRACE

DFG Deutsche Forschungsgemeinschaft

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C | S^2 | T Computational Solid-State Theory group

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